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Contents

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Editorial Remarks

This issue of the Journal is a special one. We dedicate this Journal to Katharine Gebbie, who for over twenty years, directed the National Institute of Standards and Technology's (NIST) Physical Laboratory and its successor, the Physical Measurement Laboratory (PML).

Over decades of service Gebbie accumulated an impressive list of honors recognizing her work and impact. They include two Department of Commerce (DoC) Gold Medals, a DoC Distinguished Rank Award, the NIST Equal Employment Opportunity award, a Lifetime Achievement Award from the professional society Women in Science and Engineering, the Washington Academy of Science's Physical Science Award, a special award from the American Physical Society for her leadership role in fostering excellence in Atomic, Molecular, and Optical science, and the 2002 Service to America Award from the Partnership for Public Service – the first of such recognitions given to anyone at NIST. She is also a Fellow of the Washington Academy of Sciences, the American Association of Arts and Sciences, the American Academy of Arts and Sciences, and the American Physical Society. She was Vice-President of the International Committee on Weights and Measures (CIPM) from 1993 to1999.

She was named after her aunt, Katharine Burr Blodgett, who was the first woman to earn a Ph.D. in physics from the University of Cambridge, a world-class scientist, a long-time colleague of Irving Langmuir at General Electric, and the co-discoverer of Langmuir-Blodgett thin films.

She was married to Alastair Gebbie, a pioneer in Fourier Transform Spectroscopy.

She was known for her kindness and wisdom. I came to know her late and saw that unique person who shone with excitement for science and for the people who work in it. It was a singular privilege to have known her. She strongly supported the Washington Academy of Science's awards program. We could always depend on her to submit superb candidates for awards.

In a rare move Gebbie's colleagues renamed NIST's precision measurement laboratory in Boulder, Colorado in her honor. "This renaming is our small way of saying thank you… for all [Katharine] has done for this organization over such a long period of time," said NIST Director Willie E. May.

An astrophysicist by training, Gebbie received her B.A. in Physics from Bryn Mawr College, subsequently earning a B.S. in Astronomy and Ph.D. in Physics from University College London.

For several years in the mid-1960s she trekked in Nepal, went mountaineering in Turkey, and flew around North America in her mother's airplane. Both Dr. Gebbie and her parents had taken professional flying lessons.

Please enjoy this issue in honor of Katharine Gebbie.

Sethanne Howard

Editor

A Tribute to Katharine Blodgett Gebbie

KATHARINE BLODGETT GEBBIE, a Fellow of the Washington Academy of Sciences, was born on 4 July 1932 and died on 17 August 2016. Katharine

spent most of her professional career at the National Institute of Standards and Technology (originally the National Bureau of Standards). Trained as an astrophysicist, she began her association with NBS/NIST in 1966 as a postdoc at JILA, then known as the Joint Institute for Laboratory Astrophysics, a cooperative operation of NIST and the University of Colorado at Boulder. After a distinguished career in research, Katharine was persuaded to turn her talents to scientific management. A series of increasingly responsible positions led her to become the founding Director of NIST's Physics Laboratory in 1991. She remained the director of that Laboratory for all of its 20 years and was also the founding director of its even larger successor, the NIST Physical Measurement Laboratory. Many of us believe her laboratory to be the best place in the entire world in which to do research in Physics, predominantly because of the atmosphere that Katharine created. Her creed was to hire the best people, give them the resources to do their work, and let them do it. While many other managers might have said similar things, she actually did it, and the results were astounding. Within a span of 15 years, four of her scientists received Nobel Prizes in Physics. Two of her researchers received the prestigious MacArthur awards, and many other accolades were bestowed on those under her leadership. She was a true servant, and gloried in the accomplishments of those she nurtured.

This issue of the Journal of the Washington Academy of Sciences pays tribute to Katharine Blodgett Gebbie by reprinting the "Nobel Lectures" of Katharine's four Laureates, William Phillips (1997), Eric Cornell (2001), John Hall (2005), and David Wineland (2012). These are the articles prepared by the Laureates for publication in *Reviews of Modern Physics* a few months after the award of the Nobel Prize, and are not transcripts of the

Nobel Lectures delivered in Stockholm during the events association with the 10 December prize award ceremony. The article by Cornell is co-authored with his University of Colorado colleague Carl Wieman, with whom he worked closely at JILA, and who also benefitted from Katharine Gebbie's leadership.

William D. Phillips
Gaithersburg
January 2017

Journal of the Washington Academy of Sciences

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Board of Discipline Editors

The *Journal of the Washington Academy of Sciences* has an 11-member Board of Discipline Editors representing many scientific and technical fields. The members of the Board of Discipline Editors are affiliated with a variety of scientific institutions in the Washington area and beyond — government agencies such as the National Institute of Standards and Technology (NIST); universities such as Georgetown; and professional associations such as the Institute of Electrical and Electronics Engineers (IEEE).

Laser cooling and trapping of neutral atoms[1]

William D. Phillips

National Institute of Standards and Technology,

Introduction

IN 1978, WHILE I WAS A POSTDOCTORAL fellow at MIT, I read a paper by Art Ashkin (1978) in which he described how one might slow down an atomic beam of sodium using the radiation pressure of a laser beam tuned to an atomic resonance. After being slowed, the atoms would be captured in a trap consisting of focused laser beams, with the atomic motion being damped until the temperature of the atoms reached the microkelvin range. That paper was my first introduction to laser cooling, although the idea of laser cooling (the reduction of random thermal velocities using radiative forces) had been proposed three years earlier in independent papers by Hänsch and Schawlow (1975) and Wineland and Dehmelt (1975). Although the treatment in Ashkin's paper was necessarily over-simplified, it provided one of the important inspirations for what I tried to accomplish for about the next decade. Another inspiration appeared later that same year: Wineland, Drullinger and Walls (1978) published the first laser cooling experiment, in which they cooled a cloud of Mg^+ ions held in a Penning trap. At essentially the same time, Neuhauser, Hohenstatt, Toschek and Dehmelt (1978) also reported laser cooling of trapped Ba^+ ions.

Those laser cooling experiments of 1978 were a dramatic demonstration of the mechanical effects of light, but such effects have a much longer history. The understanding that electromagnetic radiation exerts a force became quantitative only with Maxwell's theory of electromagnetism, even though such a force had been conjectured much earlier, partly in response to the observation that comet tails point away from the sun. It was not until the turn of the century, however, that experiments by Lebedev (1901) and Nichols and Hull (1901, 1903) gave

[1] The 1997 Nobel Prize in Physics was shared by Steven Chu, Claude N. Cohen-Tannoudji, and William D. Phillips. This text is based on Dr. Phillips's address on the occasion of the award. Reprinted from *Reviews of Modern Physics*, Vol. 70, No. 3, July 1998.

a laboratory demonstration and quantitative measurement of radiation pressure on macroscopic objects. In 1933 Frisch made the first demonstration of light pressure on atoms, deflecting an atomic sodium beam with resonance radiation from a lamp. With the advent of the laser, Ashkin (1970) recognized the potential of intense, narrow-band light for manipulating atoms and in 1972 the first "modern" experiments demonstrated the deflection of atomic beams with lasers (Picqué and Vialle, 1972; Schieder *et al.,* 1972). All of this set the stage for the laser cooling proposals of 1975 and for the demonstrations in 1978 with ions.

Comet tails, deflection of atomic beams and the laser cooling proposed in 1975 are all manifestations of the radiative force that Ashkin has called the "scattering force," because it results when light strikes an object and is scattered in random directions. Another radiative force, the dipole force, can be thought of as arising from the interaction between an induced dipole moment and the gradient of the incident light field. The dipole force was recognized at least as early as 1962 by Askar'yan, and in 1968, Letokhov proposed using it to trap atoms — even before the idea of laser cooling! The trap proposed by Ashkin in 1978 relied on this "dipole" or "gradient" force as well. Nevertheless, in 1978, laser cooling, the reduction of random velocities, was understood to involve only the scattering force. Laser trapping, confinement in a potential created by light, which was still only a dream, involved both dipole and scattering forces. Within 10 years, however, the dipole force was seen to have a major impact on laser cooling as well.

Without understanding very much about what difficulties lay in store for me, or even appreciating the exciting possibilities of what one might do with laser cooled atoms, I decided to try to do for neutral atoms what the groups in Boulder and Heidelberg had done for ions: trap them and cool them. There was, however, a significant difficulty: we could not first trap and then cool neutral atoms. Ion traps were deep enough to easily trap ions having temperatures well above room temperature, but none of the proposed neutral atom traps had depths of more than a few kelvin. Significant cooling was required before trapping would be possible, as Ashkin had outlined in his paper (1978), and it was with this idea that I began.

Before describing the first experiments on the deceleration of atomic beams, let me digress slightly and discuss why laser cooling is so exciting and why it has attracted so much attention in the scientific community: When one studies atoms in a gas, they are typically moving very rapidly. The molecules and atoms in air at room temperature are moving with speeds on the order of 300 m/s, the speed of sound. This thermal velocity can be reduced by refrigerating the gas, with the velocity varying as the square root of the temperature, but even at 77 K, the temperature at which N_2 condenses into a liquid, the nitrogen molecules are moving at about 150 m/s. At 4 K, the condensation temperature of helium, the He atoms have 90 m/s speeds. At temperatures for which atomic thermal velocities would be below 1 m/s, any gas in equilibrium (other than spin-polarized atomic hydrogen) would be condensed, with a vapor pressure so low that essentially no atoms would be in the gas phase. As a result, all studies of free atoms were done with fast atoms. The high speed of the atoms makes measurements difficult. The Doppler shift and the relativistic time dilation cause displacement and broadening of the spectral lines of thermal atoms, which have a wide spread of velocities. Furthermore, the high atomic velocities limit the observation time (and thus the spectral resolution) in any reasonably-sized apparatus. Atoms at 300 m/s pass through a meter-long apparatus in just 3 ms. These effects are a major limitation, for example, to the performance of conventional atomic clocks.

The desire to reduce motional effects in spectroscopy and atomic clocks was and remains a major motivation for the cooling of both neutral atoms and ions. In addition, some remarkable new phenomena appear when atoms are sufficiently cold. The wave, or quantum nature of particles with momentum p becomes apparent only when the de Broglie wavelength, given by $\lambda_{dB} = h/p$, becomes large, on the order of relevant distance scales like the atom-atom interaction distances, atom-atom separations, or the scale of confinement. Laser cooled atoms have allowed studies of collisions and of quantum collective behavior in regimes hitherto unattainable. Among the new phenomena seen with neutral atoms is Bose-Einstein condensation of an atomic gas (Anderson *et al*., 1995; Davis, Mewes, Andrews, *et al*., 1995), which has been hailed as a new state of matter, and is already becoming a major new

field of investigation. Equally impressive and exciting are the quantum phenomena seen with trapped ions, for example, quantum jumps (Bergquist *et al.,* 1986; Nagourney *et al*., 1986; Sauter *et al*., 1986), Schrödinger cats (Monroe *et al*., 1996), and quantum logic gates (Monroe *et al*., 1995).

Laser Cooling of Atomic Beams

In 1978 I had only vague notions about the excitement that lay ahead with laser cooled atoms, but I concluded that slowing down an atomic beam was the first step. The atomic beam was to be slowed using the transfer of momentum that occurs when an atom absorbs a photon. Figure 1 shows the basic process underlying the "scattering force" that results. An atomic beam with velocity v is irradiated by an opposing laser beam. For each photon that a ground-state atom absorbs, it is slowed by $v_{rec} = \hbar k/m$. In order to absorb again the atom must return to the ground state by emitting a photon. Photons are emitted in random directions, but with a symmetric average distribution, so their contribution to the atom's momentum averages to zero. The randomness results in a "heating" of the atom, discussed below.

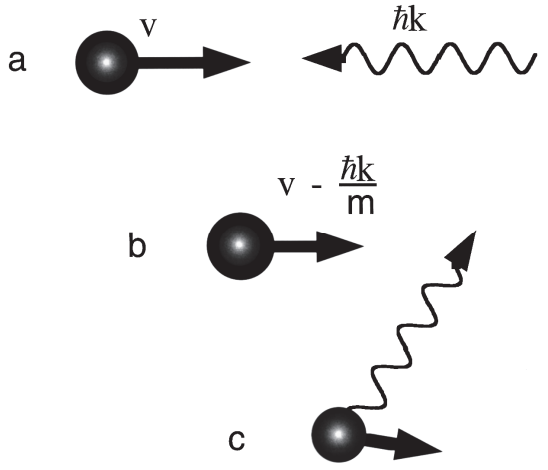

FIG. 1. (a) An atom with velocity v encounters a photon with momentum $\hbar k = h/\lambda$; (b) after absorbing the photon, the atom is slowed by $\hbar k/m$; (c) after re-radiation in a random direction, on average the atom is slower than in (a).

For sodium atoms interacting with the familiar yellow resonance light, $v_{rec} = 3$ cm/s, while a typical beam velocity is about 10^5 cm/s, so the absorption-emission process must occur about 3×10^4 times to bring the Na atom to rest. In principle, an atom could radiate and absorb photons at half the radiative decay rate of the excited state (a 2-level atom in steady state can spend at most half of its time in the excited state). For Na, this implies that a photon could be radiated every 32 ns on average, bringing the atoms to rest in about 1 ms. Two problems, optical pumping and Doppler shifts, can prevent this from happening. I had an early indication of the difficulty of decelerating an atomic beam shortly after reading Ashkin's 1978 paper. I was then working with a sodium atomic beam at MIT, using tunable dye lasers to study the scattering properties of optically excited sodium. I tuned a laser to be resonant with the Na transition from $3S_{1/2} \rightarrow 3P_{3/2}$, the $D2$ line, and directed its beam opposite to the atomic beam. I saw that the atoms near the beam source were fluorescing brightly as they absorbed the laser light, while further away from the source, the atoms were relatively dim. The problem, I concluded, was optical pumping, illustrated in Fig. 2.

Sodium is not a two-level atom, but has two ground hyperfine levels (F=1 and F=2 in Fig. 2), each of which consists of several, normally degenerate, states. Laser excitation out of one of the hyperfine levels to the excited state can result in the atom radiating to the other hyperfine level. This optical pumping essentially shuts off the absorption of laser light, because the linewidths of the transition and of the laser are much smaller than the separation between the ground state hyperfine components. Even for atoms excited on the $3S_{1/2}\ (F=2) \rightarrow 3P_{3/2}\ (F'=3)$ transition, where the only allowed decay channel is to $F=2$, off-resonant excitation of $F'=2$ (the linewidth of the transition is 10 MHz, while the separation between $F'=2$ and $F'=3$ is 60 MHz) leads to optical pumping into $F=1$ after only about a hundred absorptions. This optical pumping made the atoms "dark" to my laser after they traveled only a short distance from the source.

An obvious solution [Fig. 2(b)] is to use a second laser frequency, called a repumper, to excite the atoms out of the "wrong"

($F = 1$) hyperfine state so that they can decay to the "right" state ($F = 2$) where they can continue to cool. Given the repumper, another problem becomes apparent: the Doppler shift. In order for the laser light to be resonantly absorbed by a counter-propagating atom moving with velocity v, the frequency ω of the light must be kv lower than the resonant frequency for an atom at rest. As the atom repeatedly absorbs photons, slowing down as desired, the Doppler shift changes and the atom goes out of resonance with the light. The natural linewidth $\Gamma/2\pi$ of the optical transition in Na is 10 MHz (full width at half maximum). A change in velocity of 6 m/s gives a Doppler shift this large, so after absorbing only 200 photons, the atom is far enough off resonance that the rate of absorption is significantly reduced. The result is that only atoms with the "proper" velocity to be resonant with the laser are slowed, and they are only slowed by a small amount.

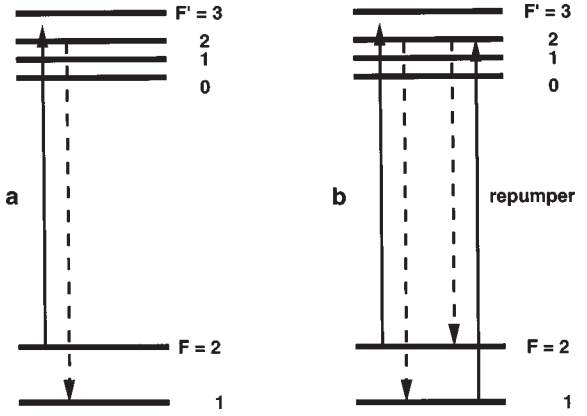

FIG. 2. (a) The optical pumping process preventing cycling transitions in alkalis like Na; (b) use of a repumping laser to allow many absorption-emission cycles.

Nevertheless, this process of atoms being slowed and pushed out of resonance results in a cooling or narrowing of the velocity distribution. In an atomic beam, there is typically a widespread of velocities around $v_{\mathrm{th}} = 3k_{\mathrm{B}}T/m$. Those atoms with the proper velocity will absorb rapidly and decelerate. Those that are too fast will absorb more slowly, then more rapidly as they come into resonance, and finally more slowly as they continue to decelerate. Atoms that are too slow to begin with will absorb little and

decelerate little. Thus atoms from a range of velocities around the resonant velocity are pushed into a narrower range centered on a lower velocity. This process was studied theoretically by Minogin (1980) and in 1981, at Moscow's Institute for Spectroscopy, was used in the first experiment clearly demonstrating laser cooling of neutral atoms (Andreev *et al.*, 1981).

Figure 3 shows the velocity distribution after such cooling of an atomic beam. The data was taken in our laboratory, but is equivalent to what had been done in Moscow. The characteristic of this kind of beam cooling is that only a small part of the total velocity distribution (the part near resonance with the laser beam) is slowed by only a small amount (until the atoms are no longer resonant). The narrow peak, while it represents true cooling in that its velocity distribution is narrow, consists of rather fast atoms.

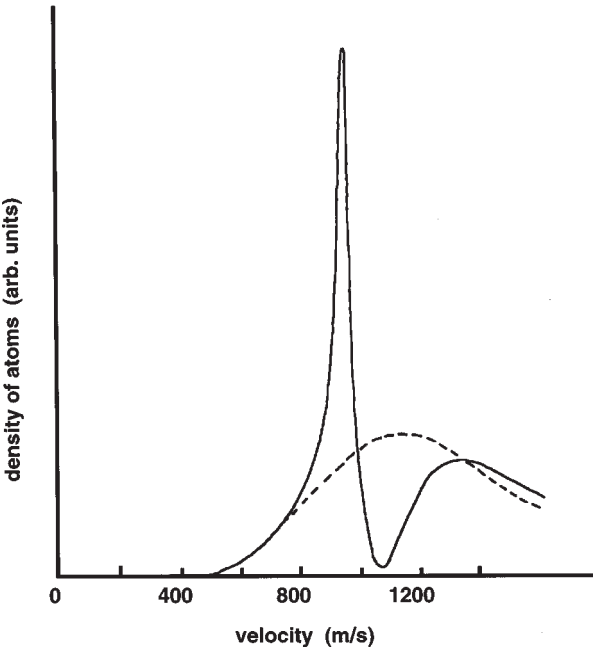

FIG. 3. Cooling an atomic beam with a fixed frequency laser. The dotted curve is the velocity distribution before cooling, and the solid curve is after cooling. Atoms from a narrow velocity range are transferred to a slightly narrower range centered on a lower velocity.

One solution to this problem had already been outlined in 1976 by Letokhov, Minogin, and Pavlik. They suggested a general method of changing the frequency (chirping) of the cooling laser so as to interact with all the atoms in a wide distribution and to stay in resonance with the atoms as they are cooled. The Moscow group applied the technique to decelerating an atomic beam (Balykin *et al.*, 1979) but without clear success (Balykin, 1980). [Later, in 1983, John Prodan and I obtained the first clear deceleration and cooling of an atomic beam with this "chirp-cooling" technique (Phillips and Prodan, 1983, 1984; Phillips, Prodan, and Metcalf, 1983a; Prodan and Phillips, 1984). Those first attempts failed to bring the atoms to rest, something that was finally achieved by Ertmer, Blatt, Hall and Zhu (1985).] The chirp-cooling technique is now one of the two standard methods for decelerating beams. The other is "Zeeman cooling."

By late 1978, I had moved to the National Bureau of Standards (NBS), later named the National Institute of Standards and Technology (NIST), in Gaithersburg. I was considering how to slow an atomic beam, realizing that the optical pumping and Doppler shift problems would both need to be addressed. I understood how things would work using the Moscow chirp-cooling technique and a repumper. I also considered using a broadband laser, so that there would be light in resonance with the atoms, regardless of their velocity. [This idea was refined by Hoffnagle (1988) and demonstrated by Hall's group (Zhu, Oates, and Hall, 1991).] Finally I considered that instead of changing the frequency of the laser to stay in resonance with the atoms (chirping), one could use a magnetic field to change the energy level separation in the atoms so as to keep them in resonance with the fixed-frequency laser (Zeeman cooling). All of these ideas for cooling an atomic beam, along with various schemes for avoiding optical pumping, were contained in a proposal (Phillips, 1979) that I submitted to the Office of Naval Research in 1979. Around this time Hal Metcalf, from the State University of New York at Stony Brook, joined me in Gaithersburg and we began to consider what would be the best way to proceed. Hal contended that all the methods looked reasonable, but we should work on the Zeeman cooler because it would be the

most fun! Not only was Hal right about the fun we would have, but his suggestion led us to develop a technique with particularly advantageous properties. The idea is illustrated in Fig. 4.

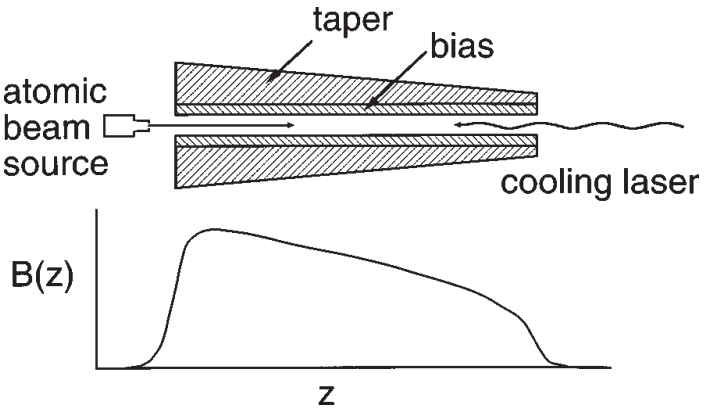

FIG. 4. Upper: Schematic representation of a Zeeman slower. Lower: Variation of the axial field with position.

The atomic beam source directs atoms, which have a wide range of velocities, along the axis (z direction) of a tapered solenoid. This magnet has more windings at its entrance end, near the source, so the field is higher at that end. The laser is tuned so that, given the field-induced Zeeman shift and the velocity-induced Doppler shift of the atomic transition frequency, atoms with velocity v_0 are resonant with the laser when they reach the point where the field is maximum. Those atoms then absorb light and begin to slow down. As their velocity changes, their Doppler shift changes, but is compensated by the change in Zeeman shift as the atoms move to a point where the field is weaker. At this point, atoms with initial velocities slightly lower than v_0 come into resonance and begin to slow down. The process continues with the initially fast atoms decelerating and staying in resonance while initially slower atoms come into resonance and begin to be slowed as they move further down the solenoid. Eventually all the atoms with velocities lower than v_0 are brought to a final velocity that depends on the details of the magnetic field and laser tuning.

The first tapered solenoids that Hal Metcalf and I used for Zeeman cooling of atomic beams had only a few sections of

windings and had to be cooled with air blown by fans or with wet towels wrapped around the coils. Shortly after our initial success in getting some substantial deceleration, we were joined by my first postdoc, John Prodan. We developed more sophisticated solenoids, wound with wires in many layers of different lengths, so as to produce a smoothly varying field that would allow the atoms to slow down to a stop while remaining in resonance with the cooling laser.

These later solenoids were cooled with water flowing over the coils. To improve the heat transfer, we filled the spaces between the wires with various heat-conducting substances. One was a white silicone grease that we put onto the wires with our hands as we wound the coil on a lathe. The grease was about the same color and consistency as the diaper rash ointment I was then using on my baby daughters, so there was a period of time when, whether at home or at work, I seemed to be up to my elbows in white grease.

The grease-covered, water-cooled solenoids had the annoying habit of burning out as electrolytic action attacked the wires during operation. Sometimes it seemed that we no sooner obtained some data than the solenoid would burn out and we were winding a new one.

On the bright side, the frequent burn-outs provided the opportunity for refinement and redesign. Soon we were embedding the coils in a black, rubbery resin. While it was supposed to be impervious to water, it did not have good adhesion properties (except to clothing and human flesh) and the solenoids continued to burn out. Eventually, an epoxy coating sealed the solenoid against the water that allowed the electrolysis, and in more recent times we replaced water with a fluorocarbon liquid that does not conduct electricity or support electrolysis. Along the way to a reliable solenoid, we learned how to slow and stop atoms efficiently (Phillips and Metcalf, 1982; Prodan, Phillips, and Metcalf, 1982; Phillips, Prodan, and Metcalf, 1983a, 1983b, 1984a, 1984b, 1985; Metcalf and Phillips, 1985).

The velocity distribution after deceleration is measured in a detection region some distance from the exit end of the solenoid.

Here a separate detection laser beam produces fluorescence from atoms having the correct velocity to be resonant. By scanning the frequency of the detection laser, we were able to determine the velocity distribution in the atomic beam. Observations with the detection laser were made just after turning off the cooling laser, so as to avoid any difficulties with having both lasers on at the same time. Figure 5 shows the velocity distribution resulting from Zeeman cooling: a large fraction of the initial distribution has been swept down into a narrow final velocity group.

One of the advantages of the Zeeman cooling technique is the ease with which the optical pumping problem is avoided. Because the atoms are always in a strong axial magnetic field (that is the reason for the "bias" windings in Fig. 4), there is a well-defined axis of quantization that allowed us to make use of the selection rules for radiative transitions and to avoid the undesirable optical pumping. Figure 6 shows the energy levels of Na in a magnetic field. Atoms in the $3S_{1/2}$ ($m_F = 2$) state, irradiated with circularly polarized σ^+ light, must increase their m_F by one unit, and so can go only to the $3P_{3/2}$ ($m_{F'} = 3$) state. This state in turn can decay only to $3S_{1/2}$ ($m_F = 2$), and the excitation process can be repeated indefinitely. Of course, the circular polarization is not perfect, so other excitations are possible, and these may lead to decay to other states. Fortunately, in a high magnetic field, such transitions are highly unlikely (Phillips and Metcalf, 1982): either they involve a change in the nuclear spin projection m_I, which is forbidden in the high field limit, or they are far from resonance. These features, combined with high purity of the circular polarization, allowed us to achieve, without a "wrong transition," the 3×10^4 excitations required to stop the atoms. Furthermore, the circular polarization produced some "good" optical pumping: atoms not initially in the $3S_{1/2}$ ($m_F = 2$) state were pumped into this state, the "stretched" state of maximum projection of angular momentum, as they absorbed the angular momentum of the light. These various aspects of optical selection rules and optical pumping allowed the process of Zeeman cooling to be very efficient, decelerating a large fraction of the atoms in the beam.

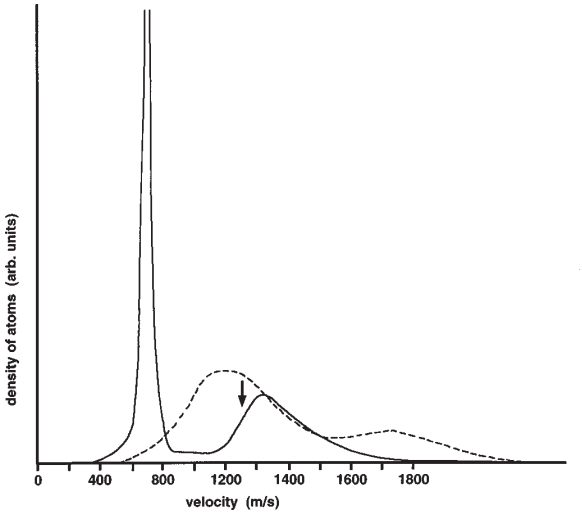

FIG. 5. Velocity distribution before (dashed) and after (solid) Zeeman cooling. The arrow indicates the highest velocity resonant with the slowing laser. (The extra bump at 1700 m/s is from $F = 1$ atoms, which are optically pumped into $F = 2$ during the cooling process.)

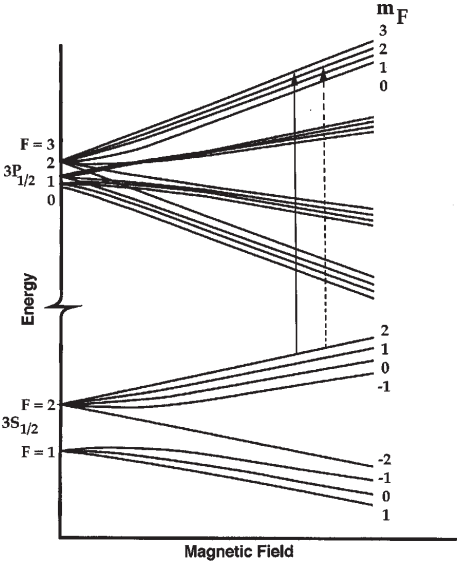

FIG. 6. Energy levels of Na in a magnetic field. The cycling transition used for laser cooling is shown as a solid arrow, and one of the nearly forbidden excitation channels leading to undesirable optical pumping is shown dashed.

In 1983 we discussed a number of these aspects of laser deceleration, including our early chirp-cooling results, at a two-day workshop on "Laser-Cooled and Trapped Atoms" held at NBS in Gaithersburg (Phillips, 1983). I view this as an important meeting in that it and its proceedings stimulated interest in laser cooling. In early 1984, Stig Stenholm, then of the University of Helsinki, organized an international meeting on laser cooling in Tvärminne, a remote peninsula in Finland. Figure 7 shows the small group attending (I was the photographer), and in that group, only some of the participants were even active in laser cooling at the time. Among these were Stig Stenholm [who had done pioneering work in the theory of laser cooling and the mechanical effects of light on atoms (Stenholm, 1978a, 1978b, 1985, 1986; Javanainen and Stenholm, 1980a, 1980b, 1980c, 1981a, 1981b)] along with some of his young colleagues; Victor Balykin and Vladimir Minogin from the Moscow group; and Claude Cohen-Tannoudji and Jean Dalibard from Ecole Normale Supérieure (ENS) in Paris, who had begun working on the theory of laser cooling and trapping. Also present were Jürgen Mlynek and Wolfgang Ertmer, both of whom now lead major research groups pursuing laser cooling and atom optics. At that time, however, only our group and the Moscow group had published any experiments on cooling of neutral atoms.

Much of the discussion at the Tvärminne meeting involved the techniques of beam deceleration and the problems with optical pumping. I took a light-hearted attitude toward our trials and tribulations with optical pumping, often joking that any unexplained features in our data could certainly be attributed to optical pumping. Of course, at the Ecole Normale, optical pumping had a long and distinguished history. Having been pioneered by Alfred Kastler and Jean Brossel, optical pumping had been the backbone of many experiments in the Laboratoire de Spectroscopie Hertzienne (now the Laboratoire Kastler Brossel). After one discussion in which I had joked about optical pumping, Jean Dalibard privately mentioned to me, "You know, Bill, at the Ecole Normale, optical pumping is not a joke." His gentle note of caution calmed me down a bit, but it turned out to be strangely prophetic as well. As we saw a few

years later, optical pumping had an important, beautiful, and totally unanticipated role to play in laser cooling, and it was surely no joke.

FIG. 7. Stig Stenholm's "First International Conference on Laser Cooling" in Tvärminne, March 1984. Back row, left to right: Juha Javanainen, Markus Lindberg, Stig Stenholm, Matti Kaivola, Nis Bjerre, (unidentified), Erling Riis, Rainer Salomaa, Vladimir Minogin. Front row: Jürgen Mlynek, Angela Guzmann, Peter Jungner, Wolfgang Ertmer, Birger Ståhlberg, Olli Serimaa, Jean Dalibard, Claude Cohen-Tannoudji, Victor Balykin.

Stopping Atoms

As successful as Zeeman cooling had been in producing large numbers of decelerated atoms as in Fig. 5, we had not actually observed the atoms at rest, nor had we trapped them. In fact, I recall a conversation with Steve Chu that took place during the International Conference on Laser Spectroscopy in Interlaken in 1983 in which I had presented our results on beam deceleration (Phillips, Prodan, and Metcalf, 1983a). Steve was working on positronium spectroscopy but was wondering whether there still might be something interesting to be done with laser cooling of neutral atoms. I offered the opinion that there was still plenty to do, and in particular, that trapping of atoms was still an unrealized goal.

It wasn't long before each of us achieved that goal, in very different ways.

Our approach was first to get some stopped atoms. The problem had been that, in a sense, Zeeman cooling worked too well. By adjusting the laser frequency and magnetic field, we could, up to a point, choose the final velocity of the atoms that had undergone laser deceleration. Unfortunately, if we chose too small a velocity, no slow atoms at all appeared in the detection region. Once brought below a certain velocity, about 200 m/s, the atoms always continued to absorb enough light while traveling from the solenoid to the detection region so as to stop before reaching the detector. By shutting off the cooling laser beam and delaying observation until the slow atoms arrived in the observation region, we were able to detect atoms as slow as 40 m/s with a spread of 10 m/s, corresponding to a temperature (in the atoms' rest frame) of 70 mK (Prodan, Phillips, and Metcalf, 1982).

The next step was to get these atoms to come to rest in our observation region. We were joined by Alan Migdall, a new postdoc, Jean Dalibard, who was visiting from ENS, and Ivan So, Hal Metcalf's student. We decided that we needed to proceed as before, shutting off the cooling light, allowing the slow atoms to drift into the observation region, but then to apply a short pulse of additional cooling light to bring the atoms to rest. The sequence of laser pulses required to do this — a long pulse of several milliseconds for doing the initial deceleration, followed by a delay and then another pulse of a few hundred microseconds, followed by another delay before detection — was provided by a rotating wheel with a series of openings corresponding to the places where the laser was to be on. Today we accomplish such pulse sequences with acousto-optic modulators under computer control, but in those days it required careful construction and balancing of a rapidly rotating wheel.

The result of this sequence of laser pulses was that we had atoms at rest in our observation region with a velocity spread corresponding to <100 mK (Prodan *et al.*, 1985). Just following our 1985 paper reporting this in *Physical Review Letters* was a report of the successful stopping of atoms by the chirp-cooling

method in Jan Hall's group (Ertmer, Blatt, Hall, and Zhu, 1985). At last there were atoms slow enough to be trapped, and we decided to concentrate first on magnetostatic trapping.

Magnetic Trapping of Atoms

The idea for magnetic traps had first appeared in the literature as early as 1960 (Heer, 1960, 1963; Vladimirskii, 1960), although Wolfgang Paul had discussed them in lectures at the University of Bonn in the mid-1950s, as a natural extension of ideas about magnetic focusing of atomic beams (Vauthier, 1949; Friedburg, 1951; Friedburg and Paul, 1951). Magnetic trapping had come to our attention particularly because of the successful trapping of cold neutrons (Kugler *et al.*, 1978). We later learned that in unpublished experiments in Paul's laboratory, there were indications of confining sodium in a magnetic trap (Martin, 1975).

The idea of magnetic trapping is that in a magnetic field, an atom with a magnetic moment will have quantum states whose magnetic or Zeeman energy increases with increasing field and states whose energy decreases, depending on the orientation of the moment compared to the field. The increasing-energy states, or low-field-seekers, can be trapped in a magnetic field configuration having a point where the magnitude of the field is a relative minimum. [No dc field can have a relative maximum in free space (Wing, 1984), so high-field-seekers cannot be trapped.] The requirement for stable trapping, besides the kinetic energy of the atom being low enough, is that the magnetic moment move adiabatically in the field. That is, the orientation of the magnetic moment with respect to the field should not change.

We considered some of the published designs for trapping neutrons, including the spherical hexapole (Golub and Pendlebury, 1979), a design comprising three current loops, but we found them less than ideal. Instead we decided upon a simpler design, with two loops, which we called a spherical quadrupole. The trap, its magnetic field lines and equipotentials are shown in Fig. 8. Although we thought that we had discovered an original trap design, we later learned that Wolfgang Paul had considered this many years ago, but

had not given it much attention because atoms were not harmonically bound in such a trap. In fact, the potential for such a trap is linear in the displacement from the center and has a cusp there.

With a team consisting of Alan Migdall, John Prodan, Hal Metcalf and myself, and with the theoretical support of Tom Bergeman, we succeeded in trapping atoms in the apparatus shown in Fig. 9 (Migdall *et al.*, 1985). As in the experiments that stopped atoms, we start with Zeeman slowing, decelerating the atoms to 100 m/s in the solenoid. The slowing laser beam is then extinguished, allowing the atoms to proceed unhindered for 4 ms to the magnetic trap. At this point, only one of the two trap coils has current; it produces a magnetic field that brings the atoms into resonance with the cooling laser when it is turned on again for 400 μs, bringing the atoms to rest. Once the atoms are stopped, the other coil is energized, producing the field shown in Fig. 8, and the trap is sprung. The atoms are held in the trap until released, or until collisions with the room-temperature background gas molecules in the imperfect vacuum knock them out. After the desired trapping time, we turn off the magnetic field, and turn on a probe laser, so as to see how many atoms remain in the trap. By varying the frequency of this probe on successive repetitions of the process, we could determine the velocity distribution of the atoms, via their Doppler shifts.

The depth of our trap was about 17 mK (25 mT), corresponding to Na atoms with a velocity of 3.5 m/s. In the absence of trapping fields, atoms that fast would escape from the region of the trap coils in a few milliseconds. Figure 10 shows a section of chart paper with spectra of the atoms remaining after 35 ms of trapping time. If the trap had not been working, we would have seen essentially nothing after that length of time, but the signal, noisy as it was, was unmistakable. It went away when the trap was off, and it went away when we did not provide the second pulse of cooling light that stops the atoms before trapping them. This was just the signature we were looking for, and Hal Metcalf expressed his characteristic elation at good results with his exuberant ‘‘WAHOO!!’’ at the top of the chart.

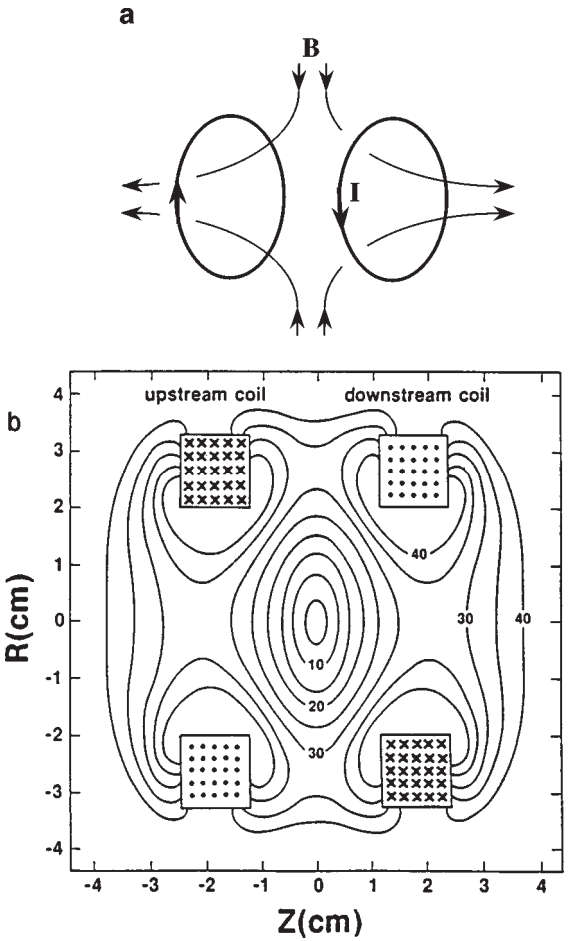

FIG. 8. (a) Spherical quadrupole trap with lines of B-field. (b) Equipotentials of our trap (equal field magnitudes in millitesla), in a plane containing the symmetry (z) axis.

As the evening went on, we were able to improve the signal, but we found that the atoms did not stay very long in the trap, a feature we found a bit frustrating. Finally, late in the evening we decided to go out and get some fast food, talk about what was happening and attack the problem afresh. When we returned a little later that night, the signal had improved and we were able to trap atoms for much longer times. We soon realized that during our supper break the magnetic trap had cooled down, and stopped outgassing, so the vacuum just in the vicinity of the trap improved considerably. With this insight we knew to let the magnet cool off

from time to time, and we were able to take a lot of useful data. We continued taking data until around 5:00 am, and it was probably close to 6:00 am when my wife Jane found Hal and me in our kitchen, eating ice cream as she prepared to leave for work. Her dismay at the lateness of our return and our choice of nourishment at that hour was partially assuaged by Hal's assurance that we had accomplished something pretty important that night.

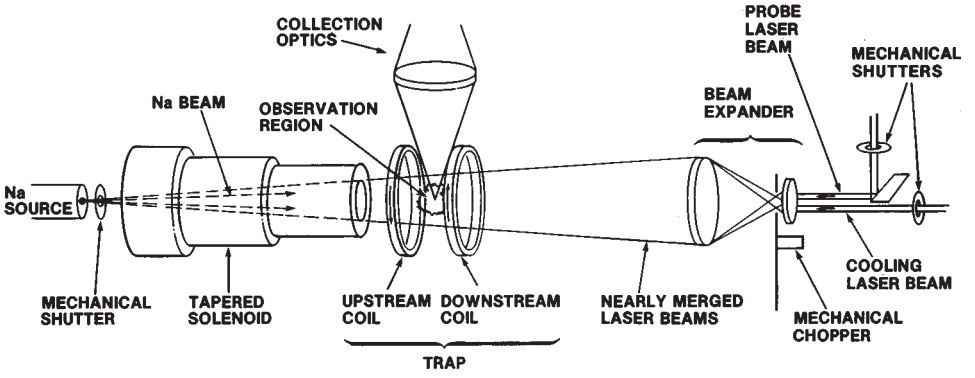

FIG. 9. Schematic of the apparatus used to trap atoms magnetically.

Figure 11(a) presents the sequence of spectra taken after various trapping times, showing the decrease in signal as atoms are knocked out of the trap by collisions with the background gas molecules. Figure 11(b) shows that the loss of atoms from the trap is exponential, as expected, with a lifetime of a bit less than one second, in a vacuum of a few times 10^{-6} pascals. A point taken when the vacuum was allowed to get worse illustrates that poor vacuum made the signal decay faster. In more recent times, we and others have achieved much longer trapping times, mainly because of an improved vacuum. We now observe magnetic trap lifetimes of one minute or longer in our laboratory.

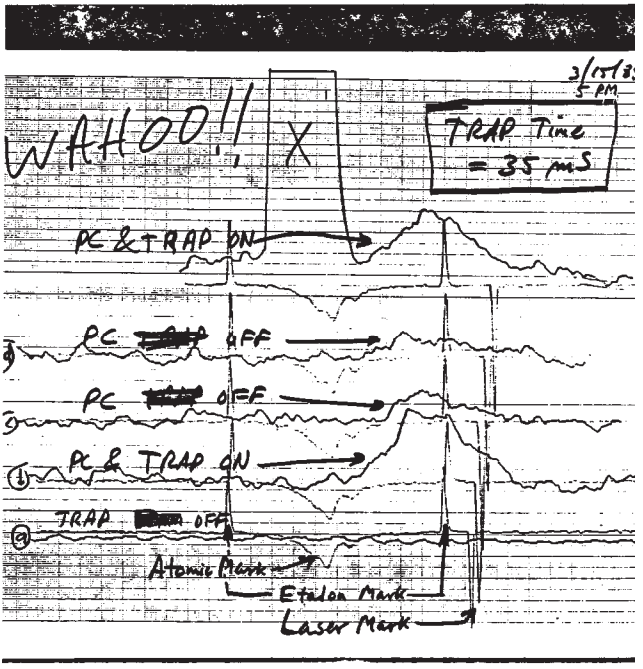

FIG. 10. A section of chart paper from 15 March 1985. "PC" and " no PC" refer to presence or absence of the ''post-cooling'' pulse that brings the atoms to rest in the trapping region.

Since our demonstration (Migdall *et al.*, 1985) of magnetic trapping of atoms in 1985, many different kinds of magnetic atom traps have been used. At MIT, Dave Pritchard's group trapped (Bagnato *et al.*, 1987) and cooled (Helmerson *et al.*, 1992) Na atoms in a linear quadrupole magnetic field with an axial bias field, similar to the trap first discussed by Ioffe and collaborators (Gott, Ioffe, and Telkovsky, 1962) in 1962, and later by others (Pritchard, 1983; Bergeman *et al.*, 1987). Similar traps were used by the Kleppner-Greytak group to trap (Hess *et al.*, 1987) and evaporatively cool (Masuhura *et al.*, 1988) atomic hydrogen, and by Walraven's group to trap (van Roijen *et al.*, 1988) and laser-cool hydrogen (Setija *et al.*, 1994). The Ioffe trap has the advantage of having a non-zero magnetic field at the equilibrium point, in contrast to the spherical quadrupole, in which the field is zero at the equilibrium point. The zero field allows the magnetic moment of the atom to flip (often called Majorana flopping), so that the atom is in

an untrapped spin state. While this problem did not cause difficulties in our 1985 demonstration, for colder atoms, which spend more time near the trap center, it can be a quite severe loss mechanism (Davis, Mewes, Joffe *et al.*, 1995; Petrich *et al.*, 1995). In 1995, modifications to the simple quadrupole trap solved the problem of spins flips near the trap center, and allowed the achievement of Bose-Einstein condensation (Anderson *et al.*, 1995; Davis, Mewes, Andrews *et al.*, 1995).

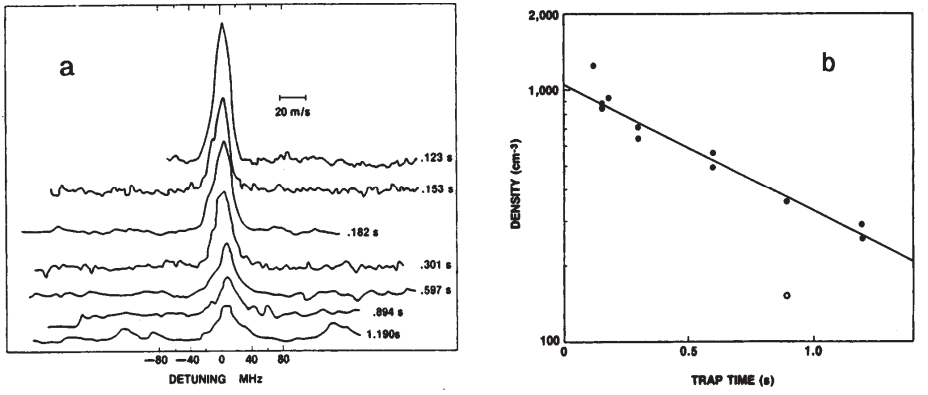

FIG. 11. (a) Spectra of atoms remaining in the magnetic trap after various times; (b) decay of number of trapped atoms with time. The open point was taken at twice the background pressure of the other points.

Optical Molasses

At the same time that we were doing the first magnetic trap experiments in Gaithersburg, the team at Bell Labs, led by Steve Chu, was working on a different and extremely important feature of laser cooling. After a beautiful demonstration in 1978 of the use of optical forces to focus an atomic beam (Bjorkholm *et al.*, 1978), the Bell Labs team had made some preliminary attempts to decelerate an atom beam, and then moved on to other things. Encouraged by the beam deceleration experiments in Gaithersburg and in Boulder, Steve Chu reassembled much of that team and set out to demonstrate the kind of laser cooling suggested in 1975 by Hänsch and Schawlow. [The physical principles behind the Hänsch and Schawlow proposal are, of course, identical to those expressed in the 1975 Wineland and Dehmelt laser cooling proposal. These

principles had already led to the laser cooling of trapped ions (Neuhauser *et al.*, 1978; Wineland *et al.*, 1978). The foci of Hänsch and Schawlow (1975) and Wineland and Dehmelt (1975), however, has associated the former with neutral atoms and the latter with ions.] In fact, the same physical principle of Doppler cooling results in the compression of the velocity distribution associated with laser deceleration of an atomic beam [see sections 2 and 3 of Phillips (1992)]. Nevertheless, in 1985, laser cooling of a gas of neutral atoms at rest, as proposed in Hänsch and Schawlow (1975), had yet to be demonstrated.

The idea behind the Hänsch and Schawlow proposal is illustrated in Fig. 12. A gas of atoms, represented here in one dimension, is irradiated from both sides by laser beams tuned slightly below the atomic resonance frequency. An atom moving toward the left sees that the laser beam opposing its motion is Doppler shifted toward the atomic resonance frequency. It sees that the laser beam directed along its motion is Doppler shifted further from its resonance. The atom therefore absorbs more strongly from the laser beam that opposes its motion, and it slows down. The same thing happens to an atom moving to the right, so all atoms are slowed by this arrangement of laser beams. With pairs of laser beams added along the other coordinate axes, one obtains cooling in three dimensions. Because of the role of the Doppler Effect in the process, this is now called Doppler cooling.

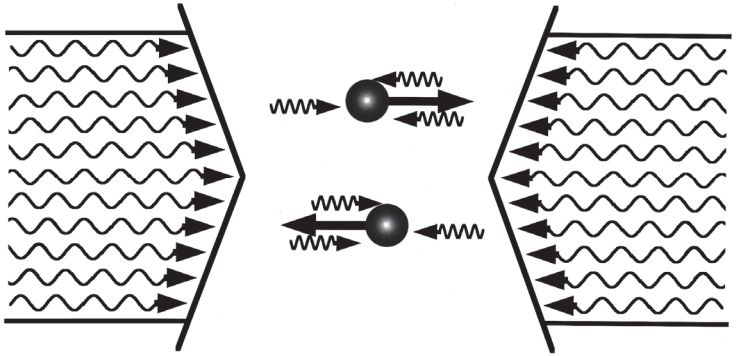

FIG. 12. Doppler cooling in one dimension.

Later treatments (Letokhov *et al.*, 1977; Neuhauser *et al.*, 1978; Stenholm, 1978a; Wineland *et al.*, 1978; Wineland and Itano, 1979; Javanainen, 1980; Javanainen and Stenholm, 1980b) recognized that this cooling process leads to a temperature whose lower limit is on the order of $\hbar\Gamma$, where Γ is the rate of spontaneous emission of the excited state (Γ^{-1} is the excited state lifetime).

The temperature results from an equilibrium between laser cooling and the heating process arising from the random nature of both the absorption and emission of photons. The random addition to the average momentum transfer produces a random walk of the atomic momentum and an increase in the mean square atomic momentum. This heating is countered by the cooling force F opposing atomic motion. The force is proportional to the atomic velocity, as the Doppler shift is proportional to velocity. In this, the cooling force is similar to the friction force experienced by a body moving in a viscous fluid. The rate at which energy is removed by cooling is $\mathbf{F}\cdot\mathbf{v}$, which is proportional to v^2, so the cooling rate is proportional to the kinetic energy. By contrast the heating rate, proportional to the total photon scattering rate, is independent of atomic kinetic energy for low velocities. As a result, the heating and cooling come to equilibrium at a certain value of the average kinetic energy. This defines the temperature for Doppler cooling, which is

$$m\left\langle v_i^2\right\rangle = k_B T = \frac{\hbar\Gamma}{4}\left(\frac{\Gamma}{2\delta}+\frac{2\delta}{\Gamma}\right), \qquad (1)$$

where δ is the angular frequency of the detuning of the lasers from atomic resonance and v_i is the velocity along some axis. This expression is valid for 3D Doppler cooling in the limit of low intensity and when the recoil energy $\hbar^2k^2/2m \ll \hbar\Gamma$. Interestingly, the equilibrium velocity distribution for Doppler cooling is the Maxwell-Boltzmann distribution. This follows from the fact that the Fokker-Planck equation describing the damping and heating in laser cooling is identical in form to the equation that describes collisional equilibrium of a gas (Stenholm, 1986). Numerical simulations of real cases, where the recoil energy does not vanish, show that the distribution is still very close to Maxwellian (Lett *et al.*, 1989). The

minimum value of this temperature is called the Doppler cooling limit, occurring when $\delta = -\Gamma/2$,

$$k_B T_{\text{Dopp}} = \frac{\hbar\Gamma}{2}. \tag{2}$$

The first rigorous derivation of the cooling limit appears to be by Letokhov, Minogin, and Pavilik (1977) [although the reader should note that Eq. (32) is incorrectly identified with the rms velocity]. Wineland and Itano (1979) give derivations for a number of different situations involving trapped and free atoms and include the case where the recoil energy is not small but the atoms are in collisional equilibrium.

The Doppler cooling limit for sodium atoms cooled on the resonance transition at 589 nm where $\Gamma/2\pi$ = 10 MHz, is 240 μK, and corresponds to an rms velocity of 30 cm/s along a given axis. The limits for other atoms and ions are similar, and such low temperatures were quite appealing. Before 1985, however, these limiting temperatures had not been obtained in either ions or neutral atoms.

A feature of laser cooling not appreciated in the first treatments was the fact that the spatial motion of atoms in any reasonably sized sample would be diffusive. For example, a simple calculation (Lett *et al*., 1989) shows that a sodium atom cooled to the Doppler limit has a "mean free path" (the mean distance it moves before its initial velocity is damped out and the atom is moving with a different, random velocity) of only 20 μm, while the size of the laser beams doing the cooling might easily be one centimeter. Thus, the atom undergoes diffusive, Brownian-like motion, and the time for a laser cooled atom to escape from the region where it is being cooled is much longer than the ballistic transit time across that region. This means that an atom is effectively "stuck" in the laser beams that cool it. This stickiness, and the similarity of laser cooling to viscous friction, prompted the Bell Labs group (Chu *et al.*, 1985) to name the intersecting laser beams "optical molasses." At NBS (Phillips, Prodan, and Metcalf, 1985), we independently used the term "molasses" to describe the cooling configuration, and the name "stuck." Note that an optical molasses is not a trap. There is no restoring force

keeping the atoms in the molasses, only a viscous inhibition of their escape.

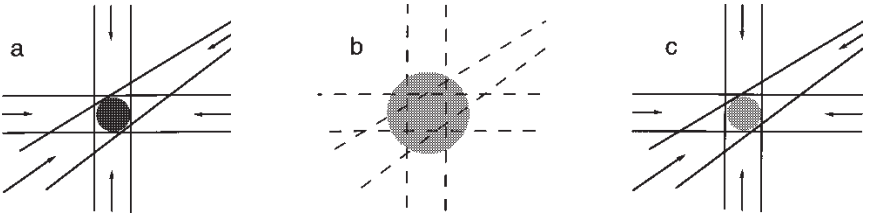

FIG. 13. Release-and-recapture method for temperature measurement.

Using the techniques for chirp cooling an atomic beam developed at NBS-JILA (Ertmer *et al.*, 1985) and a novel pulsed beam source, Chu's team at Bell Labs succeeded in loading cold sodium atoms into an optical molasses (Chu *et al.*, 1985). They observed the expected long "lifetime" (the time required for the atoms to diffuse out of the laser beams) of the molasses, and they developed a method, now called "release-and-recapture," for measuring the temperature of the atoms. The method is illustrated in Fig. 13. First, the atoms are captured and stored in the molasses, where for short periods of time they are essentially immobile due to the strong damping of atomic motion [Fig. 13(a)]. Then, the molasses laser beams are switched off, allowing the atoms to move ballistically away from the region to which they had originally been viscously confined [Fig. 13(b)]. Finally the laser beams are again turned on, recapturing the atoms that remain in the intersection (molasses) region [Fig. 13(c)]. From the fraction of atoms remaining after various periods of ballistic expansion one can determine the velocity distribution and therefore the temperature of the atoms at the time of release. The measured temperature at Bell Labs was 240^{+200}_{-60} μK. [Today one would expect a much lower temperature; the high temperature observed in this experiment has since been ascribed to the presence of a stray magnetic field from an ion pump (Chu, 1997).] The large uncertainty is due to the sensitive dependence of the analysis on the size and density distribution of atoms in the molasses, but the result was satisfyingly consistent with the predicted Doppler cooling limit.

By the end of 1986, Phil Gould and Paul Lett had joined our group and we had achieved optical molasses in our laboratory at NBS, loading the molasses directly from a decelerated beam. [Today it is also routine to load atoms directly into a magneto-optical trap (MOT) (Raab *et al.*, 1987) from an uncooled vapor (Cable *et al.*, 1990; Monroe *et al.*, 1990) and then into molasses.] We repeated the release-and-recapture temperature measurements, found them to be compatible with the reported measurements of the Bell Labs group, and we proceeded with other experiments. In particular, with Paul Julienne, Helen Thorsheim and John Wiener, we made a 2-focus laser trap and used it to perform the first measurements of a specific collision process (associative ionization) with laser cooled atoms (Gould *et al.*, 1988). [Earlier, Steve Chu and his colleagues had used optical molasses to load a single-focus laser trap—the first demonstration of an optical trap for atoms (Chu *et al.*, 1986).] In a sense, our collision experiment represented a sort of closure for me because it realized the two-focus trap proposed in Ashkin's 1978 paper, the paper that had started me thinking about laser cooling and trapping. It also was an important starting point for our group, because it began a new and highly productive line of research into cold collisions, producing some truly surprising and important results (Lett *et al.*, 1991; Lett *et al.*, 1993; Ratliff *et al.*, 1994; Lett *et al.*, 1995; Walhout *et al.*, 1995; Jones *et al.*, 1996; Tiesinga *et al.*, 1996). In another sense, though, that experiment was a detour from the road that was leading us to a new understanding of optical molasses and of how laser cooling worked.

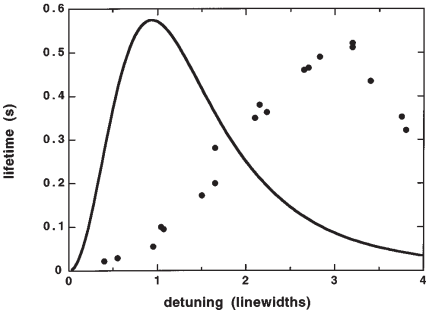

FIG. 14. Experimental molasses lifetime (points) and the theoretical decay time (curve) vs detuning of molasses laser from resonance.

Sub-Doppler Laser Cooling

During 1987 Gould, Lett and I investigated the behavior of optical molasses in more detail. Because the temperature was hard to measure and its measurement uncertainty was large, we concentrated instead on the molasses lifetime, the time for the atoms to diffuse out of the intersecting laser beams. We had calculated, on the basis of the Doppler cooling theory, how the lifetime would vary as a function of the laser frequency detuning and the laser intensity. We also calculated how the lifetime should change when we introduced a deliberate imbalance between the two beams of a counter-propagating pair. Now we wanted to compare experimental results with our calculations. The results took us somewhat by surprise.

Figure 14 shows our measurements (Lett *et al.*, 1989) of the molasses lifetime as a function of laser frequency along with the predicted behavior according to the Doppler cooling theory. The 1-D theory did not quantitatively reproduce the observed 3-D diffusion times, but that was expected. The surprise was the qualitative differences: the experimental lifetime peaked at a laser detuning above 3 linewidths, while the theory predicted a peak below one linewidth. We did not know how to reconcile this difficulty, and the results for the drift induced by beam imbalance were also in strong disagreement with the Doppler theory. In our 1987 paper, we described our failed attempts to bring the Doppler cooling theory into agreement with our data and ended saying (Gould *et al.*, 1987): "It remains to consider whether the multiple levels and sublevels of Na, multiple laser frequencies, or a consideration of the detailed motion of the atoms in 3-D can explain the surprising behavior of optical molasses." This was pure guesswork, of course, but it turned out to have an element of truth, as we shall see below.

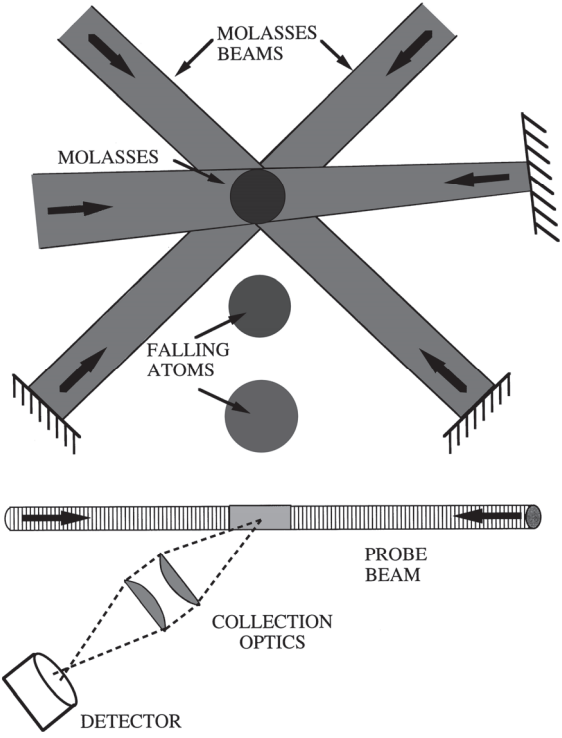

FIG. 15. Time-of-flight method for measuring laser cooling temperatures.

Having seen such a clear discrepancy between the Doppler cooling theory and the experimental results, with no resolution in sight, we, as experimentalists, decided to take more data. Paul Lett argued that we should measure the temperature again, this time as a function of the detuning, to see if it, too, would exhibit behavior different from that predicted by the theory. We felt, however, that the release-and-recapture method, given the large uncertainty associated with it in the past, would be unsuitable. Hal Metcalf suggested a different approach, illustrated in Fig. 15.

In this time-of-flight (TOF) method, the atoms are first captured by the optical molasses, then released by switching off the molasses laser beams. The atom cloud expands ballistically, according to the distribution of atomic velocities. When atoms encounter the probe laser beam, they fluoresce, and the time

distribution of fluorescence gives the time-of-flight distribution for atoms arriving at the probe. From this the temperature can be deduced. Now, with a team that included Paul Lett, Rich Watts, Chris Westbrook, Phil Gould, as well as Hal Metcalf and myself, we implemented the TOF temperature measurement. In our experiment, the probe was placed as close as 1 cm from the center of the molasses, which had a radius of about 4.5 mm. At the lowest expected temperature, the Doppler cooling limit of 240 μK for Na atoms, a significant fraction of the atoms would have been able to reach the probe, even with the probe above the molasses. For reasons of convenience, we did put the probe beam above the molasses, but we saw no fluorescence from atoms reaching the probe after the molasses was turned off. We spent a considerable time testing the detection system to be sure that everything was working properly. We deliberately "squirted" the atoms to the probe beam by heating them with a pair of laser beams in the horizontal plane, and verified that such heated atoms reached the probe and produced the expected time-of-flight signal,

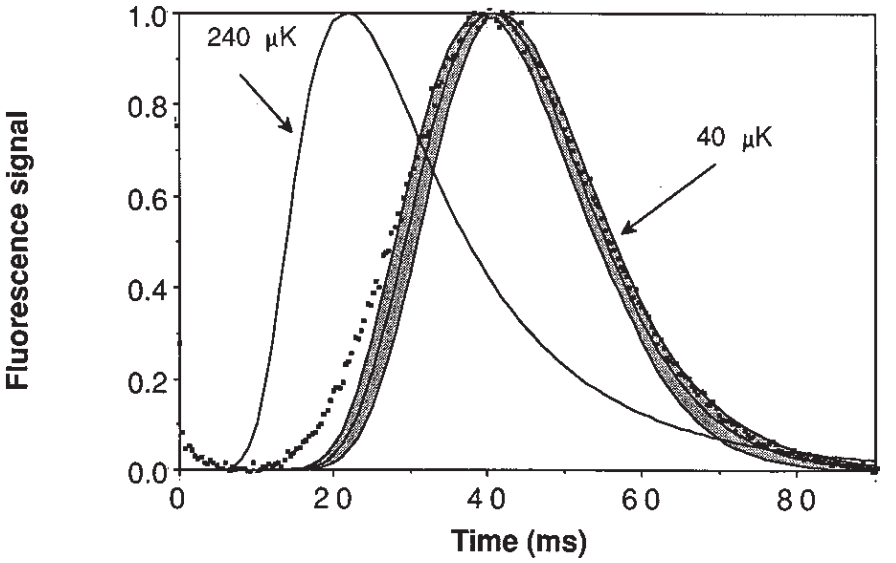

FIG. 16. The experimental TOF distribution (points) and the predicted distribution curves for 40 μK and 240 μK (the predicted lower limit of Doppler cooling). The band around the 40 μK curve reflects the uncertainty in the measurement of the geometry of the molasses and probe.

Finally, we put the probe *under* the molasses. When we did, we immediately saw the TOF signals, but were reluctant to accept the conclusion that the atoms were colder than the Doppler cooling theory predicted, until we had completed a detailed modeling of the TOF signals. Figure 16 shows a typical TOF distribution for one of the colder observed temperatures, along with the model predictions. The conclusion was inescapable: Our atoms had a temperature of about 40 μK, much colder than the Doppler cooling limit of 240 μK. They had had insufficient kinetic energy to reach the probe when it was placed above the molasses. As clear as this was, we were apprehensive. The theory of the Doppler limit was simple and compelling. In the limit of low intensity, one could derive the Doppler limit with a few lines of calculations (see for example, Lett *et al.*, 1989); the most complete theory for cooling a two-level atom (Gordon and Ashkin, 1980) did not predict a cooling limit any lower. Of course, everyone recognized that sodium was not a two-level atom, but it had seemed unlikely that it made any significant difference (our speculation in Gould *et al.*, 1987, notwithstanding). At low laser intensity the temperature depends on the laser detuning and the linewidth of the transition. Since the linewidth is identical for all possible transitions in the Na D2 manifold, and since the cooling transition [$3S_{1/2}\ (F=2) \rightarrow 3P_{3/2}\ (F=3)$] was well separated from nearby transitions, and all the Zeeman levels were degenerate, it seemed reasonable that the multilevel structure was unimportant in determining the cooling limit.

As it turned out, this was completely wrong. At the time, however, the Doppler limit seemed to be on firm theoretical ground, and we were hesitant to claim that it was violated experimentally. Therefore, we sought to confirm our experimental results with other temperature measurement methods. One of these was to refine the "release-and-recapture" method described above. The large uncertainties in the earlier measurements (Chu *et al.*, 1985) arose mainly from uncertainties in the size of the molasses and the recapture volume. We addressed that problem by sharply aperturing the molasses laser beams so the molasses and recapture volumes were well defined. We also found that it was essential to include the effect of gravity in the analysis (as we had done already for the TOF method). Because

released atoms fall, the failure to recapture atoms could be interpreted as a higher temperature if gravity is not taken into account.

Another method was the "fountain" technique. Here we exploited our initial failure to observe a TOF signal with the probe above the molasses. By adjusting the height of the probe, we could measure how high the atoms could go before falling back under the influence of gravity. Essentially, this allowed us to measure the atoms' kinetic energy in terms of their gravitational potential energy, a principle very different from the TOF method. Finally, we used the "shower" method. This determined how far the atoms spread in the horizontal direction as they fell following release from the molasses. For this, we measured the fluorescence from atoms reaching the horizontal probe laser beam at different positions along that beam. From this transverse position distribution, we could get the transverse velocity distribution and therefore the temperature.

(The detailed modeling of the signals expected from the various temperature measurement methods was an essential element in establishing that the atomic temperature was well below the Doppler limit. Rich Watts, who had come to us from Hal Metcalf's lab and had done his doctoral dissertation with Carl Wieman, played a leading role in this modeling. Earlier, with Wieman, he had introduced the use of diode lasers in laser cooling. With Metcalf, he was the first to laser cool rubidium, the element with which Bose-Einstein condensation was first achieved. He was a pioneer of laser cooling and continued a distinguished scientific career at NIST after completing his postdoctoral studies in our group. Rich died in 1996 at the age of 39, and is greatly missed.)

While none of the additional methods proved to be as accurate as the TOF technique (which became a standard tool for studying laser cooling temperatures), each of them showed the temperature to be significantly below the Doppler limit. Sub-Doppler temperatures were not the only surprising results we obtained. We also (as Paul Lett had originally suggested) measured the temperature as a function of the detuning from resonance of the molasses laser. Figure 17 shows the results, along with the

prediction of the Doppler cooling theory. The dependence of the temperature on detuning is strikingly different from the Doppler theory prediction, and recalls the discrepancy evident in Fig. 14. Our preliminary study indicated that the temperature did not depend on the laser intensity [although later measurements (Lett *et al.*, 1989; Phillips *et al.*, 1989; Salomon *et al.*, 1990) showed that the temperature actually had a linear dependence on intensity]. We observed that the temperature depended on the polarization of the molasses laser beams, and was highly sensitive to the ambient magnetic field. Changing the field by 0.2 mT increased the temperature from 40 μK to 120 μK when the laser was detuned 20 MHz from resonance [later experiments (Lett *et al.*, 1989) showed even greater effects]. This field dependence was particularly surprising, considering that transitions were being Zeeman shifted on the order of 14 MHz/mT, so the Zeeman shifts were much less than either the detuning or the 10 MHz transition linewidth. Armed with these remarkable results, in the early spring of 1988 we sent a draft of the paper (Lett *et al.*, 1988) describing our measurements to a number of experimental and theoretical groups working on laser cooling. I also traveled to a few of the leading laser cooling labs to describe the experiments in person and discuss them. Many of our colleagues were skeptical, as well they might have been, considering how surprising the results were. In the laboratories of Claude Cohen-Tannoudji and of Steve Chu, however, the response was: "Let's go into the lab and find out if it is true." Indeed, they soon confirmed sub-Doppler temperatures with their own measurements and they began to work on an understanding of how such low temperatures could come about. What emerged from these studies was a new concept of how laser cooling works, an understanding that is quite different from the original Hänsch-Schawlow and Wineland-Dehmelt picture.

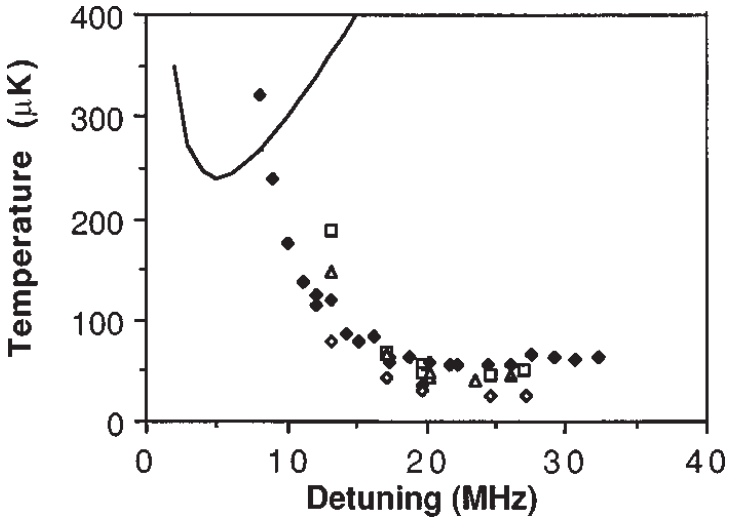

FIG. 17. Dependence of molasses temperature on laser detuning (points) compared to the prediction of Doppler cooling theory (curve). The different symbols represent different molasses-to-probe separations.

During the spring and summer of 1988 our group was in close contact with Jean Dalibard and Claude Cohen-Tannoudji as they worked out the new theory of laser cooling and we continued our experiments. Their thinking centered on the multilevel character of the sodium atom, since the derivation of the Doppler limit was rigorous for a two-level atom. The sensitivity of temperature to magnetic field and to laser polarization suggested that the Zeeman sublevels were important, and this proved to be the case. Steve Chu (now at Stanford) and his colleagues followed a similar course, but the physical image that Dalibard and Cohen-Tannoudji developed has dominated the thinking about multilevel laser cooling. It involves a combination of multilevel atoms, polarization gradients, light shifts and optical pumping. How these work together to produce laser cooling is illustrated in simple form in Fig. 18, but the reader should see the Nobel Lectures of Cohen-Tannoudji and Chu along with the more detailed papers (Dalibard and Cohen-Tannoudji, 1989; Ungar *et al.*, 1989; Cohen-Tannoudji and Phillips, 1990; Cohen-Tannoudji, 1992).

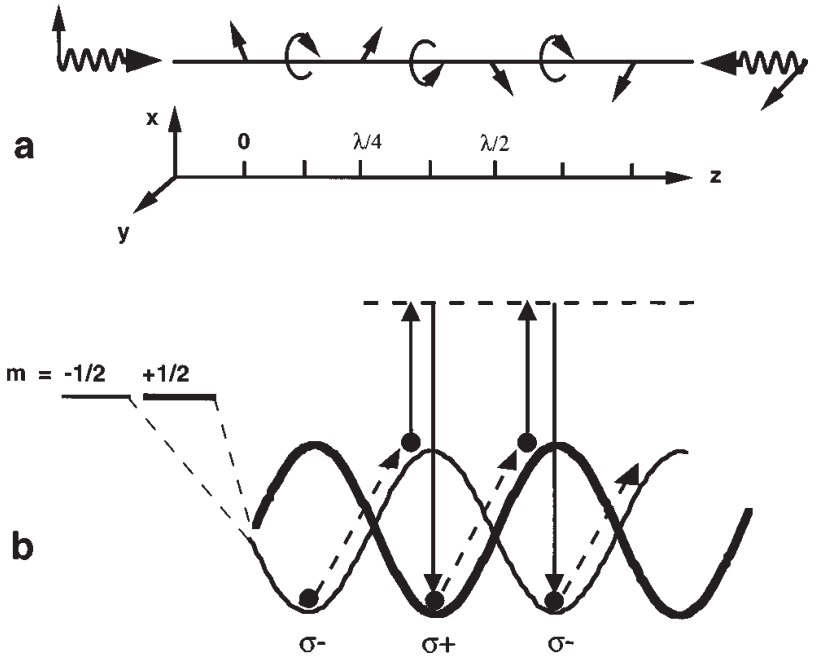

FIG. 18. (a) Interfering, counter-propagating beams having orthogonal, linear polarizations create a polarization gradient. (b) The different Zeeman sublevels are shifted differently in light fields with different polarizations; optical pumping tends to put atomic population on the lowest energy level, but non-adiabatic motion results in "Sisyphus" cooling.

Figure 18(a) shows a 1-D set of counter-propagating beams with equal intensity and orthogonal, linear polarizations. The interference of these beams produces a standing wave whose polarization varies on a sub wavelength distance scale. At points in space where the linear polarizations of the two beams are in phase with each other, the resultant polarization is linear, with an axis that bisects the polarization axes of the two individual beams. Where the phases are in quadrature, the resultant polarization is circular and at other places the polarization is elliptical. An atom in such a standing wave experiences a fortunate combination of light shifts and optical pumping processes.

Because of the differing Clebsch-Gordan coefficients governing the strength of coupling between the various ground and excited sublevels of the atom, the light shifts of the different

sublevels are different, and they change with polarization (and therefore with position). Figure 18(b) shows the sinusoidal variation of the ground-state energy levels (reflecting the varying light shifts or dipole forces) of a hypothetical $J_g=1/2 \rightarrow J_e=3/2$ atomic system. Now imagine an atom to be at rest at a place where the polarization is circular σ^- as at $z=\lambda/8$ in Fig. 18(a). As the atom absorbs light with negative angular momentum and radiates back to the ground states, it will eventually be optically pumped into the $m_g=-1/2$ ground state, and simply cycle between this state and the excited $m_e=-3/2$ state. For low enough intensity and large enough detuning we can ignore the time the atom spends in the excited state and consider only the motion of the atom on the ground state potential. In the $m_g=-1/2$ state, the atom is in the lower energy level at $z=\lambda/8$, as shown in Fig. 18(b). As the atom moves, it climbs the potential hill of the $m_g=-1/2$ state, but as it nears the top of the hill at $z=3\lambda/8$, the polarization of the light becomes σ^+ and the optical pumping process tends to excite the atom in such a way that it decays to the $m_g=+1/2$ state. In the $m_g=+1/2$ state, the atom is now again at the bottom of a hill, and it again must climb, losing kinetic energy, as it moves. The continual climbing of hills recalls the Greek myth of Sisyphus, so this process, by which the atom rapidly slows down while passing through the polarization gradient, is called Sisyphus cooling. Dalibard and Cohen-Tannoudji (1985) had already described another kind of Sisyphus cooling, for two-level atoms, so the mechanism and the name were already familiar. In both kinds of Sisyphus cooling, the radiated photons, in comparison with the absorbed photons, have an excess energy equal to the light shift. By contrast, in Doppler cooling, the energy excess comes from the Doppler shift.

The details of this theory were still being worked out in the summer of 1988, the time of the International Conference on Atomic Physics, held that year in Paris. The sessions included talks about the experiments on sub-Doppler cooling and the new ideas to explain them. Beyond that, I had lively discussions with Dalibard and Cohen-Tannoudji about the new theory. One insight that emerged from those discussions was an understanding of why we had

observed such high sensitivity of temperature to magnetic field: It was not the size of the Zeeman shift compared to the linewidth or the detuning that was important. Rather, when the Zeeman shift was comparable to the much smaller (≈1 MHz) light shifts and optical pumping rates, the cooling mechanism, which depended on these phenomena, would be disturbed. We now suggested a crucial test: the effect of the magnetic field should be reduced if the light intensity were higher. From Paris, I telephoned back to the lab in Gaithersburg and urged my colleagues to perform the appropriate measurements.

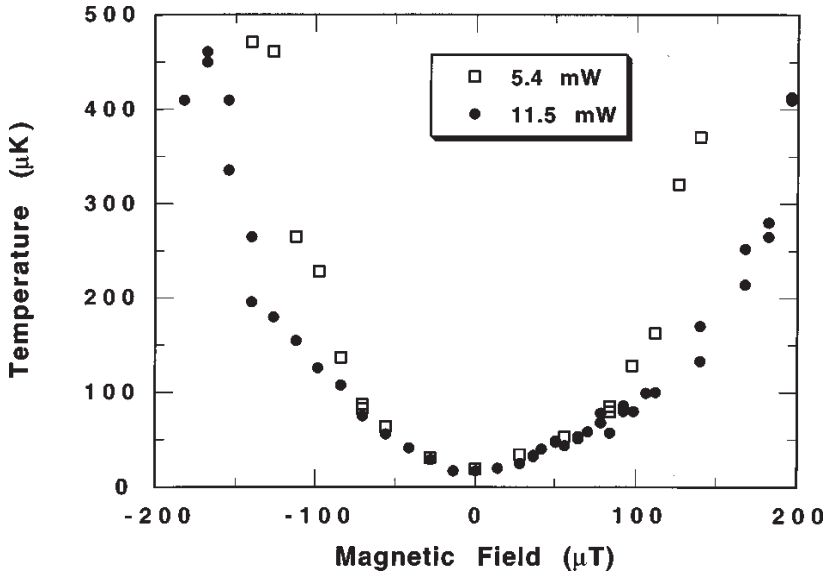

FIG. 19. Temperature vs magnetic field in a 3-D optical molasses. Observation of lower temperature at higher intensity when the magnetic field was high provided an early confirmation of the new theory of sub-Doppler cooling

The results were as we had hoped. Figure 19 shows temperature as a function of magnetic field for two different light intensities. At magnetic fields greater than 100 μT (1 gauss), the temperature was lower for higher light intensity, a reversal of the usual linear dependence of temperature and intensity (Lett *et al.*, 1989; Salomon *et al.*, 1990). We considered this to be an important early confirmation of the qualitative correctness of the new theory, confirming the central role played by the light shift and the magnetic

sublevels in the cooling mechanism. Joined by Steve Rolston and Carol Tanner we (Paul Lett, Rich Watts, Chris Westbrook, and myself) carried out additional studies of the behavior of optical molasses, providing qualitative comparisons with the predictions of the new theory. Our 1989 paper (Lett *et al.*), "Optical Molasses" summarized these results and contrasted the predictions of Doppler cooling with the new theory. Steve Chu's group also published additional measurements at the same time (Weiss *et al.*, 1989). Other, even more detailed measurements in Paris (Salomon *et al.*, 1990) (where I was very privileged to spend the academic year of 1989–1990) left little doubt about the correctness of the new picture of laser cooling. In those experiments we cooled Cs atoms to 2.5 μK. It was a truly exciting time, when the developments in the theory and the experiments were pushing each other to better understanding and lower temperatures. Around this time, Jan Hall [whose pioneering work in chirp cooling (Ertmer *et al.*, 1985) had done so much to launch the explosive activity a few years before] commented that being in the field of laser cooling was an experience akin to being in Paris at the time of the Impressionists. Figure 20 symbolizes the truth of that comment.

Optical Lattices

In 1989 we began a different kind of measurement on laser cooled atoms, a measurement that was to lead us to a new and highly fruitful field of research. We had always been a bit concerned that all of our temperature measurements gave us information about the velocity distribution of atoms *after* their release from the optical molasses and we wanted a way to measure the temperature *in situ*. Phil Gould suggested that we measure the spectrum of the light emitted from the atoms while they were being cooled. For continuous, single frequency irradiation at low intensity and large detuning, most of the fluorescence light scattered from the atoms should be "elastically" scattered, rather than belonging to the "Mollow triplet" of high-intensity resonance fluorescence (Mollow, 1969). This elastically scattered light will be Doppler shifted by the moving atoms and its spectrum should show a Doppler broadening characteristic of the temperature of the atomic sample. The spectrum will also contain the frequency fluctuations of the laser itself, but

these are relatively slow for a dye laser, so Gould suggested a heterodyne method of detection, where the fluorescent light is mixed on a photodiode with local oscillator light derived from the molasses laser, producing a beat signal that is free of the laser frequency fluctuations.

FIG. 20. (Color) Hal Metcalf, Claude Cohen-Tannoudji and the author on the famous bridge in Monet's garden at Giverny, ca. 1990.

The experiment was not easy, and it worked mainly because of the skill and perseverance of Chris Westbrook. An example of the surprising spectrum we obtained (Westbrook *et al.*, 1990) is shown in Fig. 21. The broad pedestal corresponded well to what we expected from the time-of-flight temperature measurement on a similar optical molasses, but the narrow central peak was a puzzle. After rejecting such wild possibilities as the achievement of Bose-Einstein condensation (Fig. 21 looks remarkably similar to velocity distributions in partially Bose-condensed atomic gases) we realized that the answer was quite simple: we were seeing line-narrowing from the Lamb-Dicke effect (Dicke, 1953) of atoms localized to less than a wavelength of light.

Atoms were being trapped by the dipole force in periodically spaced potential wells like those of Fig. 18(b). We knew from both theory and experiments that the thermal energy of the atoms was less than the light shifts producing the potential wells, so it was quite reasonable that the atoms should be trapped. Confined within a region much less than a wavelength of light, the emitted spectrum shows a suppression of the Doppler width, the Lamb-Dicke effect, which is equivalent to the Mössbauer effect. This measurement (Westbrook et al., 1990) marked the start of our interest in what are now called optical lattices: spatially periodic patterns of light-shift-induced potential wells in which atoms are trapped and well localized. It also represents a realization of the 1968 proposal of Letokhov to reduce the Doppler width by trapping atoms in a standing wave.

Joined by Poul Jessen, who was doing his Ph.D. research in our lab, we refined the heterodyne technique and measured the spectrum of Rb atoms in a 1-D laser field like that of Fig. 18(a). Figure 22 shows the results (Jessen *et al.*, 1992), which display well-resolved sidebands around a central, elastic peak. The sidebands are separated from the elastic peak by the frequency of vibration of atoms in the 1-D potential wells. The sideband spectrum can be interpreted as spontaneous Raman scattering, both Stokes and anti-Stokes, involving transitions that begin on a given quantized vibrational level for an atom bound in the optical potential and end

on a higher vibrational level (the lower sideband), the same level (elastic peak) or a lower level (the higher sideband). We did not see sidebands in the earlier experiment in a 3-D, six-beam optical molasses (Westbrook *et al.*, 1990) at least in part because of the lack of phase stability among the laser beams (Grynberg *et al.*, 1993). We have seen well-resolved sidebands in a 3-D, four-beam lattice (Gatzke *et al.*, 1997).

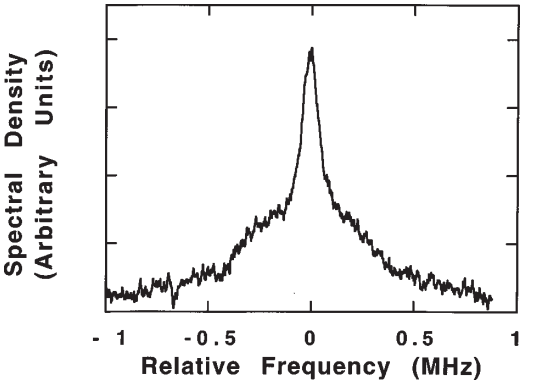

FIG. 21. Heterodyne spectrum of fluorescence from Na atoms in optical molasses. The broad component corresponds to a temperature of 84 μK, which compares well with the temperature of 87 μK measured by TOF. The narrow component indicates a sub-wavelength localization of the atoms.

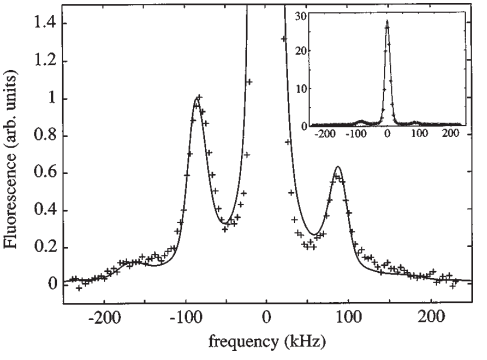

FIG. 22. Vertical expansion of the spectrum emitted by Rb atoms in a 1-D optical lattice. The crosses are the data of Jessen *et al.* (1992); the curve is a first-principles calculation of the spectrum (Marte *et al.*, 1993). The calculation has no adjustable parameters other than an instrumental broadening. Inset: unexpanded spectrum.

The spectrum of Fig. 22 gives much information about the trapping of atoms in the potential wells. The ratio of sideband intensity to elastic peak intensity gives the degree of localization, the ratio of the two sideband intensities gives the temperature, and the spacing of the sidebands gives the potential well depth. Similar, but in many respects complementary, information can be obtained from the absorption spectrum of such an optical lattice, as illustrated by the experiments performed earlier in Paris (Verkerk *et al.*, 1992). The spectrum of Fig. 22 can be calculated from first principles (Marte *et al.*, 1993) and the comparison of the experimental and theoretical spectra shown provides one of the most detailed confirmations of our ability to predict theoretically the behavior of laser cooled atoms.

In our laboratory, we have continued our studies of optical lattices, using adiabatic expansion to achieve temperatures as low as 700 nK (Kastberg *et al.*, 1995), applying Bragg scattering to study the dynamics of atomic motion (Birkl *et al.*, 1995; Phillips, 1997; Raithel, Birkl, Kastberg *et al.*, 1997; Raithel, Birkl, Phillips, and Rolston, 1997), and extending heterodyne spectral measurements to 3-D (Gatzke *et al.*, 1997). The Paris group has also continued to perform a wide range of experiments on optical lattices (Louis *et al.*, 1993; Meacher *et al.*, 1994; Verkerk *et al.*, 1994; Meacher *et al.*, 1995), as have a number of other groups all over the world.

The optical lattice work has emphasized that a typical atom is quite well localized within its potential well, implying a physical picture rather different from the Sisyphus cooling of Fig. 18, where atoms move from one well to the next. Although numerical calculations give results in excellent agreement with experiment in the case of lattice-trapped atoms, a physical picture with the simplicity and power of the original Sisyphus picture has not yet emerged. Nevertheless, the simplicity of the experimental behavior makes one think that such a picture should exist and remains to be found. The work of Castin (1992) and Castin *et al.* (1994) may point the way to such an understanding.

Conclusion

I have told only a part of the story of laser cooling and trapping at NIST in Gaithersburg, and I have left out most of the work that has been done in other laboratories throughout the world. I have told this story from my personal vantage point as an experimentalist in Gaithersburg, as I saw it unfold. The reader will get a much more complete picture by also reading the Nobel lectures of Steve Chu and Claude Cohen-Tannoudji. For the work in my lab, I have tried to follow the thread that leads from laser deceleration and cooling of atomic beams (Phillips and Metcalf, 1982; Prodan *et al.*, 1982; Phillips and Prodan, 1984; Prodan *et al.*, 1985) to magnetic trapping (Migdall *et al.*, 1985), the discovery of sub-Doppler cooling (Lett *et al.*, 1988; Lett *et al.*, 1989), and the beginnings of optical lattice studies (Westbrook *et al.*, 1990; Jessen *et al.*, 1992). Topics such as later studies of lattices, led by Steve Rolston, and collisions of cold atoms, led by Paul Lett, have only been mentioned, and other areas such as the optical tweezer work (Mammen *et al.*, 1996; Helmerson *et al.*, 1997) led by Kris Helmerson have been left out completely.

The story of laser cooling and trapping is still rapidly unfolding, and one of the most active areas of progress is in applications. These include "practical" applications like atomic clocks, atom interferometers, atom lithography, and optical tweezers, as well as "scientific" applications such as collision studies, atomic parity non-conservation, and Bose-Einstein condensation (BEC). (The latter is a particularly beautiful and exciting outgrowth of laser cooling and trapping. Since the 1997 Nobel festivities, our laboratory has joined the growing number of groups having achieved BEC, as shown in Fig. 23.) Most of these applications were completely unanticipated when laser cooling started, and many would have been impossible without the unexpected occurrence of sub-Doppler cooling.

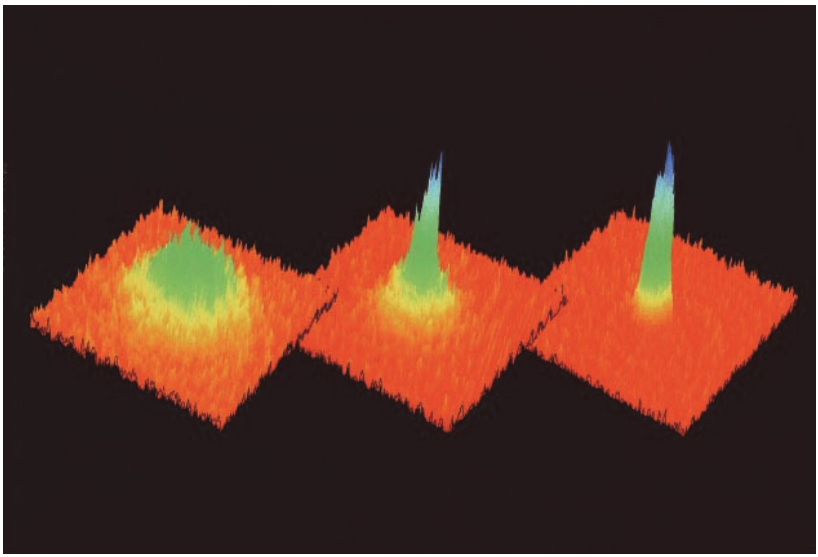

FIG. 23. (Color) One of the most recent applications of laser cooling and magnetic trapping is Bose-Einstein condensation in an atomic vapor. The figure shows a series of representations of the 2-D velocity distribution of a gas of Na atoms at different stages of evaporative cooling through the BEC transition. The velocity distribution changes from a broad thermal one (left) to include a narrow, condensate peak (middle), and finally to be nearly pure condensate (right). The data were obtained in our laboratory in February of 1998, by L. Deng, E. Hagley, K. Helmerson, M. Kozuma, R. Lutwak, Y. Ovchinnikov, S. Rolston, J. Wen and the author. Our procedure was similar to that used in the first such observation of BEC, in Rb, at NIST/JILA in 1995 (Anderson *et al.*, 1995).

Laser cooling and trapping has from its beginnings been motivated by a blend of practical applications and basic curiosity. When I started doing laser cooling, I had firmly in mind that I wanted to make better atomic clocks. On the other hand, the discovery of sub-Doppler cooling came out of a desire to understand better the basic nature of the cooling process. Nevertheless, without sub-Doppler cooling, the present generation of atomic fountain clocks would not have been possible.

I hesitate to predict where the field of laser cooling and trapping will be even a few years from now. Such predictions have often been wrong in the past, and usually too pessimistic. But I firmly believe that progress, both in practical applications and in

basic understanding, will be best achieved through research driven by both aims.

Acknowledgments

I owe a great debt to all of the researchers in the many laboratories around the world who have contributed so much to the field of laser cooling and trapping of neutral atoms. Their friendly competition and generous sharing of understanding and insights has inspired me and educated me in an invaluable way. Very special thanks go to those researchers with whom I have been privileged to work here in Gaithersburg: to Hal Metcalf, who was part of the laser cooling experiments from the start, through most of the work described in this paper; to postdocs John Prodan, Alan Migdall, Phil Gould, Chris Westbrook, and Rich Watts, whose work led our group to the discovery of sub-Doppler cooing, and who moved on to distinguished careers elsewhere; to Paul Lett, Steve Rolston, and Kris Helmerson who also were pivotal figures in the development of laser cooling and trapping in Gaithersburg, who have formed the nucleus of the present Laser Cooling and Trapping Group (and who have graciously provided considerable help in the preparation of this manuscript); and to all the other postdocs, visitors and students who have so enriched our studies here. To all of these, I am thankful, not only for scientific riches but for shared friendship.

I know that I share with Claude Cohen-Tannoudji and with Steve Chu the firm belief that the 1997 Nobel Prize in Physics honors not only the three of us, but all those other researchers in this field who have made laser cooling and trapping such a rewarding and exciting subject. I want to thank NIST for providing and sustaining the intellectual environment and the resources that have nurtured a new field of research and allowed it to grow from a few rudimentary ideas into a major branch of modern physics. I also thank the U.S. Office of Naval Research, which provided crucial support when I and my ideas were unproven, and which continues to provide invaluable support and encouragement.

There are many others, friends, family and teachers who have been of great importance. I thank especially my wife and

daughters who have supported and encouraged me and provided that emotional and spiritual grounding that makes achievement worthwhile. Finally I thank God for providing such a wonderful and intriguing world for us to explore, for allowing me to have the pleasure of learning some new things about it, and for allowing me to do so in the company of such good friends and colleagues.

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Bose-Einstein Condensation in a Dilute Gas, the First 70 Years and Some Recent Experiments[1]

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Abstract

Bose-Einstein condensation, or BEC, has a long and rich history dating from the early 1920s. In this article we will trace briefly over this history and some of the developments in physics that made possible our successful pursuit of BEC in a gas. We will then discuss what was involved in this quest. In this discussion we will go beyond the usual technical description to try and address certain questions that we now hear frequently, but are not covered in our past research papers. These are questions along the lines of: How did you get the idea and decide to pursue it? Did you know it was going to work? How long did it take you and why? We will review some our favorites from among the experiments we have carried out with BEC. There will then be a brief encore on why we are optimistic that BEC can be created with nearly any species of magnetically trappable atom. Throughout this article we will try to explain what makes BEC in a dilute gas so interesting, unique, and experimentally challenging.[2]

THE NOTION OF BOSE STATISTICS dates back to a 1924 paper in which Satyendranath Bose used a statistical argument to derive the blackbody photon spectrum (Bose, 1924). Unable to publish his work, he sent it to Albert Einstein, who translated it into German

[1] The 2001 Nobel Prize in Physics was shared by E. A. Cornell, Wolfgang Ketterle, and C. E. Wieman. Reprinted from *Reviews of Modern Physics*, **74**, 2002.

[2] This article is our ''Nobel Lecture'' and as such takes a relatively personal approach to the story of the development of experimental Bose-Einstein condensation. For a somewhat more scholarly treatment of the history, the interested reader is referred to E. A. Cornell, J. R. Ensher, and C. E. Wieman, ''Experiments in dilute atomic Bose-Einstein condensation in Bose-Einstein Condensation in Atomic Gases,'' *Proceedings of the International School of Physics "Enrico Fermi" Course CXL*, edited by M. Inguscio, S. Stringari, and C. E. Wieman (Italian Physical Society, 1999), pp. 15–66, which is also available as cond-mat/9903109. For a reasonably complete technical review of the three years of explosive progress that immediately followed the first observation of BEC, we recommend reading the above article in combination with the corresponding review from Ketterle, cond-mat/9904034.

and got it published. Einstein then extended the idea of Bose's counting statistics to the case of non-interacting atoms (Einstein, 1924, 1925). The result was Bose-Einstein statistics. Einstein immediately noticed a peculiar feature of the distribution of the atoms over the quantized energy levels predicted by these statistics. At very low but finite temperature a large fraction of the atoms would go into the lowest energy quantum state. In his words, "A separation is effected; one part condenses, the rest remains a saturated ideal gas" [3] (Einstein, 1925). This phenomenon we now know as Bose-Einstein condensation. The condition for this to happen is that the phase-space density must be greater than approximately unity, in natural units. Another way to express this is that the de Broglie wavelength, λ_{dB}, of each atom must be large enough to overlap with its neighbor, or more precisely, $n\lambda_{dB}^3 > 2.61$.

This prediction was not taken terribly seriously, even by Einstein himself, until Fritz London (1938) and Laszlo Tisza (1938) resurrected the idea in the mid-1930s as a possible mechanism underlying superfluidity in liquid helium 4. Their work was the first to bring out the idea of BEC displaying quantum behavior on a macroscopic size scale, the primary reason for much of its current attraction. Although it was a source of debate for decades, it is now recognized that the remarkable properties of superconductivity and superfluidity in both helium 3 and helium 4 are related to BEC, even though these systems are very different from the ideal gas considered by Einstein.

The appeal of the exotic behavior of superconductivity and of superfluidity, along with that of laser light, the third common system in which macroscopic quantum behavior is evident, provided much of our motivation in 1990 when we decided to pursue BEC in a gas. These three systems all have fascinating counterintuitive behavior arising from macroscopic occupation of a single quantum state. Any physicist would consider these phenomena among the most remarkable topics in physics. In 1990 we were confident that the addition of a new member to the family would constitute a major

[3] English translation of Einstein's quotes and the historical interpretation are from Pais (1982), *Subtle is the Lord . . .*

contribution to physics. (Only after we succeeded did we realize that the discovery of each of the original Macroscopic Three had been recognized with a Nobel Prize, and we are grateful that this trend has continued!) Although BEC shares the same underlying mechanism with these other systems, it seemed to us that the properties of BEC in a gas would be quite distinct. It is far more dilute and weakly interacting than liquid helium superfluids, for example, but far more strongly interacting than the non-interacting light in a laser beam. Perhaps BEC's most distinctive feature (and this was not something we sufficiently appreciated, in 1990) is the ease with which its quantum wave function may be directly observed and manipulated. While neither of us was to read C. E. Hecht's prescient 1959 paper (Hecht, 1959) until well after we had observed BEC, we surely would have taken his concluding paragraph as our marching orders:

> The suppositions of this note rest on the possibility of securing, say by atomic beam techniques, substantial quantities of electron-spin-oriented H, T and D atoms. Although the experimental difficulties would be great and the relaxation behavior of such spin-oriented atoms essentially unknown, *the possibility of opening a rich new field for the study of superfluid properties in both liquid and gaseous states would seem to demand the expenditure of maximum experimental effort*.[4]

In any case, by 1990 we were awash in motivation. But this motivation would not have carried us far, had we not been able to take advantage of some key recent advances in science and technology, in particular, the progress in laser cooling and trapping and the extensive achievements of the spin-polarized-hydrogen community.

However, before launching into that story, it is perhaps worthwhile to reflect on just how exotic a system of indistinguishable particles truly is, and why BEC in a gas is such a daunting experimental challenge. It is easy at first to accept that two atoms can be so similar one to the other as to allow no possibility of telling them

[4] Emphasis ours.

apart. However, confronting the physical implications of the concept of indistinguishable bosons can be troubling. For example, if there are ten bosonic particles to be arranged in two microstates of a system, the statistical eight of the configuration with ten particles in one state and zero in the other is exactly the same as the weight of the configuration with five particles in one state, five in the other. This 1:1 ratio of statistical weights is very counterintuitive and rather disquieting. The corresponding ratio for distinguishable objects, such as socks in drawers that we observe every day, is 1:252, profoundly different from 1:1. In the second of Einstein's two papers (Einstein, 1925; Pais, 1982) on Bose- Einstein statistics, Einstein comments that "The... molecules are not treated as statistically independent...", and the differences between distinguishable and indistinguishable state counting ". . . express indirectly a certain hypothesis on a mutual influence of the molecules which for the time being is of a quite mysterious nature." This mutual influence is no less mysterious today, even though we can readily observe the variety of exotic behavior it causes such as the well-known enhanced probability for scattering into occupied states and, of course, Bose-Einstein condensation.

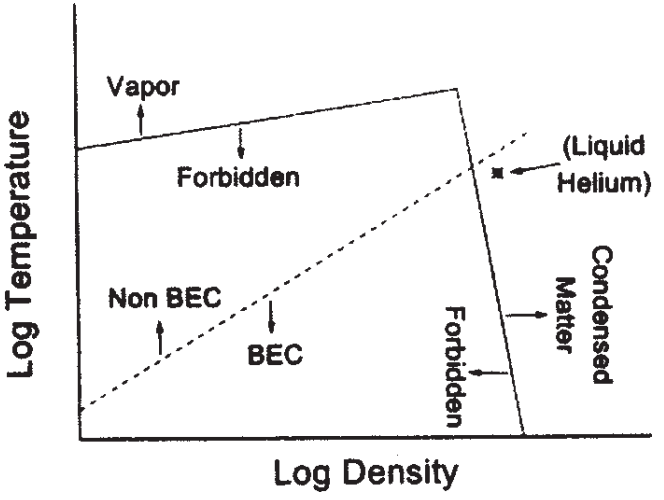

FIG. 1. Generic phase diagram common to all atoms: dotted line, the boundary between non-BEC and BEC; solid line, the boundary between allowed and forbidden regions of the temperature-density space. Note that at low and intermediate densities, BEC exists only in the thermodynamically forbidden regime.

Not only does the Bose-Einstein phase transition offend our sensibilities as to how particles ought best to distribute themselves, it also runs counter to an unspoken assumption that a phase transition somehow involves thermodynamic stability. In fact, the regions immediately above and immediately below the transition in dilute-gas experiments are both deep in the thermodynamically forbidden regime. This point is best made by considering a qualitative phase diagram (Fig. 1), which shows the general features common to any atomic system. At low density and high temperature, there is a vapor phase. At high density there are various condensed phases. But the intermediate densities are thermodynamically forbidden, except at very high temperatures. The Bose-condensed region of the n-T plane is utterly forbidden, except at such high densities that (with one exception) all known atoms or molecules would form a crystalline lattice, which would rule out Bose condensation. The single exception, helium, remains a liquid below the BEC transition. However, reaching BEC under dilute conditions (say, at densities 10 or 100 times lower than conventional liquid helium) is as thermodynamically forbidden to helium as it is to any other atom.

Of course, forbidden is not the same as impossible; indeed, to paraphrase an old Joseph Heller joke, if it were really impossible, they wouldn't have bothered to forbid it. It comes down in the end to differing time scales for different sorts of equilibrium. A gas of atoms can come into kinetic equilibrium via two-body collisions, whereas it requires three-body collisions to achieve chemical equilibrium (*i.e.*, to form molecules and thence solids). At sufficiently low densities, the two- body rate will dominate the three-body rate, and a gas will reach kinetic equilibrium, perhaps in a metastable Bose-Einstein condensate, long before the gas finds its way to the ultimately stable solid-state condition. The need to maintain metastability usually dictates a more stringent upper limit on density than does the desire to create a dilute system. Densities around 10^{20} cm^{-3}, for instance, would be a hundred times more dilute than a condensed-matter helium superfluid. But creating such a gas is quite impractical even at an additional factor of 1000 lower density, say 10^{17} cm^{-3}, when

metastability times would be on the order of a few microseconds more realistic are densities on the order of 10^{14} cm^{-3}. The low densities mandated by the need to maintain long-lived metastability in turn make necessary the achievement of still lower temperatures if one is to reach BEC.

Thus the great experimental hurdle that must be overcome to create BEC in a dilute gas is to form and keep a sample that is so deeply forbidden. Since our subsequent discussion will focus only on BEC in dilute gases, we shall refer to this simply as BEC in the sections below and avoid endlessly repeating "in a dilute gas."

Efforts to make a dilute BEC in an atomic gas were sparked by Stwalley and Nosanow (1976). They argued that spin-polarized hydrogen had no bound states and hence would remain a gas down to zero temperature, and so it would be a good candidate for BEC. This stimulated a number of experimental groups (Silvera and Walraven, 1980; Hardy *et al.*, 1982; Hess *et al.*, 1983; Johnson *et al.*, 1984) in the late 1970s and early 1980s to begin pursuing this idea using traditional cryogenics to cool a sample of polarized hydrogen. Spin-polarized hydrogen was first stabilized by Silvera and Walraven in 1980, and by the mid-1980s spin-polarized hydrogen had been brought within a factor of 50 of condensing (Hess *et al.*, 1983). These experiments were performed in a dilution refrigerator, in a cell in which the walls were coated with superfluid liquid helium as a nonstick coating for the hydrogen. The hydrogen gas was compressed using a piston-in-cylinder arrangement (Bell *et al.*, 1986) or inside a helium bubble (Sprik *et al.*, 1985). These attempts failed, however, because when the cell was made very cold the hydrogen stuck to the helium surface and recombined. When one tried to avoid that problem by warming the cell sufficiently to prevent sticking, the density required to reach BEC was correspondingly increased, which led to another problem. The requisite densities could not be reached because the rate of three- body recombination of atoms into hydrogen molecules goes up rapidly with density and the resulting loss of atoms limited the density (Hess, 1986).

Stymied by these problems, Harold Hess (Hess, 1986) from the MIT hydrogen group realized that magnetic trapping of atoms (Migdall

et al., 1985; Bagnato et al., 1987) would be an improvement over a cell. Atoms in a magnetic trap have no contact with a physical surface and thus the surface-recombination problem could be circumvented. Moreover, thermally isolated atoms in a magnetic trap would allow cooling by evaporation to far lower temperatures than previously obtained. In a remarkable paper, Hess (1986) laid out most of the important concepts of evaporative cooling of trapped atoms for the attainment of BEC. Let the highest-energy atoms escape from the trap, and the mean energy, and thus the temperature, of the remaining atoms will decrease. For a dilute gas in an inhomogeneous potential, decreasing the temperature will decrease the occupied volume. One can thus actually increase the density of the remaining atoms by removing atoms from the sample. The all-important (for BEC) phase-space density is dramatically increased as this happens because density is rising while temperature is decreasing. The Cornell University hydrogen group also considered evaporative cooling (Lovelace *et al.*, 1985). By 1988 the MIT group had demonstrated these virtues of evaporative cooling of magnetically trapped spin-polarized hydrogen. By 1991 they obtained, at a temperature of 100 °K, a density that was only a factor of 5 below BEC (Doyle, 1991a). Further progress was limited by dipolar relaxation, but perhaps more fundamentally by loss of signal-to-noise, and the difficulty of measuring the characteristics of the coldest and smallest clouds (Doyle, 1991b). Evaporative work was also performed by the Amsterdam group (Luiten *et al.,* 1993).

At roughly the same time, but independent from the hydrogen work, an entirely different type of cold-atom physics and technology was being developed. Laser cooling and trapping has been reviewed elsewhere (Arimondo *et al*., 1991; Chu, 1998; Cohen-Tannoudji, 1998; Phillips, 1998), but here we mention some of the highlights most relevant to our work. The idea that laser light could be used to cool atoms was suggested in early papers by Wineland and Dehmelt (1975), by Hänsch and Schawlow (1975), and by Letokhov's group (Letokhov, 1968). Early optical force experiments were performed by Ashkin (Bjorkholm *et al*., 1978). Trapped ions were laser-cooled at the University of Washington (Neuhauser *et al.*, 1978) and at the National Bureau of Standards (now NIST) in Boulder (Wineland *et*

al., 1978). Atomic beams were deflected and slowed in the early 1980s (Andreev *et al.*, 1981; Ertmer *et al.*, 1985; Prodan *et al.*, 1985). Optical molasses, where the atoms are cooled to very low temperatures by six perpendicular intersecting laser beams, was first studied at Bell Labs (Chu *et al.*, 1985). Measured temperatures in the early molasses experiments were consistent with the so-called Doppler limit, which amounts to a few hundred microkelvin in most alkalis. Light was first used to hold (trap) atoms using the dipole force exerted by a strongly focused laser beam (Chu *et al.*, 1986). In 1987 and 1988 there were two major advances that became central features of the method of creating BEC. First, a practical spontaneous-force trap, the magneto-optical trap (MOT) was demonstrated (Raab *et al.*, 1987); and second, it was observed that under certain conditions, the temperatures in optical molasses are in fact much colder than the Doppler limit (Lett *et al.*, 1988; Chu *et al.*, 1989; Dalibard *et al.*, 1989). The MOT had the essential elements needed for a widely useful optical trap: it required relatively modest amounts of laser power, it was much deeper than dipole traps, and it could capture and hold relatively large numbers of atoms. These were heady times in the laser-cooling business. With experiment yielding temperatures mysteriously far below what theory would predict, it was clear that we all lived under the authority of a munificent God.

During the mid-1980s one of us (Carl) began investigating how useful the technology of laser trapping and cooling could become for general use in atomic physics. Originally this took the form of just making it cheaper and simpler by replacing the expensive dye lasers with vastly cheaper semiconductor lasers, and then searching for ways to allow atom trapping with these low-cost but also low-power lasers (Pritchard *et al.*, 1986; Watts and Wieman, 1986). With the demonstration of the MOT and sub-Doppler molasses Carl's group began eagerly studying what physics was limiting the coldness and denseness of these trapped atoms, with the hope of extending the limits further. They discovered that several atomic processes were responsible for these limits. Light-assisted collisions were found to be the major loss process from the MOT as the density increased (Sesko *et al.*, 1989). However, even before that became a

serious problem, the light pressure from reradiated photons limited the density (Walker *et al.*, 1990; Sesko *et al.*, 1991). At about the same time, the sub-Doppler temperatures of molasses found by Phillips, Chu, and Cohen-Tannoudji were shown to be due to a combination of light-shifts and optical pumping that became known as Sysiphus cooling (Dalibard and Cohen-Tannoudji, 1989). Random momentum fluctuations from the scattered photons limit the ultimate temperature to about a factor of 10 above the recoil limit. In larger samples, the minimum temperature was higher yet, because of the multiple scattering of the photons. While carrying out studies on the density limits of MOT's Carl's group also continued the effort in technology development. This resulted in the creation of a useful MOT in a simple glass vapor cell (Monroe *et al.*, 1990), thereby eliminating the substantial vacuum chamber required for the slowed atomic beam loading that had previously been used.

Seeking to take advantage of the large gains in phase-space density provided by the MOT while avoiding the limitations imposed by the undesirable effects of photons, Carl and his student Chris Monroe decided to try loading the cold MOT atoms into a magnetic trap (Monroe *et al.*, 1990; see Fig. 2). This worked remarkably well. Because further cooling could be carried out as the atoms were transferred between optical and magnetic trap it was possible to get very cold samples, the coldest that had been produced at that time. More importantly, these were not optical molasses samples that were quickly disappearing but rather magnetically trapped samples that could be held and studied for extended periods. These samples were about a hundred times colder than any previous trapped atom samples, with a correspondingly increased phase-space density. This was a satisfying achievement, but as much as the result itself, it was the relative simplicity of the apparatus required that inspired us (including now Eric Cornell, who joined the project as a postdoc in 1990) to see just how far we could push this marriage of laser cooling and trapping and magnetic trapping.

Previous laser traps involved expensive massive laser systems and large vacuum chambers for atomic beam precooling. Previous magnetic traps for atoms were usually (Bagnato *et al.*, 1987; Doyle,

1991) extremely complex and bulky (often with superconducting coils) because of the need to have sufficiently large depths and strong confinement. Laser traps and magnetic traps were both somewhat heroic experiments individually, to be undertaken only by a select handful of well-equipped AMO laboratories. The prospect of trying to get both traps working, and working well, in the same room and on the same day, was daunting. However, in the first JILA magnetic trap experiment our laser sources were simple diode lasers, the vacuum system was a small glass vapor cell, and the magnetic trap was just a few turns of wire wrapped around it. This magnetic field was adequate because of the low temperatures of the laser-cooled and trapped samples. Being able to produce such cold and trapped samples in this manner encouraged one to fantasize wildly about possible things to do with such an atom sample. Inspired by the spin-polarized hydrogen work, our fantasizing quickly turned to the idea of evaporative cooling further to reach BEC. It would require us to increase the phase-space density by 5 orders of magnitude, but since we had just gained about 15 orders of magnitude almost for free with the vapor cell MOT, this did not seem so daunting.

The JILA vapor-cell MOT (Fig. 3), with its superimposed ion pump trap, introduced a number of ideas that are now in common use in the hybrid trapping business (Monroe *et al.*, 1990; Monroe, 1992): (i) Vapor-cell (rather than beam) loading, (ii) fused-glass rather than welded-steel architecture, (iii) extensive use of diode lasers, (iv) magnetic coils located outside the chamber, (v) overall chamber volume measured in cubic centimeters rather than liters, (vi) temperatures measured by imaging an expanded cloud, (vii) magnetic-field curvatures calibrated *in situ* by observing the frequency of dipole and quadrupole (sloshing and pulsing) cloud motion, (viii) the basic approach of a MOT and a magnetic trap which are spatially superimposed (indeed, which often share some magnetic coils) but temporally sequential, and (ix) optional use of additional molasses and optical pumping sequences inserted in time between the MOT and magnetic trapping stages. It is instructive to note how a modern, Ioffe-Pritchard-based BEC device (Fig. 4) resembles its ancestor (Fig. 3).

As we began to think about applying the technique of evaporative cooling with hydrogen to our very cold alkali atoms we looked carefully at the hydrogen work and its lessons. When viewed from our 1990 perspective the previous decade of work on polarized hydrogen provided a number of important insights. It was clear that the unique absence of any bound states for spin- polarized hydrogen was actually not an important issue (other than its being the catalyst for starting the entire field, of course!). Bound states or not, a very cold sample of spin-polarized hydrogen, like every other gas, has a lower-energy state to which it can go, and its survival depends on the preservation of metastability. For hydrogen the lower-energy state is a solid, although from an experimental point of view the rate-limiting process is the formation of diatomic molecules (with appropriately reoriented spins). Given that all atomic gases are only metastable at the BEC transition point, the real experimental issue becomes: How well can one preserve the requisite metastability while still cooling sufficiently far to reach BEC?

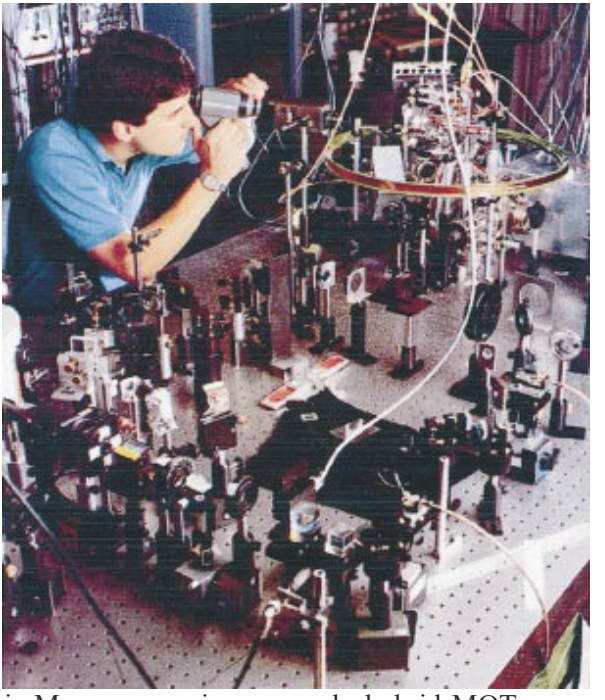

FIG. 2. Chris Monroe examines an early hybrid MOT- magnetic trap apparatus [Color].

The realization that metastability was the key experimental challenge one should focus on was probably at least as important to the attainment of BEC as any of the experimental techniques we subsequently developed to actually achieve it. The work on hydrogen provided an essential guide for evaluating and tackling this challenge. It provided us with a potential cooling technique (evaporative cooling of magnetically trapped atoms) and mapped out many of the processes by which a magnetically trapped atom can be lost from its metastable state.

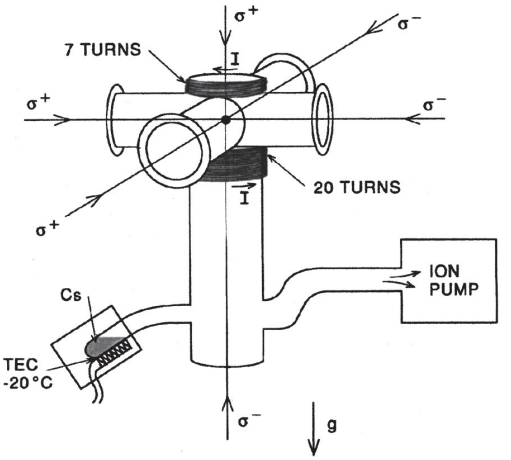

FIG. 3. The glass vapor cell and magnetic coils used in early JILA efforts to hybridize laser cooling and magnetic trapping (see Monroe *et al.*, 1990). The glass tubing is 2.5 cm in diameter. The Ioffe current bars have been omitted for clarity.

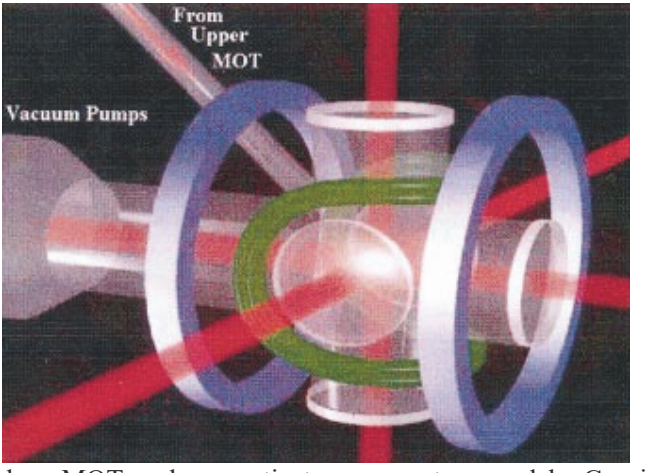

FIG. 4. Modern MOT and magnetic trap apparatus, used by Cornish *et al.*, 2000 [Color].

The hydrogen work made it clear that it was all an issue of good versus bad collisions. The good collisions are elastic collisions that re-thermalize the atoms during evaporation. The more collisions there are, the more quickly and efficiently one can cool. The bad collisions are the inelastic collisions that quench the metastability. Hydrogen had already shown that three-body recombination collisions and dipole spin-flip collisions were the major inelastic culprits. The fact that hydrogen researchers were fairly close to reaching BEC was also a strong encouragement. It meant that the goal was not ridiculously distant and that one only had to do a little better in the proportion of good to bad collisions to succeed.

The more we thought about this, the more we began to suspect that our heavy alkali atoms would likely have more favorable collision properties than hydrogen atoms and thus have a good chance of success. Although knowledge of the relevant collision cross sections was totally nonexistent at that time, we were able to come up with arguments for how the cross sections might scale relative to hydrogen. These are discussed in more detail below in the section discussing why collisional concerns make it likely that BEC can be created in a large number of different species. Here we will just give a brief summary consistent with our views circa 1990. The dipole spin-flip collisions that limited hydrogen involve spin-spin interactions and thus could be expected to be similar for the alkalis and for hydrogen because the magnetic moments are all about the same. The good collisions needed for evaporative cooling, however, should be much larger for heavy alkalis with their fat fluffy electron clouds than for hydrogen. The other villain of the hydrogen effort, three-body recombination, was a total mystery, but because it goes as density cubed while the good elastic collisions go as density squared, it seemed as if we should always be able to find a sufficiently low-density and low-temperature regime to avoid it (see Monroe, 1992).

As a minor historical note, we might point out that during these considerations we happily ignored the fact that the temperatures required to achieve BEC in a heavy alkali gas are far colder than those needed for the same density of hydrogen. The

critical temperature for ideal-gas BEC is inversely proportional to the mass. It was clear that we would need to cool to well under a microkelvin, and a large three-body recombination rate would have required us to go to possibly far lower temperatures. To someone coming from a traditional cryogenics background this would (and probably did) seem like sheer folly. The hydrogen work had been pushing hard for some years at the state of the art in cryogenic technology, and here we proposed to happily jump far beyond that. Fortunately we were coming to this from an AMO background in a time when temperatures achieved by laser cooling were dropping through the floor. Optimism was in the air. In fact, we later discovered optimism can take one only so far: There were actually considerable experimental difficulties, and further cooling came at some considerable effort and a five-year delay. Nevertheless, it is remarkable that with evaporative cooling a magnetically trapped sample of atoms, surrounded on all sides by a 300-K glass cell, can be cooled to reach temperatures of only a few nanokelvin, and moreover it looks quite feasible to reach even colder temperatures.

General collisional considerations gave us some hope that the evaporative cooling hybrid trap approach with alkali atoms would get us to BEC, or, if not, at least reveal some interesting new physics that would prevent it. Nonetheless, there were powerful arguments against pursuing this. First, our 1990-era arguments in favor of it were based on some very fuzzy intuition; there were no collision data or theories to back it up and there were strong voices in disagreement. Second, the hydrogen experiments seemed to be on the verge of reaching BEC, and in fact we thought it was likely that if BEC could be achieved they would succeed first. However, our belief in the virtues of our technology really carried the day in convincing us to proceed. With convenient lasers in the near-IR, and with the good optical access of a room- temperature glass cell, detection sensitivity could approach single-atom capability. We could take pictures of only a few thousand trapped atoms and immediately know the energy and density distribution. If we wanted to modify our magnetic trap it only required a few hours winding and installing a new coil of wires. This was a dramatic contrast with the hydrogen experiments that, like all state-of-the-art cryogenics

experiments, required an apparatus that was the better part of two stories, and the time to modify it was measured in (large) fractions of a year. Also, atomic hydrogen was much more difficult to detect and so the diagnostics were far more limited. This convinced us that although hydrogen would likely succeed first, our hybrid trap approach with easily observed and manipulated alkali samples would be able to carry out important science and so was well worth pursuing in its own right.

From the very beginning in 1990, our work on BEC was heavily involved with cold atomic collisions. This was somewhat ironic since previously both of us had actively avoided the large fraction of AMO work on the subject of atomic collisions. Atomic collisions at very cold temperatures is now a major branch of the discipline of AMO physics, but at the end of the 1980s there were almost no experimental data, and what there was came in fact from the spin-polarized hydrogen experiments (Gillaspy *et al.*, 1989). There was theoretical work on hydrogen from Shlyapnikov and Kagan (Kagan *et al.*, 1981, 1984), and from Silvera and Verhaar (Lagendijk *et al.*, 1986). An early paper by Pritchard (1986) includes estimates on low-temperature collisional properties for alkalis. His estimates were extrapolations from room- temperature results, but in retrospect, several were surprisingly accurate. As we began to work on evaporative cooling, much of our effort was devoted to determining the sizes of all the relevant good and bad collision cross sections. Our efforts were helped by the theoretical efforts of Boudewijn Verhaar, who was among the first to take our efforts seriously and attempt to calculate the rates in question. Chris Greene also provided us with some useful theoretical estimates.

Starting in 1990 we carried out a series of experiments exploring various magnetic traps and measuring the relevant collision cross sections. As this work proceeded we developed a far better understanding of the conditions necessary for evaporative cooling and a much clearer understanding of the relevant collisional issues (Monroe *et al.*, 1993; Newbury *et al.*, 1995). Our experimental concerns evolved accordingly. In the early experiments (Monroe *et al.*, 1990, 1993; Cornell *et al.*, 1991; Monroe, 1992) a number of issues came up that

continue to confront all BEC experiments: the importance of aligning the centers of the MOT and the magnetic trap, the density-reducing effects of mode-mismatch, the need to account carefully for the (previously ignored) force of gravity, heating (and not merely loss) from background gas collisions, the usefulness of being able to turn off the magnetic fields rapidly, the need to synchronize many changes in laser status and magnetic fields together with image acquisition, an appreciation for the many issues that can interfere with accurate determinations of density and temperature by optical methods, either florescence or absorption imaging, and careful stabilization of magnetic fields. The mastery of these issues in these early days made it possible for us to proceed relatively quickly to quantitative measurements with the BEC once we had it.

In 1992 we came to realize that dipolar relaxation in alkalis should in principle not be a limiting factor. As explained in the final section of this article, collisional scaling with temperature and magnetic field is such that, except in pathological situations, the problem of good and bad collisions in the evaporative cooling of alkalis is reduced to the ratio of the elastic collision rate to the rate of loss due to imperfect vacuum; dipolar relaxation and three-body recombination can be finessed, particularly since our preliminary data showed they were not enormous. It was reassuring to move ahead on efforts to evaporate with the knowledge that, while we were essentially proceeding in the dark, there were not as many monsters in the dark as we had originally imagined.

It rapidly became clear that the primary concerns would be having sufficient elastic collision rate in the magnetic trap and sufficiently low background pressure to have few background collisions that removed atoms from the trap. To accomplish this it was clear that we needed higher densities in the magnetic trap than we were getting from the MOT. Our first effort to increase the density two years earlier was based on a multiple- loading scheme (Cornell *et al.*, 1991). Multiple MOT- loads of atoms were launched in moving molasses, optically pumped into an untrapped Zeeman level, focused into a magnetic trap, then optically re-pumped into a trapped level. The re-pumping represented the necessary dissipation, so that multiple

loads of atoms could be inserted in a continuously operating magnetic trap. In practice, each step of the process involved some losses, and the final result was disappointing. Later, however, as discussed below, we resurrected the idea of multiple loading from one MOT to another to good advantage (Gibble *et al.*, 1995; Myatt *et al.*, 1996). This is now a technique currently in widespread practice.

In addition to building up the initial density we realized that the collision rate could be dramatically increased by, after loading into a magnetic trap, compressing the atoms by further increasing the curvature of the confining magnetic fields. In a harmonic trap, the collision rate after adiabatic compression scales as the final confining frequency squared (Monroe, 1992). This method is discussed by Monroe (1992) and was implemented first in early ground-state collisional work (Monroe *et al.*, 1993).

In fall of 1992, Eric's postdoctoral appointment concluded, and, after a tour through the job market, he decided to take the equivalent of an assistant professor position at JILA/NIST. He decided to use his startup money to build a new experimental apparatus that would be designed to put these ideas together to make sure evaporation worked as we expected. Meanwhile, we continued to pursue the possibility of enhanced collision cross sections in cesium using a Feshbach resonance. At that point our Monte Carlo simulations said that a ratio of about 150 elastic collisions per trap lifetime was required to achieve runaway evaporation. This is the condition where the elastic collision rate would continue to increase as the temperature decreased, and hence evaporation would continue to improve as the temperature was reduced. We also had reasonable determinations of the elastic collision cross sections.

So the plan was to build a simple quadrupole trap that would allow very strong squeezing to greatly enhance the collision rate, combined with a good vacuum system in order to make sure evaporative cooling worked as expected. Clearly, there was much to be gained by building a more tightly confining magnetic trap, but the requirement of adequate optical access for the MOT, along with engineering constraints on power dissipation, made the design problem complicated.

When constructing a trap for weak-field-seeking atoms, with the aim of confining the atoms to a spatial size much smaller than the size of the magnets, one would like to use linear gradients. In that case, however, one is confronted with the problem of the minimum in the magnitude of the magnetic fields (and thus of the confining potential) occurring at a local zero in the magnetic field. This zero represents a hole in the trap, a site at which atoms can undergo Majorana transitions (Majorana, 1931) and thus escape from the trap. If one uses the second-order gradients from the magnets to provide the confinement, there is a marked loss of confinement strength. This scaling is discussed by Petrich *et al.* (1995). We knew that once the atoms became cold enough they would leak out the hole in the bottom of the trap, but the plan was to go ahead and get evaporation and worry about the hole later. We also recognized that even with successful evaporative cooling, and presuming we could solve the issue of the hole in the quadrupole trap, there was still the question of the sign of scattering length, which must be positive to ensure the stability of a large condensate.

In setting up the new apparatus Eric chose to use rubidium. Given the modulo arithmetic that goes into determining a scattering length, it seemed fair to treat the scattering lengths of different isotopes as statistically independent events, and rubidium with its two stable isotopes offered two rolls of the dice for the same laser system. Eric then purchased a set of diode lasers for the rubidium wavelength, but of course we kept the original cesium-tuned diode lasers. The wavelengths of cesium and of the two rubidium isotopes are sufficiently similar that in most cases one can use the same optics. Thus we preserved the option of converting from one species to another in a matter of weeks. The chances then of Nature's conspiring to make the scattering length negative, for both hyperfine levels, for all three atoms, seemed very small.

Progress in cold collisions, particularly the experiment and theory of photo-associative collisions, had moved forward so rapidly that by the time we had evaporatively cooled rubidium to close to BEC temperatures a couple of years later there existed, at the 20% level, values for several of the elastic scattering lengths. In

particular, we knew that it was positive for the 2,2 state of Rb-87 (Thorsheim *et al.*, 1987; Lett *et al.*, 1993; Miller *et al.*, 1993; Abraham *et al.*, 1995; Gardner *et al.*, 1995; McAlexander *et al.*, 1995).

Our original idea for the quadrupole trap experiment was to pulse a burst of rubidium into our cell, where we would catch a large sample in the MOT and then hold it as the residual rubidium was quickly pumped away, leaving a long trap lifetime. We, particularly Eric's postdoc, Mike Anderson, spent many frustrating months discovering how difficult this seemingly simple idea was to actually implement in practice. The manner in which rubidium interacted with glass and stainless-steel surfaces conspired to make this so difficult we finally gave up. We ended up going with a far-from-optimum situation of working with extremely low rubidium pressure and doing our best at maximizing the number of atoms captured in the MOT from this feeble vapor and enhancing the collision rate for those relatively few atoms as much as possible. We recognized that this was a major compromise, but we had been trying to evaporate for some time, and we were getting impatient! We had no stomach for building another apparatus just to see evaporation. Fortunately we were able to find two key elements to enhance the MOT loading and density. First was the use of a dark-spot MOT in which there is a hole in the center of the MOT beams so the atoms are not excited. This technique had been demonstrated by Ketterle (Ketterle *et al.*, 1993) as a way to greatly enhance the density of atoms in a MOT under conditions of a very high loading rate. The number of atoms we could load in our vapor cell MOT with very low rubidium vapor was determined by the loading rate over the loss rate. In this case the loss rate was the photo-associative collisions we had long before found to be important for losses from MOT's. The dark-spot geometry reduced this two-body photo-associative loss in part because in our conditions it *reduced* the density of atoms in the MOT (Anderson *et al.*, 1994).

Using this approach we were able to obtain 10^8 atoms in the MOT collected out of a very low vapor background (so that magnetic trap lifetime was greater than 100 s). The second key element was the invention of the compressed MOT (CMOT), a

technique for substantially enhancing the density of atoms in the MOT on a transient basis. For the CMOT, the MOT was filled and then the field gradient and laser detuning were suddenly changed to greatly suppress the multiple photon scattering. This produced much higher densities and clouds whose shape was a much better match to the desired shape of the cloud in the magnetic trap. This was a very transient effect because the losses from the MOT were much larger under these conditions, but that was not important; the atoms needed only to be held for the milliseconds required before they were transferred to the magnetic trap (Petrich *et al.*, 1994; see Fig. 5). With these improvements and a quadrupole trap that provided substantial squeezing, we were able to finally demonstrate evaporative cooling in rubidium.

Cooling by evaporation is a process found throughout Nature. Whether the material being cooled is an atomic nucleus or the Atlantic Ocean, the rate of natural evaporation and the minimum temperature achievable are limited by the particular fixed value of the work function of the evaporating substance. In magnetically confined atoms, no such limit exists, because the work function is simply the height of the lowest point in the rim of the confining potential. Hess (1986) pointed out that, by perturbing the confining magnetic fields, one could make the work function of a trap arbitrarily low; as long as favorable collisional conditions persist there is no lower limit to the temperatures attainable in this forced evaporation.

Pritchard (Pritchard *et al.*, 1989) pointed out that evaporation could be performed more conveniently if the rim of the trap were defined by an rf-resonance condition, rather than simply by the topography of the magnetic field; experimentally, his group made first use of position-dependent rf transitions to selectively transfer magnetically trapped sodium atoms between Zeeman levels and thus characterized their temperature (Martin *et al.*, 1988). In our experiment we used Pritchard's technique of an rf field to selectively evaporate.

It was a great relief to see evaporative cooling of laser precooled, magnetically trapped atoms finally work, as we had been

anticipating it would for so many years. Unfortunately, it worked exactly as well, but no better, than we had anticipated. The atoms were cooled to about 40 μK and then disappeared, at just the temperature we had estimated they would be lost, through the hole in the bottom of the quadrupole trap. Eric came up with an idea that solved this problem. It was a design for a new type of trap that required relatively little modification to the apparatus and so was quickly implemented. This was the Time Orbiting Potential (TOP) trap in which a small rotating magnetic field was added to the quadrupole field (Petrich *et al.*, 1995). This moved the field zero in an orbit faster than the atoms could follow. It was the perfect solution to our problem.

Mike Anderson, another postdoc, Wolfgang Petrich, and graduate student Jason Ensher quickly implemented this design. Their efforts were spurred on by the realization that there were several other groups who had now demonstrated or were known to be on the verge of demonstrating evaporative cooling in alkalis in the pursuit of BEC. The TOP design worked well, and the samples were cooled far colder, in fact too cold for us to reliably measure. We had been measuring temperature simply by looking at the spatial size of the cloud in the magnetic trap. As the temperature was reduced the size decreased, but we were now reaching temperatures so low that the size had reached the resolution limit of the optical system. We saw dramatic changes in the shapes of the images as the clouds became very small, but we knew that a variety of diffraction and aberration effects could greatly distort images when the sample size became only a few wavelengths in size, so our reaction to these shapes was muted, and we knew we had to have better diagnostics before we could have meaningful results. Here we were helped by our long experience in studying various trapped clouds over the years. We already knew the value of turning the magnetic trap off to let the cloud expand and then imaging the expanded cloud to get a measure of the momentum distribution in the trap. Since the trap was harmonic, the momentum distribution and the original density distribution were nearly interchangeable. Unfortunately, once the magnetic field was off, the atoms not only expanded but also simply fell under the influence of gravity. We found that the atoms tended to

fall out of the field of view of our microscope before they had sufficiently expanded. The final addition to the apparatus was a supplementary magnetic coil, which provided sufficient field gradient to cancel the effects of gravity while minimizing any perturbation to the relative ballistic trajectories of the expanding atoms.

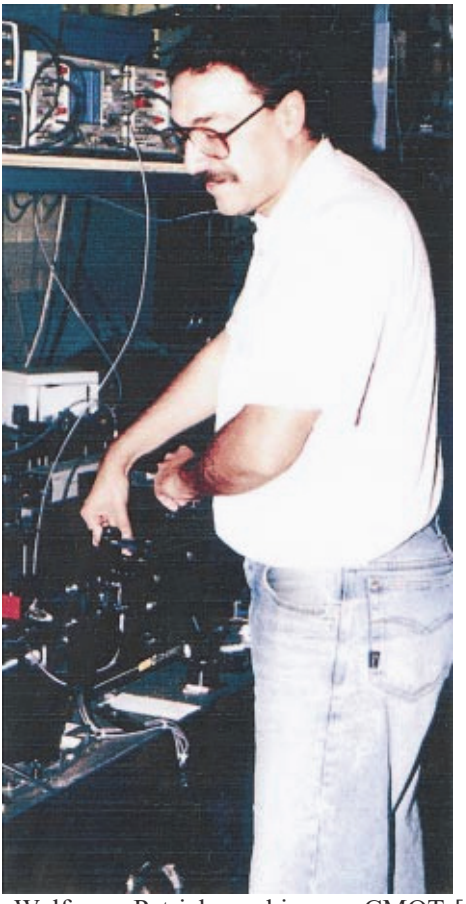

FIG. 5. Wolfgang Petrich working on CMOT [Color].

Anderson, Ensher, and a new graduate student, Mike Matthews (Fig. 6), worked through a weekend to install the antigravity coil and, after an additional day or two of trial and error, got the new field configuration shimmed up. By June 5, 1995 the new technology was working well and we began to look at the now greatly expanded clouds. To our delight, the long-awaited two-component distribution was almost immediately apparent (Fig. 7)

when the samples were cooled to the regime where BEC was expected. The excitement was tempered by the concern that after so many years of anticipating two component clouds as a signature of BEC, we might be fooling ourselves.

FIG. 6. From left, Mike Anderson, Debbie Jin, Mike Matthews, and Jason Ensher savor results of early BEC experiments [Color].

Almost from the beginning of the search for BEC, it was recognized (Lovelace and Tommila, 1987) that as the sample started to condense, there would be a spike in the density and momentum distributions corresponding to the macroscopic population of the ground state. This would show up as a second component on top of the much broader normal thermal distribution of uncondensed atoms. This was the signature we had been hoping to see from our first days of contemplating BEC. The size of the BEC component in these first observations also seemed almost too good to be true. In those days it was known that in the much higher density of the condensate, three-body recombination would be a more dominant effect than in the lower-density uncondensed gas. For hydrogen it was calculated that the condensed component could never be more than a few percent of the sample. The three-body rate constants were totally unknown for alkali atoms at that time, but because of the H results it still seemed reasonable to expect the condensate component might only

be a modest fraction of the total sample. But in our first samples we saw it could be nearly 100%! In the light of the prevailing myth of unattainability that had grown up around BEC over the years, our observations seemed too good to be true. We were experienced enough to know that when results in experimental physics seem too good to be true, they almost always are! We worried that in our enthusiasm we might confuse the long-desired BEC with some spurious artifact of our imaging system.

However, our worries about the possibility of deluding ourselves were quickly and almost entirely alleviated by the anisotropy of the BEC cloud. This was a very distinctive signature of BEC, the credibility of which was greatly enhanced to us by the fact that it first revealed itself in the experiment, and then we recognized its significance, rather than vice versa. It was a somewhat fortuitous accident that the TOP trap provided a distinctly anisotropic trapping potential, since we did not appreciate its benefits until we saw the BEC data. A normal thermal gas (in the collisionally thin limit) released from an anisotropic potential will spread out isotropically. This is required by the equipartition theorem. However, a Bose-Einstein condensate is a quantum wave and so its expansion is governed by a wave equation. The more tightly confined direction will expand the most rapidly, a manifestation of the uncertainty principle. Seeing the BEC component of our two-component distribution display just this anisotropy, while the broader uncondensed portion of the sample observed at the same time, with the same imaging system, remained perfectly isotropic (as shown in Fig. 8), provided the crucial piece of corroborating evidence that this was the long-awaited BEC. By coincidence we were scheduled to present progress reports on our efforts to achieve BEC at three international conferences in the few weeks following these observations (Anderson *et al.*, 1996). Nearly all the experts in the field were represented at one or more of these conferences, and the data were sufficient to convince the most skeptical of them that we had truly observed BEC. This consensus probably facilitated the rapid refereeing and publication of our results.

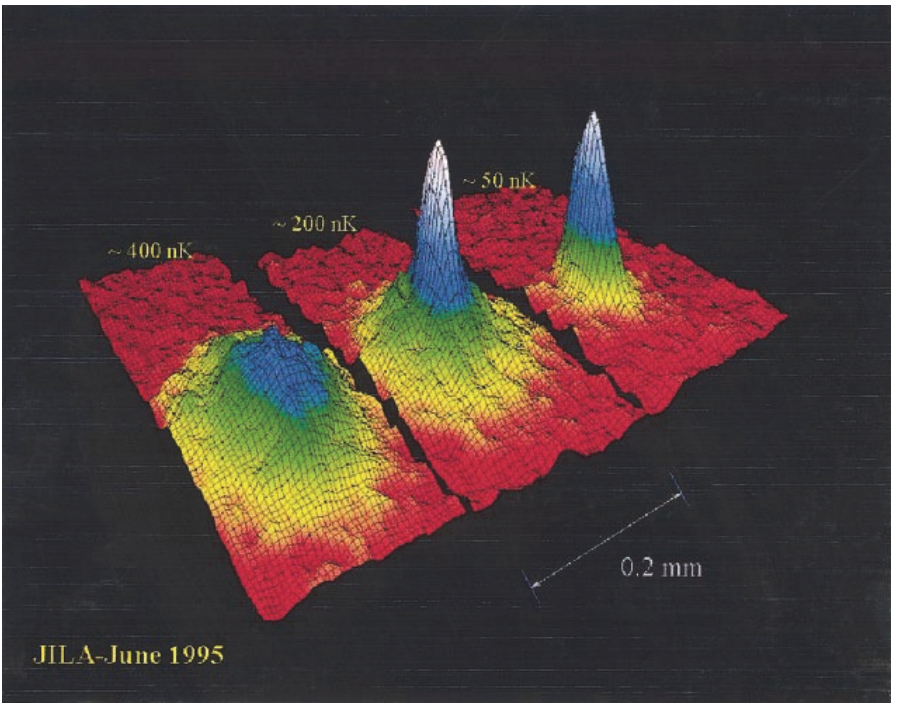

FIG. 7. Three density distributions of the expanded clouds of rubidium atoms at three different temperatures. The appearance of the condensate is apparent as the narrow feature in the middle image. On the far right, nearly all the atoms in the sample are in the condensate. The original experimental data were two-dimensional black and white shadow images, but these images have been converted to three dimensions and given false color density contours [Color].

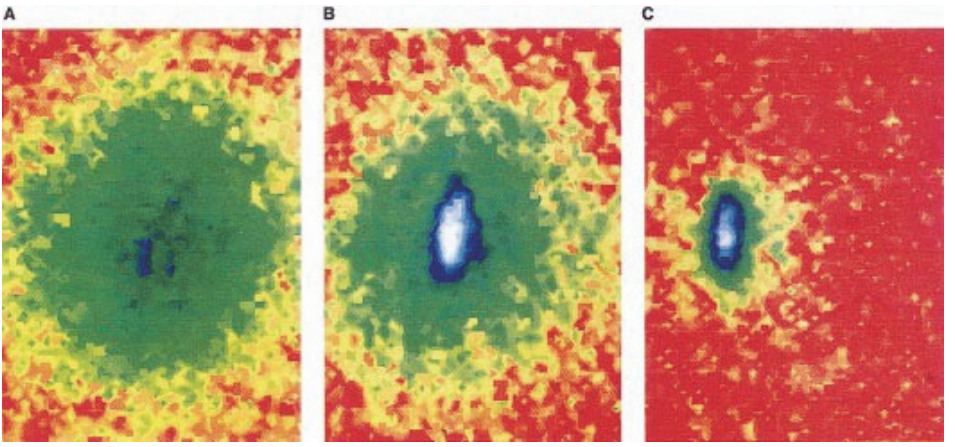

FIG. 8. Looking down on the three images of Figure 7 (Anderson *et al.*, 1995). The condensate in B and C is clearly elliptical in shape [Color].

In the original TOP-trap apparatus we were able to obtain so-called pure condensates of a few thousand atoms. By pure condensates we meant that nearly all the atoms were in the condensed fraction of the sample.

Samples of this size were easily large enough to image. Over the few months immediately following the original observation, we undertook the process of a technological shoring up of the machine, until the machine reached the level of reliability necessary to crank out condensate after reproducible condensate. This set the stage for the first generation of experiments characterizing the properties of the condensate, most notably the condensate excitation studies discussed below.

Although by 1995 and 1996 we were able to carry out a number of significant BEC experiments with the original TOP-trap machine, even by 1994, well before the original condensates were observed, we had come to realize the limitations of the single-cell design. Our efforts to modulate the vapor pressure were not very successful, which forced us to operate at a steady-state rubidium vapor pressure. Choosing the value of vapor pressure at which to operate represented a compromise between our need to fill the vapor-cell MOT with as many atoms as possible and our need to have the lifetime in the magnetic trap as long as possible. The single-cell design also compelled us to make a second compromise, this time over the size of the glass cell. The laser beams of the MOT enter the cell through the smooth, flat region of the cell; the larger the glass cell, the larger the MOT beams, and the more atoms we could herd into the MOT from the room-temperature background vapor. On the other hand, the smaller the glass cell, the smaller the radii of the magnetic coils wound round the outside of the cell, and the stronger the confinement provided by the magnetic trap. Hans Rohner in the JILA specialty shop had learned how (Rohner, 1994) to create glass cells with the minimum possible wasted area. But even with the dead space between the inner diameter of the magnetic coils and the outer diameter of the clear glass windows made as small as it could be, we were confronted with an unwelcome tradeoff.

Thus, in 1994, in parallel with our efforts to push as hard as we could toward BEC in our original, single-cell TOP trap, we began working on a new technology that would avoid this painful tradeoff. This approach was a modified version of our old multiple loading scheme in which many loads from a MOT were transferred to a magnetic trap in a differentially pumped vacuum chamber. That approach had been defeated by the difficulty in transferring atoms from MOT to magnetic trap without losing phase-space density. There was no dissipation in the magnetic trap to compensate for a slightly too hard or too soft push from one trap to the other. This made us recognize the importance of having dissipation in the second trap, and so we went to a system in which atoms were captured in a large-cell MOT in a region of high rubidium pressure, and then transferred through a small tube into a second, small-cell MOT in a low-pressure region. This eliminated the previous disadvantages while preserving the advantages of multiple loading to get much larger numbers of trapped atoms in a low-vacuum region. The approach worked well, particularly when we found that simple strips of plastic refrigerator magnet material around the outside of the transfer tube between the two traps provided an excellent guide to confine the atoms as they were pushed from one trap to the other (Myatt *et al.*, 1996).

With this scheme we were still able to use inexpensive low-power diode lasers to obtain about one hundred times more atoms in the magnetic trap than in our single MOT-loaded TOP magnetic trap and with a far longer lifetime; we saw trap lifetimes up to 1000 s in the double MOT magnetic trap. This system started working in 1996 and it marked a profound difference in the ease with which we could make BEC (Myatt *et al.*, 1997). In the original BEC experiment everything had to be very well optimized to achieve the conditions necessary for runaway evaporative cooling and thereby BEC. In the double MOT system there were orders of magnitude to spare. Not only did this allow us to routinely obtain million-atom pure condensates, but it also meant that we could dispense with the dark-spot optical configuration with its troublesome alignment. We could be much less precise with many other aspects of the experiment as well.

The first magnetic trap we used with the double-MOT BEC machine was not a TOP trap, but instead was our old baseball-style Ioffe-Pritchard trap. The baseball coil trap is rather complementary to the TOP trap in that each has unique capabilities. For example, the geometry of the TOP trap potential can be changed over a wide range, although the range of dc fields is quite limited. In contrast, the geometry of the baseball coil trap potential can be varied only by small amounts, but the dc bias field can be easily varied over hundreds of gauss. Thus in 1996, when we upgraded the original BEC machine to incorporate the double-MOT technology, we preserved the TOP trap coil design. Each is well suited to certain types of experiments, as will be evident in the discussions below.

With the double-MOT setups we were able to routinely make million-atom condensates in a highly reliable manner in both TOP and baseball-type magnetic traps. These were used to carry out a large number of experiments with condensates over the period from 1996 to the present. Some of our favorite experiments are briefly discussed below.

FAVORITE EXPERIMENTS

Collective excitations

In this section, by excitations we mean coherent fluctuations in the density distribution. Excitation experiments in dilute-gas BEC have been motivated by two main considerations. First, a Bose-Einstein condensate is expected to be a superfluid, and a superfluid is defined by its dynamical behavior. Studying excitations is an obvious initial step toward understanding dynamical behavior. Second, in experimental physics a precision measurement is almost always a frequency measurement, and the easiest way to study an effect with precision is to find an observable frequency that is sensitive to that effect. In the case of dilute-gas BEC, the observed frequency of standing-wave excitations in a condensate is a precise test of our understanding of the effect of interactions.

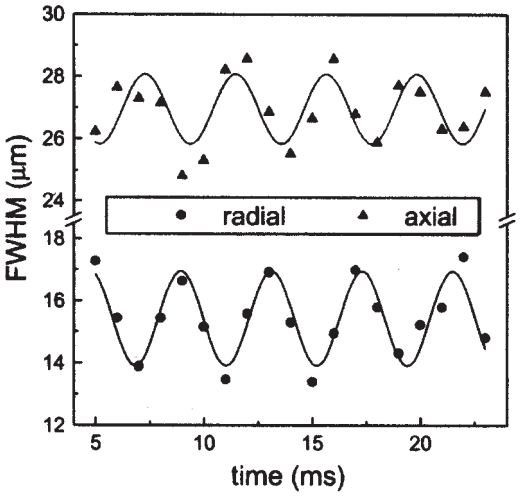

FIG. 9. Zero-temperature excitation data from Jin *et al.* (1996). A weak $m = 0$ modulation of the magnetic trapping potential is applied to a 4500-atom condensate in a 132-Hz (radial) trap. Afterward, the freely evolving response of the condensate shows radial oscillations. Also observed is a sympathetic response of the axial width, approximately 180° out of phase. The frequency of the excitation is determined from a sine wave fit to the freely oscillating cloud widths.

BEC excitations were first observed by Jason Ensher, Mike Matthews, and then-postdoc Debbie Jin, using destructive imaging of expanded clouds (Jin *et al.*, 1996). The nearly zero-temperature clouds were coherently excited (see below), then allowed to evolve in the trap for some particular dwell time, and then rapidly expanded and imaged via absorption imaging. By repeating the procedure many times with varying dwell times, the time-evolution of the condensate density profile can be mapped out. From these data, frequencies and damping rates can be extracted. In axially symmetric traps, excitations can be characterized by their projection of angular momentum on the axis. The perturbation on the density distribution caused by the excitation of lowest-lying $m = 0$ and $m = 2$ modes can be characterized as simple oscillations in the condensate's linear dimensions. Figure 9 shows the widths of an oscillating condensate as a function of dwell time.

A frequency-selective method for driving the excitations is to modulate the trapping potential at the frequency of the

excitation to be excited (Jin *et al.*, 1996). Experimentally this is accomplished by summing a small ac component onto the current in the trapping magnets. In a TOP trap, it is convenient enough to independently modulate the three second-order terms in the transverse potential. By controlling the relative phase of these modulations, one can impose $m = 0$, $m = 2$, or $m = -2$ symmetry on the excitation drive.

There have been a very large number of theory papers published on excitations; much of this work is reviewed by Dalfovo et al. (1999). All the zero-temperature, small-amplitude excitation experiments published to date have been very successfully modeled theoretically. Quantitative agreement has been by and large very good; small discrepancies can be accounted for by assuming reasonable experimental imperfections with respect to the T = 0 and small-amplitude requirements of theory.

The excitation measurements discussed above were then revisited at nonzero temperature (Jin *et al.*, 1997). The frequency of the condensate excitations was clearly observed to depend on the temperature, and the damping rates showed a strong temperature dependence. This work is important because it bears on the little-studied finite-temperature physics of interacting condensates. Connection with theory (Hutchinson *et al.*, 1997; Dodd *et al.*, 1998; Fedichev and Shlyapnikov, 1998) remains somewhat tentative. The damping rates, which are observed to be roughly linear in temperature, have been explained in the context of Landau damping (Liu, 1997; Fedichev *et al.*, 1998). The frequency shifts are difficult to understand, in large part because the data so far have been collected in a theoretically awkward, intermediate regime: the cloud of non-condensate atoms is neither so thin as to have completely negligible effect on the condensate, nor so thick as to be deeply in the hydrodynamic (HD) regime. In this context, hydrodynamic regime means that the classical mean free path in the thermal cloud is much shorter than any of its physical dimensions. In the opposite limit, the collisionless regime, there are conceptual difficulties with describing the observed density fluctuations as collective modes. Recent theoretical work suggests that good agreement with

experiment may hinge on correctly including the role of the excitation drive (Stoof, 2000; Jackson and Zaremba, 2002).

Two-component condensates

As mentioned above, the double-MOT system made it possible to produce condensates even if one were quite sloppy with many of the experimental parameters. One such parameter was the spin state in which the atoms are optically pumped before being loaded into the magnetic trap. As our student Chris Myatt was tinkering around setting up the evaporation one day, he noticed, to his surprise, that there seemed to be two different clouds of condensate in the trap. They were roughly at the locations expected for the 2,2 and 1,-1 spin states to sit, but that seemed impossible to us because these two states could undergo spin-exchange collisions that would cause them to be lost from the trap, and the spin- exchange collision cross sections were thought to be enormous. After extensive further studies to try and identify what strange spurious effect must be responsible for the images of two condensate clouds we came to realize that they had to be those two spin states. By a remarkable coincidence, the triplet and singlet phase shifts are identical and so at ultralow temperatures the spin-exchange collisions are suppressed in 87Rb by three to four orders of magnitude! This suppression meant that the different spin species could coexist and their mixtures could be studied. In early work we showed that one could carry out sympathetic cooling to make BEC by evaporating only one species and using it as a cooling fluid to chill the second spin state (Myatt *et al.*, 1997). We also were able to see how the two condensates interacted and pushed each other apart, excluding all but a small overlap in spite of the fact that they were highly dilute gases.

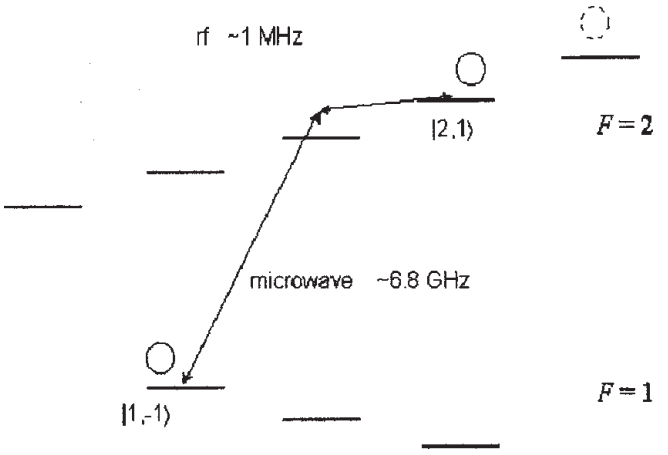

FIG. 10. Energy-level diagram for ground electronic state of ^{87}Rb. The first condensates were created in the 2,2 state. Mixtures containing the 2,2 and 1,-1 state were found to coexist. In later studies we created condensates in the 1,-1 state and then excited it to the 2,1 state using a microwave plus rf two- phonon transition.

These early observations stimulated an extensive program of research on two-component condensates. After Myatt's original measurements (Myatt *et al.*, 1997), our work in this field, led by postdoc David Hall, concentrated on the 1,-1 and 2,+1 states (see Fig. 10) because they could be coherently interconverted using two-photon (microwave plus rf) transitions and they had nearly identical magnetic moments and so saw nearly the same trapping potentials (Matthews *et al.*, 1998). When the two-photon radiation field is turned off, the rate of spontaneous interconversion between the two spin species essentially vanishes, and moreover the optical imaging process readily distinguishes one species from the other, as their difference in energy (6.8 GHz) is very large compared to the excited-state linewidth. In this situation, one may model the condensate dynamics as though there were two distinct quantum fluids in the trap. Small differences in scattering length make the two fluids have a marginal tendency to separate spatially, at least in an inhomogeneous potential, but the interspecies healing length is long so that in the equilibrium configuration there is considerable overlap between the two species (Hall *et al.*, 1998a, 1998b). On the other hand, the presence of a near-resonant two-photon coupling drive effectively brings the two energy levels quite close to one another: on

resonance, the corresponding dressed energy levels are separated only by the effective Rabi frequency for the two-photon drive. In this limit, one may in a certain sense think of the condensate as being described by a two-level, spinor field (Cornell *et al.*, 1998; Matthews *et al.*, 1999b).

We got a lot of mileage out of this system and continue to explore its properties today. One of the more dramatic experiments we did in the two-level condensate was the creation, via a sort of wave-function engineering, of a quantized vortex. In this experiment we made use of both aspects of the two-level system—the distinguishable fluids and the spinor gas. Starting with a near-spherical ball of atoms, all in the lower spin state, we applied the two-photon drive for about 100 ms. At the same time, we illuminated the atoms with an off- resonant laser beam whose intensity varied both in time and in space. The laser beam was sufficiently far from resonance that by itself it did not cause the condensate to transition from state to state, but the associated ac Stark shift was large enough to affect the resonant properties of the two-photon drive. The overall scheme is described by Matthews *et al.* (1999a) and Williams and Holland (1999). The net effect was to leave the atoms near the center of the ball of atoms essentially unperturbed, while converting the population in an equatorial belt around the ball into the upper spin state. This conversion process also imposed a winding in the quantum phase, from 0 around to two pi, in such a way that by the time the drive was turned off, the upper-spin-state atoms were in a vortex state, with a single quantum of circulation (see Fig. 11). The central atoms were nonrotating and, like the pimento in a stuffed olive, served only to mark the location of the vortex core. The core atoms could in turn be selectively blasted away, leaving the upper-state atoms in a bare vortex configuration, whose dynamic properties were shown by postdoc Brian Anderson and grad student Paul Haljan to be essentially the same as those of the filled vortex (Anderson *et al.*, 2000).

Coherence and condensate decay

One of our favorite BEC experiments was simply to look at how a condensate goes away (Burt *et al.*, 1997). The attraction of this

experiment is its inherent simplicity combined with the far-reaching implications of the results. Although it was well established that condensates lived for a finite period, fractions of a second to many seconds depending on conditions, no one had identified the actual process by which atoms were being lost from the condensate. To do this our co-workers Chris Myatt, Rich Ghrist, and Eric Burt simply made condensates and carefully watched the number of atoms and shape of the condensate as a function of time. From these data we determined that the loss process varied with the cube of the density, and hence must be three- body recombination. This was rather what we had expected, but it was nice to have it confirmed. In the process of this measurement we also determined the three-body rate constant, and this was more interesting. Although three-body rate constants still cannot be accurately calculated, it was predicted long ago (Kagan *et al.*, 1985) that they should depend on the coherence properties of the wave function. In a normal thermal sample there are fluctuations and the three-body recombination predominantly takes place at high-density fluctuations. If there is higher-order coherence, however, as one has in macroscopically occupied quantum states such as a single-mode laser, or as was predicted to exist in a dilute gas BEC, there should be no such density fluctuations. On this basis it was predicted that the three-body rate constant in a Bose-Einstein condensate would be 3 factorial or 6 times lower than what it would be for the same atoms in a thermal sample. It is amusing that such a relatively mundane collision process can be used to probe the quantum correlations and coherence in this fashion. After measuring the three-body rate constant in the condensate we then repeated the measurement in a very cold but uncondensed sample. The predicted factor of 6 (actually 7.4 ± 2.6) was observed, thereby confirming the higher-order coherence of BEC (Burt *et al.*, 1997).

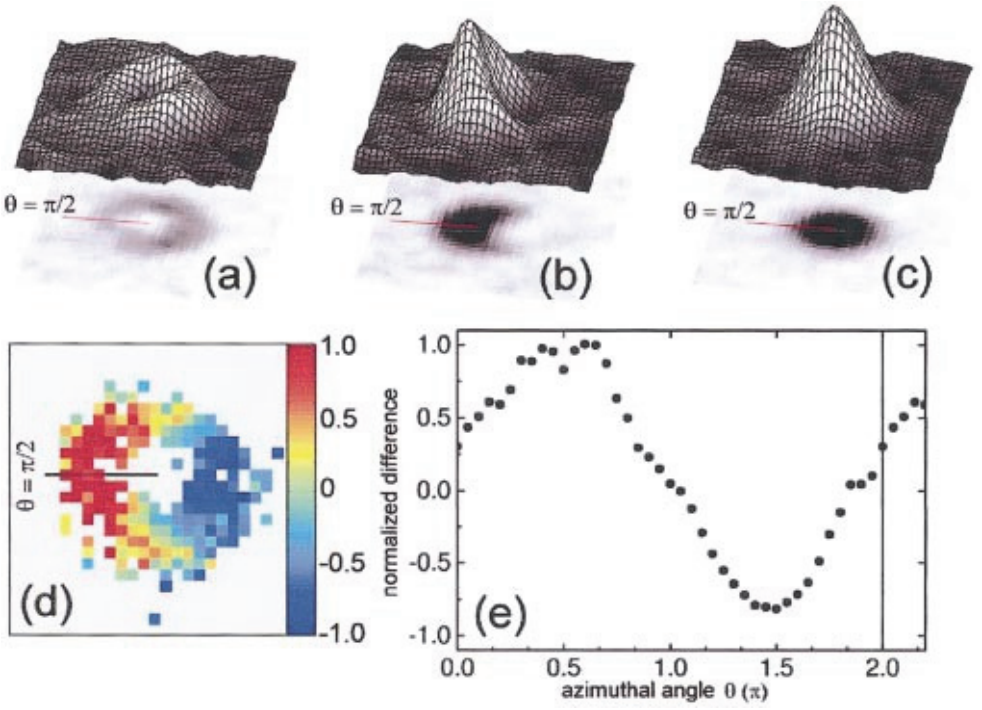

FIG. 11. Condensate images showing the first BEC vortex and the measurement of its phase as a function of azimuthal angle: (a) the density distribution of atoms in the upper hyperfine state after atoms have been put in that state in a way that forms a vortex; (b) the same state after a pi/2 pulse has been applied that mixes upper and lower hyperfine states to give an interferogram reflecting the phase distribution of the upper state; (c) residual condensate in the lower hyperfine state from which the vortex was formed that interferes with a to give the image shown in (b); (d) a color map of the phase difference reflected in (b); (e) radial average at each angle around the ring in (d). The data are repeated after the azimuthal angle 21T to better show the continuity around the ring. This shows that the cloud shown in (a) has the 21T phase winding expected for a quantum vortex with one unit of angular momentum. From Matthews *et al.*, 1999a [Color].

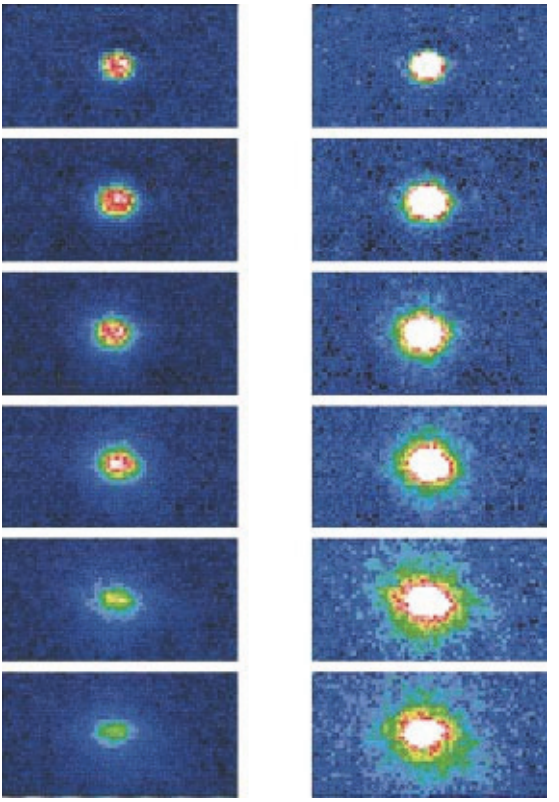

FIG. 12. Bosenova explosion from Roberts *et al.* (2001). From top to bottom these images show the evolution of the cloud from 0.2 to 4.8 ms after the interaction was made negative, triggering a collapse. On the left the explosion products are visible as a blue glow expanding out of the center, leaving a small condensate remnant that is unchanged at subsequent times. On the right is the same image amplified by a factor of 3 to better show the 200 nK explosion products [Color]

Feshbach resonance physics

In 1992 Eric Cornell and Chris Monroe realized that dipole collisions at ultralow temperatures might have interesting dependencies on magnetic field, as discussed in the Appendix. With this in mind we approached Boudwijn Verhaar about calculating the magnetic-field dependencies of collisions between atoms in the lower F spin states. When he did this calculation he discovered (Tiesinga *et al.*, 1993) that there were dramatic resonances in all the cross sections as a function of magnetic field that are now known as Feshbach resonances because of their similarity to scattering

resonances described by Herman Feshbach in nuclear collisions. From the beginning Verhaar appreciated that these resonances would allow one to tune the *s*-wave scattering length of the atoms and thereby change both the elastic collision cross sections and the self-interaction in a condensate, although this was several years before condensates had been created. In 1992 we hoped that these Feshbach resonances would give us a way to create enormous elastic collision cross sections that would facilitate evaporative cooling. With this in mind we attempted to find Feshbach resonances in the elastic scattering of first cesium and then, with postdoc Nate Newbury, rubidium. These experiments did provide us with elastic scattering cross sections (Monroe *et al.*, 1993; Newbury *et al.*, 1995), but were unable to locate the few-gauss-wide Feshbach resonances in the thousand-gauss range spanned by then theoretical uncertainty.

By 1997 the situation had dramatically changed, however. A large amount of work on cold collisions, BEC properties, and theoretical advances provided accurate values for the interaction potentials, and so we were fairly confident that there was likely to be a reasonably wide Feshbach resonance in rubidium 85 that was within 20 or 30 gauss of 150 G. This was a quite convenient bias field at which to operate our baseball magnetic trap, so we returned to the Feshbach resonance in the hope that we could now use it to make a Bose-Einstein condensate with adjustable interactions.

The time was clearly ripe for Feshbach resonance physics. Within a year Ketterle (Inouye *et al.*, 1998) saw a resonance in sodium through enhanced loss of BEC, Dan Heinzen (Courteille *et al.*, 1998) detected a Feshbach resonance in photoassociation in ^{85}Rb, we (Roberts *et al.*, 1998; notably students Jake Roberts and Neil Claussen) detected the same resonance in the elastic scattering cross section, and Chu (Vuletic *et al.*, 1999) detected Feshbach resonances in cesium. Our expectations that it would be as easy or easier to form BEC in ^{85}Rb as it was in ^{87}RB and then use this resonance to manipulate the condensate were sadly naive, however. Due to enhancement of bad collisions by the Feshbach resonance, it was far more difficult and could only be accomplished by following a

complicated and precarious evaporation path. However, by finding the correct path and cooling to 3 nK we were able to obtain pure ^{85}Rb condensates of 16 000 atoms (Roberts *et al.*, 2001).

The scattering length of these condensates could then be readily adjusted by varying the magnetic field over a few gauss in the vicinity of the Feshbach resonance (Cornish *et al.*, 2000). This has opened up a wide range of possible experiments, from studying the instability of condensates when the self-interaction is sufficiently attractive (negative a) to exploring the development of correlations in the wave function as the interactions are made large and repulsive. This regime provides one with a new way to probe such disparate subjects as molecular Bose-Einstein condensates and the quantum behavior of liquids, where there is a high degree of correlation. This work represents some of the most recent BEC experiments, but almost everything we have explored with this system has shown dramatic and unexpected results. Thus it is clear that we are far from exhausting the full range of interesting experiments that are yet to be carried out with BEC.

In the first of these Feshbach resonance experiments our students Jake Roberts, Neil Claussen, and postdoc Simon Cornish suddenly changed the magnetic field to make a negative. We observed that, as expected, the condensate became unstable and collapsed, losing a large number of atoms (Roberts *et al.*, 2001). The dynamics of the collapse process were quite remarkable. The condensate was observed to shrink slightly and then undergo an explosion in which a substantial fraction of the atoms were blown off (Donley, 2001). A large fraction of the atoms also simply vanished, presumably turning into undetectable molecules or very energetic atoms, and finally a small cold stable remnant was left behind after the completion of the collapse. This process is illustrated in Fig. 12. Because of its resemblance (on a vastly lower energy scale) to a core collapse supernova, we have named this the Bosenova. There is now considerable theoretical effort to model this process and progress is being made. However, as yet there is no clear explanation of the energy and anisotropy of the atoms in the explosion, the fraction of vanished atoms, and the size of the cold

remnant. One of the more puzzling aspects is that the cold remnant can be far larger than the condensate stability condition that determines the collapse point would seem to allow (Donley, 2001).

Another very intriguing result of Feshbach resonance studies in ^{85}Rb was observed when our students Neil Claussen and Sarah Thompson and postdoc Elizabeth Donley quickly jumped the magnetic field close to the resonance while keeping the scattering length positive. They found that they could observe the sample oscillate back and forth between being an atomic and a molecular condensate as a function of time after the sudden perturbation (Donley *et al.*, 2002). This curious system of a quantum superposition of two chemically distinct species will no doubt be a subject of considerable future study.

An optimistic appendix

Until a new technology comes along to replace evaporative cooling, the crucial issue in creating BEC with a new atom is collisions. In practice, this means that planning a BEC experiment with a new atom requires learning to cope with ignorance. It is easy to forget that essentially nothing is known about the ultralow-temperature collisional properties of any atomic or molecular species that is not an atom in the first row of the Periodic Table. One cannot expect theorists to relieve one's ignorance. Interatomic potentials derived from room-temperature spectroscopy are generally not adequate to allow theoretical calculations of cold elastic and inelastic collision rates, even at the order-of- magnitude level. Although the cold collisional properties of a new atom can be determined, this is a major endeavor, and in most cases it is easier to discover whether evaporation will work by simply trying it.

Launching into such a major new project without any assurances of success is a daunting prospect, but we believe that, if one works hard enough, the probability that any given species can be evaporatively cooled to the point of BEC is actually quite high. The scaling arguments presented below in support of this assertion are largely the same as those that originally encouraged us to pursue

BEC in alkalis, although with a bit more refinement provided by age and experience.

Although there is an extensive literature now on evaporative cooling, the basic requirement is simply that there be on the order of 100 elastic collisions per atom per lifetime of the atoms in the trap. Since the lifetime of the atoms in the trap is usually limited by collisions, the requirement can be restated: the rate of elastic collisions must be about two orders of magnitude higher than the rate of bad collisions. As mentioned above, there are three bad collisional processes, and these each have different dependencies on atomic density in the trap, n: background collisions (independent of n), two- body dipolar relaxation (αn), and three-body recombination (αn^2). The rate for elastic collisions is $n\sigma v$, where n is the mean density, σ is the zero-energy s-wave cross section, and v is the mean relative velocity. The requirement of 100 elastic-to-inelastic collisions must not only be satisfied immediately after the atoms are loaded into the trap, but also as evaporation proceeds toward larger n and smaller v. With respect to evaporating rubidium 87 or the lower hyperfine level of sodium 23, Nature has been kind. One need only arrange for the initial trapped cloud to have sufficiently large n, and design a sufficiently low-pressure vacuum chamber, and evaporation works. The main point of this section, however, is that evaporation is likely to be possible even with less favorable collision properties.

Considering the trap loss processes in order, first examine background loss. Trap lifetimes well in excess of what are needed for ^{87}Rb and Na have been achieved with standard vacuum technology. For example, we now have magnetic trap lifetimes of nearly 1000 s. (This was a requirement to achieve BEC in ^{87}Rb with its less favorable collisions.) If one is willing to accept the added complications of a cryogenic vacuum system, essentially infinite lifetimes are possible. If the background trap loss is low enough to allow evaporative cooling to begin, it will never be a problem at later stages of evaporation because nv increases.

If dipolar relaxation is to be a problem, it will likely be late in the evaporative process when the density is high and velocity low. There is no easy solution to a large dipolar relaxation rate in

terms of changing the spring constant of the trap or the pressure of the vacuum chamber. Fortunately, one is not required to accept the value of dipolar collisions that Nature provides. In fact, all one really has to do is operate the trap with a very low magnetic bias field in a magnetic trap, or if one uses an optical trap very far off-resonance (such as CO_2 laser), trap the atoms in the lowest spin state, for which there are no dipole collisions. The bias field dependence comes about because below a field of roughly 5 G, the dipolar rate in the lower hyperfine level drops rapidly to zero. This behavior is simple to understand. At low temperature, the incoming collisional channel must be purely *s* wave. Dipolar relaxation changes the projection of spin angular momentum, so to conserve angular momentum the outgoing collisional channel must be *d* wave or higher. The nonzero outgoing angular momentum means that there is an angular momentum barrier in the effective molecular potential, a barrier of a few hundred microkelvin. If the atoms are trapped in the lower hyperfine state (F=1, m_F=-1, in rubidium 87) the outgoing energy from a dipolar collision is only the Zeeman energy in the trapping fields, and for B less than about 5 G this energy is insufficient to get the atoms back out over the angular momentum barrier. If relaxation is to occur, it can happen only at interatomic radii larger than the outer turning point of the angular momentum barrier. For smaller and smaller fields, the barrier gets pushed further out, with correspondingly lower transition rates.

It is unlikely that the three-body recombination rate constant could ever be so large that three-body recombination would be a problem when the atoms are first loaded from a MOT into the evaporation trap. As evaporation proceeds, however, just as for the dipolar collisions, it becomes an increasingly serious concern. Because of its density dependence, however, it can always be avoided by manipulating the trapping potential. Adiabatically reducing the trap confinement has no effect on the phase-space density but it reduces both the density and the atom velocity. The ratio of three-body to elastic collisions scales as $1/nv$. Therefore, as long as one can continue to turn down the confining strength of one's trap, one can ensure that three-body

recombination will not prevent evaporative cooling all the way down to the BEC transition.

To summarize, given (i) a modestly flexible magnetic trap, (ii) an arbitrarily good vacuum, (iii) a true ground state with F ≠. 0, and (iv) non-pathological collisional properties, almost any magnetically trappable species can be successfully evaporated to BEC. If one is using a very far off-resonance optical trap (such as a CO_2 dipole trap) one can extend these arguments to atoms that cannot be magnetically trapped. In that case, however, current technology makes it more difficult to optimize the evaporation conditions than in magnetic traps, and the requirement to turn the trap down sufficiently to avoid a large three-body recombination rate can be more difficult. Nevertheless, one can plausibly look forward to BEC in a wide variety of atoms and molecules in the future.

Acknowledgments

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Defining and Measuring Optical Frequencies[1] – the Optical Clock Opportunity – and More -

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Abstract

Four long-running currents in laser technology met and merged in 1999-2000. Two of these were the quest toward a stable repetitive sequence of ever-shorter optical pulses and, on the other hand, the quest for the most time-stable, unvarying optical frequency possible. The marriage of UltraFast- and UltraStable lasers was brokered mainly by two international teams and became exciting when a special "designer" microstructure optical fiber was shown to be nonlinear enough to produce "white light" from the femtosecond laser pulses, such that the output spectrum embraced a full optical octave. Then, for the first time, one could realize an optical frequency interval equal to the comb's lowest frequency, and count out this interval as a multiple of the repetition rate of the femtosecond pulse laser. This "gear-box" connection between the radio frequency standard and any/all optical frequency standards came just as Sensitivity-Enhancing ideas were maturing. The four-way Union empowered an explosion of accurate frequency measurement results in the standards field and prepares the way for refined tests of some of our cherished physical principles, such as the time-stability of some of the basic numbers in physics (*e.g.*, the "fine-structure" constant, the speed of light, certain atomic mass ratios ...), and the equivalence of time-keeping by clocks based on different physics. The stable laser technology also allows time-synchronization between two independent femto-second lasers so exact they can be made to appear as if the source were a single laser. By improving pump/probe experiments, one important application will be in bond-specific spatial scanning of biological samples. This next decade in optical physics should be a blast!

Overview and Summary

THE VIEW BACKWARD over some momentous developments often suggests a kind of certainty and inevitability that may not have been evident, even in the slightest form, when the story was going on. One modern trend is to focus on some particular research project – one which is so simple and transparent that the Manager can expect to be successful in the chosen

[1] The 2005 Nobel Prize for Physics was shared by Roy J. Glauber, John L. Hall, and Theodor W. Hänsch. This lecture is the text of Dr. Hall's address on the occasion of the award. Reprinted from RevModPhys.78.1279.

research task. But such a project will likely have modest consequences: Surely its consequences were at least dimly visible from the beginning. By contrast, this "Optical Frequency Comb" capability has come "out of the blue" from a remarkable synthesis of independent "state-of-the-art" developments in four distinct fields: UltraStable Lasers, UltraFast Pulse Lasers, Ultra-NonLinear Materials and Responses, and UltraSensitive Laser Spectroscopy. These separate fields were alike in their shared – but independent – pursuit of advancing simple and effective technology for using electromagnetic signals for their own spectroscopic and other optical physics interests in the visible domain. After the Great Laser Technology Synthesis of 1999-2000, celebrated by the brief name of "Optical Frequency Comb," the Optical Toolbox has really blossomed. In respecting our Patron, Dr. Nobel, we may be more expansive and clear: the field of optics has blossomed explosively!

The resulting new capabilities are unbelievably rich in terms of the tools and capabilities that have been created, and these in turn are reinforcing progress in these related contributing fields. For example, after the frenzy of the first generation frequency measurements, some of the Generation II comb applications now include: low-jitter time synchronization between ultrafast laser sources, coherent stitching-together of the spectra of separate fs laser sources so as to spectrally broaden and temporally shorten the composite pulse, optical waveform synthesis for Coherent Control experiments, precision measurement of optical nonlinearities using the phase measurement sensitivity of rf techniques, coherently storing a few hundred sequential pulses and then extracting their combined energy to generate correspondingly more intense pulses at a lower repetition rate… . Attractive topics of research for Generation III applications include precise remote synchronization of accelerator cavity fields and the stable reference oscillators for Large Array Microwave Telescopes; and potential reduction of the relative phase-noise of the oscillator references used for deep space telescopes. (NASA, VLBI …) That's just part of the first five years.

So in the precision metrology field, what exactly could one say is different now? In the same way we have enjoyed for the last half-century powerful spectroscopy methods with radiofrequency signals (consider Magnetic Resonance Imaging as one of its useful forms!), we now can use

frequency-control methods for optical spectroscopy. But there is a really important difference: the number of cycles per second in the optical domain is roughly 10-million-fold greater than in the rf domain, even as the rf processes themselves are still a few million-fold faster than human perception scales. In essence, these large factors map into a corresponding improvement in resolution – our measurement capability. See the discussion below. With human senses we can perceive halves and quarters and tenths, and perhaps a little better. These capabilities are enhanced approximately by the product of these two large numbers, bringing us immediately into the garden of a few parts in 10^{14} metrology. We can do even better by averaging independent measurements.

Metrological Standards and Science

A Close and bi-directional Connection

On occasion, accumulation of progress in the details of some scientific enquiry leads us to a glorious new vision of some parts of our experience: basically a new insight or organizing principle becomes available. But behind this revelation normally is a huge amount of painstaking work, quantitatively stating experimental results, which normally are expressed in absolute units. Sometimes an experiment can provide its own internal calibration, but in the main we really need to have practical standards to reference the measurements against. Of course the Standards must themselves be reproduced and distributed before the scientific results can be confirmed by several labs. The best case is that the needed Standard is based on some fundamental physical effect, ideally a quantum effect, so it can be *independently realized* by different laboratories at the same accuracy. This standards-realization process is in a revolution itself! [1]

The Length Standard and its Relationship to Frequency/Time

It's useful to discuss a bit about metrological Standards, which we can initially take to be the seven base quantities of the **Système International d'Unités** (International System of Units), or more briefly the SI, or "the Metric System" These are Mass, kg; Time, s; Length, m; Current, A; Temperature, K; Quantity of Matter, mol; Unity of Light Intensity (Candela), cd.. From these seven base units, another ~30 useful derived units can be defined. For our purposes of *stretching measurement precision*

to the ultimate limits, clearly **Time** and **Length** are the two quantities offering the highest potential precision. For eons the day was a natural unit for Time, but standards for Length have seemed artificial and arbitrary. In 1791 the Metric System was first discussed but, lacking serious metrology experience, these Age of Enlightenment gentlemen of the French Academy of Sciences decided that the Metre would be defined as some small fraction ($¼ \times 10^{-7}$) of the Earth's circumference on a great circle passing through the poles and France. Of course, having the standard based on surveying had some limitations in practical lab work, but at least the unit of length was finally a definite and basically absolute distance. This was welcome change since public exhibits in places such as Braunschweig, Germany and on Santorini Island, Greece show there was a succession of length standards in sequential use, as a new Duke of different personal arm length came into power. But by 1875, with the Treaty of the Metre Convention, a stable metal bar began to look like a good idea. While not fully universal and independently realizable, the factory could make many of these prototype Metre bars, and could confirm their equivalence.

The community of Metric countries in 1889 welcomed the improved X-cross-section meter bars known as the "International Prototype Metre" length standard. This design used graduations (lines) engraved onto a platinum-iridium bar, with a Meter defined as the separation between two graduation lines at 0 C, measured with a specified mounting arrangement, and under atmospheric pressure. The 30 new bars were calibrated using an optical comparator technique, before dissemination of two to each country.

By 1890 A. A. Michelson had identified the exceptional coherence of the Cd red line, and by 1892 had used it with his new interferometer to determine the length of the International Prototype Metre. His measurements showed the defined Metre contained 1,553,164.13 units of the wavelength of the cadmium red line, measured in air at 760 mm of atmospheric pressure at 15 C. For this and other contributions, Michelson was awarded the Nobel Prize in 1907. Of course thermal expansion was a limiting problem, such that when the low-expansion steel alloy *Invar* was invented, the creator (and Director of the BIPM), C. D. Guillaume, was awarded the Nobel Prize for 1920. However, the SI Metre definition was unchanged for 85 years: the Meter Bars worked well and optical comparators got fatigueless photo-electric eyes.

Spectroscopic experiments and supporting Quantum Theory led to improved understanding and improved light sources. The metrological needs of the World Wars changed the Science climate, and transportation disruptions emphasized the advantage of having *independently-reproducible standards* based on quantum physics. Eventually, in 1960 the Eleventh General Conference on Weights and Measures was able to redefine the International Standard of Length as 1,650,763.73 vacuum wavelengths of orange light resulting from transitions between specified atomic energy levels of the krypton isotope of atomic weight of 86. Going forward with a new definition, one would say the Kr wavelength is $\lambda = 1$ m / 1,650,763.73 = 0.605,780,211 μm. While the adopted Definition speaks about unperturbed atoms, in fact several shifts were observed in light from the discharge lamp used for realizing this Metre in practice. Pressure shifts and discharge operating conditions were stabilized by operating the lamp at a specified discharge current and at a fixed pressure and temperature (using the triple-point of liquid nitrogen). A field-induced gas flow of Kr^+ led to a wavelength difference of light viewed from the two cell ends. When laser comparisons with this standard were performed, the additional problem of radially-dependent Doppler shifts of the emitted light was discovered.

The 1960's and 1970's saw a number of different stabilized lasers systems introduced, refined, and the wavelengths measured and compared between various national labs. Basically, all these laser systems were entered into the competition to be the next International Length Standard. There were then 48 nations involved in the Metre Convention, so politically speaking, choosing one out of the many offered candidate lasers would be difficult. In addition, none of these approaches were overwhelmingly superior, when performance, cost and complexity were all considered. And scientifically, it seemed attractive for the new Length Standard definition to be based on the Speed of Light, introduced as a defined quantity. On the basis of a number of laser-based measurements, this value was taken as 299,792,458 m/s exactly, a rounded value in accord with the measurements of the several standards labs. This redefinition of 1983 took the form:

"*the Meter is the length of the path traveled by light in vacuum during a time interval of 1/299,792,458 of a second*. The *speed of light* is

c = 299,792,458 m/s, exactly.

The *second* is determined to an uncertainty, U = 1 part in 10^{14} by the Cesium clock."

The General Conference also suggested several recommended radiations for realizing the meter at that time, *e.g.*: "The wavelength of the iodine-stabilized Helium-Neon laser is

$$\lambda_{HeNe} = 632.99139822 \text{ nm},$$

with an estimated relative standard uncertainty (U) of $\pm$ 2.5 x 10^{-11}."

In all of these changes in definition, the goal was not only to improve the precision of the definition, but also to change its actual length as little as possible. See [3]. With the speed of light defined, an optical frequency (linked to time) can thus serve as a length unit.

Fundamental Physics Issues in the Re-Definition of Length

At the times of these redefinitions, there were some concerns that we were switching the physical basis for the Metre definition. For example, if in the future we discover that some of the "constants of Physics" actually are slowly changing, one could worry that the new definition might impact or even limit our discovery process. In any case, we would be unaware of a global change that would conserve the physical relationships we have discovered. But could there be a differential effect that might be observable? Before 1960 we were accepting the spacing of some lattice planes in the Pt-Ir alloy of the Meter Bar as our measurement basis for length: this length certainly would fundamentally involve Quantum Mechanics, and Electricity and Magnetism. And, considering the thermal vibration of molecules in the somewhat-anharmonic interatomic potentials, we can suppose that the nuclear masses – and thus the Strong Interactions – will also play a role in length via the thermal expansion. With the 1960 redefinition of the Metre in terms of a Krypton atom's radiation's wavelength, perhaps we were opening some opportunity for confusion? Now Quantum Mechanics and Electricity and Magnetism are still fundamentally involved, but the atom's mass is involved only in a reduced-mass correction, rather than via thermal effects. Certainly a new "constant," the speed of light, is linearly serving as

the dimensioned scale constant. Initially the 1983 redefinition appears to be still a different sort compared with the 1960 Kr definition, but really it just repeats the energy level difference idea (now it is Cs in defining the second rather than Kr defining an optical energy) followed by a conversion of dimensions. Who knows if there is some fun hidden in here?

Where we have come to is that the SI is now functioning with six, rather than seven, basic units. The Metre has been demoted to a derived unit, and the significance of Time and Frequency have been further elevated. This begins a long story, with the SI base units being challenged by spectacular advances "at the bottom of a Dewar" [4], giving us a Josephson-effect based voltage standard (Nobel Prize of 1973), while the von-Klitzing-effect defines a quantum resistance standard (Nobel Prize 1985). Taken together as V^2/R, an electrical Watt unit is apparent, while an SI Watt – defined as a Joule per second – would be represented as ½ kg $(m/s)^2$ /s. The relationship between these is established by a "Watt Balance" experiment [5]. Recently the Single Electron Transistor begins to enable digital counting of electron charges per second, contacting the SI Ampere, the unit of electric current. This interface between metrology and quantum physics is becoming a "Hot Topic" of our time [1, 6]. The remarkable advances in Metrology also allow – and advances in Cosmology and Astronomy strongly motivate – curiosity about the "exactness" and "time-invariance" of the various physics numbers used in our description of physical reality.

Clocks and Time

Time represents our most precisely measurable quantity and so it always has attracted certain kinds of devoted researchers. But also, now with various sensors and microprocessor control, many physical parameters can be read out by frequency measurements, and so we add a huge number of scientists in other fields who want to recover the finest details within their measurements. (Still, many really important research subjects are not yet so well developed that these frequency tools are useful: for example, world-changing decisions about air pollution management are being made even though we scarcely are sure about the *sign* of some effects.)

But for technology people, the improvement of time measurement precision grows as a field of intense interest and competition worldwide. In no small part this is because of the very advances singled out by this year's

Prize: a capability jump by several decades is uncommon in any field, let alone *the* field where the precision of measurement was already at the highest level, and had already been driven to near its apparently basic limits.

Of course interest in time has been part of man's history from our beginnings, but only in the last several recent centuries have some lucky subsets of people been somewhat isolated from seasonal variations, with leisure to think about Nature, and so *time* as an experimental parameter began to emerge. Nowadays we can look from the scientific and experimental point at the question: why would one be interested in time? For those who love precision, the clear reason is that *time is the most powerful metrological variable*.

Scaling of Precision Attainable when we are Measuring Time

The precision of time measurements can be increased essentially without limit, by increasing the measurement duration and simply counting the increased number of cycles of some regularly-spaced events. However a stronger information growth with measurement duration is possible if we have a nice source that has coherence from the beginning of the measurement until the end. (For the present purpose we may take this "coherence" to mean that if we know the oscillation cycle's phase early in the measurement, the *coherent* source is so steady that the oscillation phase could be predicted at later times near the measurement's end to a precision of 1 radian of phase.) In this case we can have a measurement precision which will grow with the measurement interval τ according to $\tau^{3/2}$. A simple way to explain this assertion is to suppose we divided the measurement duration into 3 equal sections, each with N/3 measurements. In the starting zone we compare the reference clock and the unknown clock, with a relative phase precision which scales as $(N/3)^{1/2}$. Next, in the middle section, we merely note the number of events, N/3. In the last section we again estimate the analog phase relationship between test and reference waves, with a relative imprecision which is again $(N/3)^{1/2}$. Subtracting the two analog phases increases the uncertainty of one measurement by a factor $2^{1/2}$ so, altogether, the relative precision increases as $(1/2^{1/2}) \times (N/3)^{3/2}$. Thinking of a microwave frequency measurement, with a base frequency of 10^{10} Hz, in a 1 s measurement we have a factor of 10^5 potentially to win. Commercial counters already can register 12 digits in 1 s for a reasonable input signal. One can see there is just a huge gain in measurement precision if we can

measure a coherent frequency source in a proper way: No wonder we have the situation where metrology scientists as well as philosophers, sailors, and farmers are interested in clocks and time and seasons [7]. Indeed our most powerful test of the existence of Einstein's predicted gravitational radiation comes from the observed shortening of the year of the Hulse-Taylor binary pulsar: orbital clock physics vs. quantum frequency standard physics on the earth. This marvelous work was celebrated by the Nobel Prize of 1993.

What makes a clock?

The three essentials of clocks are: a source of regular events, a counter/integrator to totalize the events, and a suitable readout mechanism to present the current result to an interested human or machine. In many ways the frequency source is the most interesting part since it is intrinsically an analog system, where the design goal is to diminish as little as possible the intrinsic stability of some physical oscillation, in the course of reading out its information. In this game, nuance and subtlety count for a lot. It is customary that the performance of clocks based on some well-known source of regular "clicks" will be improved several orders of magnitude by the work of many people over many years, with the ultimate fate of becoming suddenly obsolete due to the introduction of a better kind of stable oscillator. The new idea must be a serious advance, since it must be competitive at the start of its life with the previous technology which has been enhanced and improved in many stages. Still, some technologies have had a long lifetime – for example one can still buy a good wristwatch based on a torsional oscillator, even though this balance wheel concept was used by Ch. Huygens in 1675.

Keeping time has been of serious interest since man turned agrarian, but became of critical interest with the expansion of lucrative international trade: "inevitable" shipwrecks could be avoided by better knowledge of position (mainly longitude) at sea. Parliament's Longitude Prize of 1714 (above $10 M in current terms) attracted John Harrison's attention and some *40 years* of his inventive work. In 1761 his H-4 clock demonstrated 1/5 s/d, $\delta\nu/\nu \sim 2.5 \times 10^{-6}$ even while at sea. This was several-fold better that the requirement, but only half the Prize was initially paid: in part the controversy was about the Intellectual Property! A second problem was conflict of interest within the judging Committee. (This story is well-told in [7].) Present customers of precise timekeeping include TV Networks (for

synchronization), cellular telephone companies, the GPS users who need the limiting performance, radio astronomers, NASA Deep Space Tracking, and various other branches of Science in which a physical variable has been read out by frequency methods.

Evolution of Frequency Sources: Distinguishing Precision and Accuracy

In discussing the performance of a mechanical clock, or the electronic oscillators based on vibrational modes of quartz crystals, it is clear that the basic frequency is set by mechanical dimensions. Such a device could be stable and have good precision, in that its readout could be determined with many digits, but there can be no claim to any particular fixed or natural frequency. Still the stability of any particular crystal device could be remarkable: a drift of $>10^{-6}$ /day gradually improved to the present $<\sim 1 \times 10^{-10}$ /day, while the shift with acceleration remains near 10^{-9} per "g". The high frequency of electronic oscillators served well for convenient interpolation between "clicks" of the absolute standard, provided by zenith sightings of the daily motion of the Sun, as codified by the 1875 Metre Convention. (Later the Earth rotation data series were based on telescopic observations of the lunar occultation starts of various stars and planets.) By the 1950's the electronic oscillators were refined enough that variability $\sim 10^{-8}$ was inferred in the earth's spin rate, and was associated with changes of the earth-atmosphere system's moment of inertia due to North-South ocean tides, and large storms. The community wished to eliminate the variability, but still needed an absolute and universal (rather than local artifact) standard. The new choice in 1960 adopted a stated number of seconds in the "Tropical Astronomical Year 1900". Perhaps this was good in its motivation, in that the rotation of the earth around the sun would have a lower level of perturbation. However a clock/oscillator that has only a single click per year will be hard to enjoy at its full precision. As a metrology principle we rather would prefer the basic frequency source to be at a very high frequency so that the integer multiple of the standard's clicks will be a huge digital number in our measurement of some interesting phenomenon, and the unavoidable noise and uncertainty of the remaining analog subdivision of the unit will be as insignificant as possible.

Electronic Clocks based on Quantum Transitions

Based on Otto Stern's atomic beam method, which had resulted in his Nobel Prize of 1943, I. I. Rabi introduced atomic beam resonance methods which allowed probing internal (hyperfine) quantum energy states of atoms such as Cesium with greater precision. This work was recognized by the Nobel Prize of 1944. Using atoms in this way, the *independent realizability* and *universality* requirements for a Primary Standard could be well addressed. In addition, the transition frequencies were near the high-frequency-end of the usable rf spectrum, so the Metrology aspects were optimized as well. The first Atomic Beam Clock was developed at NBS in 1949 based on microwave transitions in Ammonia, and by 1955 Cs beam clocks were in operation at the NPL and NBS. The powerful Method of Separated Oscillating Fields was invented by N. F. Ramsey, reported in 1955, and later recognized by the Prize in 1989. In this dual-excitation concept, suitable atoms were excited once, and then left to evolve their internal phase (ideally) free of perturbation, until a second excitation pulse effectively completed the interferometric comparison of the phase evolution rates between the atomic and laboratory oscillating systems. Progress on the Cs beam atomic frequency standard was widespread and rapid, allowing redefinition in 1967 of the SI Second as 9 192 631 770 units of the Cs hyperfine oscillation period. Correspondingly, the Cs oscillation frequency is defined as (exactly) 9 192 631 770 Hertz (cycles of per second). The specialists involved in this redefinition of Time and Frequency wisely did not specify exact details of the measurement process, leaving room for considerable progress. For example when laser-based optical pumping of atoms between hyperfine states became feasible and popular in the early 1990's, NIST colleagues built a new atomic beam cesium standard, NIST-7, based on optically transferring most of the population from the 16 available hyperfine levels into the special ($|3,0\rangle$) lower state involved in the clock transition. Along with this factor, ~16x, improvements of the atom source itself, and better frequency source and readout electronics were helpful. Above all, computer-based signal processing and active control of measurement systematic offsets made it possible to reduce the inaccuracy of realizing the Cs second at NIST to $\sim 5 \times 10^{-15}$. But as usual in the art-form of Precision Measurement, this "tour de force" system was soon made obsolete in a single step by a qualitatively better technology.

As shown by Kasevich and Chu [8], laser cooling of the Cs atoms made it possible to successfully implement the "atomic fountain" concept for the realization of the Cs-based frequency definition. By shifting laser frequencies or powers, a slowly moving ball of atoms could be dispatched vertically upward through the excitation rf cavity, reaching apogee a good part of a meter above the cavity, and then beginning the return trip to pass through the excitation cavity a few 100 ms later. With such a long coherent interaction time, instantly the resonance linewidth dropped to ~ 1 Hz, down from ~300 Hz in the previous epoch of thermal beam of atoms. Optical probing of the atoms below (and temporally after) the cavity could yield the excitation-probability vs. probe-frequency-tuning curve needed to control the source oscillator's frequency. By using suitably-closed optical transitions for readout, one can have many photons emitted per atom so that, even after solid angle and detection inefficiencies are considered, the measurement noise is not much larger than the minimum associated with the finite number of atoms. Andre Clairon and his colleagues made the first real Cs Fountain Frequency Standard, in 1995 [9] at the Paris Institute now known as LNE-SYRTE (Laboratoire National de Métrologie et d'Essais – Systèmes de Références Temps/Espace). Even without the contemporary schemes to break this atom-shot-noise limit, the fountain Cs clocks at NIST and SYRTE now achieve accuracy levels below 1 $x10^{-15}$ when all the known measurement and perturbation issues are taken into account [10]. Of course with the resolution improvement one hopes for more potential accuracy, but will have beforehand an expanded list of small shifts and niggling concerns to consider. After all, even with the extended interaction time, fewer than 10^{10} oscillation cycles are counted, so the achieved inaccuracy of 1 $x10^{-15}$ already corresponds to 10 ppm splitting of the atomic fountain's resonance linewidth. Fountain Cs clocks are limited by two newly important effects, collisionally induced frequency shifts due to the hugely increased atom density [11], and shifts due to the effects of the ambient thermal radiation associated with the vacuum system's walls. Attempting to split lines further always brings a diverging list of new small problems, leading to an effective barrier.

An important observation is that for many types of Quantum Absorber samples the line broadening processes will be the same for both radio and optical frequency domains. For example, the atomic fountain apparatus could explore optical transitions, rather than microwave ones,

with the same interaction time. Clearly we would prefer the higher base frequency of the optical world, since the resonance feature of interest will then display a relative sharpness increased by roughly the same huge factor of optical/microwave frequency ratio. With sharper line shapes we can expect more precise measurements that will let us better see the small effects of various experimental parameters, leading to better *independent reproducibility* which, with major investment of efforts, can often be parlayed into nearly a corresponding increase in measurement *accuracy capability* as we come more fully to characterize the offset processes. But before the Millennium Year of the Optical Comb, just how did you plan to measure the absolute optical frequency?

This *repeatability* idea seems weaker than the gold standard of *accuracy*, which additionally conveys our being able to connect the measured result with the base units of the Systeme International. But in fact we now know several optical clock systems that have 10-fold smaller uncertainty than the Cs standard. So before a redefinition is appropriate, their comparisons will be most interesting, especially as an entry point for one of the most interesting branches of Science, trying to figure out which physical "laws" are essentially exact, which ones are ignoring some details to have a tidy presentation, and which are in fact stating "facts" about Nature which are not exactly actually true. Celestial mechanics, ideal gas laws ignoring molecular volumes, and parity conservation in atomic physics could be my examples.

Starting the Dream of Optically-based Clocks

The Laser Arrives

The future of metrology was changed fundamentally on 12 December 1960 when a small team at Bell Labs, led by Ali Javan, eventually found the right conditions for their Optical Maser to generate self-sustained optical oscillations. Their specially crafted gas discharge tube had the improbable situation in which the populations in two particular Neon atomic levels were reversed from the thermal norm: by means of the discharge in the more-abundant He gas, collisional energy transfer set up a population inversion, whereby more atoms were in the Ne's higher energy state. It is impressive that these conditions were established on the basis of careful measurements and modeling of the discharge conditions! Having the

populations inverted from the usual case reverses the sign of the absorption that experience teaches us is a universal property of (normal) matter. Accordingly, with an inverted population, rather than absorption, Javan's group had optical emission. The atoms would provide amplification of any resonant optical signal passing down the discharge cell. A few percent gain wouldn't be very exciting normally, except that the utilized multilayer mirrors were designed and fabricated to have reflection losses that could be even smaller, setting the stage for a buildup of power on every pass. So finally they did obtain a self-sustained continuous optical oscillation, and observed the collimated beam that was anticipated by Charles Townes and Arthur Schawlow in a classic paper of 1958. Similar ideas were also considered in the Former Soviet Union, leading to the Nobel Prize of 1964 being shared by N. Basov, A. Prokhorov, and Townes.

Connection to Glauber's Coherent States of Light

In planning a theoretical study of optical fields, perhaps one can understand starting with known results for single-photon fields, then adding a few photons cautiously to see what happens. Actually, for all of us following Professor Glauber's work it was surprising just how few photons were needed for the new photon density distribution functions to change fundamentally from the customary Poisson limit: with increasing number of photons in a mode the fields start showing the small fractional fluctuations that would characterize a classical field. On the experimental side, for Javan's very first laser, the output laser power was ~ 1 milliWatt, about 10^{16} photons per second! We can proceed to estimate the expected fractional variation of $1/\sqrt{N}$, but with such an incredibly large number of coherent photons in one mode, the result is an unphysically small variation. Thousands of merely technical processes would cause fluctuations larger than the predicted $1{:}10^{8}$! An equivalent statement is that these lasers were operating strongly, far into the domain of classical fields, and quantum fluctuations would be very hard to observe. Indeed it was not until the end of the 1970's that people began again to appreciate how to study manifestly quantum fields with just a few photons in them. At this vastly-reduced intensity, quantum correlations are challenging to observe, but they are very interesting, since they correspond to rather significant fractional effects. For example, H. J. Kimble's group used phase-dependent Squeezed Light to make a spectroscopic measurement with about 2-fold better Signal/Noise

than the naïve shotnoise limit [12]. To observe strong Squeezed Light effects, it is essential to minimize optical losses, as they work to revert the statistics toward the thermal limit. Regrettably, noise from technical sources will grow linearly in the laser power, while the advantage due to squeezing will grow more slowly. It seems that getting a factor 10 amplitude S/N improvement will be incredibly difficult.

Coherence of the Laser Field Enables Frequency-Diagnostics

The Bell Labs laser design success had grown out a semiclassical view of how Optical Masers would operate. Yes, amplification would be provided by quantum mechanical atomic systems, rather than radio tubes or klystrons, and yes each atom could contribute just one photon to the field in each event. But still, considering how huge is the number of photons in the field, the discreteness probably will hardly matter. Almost immediately the Bell-Labs team was testing this understanding by combining two separate laser beams into a single coaxial beam, and shining this onto the sensitive surface of a high-speed photodetector. They already were thinking of each laser oscillation as being an essentially classical field, satisfying reflection boundary conditions at the two mirrors. So this stable-and-repeating bouncing specification would define the possible wavelength(s) of the generated laser light. By luck and design the discharge was wonderfully calm, so one could expect the gas' refractive index would be essentially constant. Thus the interferometric boundary conditions would essentially define the oscillation *frequency* and, accordingly, one would expect to see a sharp optical frequency come out of this device. With two lasers' sharp frequencies on the nonlinear detector's surface, one should expect the difference frequency to be generated, which it was. I can still remember hearing the audio beat whistle that Javan had recorded when his two lasers were tuned almost to the same optical frequency. It was a ~1 kHz difference between two sources at 260 THz!

Actually the linewidth of these beats was remarkably narrow. We already expected that based on the numbers noted above: a stream of $\sim 10^{16}$ photons/s would have random power fluctuation of $\sim 10^{-8}$ relative to the full power. So the optical phase could be extremely well defined. However the laser's Schawlow-Townes linewidth calculation includes the role of optical loss, which actually limits the laser coherence, giving ~ milliHz linewidth expectation.

In principle then we have a radiation of incredible sharpness, and should be ready to seek interesting physical effects. The immediate disappointing truth is that this tiny predicted laser phase fluctuation will be completely masked by noise of technical origins. We already noted that the strongest definition of the oscillation's frequency is fixed by the interferometric standing-wave condition bouncing on the laser cavity mirrors. But the lab is a noisy place, seismically speaking, with a quiet lab having a ground noise of ~3 x10^{-9} m/$\sqrt{\text{Hz}}$ in the vibration frequency band say 1 – 30 Hz. A laser cavity is some fraction of a meter in length, so it will be difficult to make a system arbitrarily stiff. Rather, some important fraction of the ground noise will appear as cavity length variations, and therefore laser frequency variations. Suppose we say only 1% couples in to relative length changes. One can instantly see the scale of the problem: ~10^{-10} fractional frequency variations will be our *a priori* scale. Even temperature variations will be painful, since the 10^{-10} scale already corresponds to a few milliKelvin temperature change for low expansion materials like fused silica. We can make progress by locking the laser to a stable reference cavity [13]. Optimizing for vibrational integrity, we will use a stiff structure for mounting the reference cavity mirrors, and then mount the assembly with a horizontally soft suspension. By focusing on the vibration isolation, Bergquist has obtained [14, 15] a record narrow laser linewidth ~0.16 Hz! Another approach seeks to minimize the cavity acceleration sensitivity. By use of a vertically-symmetric mounting [16] of the reference cavities, our group recently reported Hz-level laser linewidths.

Coherence of the Laser Beats Enables Frequency- Based Laser Control

Considering the small intrinsic phase noise of the laser source, and the rather high power ~ mW, heterodyne detection of the beat frequency between two laser sources yields an interestingly high Signal/Noise ratio. Even with very short averaging times, say 1 μs, we have generous S/N performance. Additionally, for such short times a well-engineered laser will scarcely respond to the "garbage effects" of real life in the lab (temperature variations, power-supply variations, vibrations …) – within 1 μs these have not changed the system very much. The duration of the perturbations is too small for them to begin to wreak havoc with the stability of the frequency-defining cavity. So we actually can make useful measurements of the laser's phase in such a quick time frame that the problems are not yet apparent!

One begins to see a strategy coming up: We will quickly *measure* what the laser actually is doing, compared with our desired behavior, and then use feedback onto suitable actuators to *control* the laser's frequency. If we can make the corrections quickly enough and accurately enough, then the controlled laser will very closely approximate the ideal frequency-stable laser we need.

Implementation of this servo-control feedback concept is a multi-nuanced thing, in the perfection of which this author has invested something over 40 years of active work. It has led to a lot of interesting and useful electro-optic tools and techniques.

The Relative High Power of Lasers Empowers Nonlinear Spectroscopy and Sharp Resonances

Let's begin with the first approach to observation of narrow atomic resonances, using Saturated Absorption Spectroscopy. These phenomena were studied first within a laser cavity by Bill Bennett using the dispersion effects associated with the active Neon laser gas. Owing to the Doppler Effect, the Neon atom's natural resonance linewidth of ~10 MHz becomes masked and broadened to ~ 1500 MHz. Thus most of the gas atoms are detuned, and in a velocity-specific way. Some atoms have velocities near the special one giving the Doppler shift that will bring them into resonance with the intracavity laser field. Actually there are two such velocities to consider, since the laser beam goes both directions as it is bouncing back and forth between the mirrors. These resonant atoms will interact rather strongly with the field, leading to an increased decay rate for excited state atoms of that velocity – their inverted population gets converted into cavity photons! If we imagine a plot of the population difference (upper state minus lower state populations) we can expect to see a local and rather narrow dip around the velocity which is being converted from population inversion into light quanta. Actually there are the two mirror-symmetric dips as noted before. The interesting effects come when we let the laser frequency be tuned toward the atom's rest-frame frequency. Then the resonant atoms will have lower and lower Doppler velocities, until finally the selected velocity is zero. Now a new thing happens: when detuned, we had two groups of active atoms contributing their power to the laser output. When we reach the central tuning, both running-wave fields interact with a single atom velocity group. So with fewer atoms contributing, the laser

power decreases conspicuously, but only at the central tuning. This feature in the power output with laser tuning could be used for locking the laser to this central tuning dip, which is called "Lamb's dip" after Willis Lamb whose early theoretical work made clear this origin of the experimentally-observed effect. (His Nobel Prize in 1955 was for his work on the new sub-hyperfine structure in the Hydrogen spectrum.) As it turns out, operating pressures for optimum laser operation were rather large (~3 Torr, 400 Pa), which led to substantial probability of atom-atom collisions, even during the few 10's of ns optical lifetimes. So the Lamb dips would be broader and less deep, and had to be observed against a somewhat-peaked Doppler profile representing the distribution of available atom velocities. In addition to reducing the Lamb-dip contrast, significant frequency shifts were generated [17]. One could not arbitrarily reduce the gas pressure since the discharge pumping mechanism actually populated a metastable He* level, and collisions were needed to transfer this excitation to the Neon atom component in the discharge. So even though the wavelength of the laser's characteristic coherent light was more-readily-measurable than the incoherent light from the krypton discharge lamp (the existing wavelength standard), in fact the lasers' pressure shifts were simply too large to accept. Particularly this was the case since the discharge technology of the day led to important change of the fill gas pressure and species ratio with operation, due to electrode sputter-pumping.

The clearly important idea of separating the amplifier and the reference gas cells' functions was soon introduced by Lee and Skolnick. More discussion of those interesting developments is available elsewhere [18, 19], but for our present purposes we do need to consider some of the essentials. Since the purpose was to have a sheltered life for our reference atoms, it was attractive to be thinking in terms of *absorption*, rather than amplification. Then we didn't need any discharge or optical pumping of the reference quantum resonators. Of course, to be able to use Lamb's nonlinear resonance for frequency stabilization, we certainly needed to be able to tune the laser to the reference cell's resonance frequency. Nowadays, this is no big problem, by just using tunable lasers. At that time, the best idea to get a wavelength coincidence would be to use molecules as the absorbers – then we would have zillions of absorption lines to choose from. The modern champion for this approach is molecular Iodine, with narrow useful absorption lines from the Near IR down to ~500 nm. For other molecules,

utilizing transitions only between vibrational-rotational states, typical wavelengths are in the IR from ~2-10 micrometers range.

The first such dual-component optical frequency reference system, and still one of the better ones, uses a HeNe discharge cell to provide gain and laser oscillation at 3392 nm [18]. Also contained in the laser cavity is a cell containing CH_4 molecules, plain old tetrahedrally-symmetric methane, which has interesting lines that can be reached with the HeNe laser. To be brief, the necessary emitter/absorber spectral overlap is arranged by selection, based on good luck! The IR absorption band utilized, ν_3, is a strong fundamental vibration band, providing 0.18 cm^{-1} absorption coefficient per Torr. Of course having the absorber gas inside the cavity means we don't need very much absorption to have an impact on the laser dynamics – just a few percent would be fine, since it would then be roughly ½ the loss associated with the output-coupling mirror. At 10 milliTorr, the associated pressure broadening of the CH_4 resonance would then be ~160 kHz, similar to the 130 kHz broadening associated with the molecular free-flight through the intracavity light beam, of 0.3 mm typical diameter

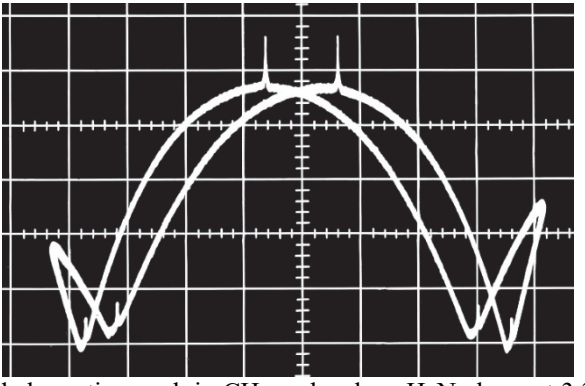

Figure 1. Saturated absorption peak in CH_4 molecules. HeNe laser at 3.39 μm is excited by rf discharge. CH_4 cell at 12 mTorr (16 mBar) is located inside laser cavity. Power output is 300 ~μW and peak contrast is ~12%. Peak width is ~270 kHz HWHM. At maximum power (~0.8 mW) contrast is ~15%. Cavity free spectral range is 250 MHz. Note cross-over resonances in two-mode region near cusps. Hysteresis of scan causes trace doubling.

Importantly, the pressure-induced *shift* turns out to be *very* small for these transitions, only ~1 kHz under these conditions.

So we are talking about a system with a resonance in the power curve of ~0.6 MHz FWHM, with perhaps 5% relative contrast on the total laser output of say 200 μW. A little calculation leads one to a

Signal/ShotNoise ratio ~10^6 in a 1 Hz measuring BW, while we're looking at the sub-MHz –wide peak produced at the central tuning, when both cavity running waves are bleaching the same absorbing molecules and thereby reducing the intracavity absorption losses. If this S/N were optimally used, the laser could be stabilized to have sub-Hz frequency deviations measured in 1 s intervals. In 1968 when this Saturated Absorption Optical Frequency Reference business began, our detectors and preamplifiers were not so good, and we didn't begin to approach the shot noise limit – that would have been a frequency (in)stability of ~2 x10^{-14} at 1s. Early on, we did get $\delta\nu/\nu$ ~1 x10^{-12}, which was soon improved to 3 x10^{-13} with better detectors and signal processing.

By locating the sample cell outside the laser resonator, the physical situation could be more-readily analyzed, and this arrangement was employed by Bordé, and by Hänsch, and by Chebotayev's group in early experiments. The interesting details are discussed in textbooks: see e.g. Letokhov & Chebotayev [20], Stenholm [21],and Levenson & Kano [22]. Now we consider the transit-time linewidth issue.

Free-flying Molecules see a Light Pulse: two views of the Uncertainty Principle

For these transitions, the radiative lifetime (~ ms) was much larger than the transit time of the essentially free-flying molecules in crossing the laser beam. At low pressure the saturated absorption linewidth was not collisionally nor Doppler limited, so it could be immediately observed that the resonance linewidths could be reduced by increasing the field/molecule interaction time. Larger beams helped. So did liquid Nitrogen cooling of the glass cell. So a serious study began to really understand the lineshape in the free-flight regime. Chebotayev and his colleagues developed the theory analytically near the low-pressure, low optical power limit [23]. The JILA theory was based on computer integrations of the Density Matrix for absorbers making a free transit through the assumed Gaussian light beam mode [24]. Low intensity and weak interactions were assumed to simplify the calculations, but soon it became clear that most of the observed signal would be contributed by a very small number of slow molecules. The theoretical result is a logarithmic cusp at the exact line center. With long interaction times, even a "weak" power would lead to saturation and other strong-field effects.

We need low velocity in the longitudinal direction so that the molecule wouldn't cross wavefronts axially, and thereby begin to develop Doppler-related phase modulation. Effectively molecules should fly perpendicular to the axis, and leave the wavefront after the transit with only <1 radian geometrical phaseshift. We also need low transverse velocities, since a longer transit time will be directly imaged into a narrower line. We can see $\delta\nu \bullet \tau \approx 1$ will yield $\delta\nu = \beta\, v_{th}/w_0$, $\delta\nu$ is the HWHM of the observable resonance, v_{th} is the thermal velocity, w_0 is the Gaussian beam radius, and β is a measured parameter. Experimentally we found $\beta\, v_{th} = 88$ kHz mm for Methane at room temperature. Laser mode radius w_0 values from 56 μm to 9 cm were measured, with corresponding HWHM values from 1.6 MHz down to 940 Hz. (The interesting substructure will be addressed momentarily.) First it is useful to consider the transit-time broadening in the Fourier-dual domain: angular divergence. Corresponding to a Gaussian beam radius w_0 there is a minimum angular divergence of the collimated laser beam of $\delta\theta = \lambda / 2\pi w_0$. The k-vector spread, particularly the non-axial components lead to a velocity-dependent Doppler shift of the *same* sign for both running waves, which will appear as broadening and shift of the resonance. Of course with a smaller mode diameter, the angular content is increased, and more broadening will appear spectrally.

While molecules typically do not have the "closed" optical transitions analogous to those needed for normal laser atom cooling, polar molecules *do* have a dipole moment. So with some electrical effort, one can arrange Sisyphus-like molecule slowing by switching the sign of the strong applied electric field, as shown by Meijer's group [25]. More recently Ye's group has achieved unprecedented high resolution microwave spectroscopy on Stark-slowed OH free radicals [26]. Certainly this will be an interesting frontier!

Other important directions are high sensitivity detection and improving the accuracy of locking to the molecular signals. For example some JILA work ("NICE-OHMS") shows a road to sensitivity increase by combining cavity enhancement and rf sideband techniques [27]. A fascinating physics avenue is the search for a parity-related frequency shift between suitable enantiomers [28]. Other important laser applications are considered in Svanberg's book. [29]

Momentum transfer from Light to Molecules – the Recoil Splitting

A full treatment of radiative interactions must include the field and molecular momenta, as well as the photon numbers and internal states of the quantum system. Such a treatment is essential for the case of pumping atoms with closed energy levels, which can allow the repeated interactions and deep velocity cooling celebrated by the 1997 Atom Cooling Prize of Phillips, Chu, and Cohen-Tannoudji. For the molecular sample of interest here, there are many decay channels, and likely even impact on the vacuum chamber walls before any particular molecule reappears in the laser fields: so a single interaction picture is reasonable. A clear observation of the transfer of momentum from field to atomic system is available with Saturated Absorption Spectroscopy, basically because it is a two-step process. Let's consider absorbers that initially have essentially zero velocity along the light beam. Then the left-running light beam can be tuned to $\nu = \nu_0\left(1+\frac{h\nu}{2Mc^2}\right)$, the extra (recoil) energy being needed beyond the transition energy ν_0 to provide the kinetic energy associated with the recoil momentum the molecule will have after the transition occurs. The opposite-running beam will also deplete this zero-velocity group. So at this resonance tuning, the resulting nonlinear decrease of molecular opacity will lead to a peak in the transmission spectrum, and it is shifted slightly to the blue of the rest frequency. Another interesting case occurs when the molecules have a velocity $v = h/M\lambda$, *i.e.*, there is enough molecular momentum initially so that when the red-detuned laser interacts with this molecule, the photon and molecular momenta just cancel, and the original kinetic energy can make up for the photon's energy deficit. The result is an excited molecule with zero axial velocity. Now the laser beam in the other running direction will experience amplification from this particular tuning condition, again leading to a relative peak in the sample's transmission. With the molecule initially possessing some kinetic energy, the laser tuning for this upper-state resonant condition will be $\nu = \nu_0\left(1-\frac{h\nu}{2Mc^2}\right)$. So considering photon recoil, the nonlinear interaction is associated with either the ground or excited state population being accessed by both beams for the same detuning, namely zero velocity in either one of the two states. For methane

the splitting between the two peaks is 2.163 kHz and may be seen clearly in Figure 2 [30].

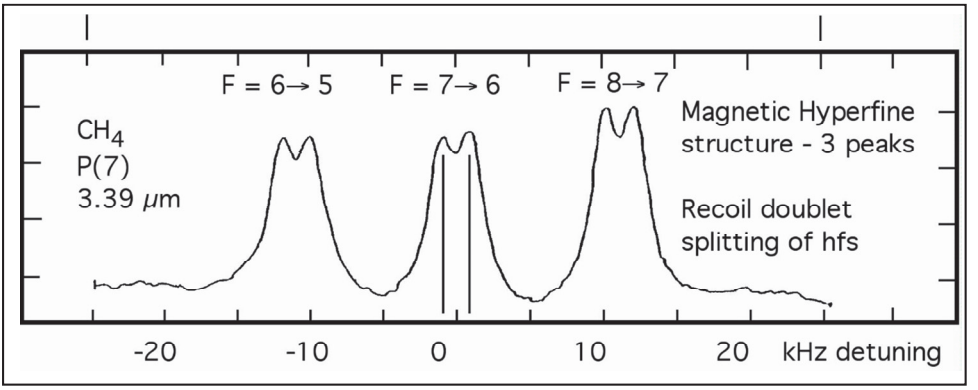

Figure 2. Recoil Splittings of Hyperfine-Structure Peaks in free-flight Methane Molecules. The vertical strokes indicate the positions of the two recoil components in one of the Hyperfine components.

While the JILA and University of Paris Nord work exploited mainly the large diameter optical beams to gain a longer molecular interaction time, Chebotayev, Bagayev, and colleagues in the Novosibirsk group made good use also of another physical idea, namely the use of super-slow molecules to contribute the main part of the observed signal. In this way an additional 20-fold linewidth reduction to <50 Hz was achieved [31]. An important aspect of this approach is that the total 3-D effective molecular temperature is below 0.1 K, leading to a much-reduced second-order Doppler shift, of << 1 Hz. An average velocity 13x below thermal for slow C_2HD molecules was shown by Ye et al. [27], and was feasible only because of the very large sensitivity provided by the NICE-OHMS technique.

Other Optical Frequency References Based on NonLinear Spectroscopy

Many research groups have been attracted to working with laser stabilization for Measurement Standards applications, such as interferometric calibration of gage blocks that serve to check reference standards used by industry. For this kind of application it is highly desirable that the reference laser beam be visible, as well as stable enough and reproducible enough. A huge success in this area is the 633 nm HeNe laser with an intracavity Iodine cell, and well developed systems of this type are even available commercially. This HeNe/I_2 system was the one whose frequency was measured by the NBS efforts in the early 1980's, with an

uncertainty of 70 kHz. (Being the first measurement of such a visible system, it is perhaps understandable that several of the uncertainties were far from fundamental in their origins.) Other labs joined in and over the next decade many labs gained experience and a few had frequency measurements confirming the NBS result. Slowly it became acceptable to reconsider the definition of the International Unit of Length, the SI Metre.

As may be seen, the world of spectroscopy offers us an unending garden of fascinating details. Presumably Parity-Non-Conservation will lead to a next generation of fine structures in chiral molecules, particularly with the development of cold-molecule techniques. But enough about the "ticks" of the clock: now we must return to the main story, the development of frequency stabilization and cycle-counting measurement tools – The inside Gear-Works of the Optical Clock!

Measuring Optical Frequencies with Optical Combs

The Metre redefinition of 1983 was not really a kindness to metrologists tasked with actually measuring some physical parts, because the practical methods for application to measurements were not yet spelled out. But it was a *boon* to the metrology researchers: it became their task to explore just which good stabilized laser system would have the optimal properties for precision interferometry, for outdoor surveying, for servo-loop guidance of milling machines, for … ? So within a dozen years after the redefinition there were at least 10 well-developed optical frequency standards, as illustrated in Fig. 3.

As may be seen in Fig. 3, there are stable frequency sources available from roughly 10 μm (30 THz) to ~280 nm (~1PHz), well beyond the visible range. It was striking that the difference between lines were surprisingly similar frequency intervals, ~88 THz, approximately the frequency of the CH_4 - stabilized laser. This led to schemes where doubled frequency of one laser would be compared with the sum of the two straddling lasers. Some "pocket change" of frequency, a few THz, could be synthesized as sidebands using a Kourogi comb, based on a microwave modulator in a cavity whose length provided resonance enhancement of all the generated sidebands [32]. In such a way we measured the 532 nm Iodine standard in terms of the difference of frequency between twice the HeNe Iodine system at 633 nm, and the Rb two photon line at 782 nm.[33]

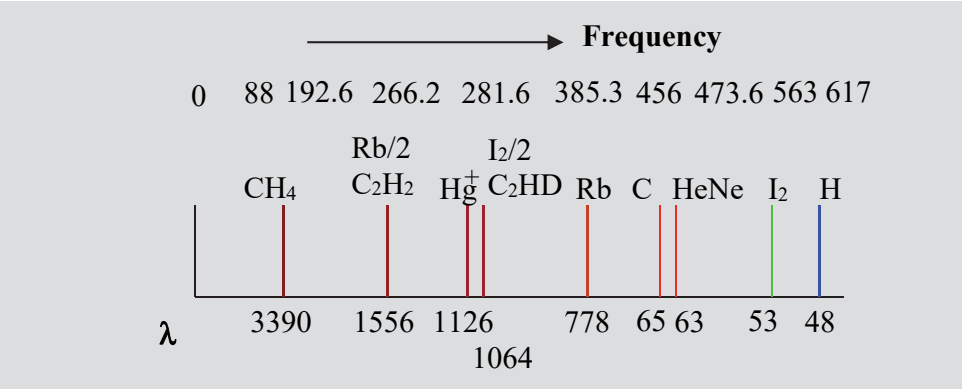

Figure 3. Stable Lasers based on NonLinear Doppler-free Resonances in Gases (1995). The frequency axis (above) is in THz units, the wavelength scale (below) is in nm.

This was our introduction to the elegance of having an optical comb – a coherent ensemble of spectral lines whose frequencies are accurately represented by a simple formula. Our system covered just a few nm wavelength. How sweet it would be to cover the entire visible band, giving several million accurately known frequency reference lines all at once!

One way to broaden this Kourogi comb's spectral width would be to provide intracavity gain, to compensate the modulator's optical losses, a scheme which was demonstrated by Diddams using an OPO crystal also inside the resonator. Oscillation and generation of hundreds of FM sidebands were easily observed [34]. For some tuning conditions the phase of the several spectral components led to pulse generation, rather than pure FM emission. In many ways this was just the hard way to do what the Ultra-Fast Laser scientists appreciated about the Ti:Sapphire self-mode-locked lasers: stable, self-organized, ultra-short high repetition rate pulse trains. Elsewhere our group's papers discuss the technical richness of these lasers and the comb business [35]. This is just one further note about the mutual coupling between "independent" research streams: we switched to Ti:Sapphire fs lasers and never looked back.

Coincidentally, in these final days of the last Millennium, this laser community received a fundamentally-important gift from the laser industry. There would probably be no widely-used frequency combs without it. This "gift" was the introduction of high-power visible lasers, based on frequency-doubling the output of a laser-diode-pumped Nd solid state laser. These were immediately put to use replacing the fussy and quite noisy

Argon Ion laser in wide use for pumping the Solid State lasers. Competitive forces led these new pump lasers to be well engineered, with intensity stabilization to yield exceedingly low levels of residual amplitude noise. This property is crucial because of the way a self-mode-locked laser operates – these Ti:Sapphire lasers are self-mode-locked by a self-induced optical lens which makes the cavity less lossy when the laser modes are all synchronized to form an "optical bullet" in the laser medium [36]. This temporary lens is formed by the radial index gradient, induced and present only if a light bullet is present. So the laser cavity is originally set up to need this extra focusing to produce low-loss cavity modes, and after the laser is started in the pulse regime, stable self-mode-locking is maintained. Consider that the pulse lengths are only ~10 fs, while the repetition periods are ~10 ns. With ideal synchronization, the peak power/average power ratio is $\sim 10^6$. A typical laser will emit ~0.5 W through an output mirror of 5% transmission. So we have 10 W average internal power, and 10 MW peak power, which is focused to a ~14 μm radius spot in the Ti:Sapphire laser crystal. This active area is only 3×10^{-6} cm^2, so with 10 MW peak power we have 3 TW/cm^2! The associated electric field is ~10% of the interatomic fields in the crystal, so it is not so surprising that a significant optically-induced increase of the index of refraction occurs (optical Kerr effect). The low amplitude noise of the pump laser is now seen to be critical: an intensity-dependent phase-shift though the laser crystal will produce amplitude → frequency conversion and thus unacceptable phase noise if the pump is noisy. In a good case the linewidth of laser comb-lines without frequency control is ~3 – 10 kHz due to this cause, before the servo is used. Details of the process have been studied [37].

So the pulse train leaving the laser is of ~500 kW peak power, much of which we will focus into the special nonlinear fibers that brought in the age of the Optical Comb. Because of the microstructure design of the fiber, full light guiding is possible even with fiber core sizes of 1.5 -2 μm diameter. So now when we estimate the fiber's active area, it is roughly 200-fold smaller than the laser's, while the power level is ~20-fold lower. The 10-fold higher intensity produces a 3-fold higher electric field in the silica fiber, being now essentially comparable with interatomic field and setting the stage for SERIOUS NonLinear interactions. Forget Mr. Taylor's expansion here: this is strong signal NonLinear physics! All frequency components from the laser are mixed with each other, resulting in a drastic

spectral broadening. By the fiber's optical design, a broad range of optical frequencies can travel through the fiber with little speed variation, which allows these frequency conversion processes to remain phase-matched and accumulate power into the newly created frequencies. Essentially, in a few cm of length, the input spectrum is converted to white light and covers an octave or more of optical bandwidth. Actually the light is not quite "white" since it still carries the basic heartbeat of the original fs laser, for example 100 MHz. As explained previously, this intrinsically generates a comb spectrum with component widths just connected to the spectral resolving power employed. Eventually, at the kHz level and below, the broadly-active phase modulation processes that affect all lasers will broaden these lines also (before the servo control is ON).

Complementarity, Cooperation, and Competition

The Basics

The remarkable insights of Professor Hänsch's Stanford work [38] were published in ~1978, and already demonstrated using a repetitively-pulsing laser to generate an optical comb which could serve as a spectral ruler. However the bandwidth of the covered spectrum was too small for general frequency measurements – only a GHz or two. Since these intervals could be spanned in other ways, the methods were not widely adopted. Basically there was not a technical growth path available at the time. Principle, yes; Tool, no.

The hard work, straight-ahead "government" approach to frequency measurement had been demonstrated at NBS in 1972 [39], following the pioneering work of Ali Javan's MIT frequency measurement group (See references in [40]). But this was a heroic effort and mainly only national standards laboratories took much interest. Laser after different laser had to be lined up and frequency-related to the doubled frequency of its predecessor, to step-by-step build up the frequency measurement chain. This kind of work required development of frequency- and phase-locking schemes now in wide use. We also got a "one-of-a-kind" physical result, a single laser frequency was measured by the cooperative and extended work of the NBS group [41]. But it was enough to get the Metre redefinition process started.

The Divide and Conquer Scheme

In a notable paper (1990), Professor Hänsch and his colleagues suggested an excellent way to simplify the frequency chains: one should use the *difference frequencies* between lasers as the entities that were harmonically marching up the spectrum [42]. In this way, the ensemble of lasers would all have nearly the same wavelength, and could be built essentially by duplication of a basic diode laser unit. Then with nonlinear crystals, fast photodetectors, and suitable phase-locking electronics one could progress from microwaves to optical frequencies. This system also felt rather elaborate and specialized, but was used with good results in Garching. A related strategy was developed at NRC [43], based on difference frequencies, using CO_2 lasers. Inspecting such a system, one came to see that the first 9 or 10 of the 14 stages served only to get the frequency up into the low THz range.

Then in 1994 came Kourogi and Ohtsu's multiply-resonant cavity approach, allowing one to reach a few THz in a single step [32]. Eventually the buildup of phase noise – according to the high harmonic of the original microwave source – would have been a problem in going into the visible range. But the fs laser Comb arrived and offers an easier and better way. See below.

A Brief History of the Optical Miracle of 1999 - 2000

Fibers for Spectral Broadening

By now the JILA group had accepted the fs laser as a great source of pulsed laser light. Ours had ~ 80 nm bandwidth at 800 nm. But the optical frequency standards we wanted to connect were at 1064 nm (fundamental of Iodine-stabilized Nd laser) and 778 nm (Rb two-photon-stabilized diode laser). An ordinary communication fiber was found to be just barely capable of spectral broadening the necessary amount – 104 THz. This paper was submitted at the end of September 1999 [44].

MicroStructure Fibers for Serious Nonlinearity

The Conference on Laser and Electro-Optics of June 1999 had a spectacular post deadline presentation by a Bell Labs team [45], wherein a normal fs laser pulse evolved its color in a dramatic way in propagating through a few meters of a special fiber. Such a fiber *did* make collimated

white light, in the form of stably-repeating pulses, just as Ted Hänsch had postulated for his (unpublished) frequency measurement proposal. Using that previously-unknown light source, most of the rest should be possible. (Seeing the repetitively-pulsed laser-like white light the fiber generated instantly convinced me that Ted's Concept actually could be a real and physical possibility! Without a repetitive white-light laser, there was no chance.) Lengthy appeals for scientific collaboration with the fiber owners' organization ultimately became irrelevant due to the miraculous appearance in JILA of a sample of this Magic Fiber. The concept of "band-gap" or "Photonic-Crystal" fibers was introduced in 1996 by Knight et al., pointing out the possibility of controlling the spatial modes and effective group velocity dispersion by the mechanical design of the air holes [46]. Our first JILA experiments were made using microstructured fiber drawn from a preform prepared on September 10, 1997 by Robert S. Windeler of Bell Labs [47], using a construction technique of his own devising. A broad range of fiber designs was investigated in Bath, UK, by P St. J Russell and colleagues.

The Race is ON

Of course in JILA we didn't know that the Garching team had already gone from a plan to the first demonstration of a comb-based phase coherent link from microwaves to the visible, and had submitted their *Phys. Rev. Letter* in November 1999. Even before we got the Magic Fiber! They used a comb of somewhat limited bandwidth, 44 THz, but their divider stages could connect the optical frequency with the 28^{th} harmonic of the difference between the comb's edges. It is a beautiful result, and appeared finally on 10 April 2000 [48]. In the meantime the JILA team was working hard with the Magic Fiber's white light output to implement and demonstrate our phase-coherent locking of the carrier-envelope offset frequency in terms of the laser's repetition rate. Our Disclosure of the scheme called this "Self-Referencing". The control electronics we built had a digital click switch so the phase could be set on any integer multiple of 1/16 of a phase-slip cycle per pulse. The JILA experimental demonstration was based on interferometrically determining the carrier-envelope phase difference between two optical pulses, separated by one intervening pulse. Finally the new electronics worked, the experimental data were clear and our report [49] appeared in *Science* on 28 April 2000. A *PRL* joint article

celebrating the success of the combined Garching, Bell Labs, and JILA teams appeared on 29 May 2000 [50]. Within the next year there was an avalanche of absolute optical frequency measurements from labs all over the world. This was a glorious chapter in optical physics history, in no small part because of the high mutual respect of the two teams for each other, aided by the complete openness fostered by the frequent exchange of postdocs Scott Diddams and Thomas Udem between the two hotly-competing groups.

Some Frequency Measurement Results

Many laser frequency standards were being actively studied worldwide so that, when the Comb breakthrough came, there were many things to be accurately measured – many for the first time. A few of the world-wide results include the work shown in the following Table.

Ca	657 nm	Schnatz	PTB	PRL	1 Jan ‘96
Rb	780 nm	Ye	JILA	Opt. Lett.	August ‘96
C_2H_2	1500 nm	Nakagawa	NRLM	JOSA-B	Dec ‘96
I_2	532 nm	Hall	JILA	IEEE Instr,Meas	April ‘99
Sr^+	674 nm	Bernard	NRC	PRL	19 Apr ‘99
In^+	236 nm	v. Zanthier	MPQ	Opt.Comm.	Aug’99
H	243 nm	Reichert	MPQ	PRL	10 Apr ’00
Rb	778 nm	D. Jones	JILA	Science	28 Apr '00
I_2	532 nm	Diddams	JILA	PRL	29 May ’00
H	243 nm	Niering	MPQ	PRL	12 June ‘00
Yb^+	467 nm	Roberts	NPL	PRA	7 July ‘00
In^+	236 nm	v. Zanthier	MPQ	Opt. Lett.	1 Dec.‘00
Ca	657 nm	Stenger	PTB	PRA	17 Jan ‘01
Hg^+	282 nm	Udem	NIST	PRL	28 May ‘01
Ca	657 nm	Udem	NIST	PRL	28 May ‘01
Yb^+	435 nm	Stenger	PTB	Opt. Lett.	5 Oct ‘01

Table I. Measured Optical Frequencies. The reference atom/molecule and its transition wavelength are indicated, followed by lead author and institution, the journal name and date. The first fs Comb measurement was Hydrogen, by Reichert *et al*. The first direct fs optical measurements were by the JILA team (Jones). Note the brevity of time between publications!

The comb technology spread explosively in 2000, bringing vast simplification of optical frequency measurements, along with a steady improvement in the accuracy. Very soon after the initial measurements, it has become the case that the comb’s measurement precision can exceed that of the standards being measured. Recent tests at NIST, BIPM, and ECNU

[51] confirm the earlier MPQ experiments [52] showing that the comb principle is strictly correct up to a measurement precision of more than 18 digits.

Molecular Iodine Optical Frequency Standard

The Iodine-stabilized Nd:YAG laser is a sweet spot in the stabilized laser domain, counting on its excellent performance and relative simplicity. One system was made in Japan that met airlines cabin baggage limitations and still delivered excellent performance [53]. Because of the Iodine's great atomic mass, the second-order Doppler correction for this system is only ~5 $x10^{-13}$ and it is likely that independent reproducibility perhaps 5-fold superior to this can come with improved technical realizations. In particular, providing an offset-free modulation strategy is still a challenge. The advantage of this system is its compactness and potentially reasonable cost. Taken with an optical comb, one can have an attractive clock [54]. See Fig. 4. The frequency (in-)stability of all the 1 million optical comb lines is ~ 4 $x10^{-14}$ /$\sqrt{\tau}$.

Recently stable Yb:YAG single frequency lasers became available, with output tunable to 1029 nm. When frequency doubled, excellent stabilization performance should be possible with the I_2 transitions at 514.5 nm, considering that the linewidth is at least 5-fold smaller than for the 532 nm line [55]. Single frequency fiber systems can also offer this wavelength.

So What Comes Next?

In addition to the simplification of optical frequency measurements, the resulting new capabilities are unbelievably rich in terms of the tools and capabilities that have been created, and these in turn are reinforcing progress in these contributing fields. This paper can't even attempt to present a myriad of delicious physical effects, which are normally understood as being in different fields, but which in their now-unified relationships can be seen as creating a truly remarkable and enabling advance of the research tools available in optical science. But let me still give a few examples.

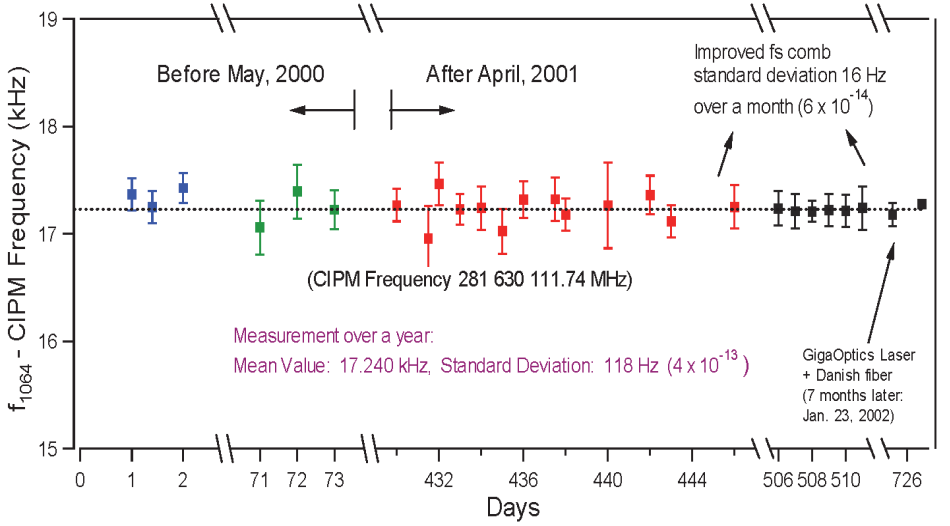

Fig 4. Long-term frequency stability of Iodine-based Optical Clock. This figure conveys the refinement and small frequency offset of this stable optical clock's frequency from previous, much less accurate measurements. With improved technology in 2002 the uncertainty was further reduced to ~6 x10^{-14}.

After the frenzy of Generation I frequency measurements of Table I, some of the Generation II comb applications in Jun Ye's group include: low-jitter time synchronization (~fs) between ultrafast laser sources [56]; coherent stitching-together the spectrum of separate fs laser sources so as to spectrally broaden and temporally shorten the composite pulse [57]; precision measurement of optical nonlinearities using the phase measurement sensitivity of rf techniques [58] ; coherently storing a few hundred sequential pulses and then extracting their combined energy to generate correspondingly more intense pulses at a lower repetition rate [59]; and searching for a change in the physical constants by the Garching team [60]. Exciting topics of research for Generation III applications now include connecting optical frequency interim standards at the sub-Hz level (in spite of their different locations spectrally and physically), allowing precise remote synchronization of accelerator cavity fields, providing stable reference oscillators for Large Array Microwave Telescopes, and potentially reducing the relative phase-noise of the oscillator references used for deep space telescope arrays (NASA, VLBI …) That's part of the first five years.

And the next projects? What about 14.4 keV comb-line harmonics to look at Mössbauer ^{57}Fe nuclear resonances? Another sharp line is in ^{181}Ta at 6.2 keV. How about parallel processing to determine biological activity of a candidate drug, by means of CARS using synchronized pulse lasers to excite specific ligand Raman resonances of a single molecule that was attracted to and stuck by a particular test protein patch on a surface?

In a larger framework, we now find ourselves at an almost unique point in the development of Science, where we the have remarkable ability to "understand" practically all phenomena, to compute accurate predictions from our equations, and to integrate a variety of details into our models. Consider for example the GPS system, in which different kinds of physics such as gravity and relativity are successfully merged with our sophisticated atomic clocks – not to forget satellite dynamics and radio engineering and computer software – so that in the total we have a coherent and highly useful practical tool. Remarkably, the system is simple for the end user to apply. We must count this GPS achievement as one of the all-time ultimate technical success levels ever achieved.

The work recognized by the 2005 Nobel Physics Prize represents entry of another dramatic, major and enabling advance, and one which we can expect to show some flavors of the same breadth and character just noted regarding GPS. But in these first moments after its birth, our opto-electronic technology is new and is barely illustrated, not much beyond the first cases of interest to frequency-standards people and metrologists. We know that the *accuracy* of optical frequency measurements is now limited to "just" 15 digits by the present microwave standard of frequency, but the "Comb" technology actually allows two optical frequencies to be compared with several orders of magnitude more precision. If the history of physics is any guide, we realistically can expect to find some nice surprises ahead as these capabilities become even more widespread, and are applied to ingenious fundamental measurements by a growing and imaginative community of "fundamental physics" scientists. After considering all the known progress in Science, would *you* bet that we have already opened the Russian Matryoshka doll of Nature and already found the ultimate inside limit?

Acknowledgements

The joy of interacting with superb young scientists is clearly one of life's treasures. Among these many I must single out for special thanks Jim Bergquist, Leo Hollberg, Miao Zhu, and Jun Ye for their enthusiasm and unique contributions to the JILA program. The NIST management is enthusiastically thanked for accepting and sponsoring over the years a series of risky proposals in Laser Spectroscopy. Particularly Leo Hollberg, Steve Cundiff and I were glad they accepted the fs comb frequency synthesizer proposal in Spring 1999. Scott Diddams and David Jones were the excellent colleagues additionally involved in these experiments, and JILA's research force was hugely expanded when Jun Ye rejoined JILA in 1999 to start his own group. As always, whenever Professor Long Sheng Ma was visiting us from Shanghai our pace was strongly advanced. Visiting Scientists such as Christian Bordé continue to be collaborators even three decades after their JILA time. I am pleased and grateful to acknowledge that the work has been supported in part by the NSF, ONR, AFOSR, and NASA, and for over 4 decades by the NIST. I benefited greatly from the knowledge and generous sharing of ideas and opportunities by my NBS mentor, Peter L. Bender. Above all I am indebted to my patient and insightful lifetime friend Lindy Hall for her understanding, her great efforts and contributions to this scientific work and, more importantly, to our joyous and fun life together. It has been wonderful in the course of these 45 years to see a progression of experiments and technical advances make possible this ultimate payoff in the optical comb. Now we're discussing if it's almost time to clean my office at JILA and pack up into a new camper unit to go out and further explore another domain in the world.

Appendix: The Full Comb Story in an Undergraduate's World

I'm glad you asked how to think about frequency combs. Suppose you have a sinewave voltage or field. Then a plot in time shows a smooth oscillation and a plot in frequency shows a single Fourier component, namely a sharp line. Now add a few harmonics onto this wave. The spectrum now has a few more lines at exact harmonic frequencies, while the time picture has a rather complicated shape. By adjusting the phases of the harmonics, we can begin to synthesize some disturbance in time that begins to remind one of a pulse, or more exactly, a series of identical pulses. Carry this a step forward by having a large number of harmonics. The more we

add, the sharper is the pulse we can synthesize, and of course the richer is the spectrum of this wave. Going further in this direction of adding coherent harmonics, the spectrum now has zillions of spectral lines, all at the harmonics of our original sinewave. Carrying this concept to the visible will require a few million (10^6) harmonics for a source with 100 MHz basic repetition rate. With the proper phase adjustments, the time domain pulse can be 10^6 times sharper in time than the original sinewave. So we can expect really narrow temporal pulses, and really wide spectral bandwidths.

This situation fits well with what we would expect from Fourier analysis of a single pulse: such an impulse will have Fourier components at all frequencies, with their nearly-constant amplitudes gradually decreasing for frequencies above the reciprocal temporal pulsewidth. If we have a repetitive pulse train in time, but insist to ask about its spectrum, we will need an analyzer with a narrower passband compared with the repetition frequency, otherwise it couldn't resolve the harmonic structure. But a narrow spectral passband corresponds to a long temporal response time. So the output of the spectrometer at any particular wavelength or frequency setting will be the result of coherent addition of the contributions of many pulses. While an individual pulse has a broad and continuous spectrum, when we coherently add their spectral amplitudes we can expect to have interferences that will modulate the spectrum. Adding more pulses temporally (narrower spectral resolution) will give deeper modulation. Eventually we arrive at very sharp spectral lines, evenly arranged as Fourier harmonics. Until we encounter technical issues such as phase-noise of the repetition rate, the sharper an analysis resolution we apply to the waveform, the sharper will be the spectral lines we observe. So the spectrum does indeed remind one of a "comb." You can demonstrate these ideas safely at home for yourself easily in the electronics domain, but of course the optical and electronics worlds *should* work the same …

In fact, with the fs lasers used to generate these pulses, there is one more little item of interest. That is that the laser can oscillate in any one of its cavity modes, defined by having a repeating phase after going one loop around the cavity. All the many modes involved have their own longitudinal quantum numbers, essentially how many full optical cycles are contained in the closed loop. This calculation clearly involves the wavelength-dependent phase velocity, and some average of the propagation through the many

optical components. Another reality is that the laser operates in a self-organized repetitive pulsing mode. Effectively the laser's optical losses can be made large enough to inhibit laser action, unless all the cavity modes can adjust their phases to synthesize a delta-function spatially. The critical thing is to have a short pulse when passing through the Ti:Sapphire crystal, since the short pulse will correspond to very high peak power, and that will interact with the laser rod's material in a quadratic way (optical Kerr effect) to produce a positive lens: a bigger index on the axis where the intensity is maximum. So the self-organized pulse situation is stable in which the laser's cavity has a high diffraction-loss (doesn't have quite enough positive lens power), but the losses are periodically remedied by a bullet of light which uses its self-action on the crystal to produce the needed extra refraction that makes the cavity losses be suitably less.

Now the pulse envelope that describes this light "bullet" results from superposition of many cavity modes, and the shape will evolve if there are temporal delay differences with wavelength. We are now just discussing the group velocity concept, whereby the shape of a disturbance will evolve unless all the frequencies have the same propagation speed. In the physical laser we must include some optical elements specifically to deal with the fact that blue light in the laser crystal will travel more slowly than red light. To get the shortest pulses the time delays around the loop need to be essentially the same, although you can see this becomes a little complicated in that the laser pulses themselves act to influence the time delays. In any case, the light which comes out of the laser's coupling mirror will be a regular time series of sharp pulses, and will display a comb-like structure under frequency analysis. However the underlying fast optical oscillations will in general have a different phase each time the pulse hits the mirror's surface. The fast oscillation's phase will shift a bit forward or backward from one pulse to the next, and so the optical frequency comb may be offset a bit from the strictly Fourier harmonic case we first imagined. The usual case is a constant phase shift for each pulse, and so a constant rate of accumulating a phase beyond the repetition-rate's harmonic. We have developed an electro-optic scheme called "self-referencing" in which this additional frequency, the Carrier-Envelope Offset Frequency, is stably locked to the repetition frequency in a digital ratio. For example one could choose zero for the setpoint ratio and thereby have a strictly harmonic comb. With the offset = 1/2, one generates a comb offset by 1/2 the basic repetition

rate, which itself is of course the frequency comb's tooth spacing. See Refs [48-52].

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Nobel Lecture: Superposition, Entanglement, and Raising Schrödinger's Cat[1]

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Contents

I. Introduction

EXPERIMENTAL CONTROL OF QUANTUM SYSTEMS has been pursued widely since the invention of quantum mechanics. In the first part of the 20th century, atomic physics helped provide a test bed for quantum mechanics through studies of atoms' internal energy differences and their interaction with radiation. The advent of spectrally pure, tunable radiation sources such as microwave oscillators and lasers dramatically improved these studies by enabling the coherent control of atoms' internal states to deterministically prepare superposition states, as, for example, in the

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Ramsey method (Ramsey, 1990). More recently this control has been extended to the external (motional) states of atoms. Laser cooling and other refrigeration techniques have provided the initial states for a number of interesting studies, such as Bose-Einstein condensation. Similarly, control of the quantum states of artificial atoms in the context of condensed-matter systems is achieved in many laboratories throughout the world. To give proper recognition to all of these works would be a daunting task; therefore, I will restrict these notes to experiments on quantum control of internal and external states of trapped atomic ions.

The precise manipulation of any system requires low-noise controls and isolation of the system from its environment. Of course the controls can be regarded as part of the environment, so we mean that the system must be isolated from the uncontrolled or noisy parts of the environment. A simple example of quantum control comes from nuclear magnetic resonance, where the spins of a macroscopic ensemble of protons in the state $|\downarrow\rangle$ (spin antiparallel to an applied magnetic field) can be deterministically placed in a superposition state $\alpha|\downarrow\rangle+\beta|\uparrow\rangle\left(|\alpha|^2+|\beta|^2=1\right)$ by application of a resonant rf field for a specified duration. Although the ensemble is macroscopic, in this example each spin is independent of the others and behaves as an individual quantum system.

But already in 1935, Erwin Schrödinger (Schrödinger, 1935) realized that, in principle, quantum mechanics should apply to a macroscopic system in a more complex way, which could then lead to bizarre consequences. In his specific example, the system is composed of a single radioactive particle and a cat placed together with a mechanism such that if the particle decays, poison is released, which kills the cat. Quantum mechanically we represent the quantum states of the radioactive particle as undecayed $=|\uparrow\rangle$ or decayed $=|\downarrow\rangle$ and live and dead states of the cat as $|L\rangle$ and $|D\rangle$. If the system is initialized in the state represented by the wave function $|\uparrow\rangle|L\rangle$, then after a duration equal to the half-life of the particle, quantum mechanics says the system

evolves to a superposition state where the cat is alive and dead simultaneously, expressed by the superposition wave function

$$\Psi = \frac{1}{\sqrt{2}}\left[\left|\uparrow\right\rangle\left|L\right\rangle + \left|\downarrow\right\rangle\left|D\right\rangle\right]. \quad (1)$$

Schrödinger dubbed this an entangled state because the state of the particle is correlated with the state of the cat. That is, upon measurement, if the particle is observed to be undecayed, one can say with certainty that the cat is alive, and *vice versa*. But before measurement, the particle and cat exist in both states. This extrapolation of quantum mechanics from individual quantum systems to the macroscopic world bothered Schrödinger (and a lot of other people). As one way out of the dilemma, in 1952, Schrödinger (Schrödinger, 1952b) wrote

> "… we never experiment with just one electron or atom or (small) molecule. In thought experiments, we sometimes assume that we do; this invariably entails ridiculous consequences..."

But of course these days, this argument doesn't hold and we can in fact experiment with individual or small numbers of quantum systems, deterministically preparing superpositions and entangled superpositions. Our control is best when we deal with very small numbers of particles, which enables us to realize many of the gedanken experiments that provided the basis for discussions between Schrödinger and the other founders of quantum mechanics. And, we can also make small analogs of Schrödinger's cat, which are by no means macroscopic but have the same basic attributes. So far, it appears that our inability to make macroscopic "cats" is due just to technical, not fundamental, limitations. Admittedly, these technical limitations are formidable, but one can be optimistic about increasing the size of these states as technology continues to improve.

This contribution is based on the lecture I gave at the Nobel ceremonies in 2012. It is mostly a story about our group at the National Institute of Standards and Technology (NIST) in Boulder, Colorado, whose combined efforts were responsible for some of the contributions to the field of trapped-ion quantum control. It will be a somewhat personal tour, giving my perspective of the development of the field, while trying to

acknowledge some of the important contributions of others. For me, the story started when I was a graduate student.

II. Some Early Steps toward Quantum Control

From 1965 to 1970, I was a graduate student in Norman Ramsey's group at Harvard. Norman, with his close colleague Dan Kleppner and student Mark Goldenberg, had recently invented and demonstrated the first hydrogen masers (Goldenberg, Kleppner, and Ramsey, 1960; Kleppner, Goldenberg, and Ramsey, 1962). As part of this program, Norman wanted to make precise measurements of the hyperfine frequencies of all three isotopes of hydrogen, so I chose to work on deuterium. The experiment was relatively straight- forward, complicated a bit by the relatively long wavelength (~ 92 cm) of deuterium's hyperfine transition relative to that of hydrogen (~ 21 cm) (Wineland and Ramsey, 1972). Most importantly, this experiment taught me to pay close attention to, and control as best as possible, all environmental effects that would shift the measured transition frequency from that found for an isolated atom. In addition to enjoying the detective work involved in this, I also became hooked on the aesthetics of long coherence times of superposition states (~ 1 s in the masers), and their importance in atomic clocks. Norman received the 1989 Nobel Prize in physics for his invention of the separated-fields method in spectroscopy and development of the hydrogen maser (Ramsey, 1990).

During my time as a graduate student, I also read about and was intrigued by the experiments of Hans Dehmelt and his colleagues Norval Fortson, Fouad Major, and Hans Schuessler at the University of Washington. The trapping of ions at high vacuum presented some nice advantages for precision spectroscopy, including the elimination of the first-order Doppler shifts and relatively small collision shifts. The Washington group made high-resolution measurements of the $^3He^+$ hyperfine transition, which has internal structure analogous to hydrogen, by storing the ions in an rf (Paul) trap. One challenge was that detection by optical pumping was (and still is) not feasible because of the short wavelengths required. Therefore, in a heroic set of experiments, state preparation was accomplished through charge exchange with a polarized Cs beam that passed through the ions. Detection was accomplished through a charge-transfer process $\left(^3He^+ + Cs \rightarrow {}^3He + Cs^+\right)$ that depended on the internal

state of $^3He^+$, followed by detection of the depleted $^3He^+$ ion number by observing the ions' induced currents in the trap electrodes (Fortson, Major, and Dehmelt, 1966; Schuessler, Fortson, and Dehmelt, 1969).

Although these experiments were what first attracted me to ion trapping, my postdoctoral research with Dehmelt, starting in the fall of 1970, was focused on experiments where collections of electrons were confined in a Penning trap for a precise measurement of the electron's magnetic moment or *g* factor. These experiments were started by Dehmelt's graduate student, Fred Walls, who later became a colleague at the National Bureau of Standards. After a while, it became clear that systematic effects would be much better controlled if the experiment could be performed on single electrons. Therefore, the first task was to isolate a single trapped electron. This was accomplished by first loading a small number of electrons into the trap and driving their nearly harmonic motion (~ 60 MHz) along the magnetic field direction. This motion could be detected by observing the currents induced in the electrodes (proportional to the number of electrons). By adjusting the strength of the drive to a critical level, occasionally one of the electrons would gain enough energy to strike a trap electrode and be lost. Steps in the induced current level could then be used to determine when one electron was confined in the trap (Wineland, Ekstrom, and Dehmelt, 1973). Subsequent experiments on single electrons by Robert Van Dyck, Paul Schwinberg, and Dehmelt were used to make precision measurements of the electron's *g* factor (Van Dyck, Schwinberg, and Dehmelt, 1977; Dehmelt, 1990). For this and the development of the ion-trapping technique, Dehmelt and Wolfgang Paul shared the Nobel Prize in 1989, along with Ramsey.

The modes of motion for a single charged particle in a Penning trap include one circular mode about the trap axis called the magnetron mode. For the electron *g*-factor experiments, it was desirable to locate the electron as close to the trap axis as possible by reducing the amplitude of this mode. This could be accomplished with a form of "sideband cooling" (Wineland and Dehmelt, 1975a, 1976) as demonstrated by Van Dyck, Schwinberg, and Dehmelt (1978). Around this time, I was also stimulated by the papers of Arthur Ashkin (Ashkin, 1970a, 1970b) on the possibilities of radiation pressure from lasers affecting the motion of atoms. In analogy with the electron sideband cooling, Dehmelt and I came up with a scheme

for cooling trapped-ion motion with laser beams (Wineland and Dehmelt, 1975b) (see below). The cooling could also be explained in terms of velocity-dependent radiation pressure as in a concurrent proposal by Ted Hänsch and Art Schawlow (Hänsch and Schawlow, 1975). We didn't anticipate all of the uses of laser cooling at the time, but it was clear that it would be important for high-resolution spectroscopy of trapped ions. For example, the largest systematic uncertainty in the $^3He^+$ experiment (Schuessler, Fortson, and Dehmelt, 1969) was the uncertainty in the time dilation shift, which would be reduced with cooling.

In the summer of 1975, I took a position in the Time and Frequency Division of NIST (then NBS, the National Bureau of Standards). My first task was to help make a measurement of the cesium hyperfine frequency, the frequency reference that defines the second. The apparatus, NBS-6, had been built by David Glaze of the Division. It was a traditional atomic beam apparatus but had a relatively long distance between Ramsey zones of 3.75 m. With it, we realized a fractional accuracy of 0.9×10^{-13} (Wineland *et al*., 1976). At that time, the Division was more service oriented, with very little basic research. Fortunately my group leader, Helmut Hellwig, had a progressive view of the Division's future and was able to obtain NBS support to initiate laser-cooling experiments. That support, along with some seed money from the Office of Naval Research (ONR), enabled us to start a project on laser cooling in the fall of 1977. With Robert Drullinger (a local laser expert) and Fred Walls, we chose to use $^{24}Mg^+$ because of its simple electronic structure and Penning traps, because of our prior experience with them. This was a very exciting time, being able to work on a project of our choosing, and by the spring of 1978, we had obtained our first cooling results (Wineland, Drullinger, and Walls, 1978). In our experiments we observed currents in the trap electrodes induced by the ions' thermal motion and hence had a direct measurement of the ions' temperature. Meanwhile, Peter Toschek's group in Heidelberg (joined by Dehmelt, who was on sabbatical) was working toward the same goal, using Ba^+ ions confined in an rf-Paul trap. They, with colleagues Werner Neuhauser and Martin Hohenstatt, also observed the cooling at about the same time (Neuhauser *et al.*, 1978), through the increased trapping lifetime of ions. In a near coincidence, although there was no contact between the groups, the manuscripts were received by *Physical Review Letters* within

one day of each other (Peter Toschek's group ''won'' by one day!). The cooling observed in both experiments is typically called Doppler cooling, where the oscillation frequency of the ions' motion is less than the linewidth of the cooling transition. Theoretical groups were becoming interested in the cooling, and some of the earlier work is discussed in Letokhov, Minogin, and Pavlik (1977), Kazantsev (1978), and Stenholm (1986).

To us, the cooling of course provided a start toward improving clocks and in 1985, working with John Bollinger, John Prestage, and Wayne Itano, we demonstrated the first clock that utilized laser cooling (Bollinger *et al*., 1985). But as physicists, we were excited by just the cooling process itself. So, in addition to clock applications, it would eventually lead to reaching and controlling the lowest quantized levels of motion for a trapped particle (below).

III. Controlling the Quantum Levels of Individual Trapped Ions

One of the obvious next steps was to isolate single ions. In addition to the aesthetic appeal of this, as for single electrons, the systematic errors in spectroscopy would be smallest in this case (Dehmelt, 1982). By observing steps in the ion laser fluorescence, the Heidelberg group was able to isolate Ba^+ single ions (Neuhauser *et al*., 1980). With Wayne Itano, we subsequently used this fluorescence "steps" method to observe single $^{24}Mg^+$ ions (Wineland and Itano, 1981). The Heidelberg group also made photographs of a single ion, and because of its relatively long fluorescence wavelength (493 nm), with a magnifier, a single Ba^+ ion can be observed with the human eye!

In NIST single-ion experiments we chose to focus on Hg^+ because for frequency-standard applications, $^{199}Hg^+$ has a relatively high ground-state hyperfine clock transition frequency of 40.5 GHz (Major and Werth, 1973; Cutler, Giffard, and McGuire, 1982; Prestage, Dick, and Maleki, 1991) and also a narrow $^2S_{1.2} - {}^2D_{5.2}$ optical transition $\left[\tau\left(^2D_{5/2}\right) \cong 86\,\text{ms}\right]$, which could potentially be used as an optical frequency standard (Bender et al., 1976). Although optical pumping of $^{199}Hg^+$ could be achieved with radiation from isotopically selected Hg^+ fluorescence lamps (Major and Werth, 1973; Cutler, Giffard, and McGuire, 1982; Prestage, Dick, and

Maleki, 1991), laser excitation was made difficult because of the short (194 nm) wavelength required. Jim Bergquist in our group, with colleagues Hamid Hemmati and Wayne Itano, first developed the required source by sum-frequency mixing a doubled Ar^+ laser at 515 nm with 792 nm from a dye laser in a potassium pentaborate crystal (Hemmati, Bergquist, and Itano, 1983). We used an rf trap with a simple ring-and-end-cap structure shown in Fig. 1, similar to that used by the Heidelberg group.

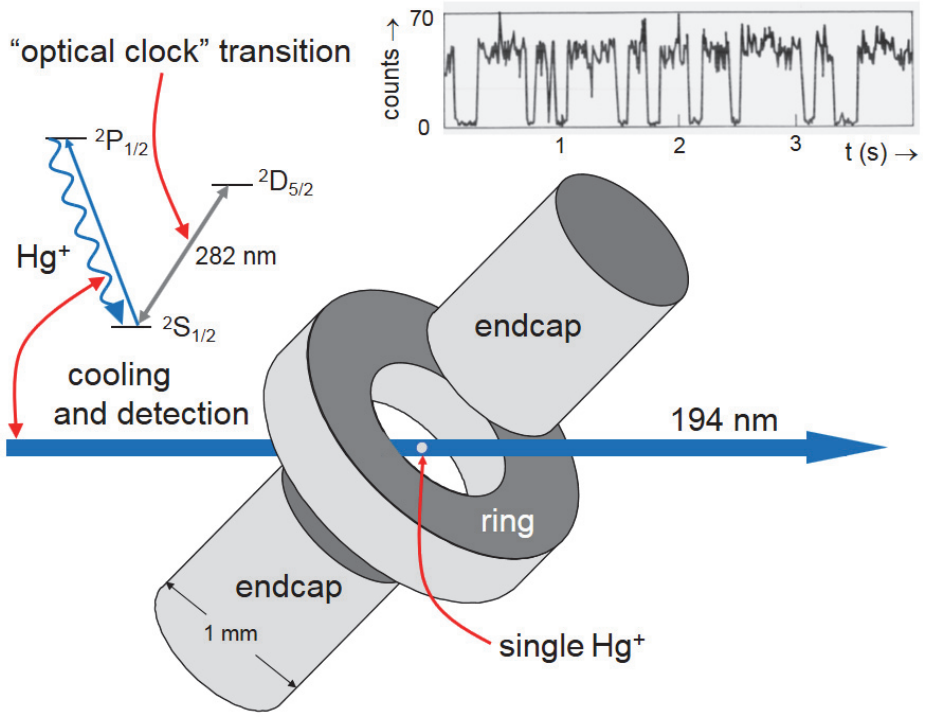

FIG. 1 (color). Schematic of the trap for single Hg^+ ion studies. An rf potential is applied between the ring electrode and endcap electrodes (which are in common), forming an rf "pseudopotential" for the ion. The relevant Hg^+ energy levels are indicated, including the narrow $^2S_{1/2} \rightarrow {}^2D_{5/2}$ "optical clock" transition. The data in the upper right-hand part of the figure show the number of 194 nm fluorescence photons detected in 10 ms detection bins vs time when both transitions are excited simultaneously (Bergquist *et al*., 1986).

By the mid-1980s ion trappers were able to directly address one of Schrödinger's questions, which formed the title for his publication "Are there quantum jumps?" (Schrödinger, 1952a, 1952b). Three similar

demonstrations were made in 1986 (Bergquist *et al*., 1986; Nagourney, Sandberg, and Dehmelt, 1986; Sauter *et al*., 1986; Blatt and Zoller, 1988); for brevity, we describe the experiment of Bergquist *et al*. Referring to Fig. 1, a nearly harmonic binding potential called a pseudopotential (Paul, 1990) is formed by applying an rf potential between the ring electrode and the endcap electrodes (held in common). The relevant optical energy levels of a Hg^+ ion are indicated in the upper left-hand part of the figure. The $^2S_{1/2} \rightarrow {}^2P_{1/2}$ electric-dipole transition $\left[\lambda = 194\,\text{nm},\ \tau\left({}^2P_{1/2}\right) \cong 2.9\,\text{ns}\right]$ was used for Doppler laser cooling. If continuously applied, a steady fluorescence from the ion would be observed and could be used to produce images of the ion. If $^2S_{1/2} \rightarrow {}^2D_{5/2}$ resonance radiation was applied simultaneously, one would expect the 194 nm fluorescence to decrease because of excitation to the $^2D_{5/2}$ state.

A density-matrix description, valid for an ensemble of atoms, would predict a reduced but steady fluorescence rate. But what would be observed for a single ion? (Cook and Kimble, 1985; Erber and Putterman, 1985; Cohen-Tannoudji and Dalibard, 1986; Javanainen, 1986; Kimble, Cook, and Wells, 1986; Pegg, Loudon, and Knight, 1986; Schenzle, DeVoe, and Brewer, 1986). In fact the ion's fluorescence does not steadily decrease, but switches between the full value and no fluorescence, effectively indicating quantum jumps between the $^2S_{1/2}$ and $^2D_{5/2}$ states. For the data shown in the upper right-hand corner of Fig. 1, the 194 nm fluorescence photon counts registered by a photomultiplier tube were accumulated in 10 ms time bins and plotted as a function of elapsed time to show the jumps. In a more general context, a measurement of the quantum system composed of the $^2S_{1/2}$ and $^2D_{5/2}$ states can be made by applying the 194 nm ''measurement'' beam for 10 ms and observing the presence or absence of fluorescence. The $^2S_{1/2} \rightarrow {}^2P_{1/2}$ transition is some-times called a "cycling transition" because when the $^2S_{1/2}$ state is excited to the $^2P_{1/2}$ state, the ion decays back to the $^2S_{1/2}$ state, emitting a photon, and the excitation/decay process is then repeated. Neglecting the occasional decays of the $^2P_{1/2}$ to the $^2D_{3/2}$ state (Itano *et al*., 1987), this procedure approximates an ideal measurement in quantum mechanics because the detection of the state is nearly 100% efficient and because the state of the Hg^+ ion, either the $^2S_{1/2}$ or $^2D_{5/2}$ state, remains in its original condition after the measurement.

Dehmelt dubbed this "electron shelving" detection (Dehmelt, 1982), where in this example the ion is shelved to the $^2D_{5/2}$ state. Such measurements are also called quantum nondemolition (QND) measurements (Braginsky and Khalili, 1996; Haroche and Raimond, 2006). The method of detection by state-dependent fluorescence has now become rather ubiquitous in atomic physics.

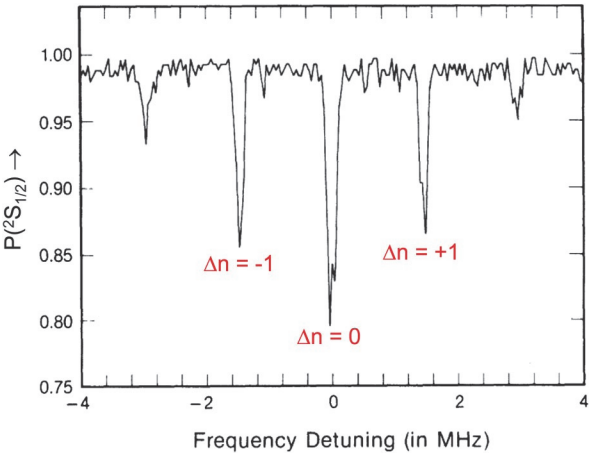

FIG. 2 (color). Spectroscopy of the $^2S_{1/2} \rightarrow {}^2D_{5/2}$ transition on a single $^{198}Hg^+$ ion. Referring to Fig. 1, for each measurement cycle, the ion is prepared in $^2S_{1/2} \equiv \left|\downarrow\right\rangle$ state by allowing it to decay to that level. Then, application of a 282 nm "probe" laser beam is alternated with a 194 nm measurement beam. The $\left|\downarrow\right\rangle$ and $^2D_{5/2} \equiv \left|\uparrow\right\rangle$ states are detected with nearly 100% efficiency by observing the presence or absence of 194 nm scattered light. By stepping the frequency of the probe beam and averaging over many measurements, we obtain the spectrum shown where we plot the probability of the ion remaining in the $^2S_{1/2}$ state P($^2S_{1/2}$) vs the 282 nm laser beam frequency. In a quantum picture of the motion, the central feature or "carrier" state denotes transitions of the form $\left|\downarrow\right\rangle\left|n\right\rangle \rightarrow \left|\uparrow\right\rangle\left|n\right\rangle$, where n denotes the motional Fock state. "Red" and "blue" side-bands correspond to $\left|\downarrow\right\rangle\left|n\right\rangle \rightarrow \left|\uparrow\right\rangle\left|n+\Delta n\right\rangle$ transitions with Δn = −1 or +1, respectively. The central feature or carrier is essentially unshifted by photon recoil, since the recoil is absorbed by the entire trap apparatus as in the Mössbauer effect; see, *e.g.*, Dicke (1953), Lipkin (1973), and Wineland *et al*. (1998).

To perform spectroscopy on the $^2S_{1/2} \rightarrow {}^2D_{5/2}$ transition ($\lambda \sim 282$ nm), radiation was first applied near the transition frequency in the absence of the 194 nm beam; this avoids perturbations of the energy levels

from the 194 nm beam. The 282 nm beam was then switched off, followed by measurement of the ion's state with the 194 nm beam. This process was repeated many times, and by stepping the frequency of the 282 nm beam, spectra like that shown in Fig. 2 are obtained (Bergquist, Itano, and Wineland, 1987). To interpret this spectrum, we must consider the motion of the ion. Along any mode axis the motion is nearly harmonic, so in the frame of the ion, the laser beam appears to be sinusoidally frequency modulated due to the first-order Doppler shift. Thus the central feature or "carrier," which corresponds to the transition frequency, is surrounded by frequency-modulation sidebands spaced by the motional frequency of the ion (Dicke, 1953). An equivalent picture is that the ion can absorb radiation while simultaneously gaining or losing one quantum of motion, which leads to absorption features spaced by the frequency of motion around the carrier.

As in many atomic physics experiments, by using highly coherent radiation, we can initialize an ion in an eigenstate and deterministically prepare superpositions; *e.g.*, $|\downarrow\rangle \rightarrow \alpha|\downarrow\rangle + \beta|\uparrow\rangle$. To extract the values of $|\alpha|$ and $|\beta|$, we detect as described above. A single measurement indicates either the $|\downarrow\rangle$ or $|\uparrow\rangle$ state with respective probabilities $P = |\alpha|^2$ and $1 - |\alpha|^2$. Quantum fluctuations or "projection noise" in the measurements are characterized with a variance $\sqrt{P(1-P)/M}$, where M is the number of measurements on identically prepared atoms (Itano *et al*., 1993). Therefore, accurate measurements of P generally require many repeated experiments. Similarly, Ramsey-type experiments where the two pulses are separated in time can measure the relative phase between α and β. From these types of measurements, many ion trap groups now routinely produce and verify superposition states of single ions that have coherence times exceeding 1 s. [For ion ensembles, coherence times exceeding 10 min have been demonstrated (Bollinger *et al*., 1991; Fisk *et al*., 1995).]

The Hg^+ clock project at NIST, led by Jim Bergquist, has been a long but very successful story. First, an accurate clock based on the 40.5 GHz hyperfine transition of a few $^{199}Hg^+$ ions confined in a linear Paul trap achieved systematic errors of about 4×10^{-14} (Berkeland *et al*., 1998). Although we felt these errors could be substantially reduced, we also

realized that the future of high-performance clocks was in the optical domain, so we focused on the $^2S_{1/2} \rightarrow {}^2D_{5/2}$ optical clock transition. For many years it had been appreciated that higher frequency was advantageous in terms of measurement precision; basically the higher oscillation frequencies allows one to divide a time interval into finer units. But two things were needed: a laser with high enough spectral purity to take advantage of narrow optical transitions, and a practical means to count cycles of the "local oscillator," in this case the laser that would excite the clock transition. In our lab, Brent Young, Bergquist, and colleagues were able to make a cavity-stabilized laser at 563 nm, which was doubled to produce the clock radiation. The 563 nm source had a line- width of less than 0.2 Hz for an averaging time of 20 s (Young *et al*., 1999). It is now understood that the linewidth was limited by thermal fluctuations in the mirror surface, currently still the limit for the most stable lasers. The solution to the second problem is by now well known. The relatively rapid development of optical combs by Jan Hall (Hall, 2006), Ted Hänsch (Hänsch, 2006), their colleagues, and other researchers meant that it was now possible to effectively count optical cycles. Including these developments, in 2006, Bergquist and colleagues demonstrated a $^{199}Hg^+$ optical clock with a systematic uncertainty of 7.2×10^{-17}, the first clock since the inception of atomic clocks that had smaller systematic errors than a cesium clock (Oskay *et al*., 2006).

IV. Manipulating Ion Motion at the Quantum Level

An interesting next step would be to control an ion's motion at the quantum level. Since a cold trapped ion's motion along any mode axis is harmonic to a very good approximation, in a quantum description

(Neuhauser *et al*., 1978; Wineland and Itano, 1979; Stenholm, 1986), we express its Hamiltonian in the usual way as $\hbar\omega_z a^\dagger a$ with ω_z the oscillation frequency (along the z axis here) and a and $a^\dagger$ the lowering and raising operators for the ion motion. The operator for the ion's position about its mean value is $z = z_0\left(a + a^\dagger\right)$, where $z_0 = \sqrt{\hbar / 2m\omega_z}$ is the spread of the ground-state wave function, with m the ion's mass. In principle, we could detect the ion's motion through the current it induces in the trap electrodes, as was done for electrons. In practice, however, a far more

sensitive method is to map information about the motional states onto internal states of the ion and read those out as described above. For this, we need to efficiently couple an ion's internal states to its motion. To see how this works, consider a single trapped ion that has a single-electron electric-dipole transition with resonance frequency ω_0. If this transition is excited by a laser beam of frequency ω_L propagating along the z axis, the interaction is given by

$$\begin{aligned} H_I &= -e\vec{r}\cdot\hat{\epsilon}\, E_0 \cos\left(kz - \omega_L t + \phi\right) \\ &= \hbar\Omega\left(\sigma_+ + \sigma_-\right)\left(e^{i(kz-\omega_L t+\phi)} + e^{-i(kz-\omega_L t+\phi)}\right), \end{aligned} \tag{2}$$

where $\vec{r}$ is the electron coordinate relative to the ion's core, e is the electron charge, $\hat{\epsilon}$, E_0, and k are, respectively, the laser beam's electric-field polarization, amplitude, and wave vector, and ϕ is the electric-field phase at the mean position of the ion. The operators $\sigma_+ \left(=\left|\uparrow\right\rangle\left\langle\downarrow\right|\right)$ and $\sigma_- \left(=\left|\downarrow\right\rangle\left\langle\uparrow\right|\right)$ are the internal state raising and lowering operators, and $\Omega \equiv -eE_0\left\langle\uparrow\left|\vec{r}\cdot\hat{\epsilon}\right|\downarrow\right\rangle/2\hbar$, with $\left|\downarrow\right\rangle$ and $\left|\uparrow\right\rangle$ denoting the ion's ground and optically excited states as above. If we transform to an interaction picture for the ion's internal states $\left(\sigma_+ \to \sigma_+ e^{i\omega_0 t}\right)$ and motion states $\left(a^\dagger \to a^\dagger e^{i\omega_z t}\right)$ and assume $\omega_L \cong \omega_0$, then neglecting terms that oscillate near 2 ω_0 (rotating wave approximation), Eq. (2) becomes

$$\begin{aligned} H_I &\cong \hbar\Omega\sigma_+ e^{i\left[kz-(\omega_L-\omega_0)t+\phi\right]} + \text{H.c.} \\ &\cong \hbar\Omega\sigma_+ e^{-i\left[(\omega_L-\omega_0)t+\phi\right]}\left[1+i\eta\left(ae^{-i\omega_z t} + a^\dagger e^{i\omega_z t}\right)\right] + \text{H.c.} \end{aligned} \tag{3}$$

Here, H.c. stands for Hermitian conjugate and $\eta \equiv kz_0 = 2\pi z_0/\lambda$ is the Lamb-Dicke parameter, which we assume here to be much less than 1. For an ion of mass 40 u (*e.g.*, $^{40}Ca^+$) in a well with $\omega_z/2\pi = 3\,\text{MHz}$ and $\lambda = 729\,\text{nm}$, we have $z_0 = 6.5$ nm and η = 0.056. For $\omega_L = \omega_0$ and $\eta\Omega << \omega_z$, to a good approximation we can neglect the non-resonant η term in Eq. (3) and obtain $H_I \cong \hbar\Omega e^{j\phi} S_+ + \text{H.c.}$. This is the Hamiltonian for carrier transitions or, equivalently, spin-vector rotations about an axis in the x-y plane of the Bloch sphere. If we assume

$\omega_L = \omega_0 - \omega_z$ (laser tuned to the ‘‘red sideband’’), and absorb phase factors in the definition of Ω, the resonant term gives

$$H_I \cong \hbar\eta\left(\Omega\sigma_+ a + \Omega^*\sigma_- a^\dagger\right). \tag{4}$$

This Hamiltonian describes the situation where a quantum of motion is exchanged with a quantum of excitation of the ion's internal state. It is most commonly known as the Jaynes- Cummings Hamiltonian from cavity QED, which expresses the exchange of energy between the internal states of an atom in a cavity and the photons confined by the cavity (Jaynes and Cummings, 1963; Haroche and Raimond, 2006). In the cavity-QED experiments of Serge Haroche, Jean-Michel Raimond, Michel Brune, and their colleagues in Paris, the atoms play much the same role as they do in the ion experiments; however, in the cavity-QED experiments, the relevant harmonic oscillator is that which describes a field mode of the cavity, whereas in the ion case, the relevant harmonic oscillator is that associated with the ion's motion (Sauter *et al.,* 1988; Blockley, Walls, and Risken, 1992). Over the years, this connection has led to some interesting and complementary experiments between the two types of experiments (Haroche and Raimond, 2006).

In the trapped-ion world, this type of exchange at the quantum level was first used in the electron *g*-factor experiments of Dehmelt and colleagues, where a change of the electron's cyclotron quantum number was accompanied by spin flip of the electron, which could be detected indirectly (Dehmelt, 1990). If we apply H_I of Eq. (4) to an atomic ion in the state $|\downarrow\rangle|n\rangle$, where n denotes the harmonic oscillator's quantum state (Fock state), we induce the transition $|\downarrow\rangle|n\rangle \rightarrow |\uparrow\rangle|n-1\rangle$. This corresponds to the absorption feature labeled $\Delta n = -1$ in Fig. 2, and reduces the energy of motion by $\hbar\omega_z$. When the ion decays, on average, the motion energy increases by the recoil energy $R = (\hbar k)^2/(2m)$, where $k = 2\pi/\lambda$. Typically, we can achieve the condition $R << \hbar\omega_z$, so that in the overall scattering process the motional energy is reduced. In Fig. 2, the carrier absorption feature is labeled $\Delta n = 0$, indicating photon absorption without changing the motional state. This is a manifestation of the “recoilless” absorption of the

Mössbauer effect [see, *e.g.*, Dicke (1953), Lipkin (1973), and Wineland *et al.* (1998)], but in the visible wavelength region.

Continuous application of the red-sideband transition provides a relatively straightforward way to laser cool the ion to near the ground state of motion. After many scattering events, the ion reaches the $|\downarrow\rangle|n=0\rangle$ state, a "dark state" in which scattering stops, since the $|\uparrow\rangle|n=-1\rangle$ state does not exist. The process is not perfect, since scattering in the wings of $\Delta n = 0, +1$ transitions leads to some residual recoil heating, but the condition $\langle n\rangle << 1$ can be achieved. This is easily verified because absorption on the $\Delta n = -1$ red sideband nearly disappears, but the $\Delta n = +1$ blue-sideband absorption remains. In 1989, with Frank Diedrich, who was a postdoc in our lab, we achieved near-ground-state laser cooling in two dimensions, in essentially the way described here (Diedrich *et al*., 1989). Later in an experiment led by Chris Monroe, we achieved near-ground-state cooling in 3D using two-photon stimulated-Raman transitions (Monroe, Meekhof, King, Jefferts *et al*., 1995).

In addition to suppressing Doppler shifts in spectroscopy to the highest degree possible (Wineland *et al.*, 1987), one motivation for sideband cooling was the intrinsic appeal of (actively) placing a bound particle in its ground state of motion, the lowest energy possible within the limitations imposed by quantum mechanics. Here, the ground state is a Gaussian-shaped wave packet with spread $\sqrt{\langle z^2\rangle} = \sqrt{\hbar/2m\omega_z} \equiv z_0$ and energy $\hbar\omega_z/2$. We were also interested in generating non-classical states of motion (Heinzen and Wineland, 1990; Cirac, Blatt *et al.*, 1993; Cirac, Parkins *et al*., 1993; Cirac *et al*., 1996) or entangled states of spins (Wineland *et al*., 1992; Bollinger *et al*., 1996). For these experiments, cooling to the ground state of motion provides a clean starting point for motional state manipulation. [In the Paris experiments, the ground state of the cavity mode can be achieved either by thermally cooling to $\langle n\rangle << 1$ by operating at low temperature or by extracting photons with atoms sent through the cavity in a process analogous to ion sideband cooling (Haroche and Raimond, 2006).]

The red-sideband interaction of Eq. (4) and the "blue- sideband" interaction $\left(H_I \cong \hbar\eta\Omega\sigma_+ a^\dagger + \text{H.c., for } \omega_L = \omega_0 + \omega_z\right)$ that induces $\left|\downarrow\right\rangle\left|n\right\rangle \rightarrow \left|\uparrow\right\rangle\left|n+1\right\rangle$ transitions, provide simple tools for the manipulation of an ion's motional states. For example, starting from $\left|\downarrow\right\rangle\left|n=0\right\rangle$, and applying a series of blue-sideband, red-sideband, and carrier π pulses, Fock states for a selected value of *n* can be deterministically prepared (Meekhof *et al.*, 1996). From $\left|\downarrow\right\rangle\left|n=0\right\rangle$, we can also make coherent states ion motion by forcing the ion at its motion frequency with an oscillating classical uniform field (Carruthers and Nieto, 1965) or by applying an oscillating optical-dipole force (Meekhof *et al*., 1996), which results from spatial gradients of laser-beam-induced ac Stark shifts. A coherent state of a quantum particle is very much like an oscillating classical particle but, as opposed to a classical particle that can be point-like, the shape of the quantum particle's wave packet is the same as it is in the ground state. In a clever but straightforward scheme suggested by Chi Kwong Law and Joe Eberly (Law and Eberly, 1996) arbitrary motional state superpositions can be prepared (Ben-Kish *et al*., 2003). As a final example, the red-sideband interaction applied for a "π-pulse" duration $t = \pi/\left(2\eta\Omega\right)$ provides internal-state to motion-state transfer

$$\left(\alpha\left|\downarrow\right\rangle + \beta\left|\uparrow\right\rangle\right)\left|0\right\rangle \rightarrow \left|\downarrow\right\rangle\left(\alpha\left|0\right\rangle + \beta\left|1\right\rangle\right). \qquad (5)$$

V. Schrödinger's Cat

The optical-dipole force is interesting because the strength of the force can depend on the ion's internal state. In 1996 (Monroe *et al*., 1996), using state-dependent optical-dipole forces, we were able to produce an analog to the Schrödinger's cat state in Eq. (1), which had the form

$$\Psi = \frac{1}{\sqrt{2}}\left[\left|\uparrow\right\rangle\left|\alpha\right\rangle + \left|\downarrow\right\rangle\left|-\alpha\right\rangle\right], \qquad (6)$$

where $\left|\alpha\right\rangle$ denotes a coherent state. The amplitude of the particle's oscillatory motion is equal to $2\alpha z_0$. The spatial part of the state in Eq. (6)

represents two wave packets that oscillate back and forth but are 180° out of phase with each other and therefore pass through each other at the center of the trap every half cycle of oscillation. Here, the analogy to Schrödinger's cat is that the spin states of the ion are like the states of the single radioactive particle and the coherent states of the ion, which follow more macroscopic classical trajectories, are like the state of the cat; *e.g.*, the ion at its left extremum point ≅ live cat, ion at its right extremum ≅ dead cat. Figure 3 describes how this state was produced.

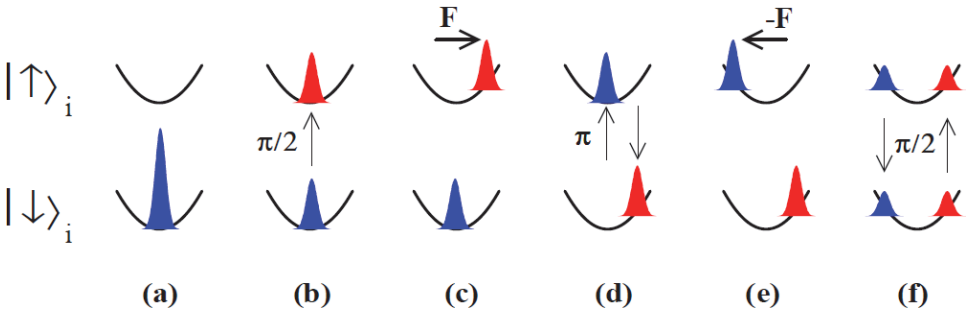

FIG. 3 (color). Depiction of the harmonic oscillator potential and the wave packets for each component of the ion's internal states, denoted $|\uparrow\rangle$ and $|\downarrow\rangle$. The images are snapshots in time; for images (c) through (f) the wave packets are shown at the extremes of their motion. The areas of the wave packets correspond to the probability of finding the atom in the given internal state. (a) The initial wave packet corresponds to the ground state of motion after laser cooling and preparation of the $|\downarrow\rangle$ internal state. (b) A $\pi/2$ carrier pulse creates the internal-state superposition $\frac{1}{\sqrt{2}}(|\downarrow\rangle+|\uparrow\rangle)$. (c) An oscillating optical-dipole force is applied that excites only the $|\uparrow\rangle$ component of the superposition to a coherent state of amplitude α, creating the state $\frac{1}{\sqrt{2}}(|\downarrow\rangle|n=0\rangle+|\uparrow\rangle|\alpha\rangle)$. (d) The spin states are flipped by applying a carrier π pulse. (e) The wave packet associated with the $|\uparrow\rangle$ state is excited by the optical-dipole force to an amplitude of $-\alpha$, that is, out of phase with respect to the first excitation. This is the state of Eq. (6). (f) To analyze the state produced in step (e) and verify phase coherence between the components of the cat wave function, we apply a final $\pi/2$ carrier pulse and then measure the probability $P(\downarrow)$ of the ion to be in state $|\downarrow\rangle$ (see text). From Monroe *et al.*, 1996.

To analyze the experiment, in Fig. 3, we can control the phase of the amplitude such that the coherent state is $e^{i\phi}\alpha$ rather than $-\alpha$. Near the condition $\phi = 0$, the probability $P(\downarrow)$ of the ion to be in state $|\downarrow\rangle$ oscillates as a function of ϕ due to interference of the two wave packets. This verifies the coherence between the two components of the cat superposition state. These interference oscillations are very analogous to the fringe oscillations observed in Young's-slit-type experiments performed on individual photons, electrons, neutrons, or atoms, but in those experiments the particle wave packets disperse in time, whereas the wave packets in a harmonic oscillator do not, and in principle last arbitrarily long.

In Monroe *et al*. (1996), for the condition described by Eq. (6), the maximum separation of the wave packets was $4\alpha z_0 \cong 83$ nm, while the size of the wave packets z_0 was 7.1 nm [see also McDonnell *et al.* (2007) and Poschinger *et al*. (2010)]. Of course, one might object to dignifying the state produced by calling it a Schrödinger cat since it is so small. In fact as we tried to make $|\alpha|$ larger, the quality of the superposition became more susceptible to decoherence caused by noisy ambient electric fields (Myatt *et al*., 2000a, 2000b; Turchette *et al*., 2000), limiting the size that was obtained. However, as far as we know, this is just a technical, not fundamental limitation and we should eventually be able to make a cat with $|\alpha|$ large enough that the wave packets are separated by macroscopic distances.

VI. Enter Quantum Information

Following Peter Shor's development of a quantum- mechanical algorithm for efficient number factoring (Shor, 1994), there was a dramatic increase of activity in the field of quantum information science. The potential realization of general-purpose quantum information processing (QIP) is now explored in many settings, including atomic, condensed-matter, and optical systems.

At the 1994 International Conference on Atomic Physics held in Boulder, Colorado, Artur Ekert presented a lecture outlining the ideas of quantum computation (Ekert, 1995), a subject new to most of the audience. This inspired Ignacio Cirac and Peter Zoller, who attended the conference

and were very familiar with the capabilities (and limitations) of trapped- ion experiments, to propose a basic layout for a quantum computer utilizing trapped ions (Cirac and Zoller, 1995). This seminal paper was the first comprehensive proposal for how a quantum information processor might be realized. In their scheme, quantum bits or ‘‘qubits’’ are realized with two internal states of the ion, e.g., the $|\downarrow\rangle$ and $|\uparrow\rangle$ states above. The ion qubits are held in a trap shown schematically in Fig. 4. The motion of the ions is strongly coupled by the Coulomb

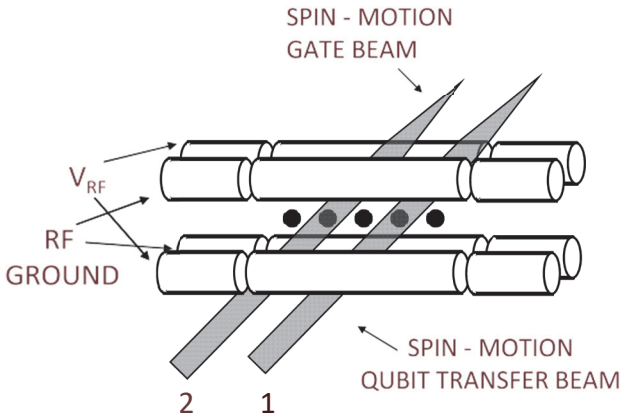

FIG. 4 (color online). Scheme for quantum computation proposed by Cirac and Zoller (Cirac and Zoller, 1995). Quadrupolar electrodes are configured to produce a linear array of trapped-ion qubits (filled black circles). Two diagonally opposite rods support an rf potential to realize a ponderomotive pseudopotential transverse to the trap’s (horizontal) axis. Static potentials applied to the end segments of the electrodes confine ions along the axis. Ideally, all motional modes are laser cooled to the ground state before logic operations. The quantized modes of motion can be used as a data bus to share information between the internal-state qubits of ions that are selected by focused laser beams (see text).

interaction and is best described by the normal modes of a kind of pseudomolecule. Typically, the motion of each mode is shared among all the ions and can act as a data bus for transferring information between ions. A single-qubit gate or rotation (the relatively easy part) is implemented by applying a focused laser beam or beams onto that ion and coherently driving a carrier transition as described above. The harder part is to perform a logic gate between two selected ions. This can be accomplished by first laser cooling all modes to the ground state. The

internal qubit state of one ion is then transferred onto the qubit formed from the ground and first excited state of a particular mode of motion (laser beam 1 in Fig. 4), as indicated in Eq. (5). Laser beam 2 then performs a logic gate between the (shared) motion qubit state and a second selected ion. Since the second ion is generally in a superposition state, before the gate operation is performed, the wave function for the spin and motional state of the second qubit can be written as $\alpha|\downarrow\rangle|0\rangle+\beta|\downarrow\rangle|1\rangle+\xi|\uparrow\rangle|0\rangle+\zeta|\uparrow\rangle|1\rangle$. One type of logic gate imparts a minus sign to the $|\uparrow\rangle|1\rangle$ component of the wave function by coherently driving a 2π transition $|\uparrow\rangle|1\rangle\rightarrow|\text{aux}\rangle|0\rangle\rightarrow-|\uparrow\rangle|1\rangle$, where $|\text{aux}\rangle$ is a third ''auxiliary'' internal state of the ion (Cirac and Zoller, 1995). Flipping the sign of the $|\uparrow\rangle|1\rangle$ component of the wave function realizes an entangling two- qubit "π-phase" gate and is universal for computation. Finally, the initial transfer step on the first ion is reversed, restoring the motion to the ground state and effectively having performed the logic gate between the internal qubit states of the two laser-beam-selected ions. At NIST, since we had recently achieved ground-state cooling with stimulated- Raman transitions on hyperfine qubit states, we were able to quickly demonstrate a universal gate between a hyperfine qubit and a motional mode qubit (Monroe, Meekhof, King, Itano, and Wineland, 1995). The complete Cirac-Zoller gate between two selected qubits was subsequently demonstrated by the Innsbruck group, led by Rainer Blatt (Schmidt-Kaler *et al*., 2003).

More streamlined gates were subsequently devised in which multiple ions are addressed simultaneously by the same laser beams (Sørensen and Mølmer, 1999, 2000; Solano, de Matos Filho, and Zagury, 1999; Milburn, Schneider, and James, 2000; Wang, Sørensen, and Mølmer, 2001). These gates also have the advantage that it is not necessary to prepare all modes in the ground state; it is only necessary that each ion is maintained well within the Lamb-Dicke regime $\left[\left\langle z^2\right\rangle<<\left(\lambda/2\pi\right)^2\right]$. These "geometric" gates can be viewed as arising from quantum phases that are acquired when a mode of the ions' motion is displaced in phase space around a closed path; the phases accumulated are

proportional to the enclosed area in phase space. The different gates can be viewed in a common framework, the main difference being whether or not the forces act on the spin states in the z basis (eigenstates $|\downarrow\rangle|\uparrow\rangle$) or in the x, y basis [eigenstates of the form $\frac{1}{\sqrt{2}}(|\downarrow\rangle + e^{i\xi}|\uparrow\rangle)$, $\frac{1}{\sqrt{2}}(|\downarrow\rangle - e^{i\xi}|\uparrow\rangle)$] (Lee *et al*., 2005). The forces required for the displacements are usually implemented with optical-dipole forces as in the Schrödinger cat example. Since the forces are state dependent, the differential geometric phases generate entangling gates. Two-qubit phase gates have been implemented in the z basis (Leibfried *et al*., 2003; Home *et al*., 2006) and in the x, y basis (Sackett *et al*., 2000; Haljan *et al*., 2005; Benhelm *et al.*, 2008; Kim *et al.*, 2009). In the Innsbruck experiment of Benhelm *et al*. (2008), a Bell state with fidelity 0.993(1) was produced, setting a standard for all QIP experiments. The use of single- and multiqubit gates has enabled the demonstration of several ion-based QIP algorithms; see, for example, Blatt and Wineland (2008) and Blatt and Roos (2012). At NIST most such demonstrations were led by Didi Leibfried. Chris Monroe's group at the University of Maryland is leading efforts on an entirely different scheme for ion entanglement generation based on performing joint measurements on photons that are first entangled with ion qubits (Moehring *et al*., 2007; Olmschenk *et al.*, 2010; Monroe *et al*., 2012). This scheme has the advantage that the ions don't have to be in the Lamb-Dicke regime, and it also enables entanglement of widely separated qubits because of the relative ease of transferring photons over large distances.

The basic elements of the Cirac-Zoller proposal are carried forward in the different variations of trapped-ion QIP. This proposal rejuvenated the field of trapped ions and today there are over 30 groups in the world working on various aspects of quantum information processing. These include groups at the University of Aarhus; Amherst College; University of California, Berkeley; University of California, Los Angles; Duke University; ETH Zürich; University of Freiburg; Georgia Tech; Griffiths University; Imperial College; University of Innsbruck; Lincoln Laboratories; Mainz University; University of Hannover and PTB (Germany); MIT; NIST (USA); NPL (UK); Osaka University; Oxford University; Joint Quantum Institute at the University of Maryland; Université de Paris; Saarland University (Saarbrücken); Sandia National

Laboratory (USA); Siegen University; Simon Fraser University; National University of Singapore; Sussex University; University of Sydney; Tsinghua University; University of Ulm; University of Washington; Wabash College; and the Weizmann Institute.

VI(a). Quantum Simulation

In the early 1980s, Richard Feynman proposed that one quantum system might be used to efficiently simulate the dynamics of other quantum systems of interest (Feynman, 1982; Lloyd, 1996). This is now a highly anticipated application of QIP and will likely occur well before useful factorization is performed. Of course, the universality of a large-scale quantum computer will allow it to simulate any quantum system of interest. However, it is also possible to use the built-in available interactions in a quantum processor to simulate certain classes of physical problems. For trapped ions, it has been possible to use the interactions employed in the various gates to simulate other systems of interest, for example, nonlinear optical systems (Leibfried *et al*., 2002), motional quantum dynamics as in an electron's Zitterbewegung (Gerritsma *et al*., 2010), or the properties of a ''quantum walk'' (Schmitz, Matjeschk *et al.*, 2009; Zähringer *et al*., 2010). Currently, efforts are underway in several laboratories to use QIP interactions to simulate various dynamics including those of condensed-matter systems. Some of the basic ideas for how this might work with ions have been outlined in Wunderlich and Balzer (2003), Porras and Cirac (2004, 2006), Deng, Porras, and Cirac (2005), Pons *et al*. (2007), Schätz *et al*. (2007), Chiaverini and Lybarger (2008), Taylor and Calarco (2008), Clark *et al*. (2009), Johanning, Varon, and Wunderlich (2009), Schmitz, Friedenauer *et al*. (2009), Schmied, Wesenberg, and Leibfried (2011), Blatt and Roos (2012), Britton *et al.* (2012), Korenblit *et al.* (2012), and Schneider, Porras, and Schaetz (2012). Here, logic gate interactions between ions i and j invoke a spin-spin–like interaction of the form $\sigma_{\hat{u}i}\sigma_{\hat{u}j}$ where $\hat{u} \in \{\hat{x}, \hat{y}, \hat{z}\}$. Spin rotations about a direction $\hat{u}$ act like magnetic fields along $\hat{u}$. These basic interactions have been implemented on up to 16 ions in an rf trap (Schätz *et al*., 2007; Friedenauer *et al*., 2008; Kim *et al*., 2009, 2010; Edwards *et al*., 2010; Islam *et al*., 2012; Korenblit *et al*., 2012). One interesting aspect of this work is the study of quantum phase transitions by varying the relative

strengths of the (simulated) spin-spin and magnetic field interactions. Under appropriate conditions, the effects of spin "frustration" are now becoming apparent. The basic interactions have also been implemented on over 100 spins in a Penning trap experiment led by John Bollinger at NIST (Britton *et al.*, 2012), where the ions naturally form into a triangular array. In the Innsbruck group, simulations including engineered dissipation have also been implemented (Barreiro *et al.*, 2011; Blatt and Roos, 2012), and a striking demonstration of a digital quantum simulator has been made (Lanyon *et al.*, 2011; Blatt and Roos, 2012), in essence the first universal quantum computer.

VI(b). Spectroscopy and Quantum Metrology

Some potential applications of quantum control and QIP are motivated by the idea of using entangled states to improve spectroscopic sensitivity (Wineland *et al*., 1992, 1994; Bollinger *et al*., 1996; Leibfried *et al*., 2004; Roos *et al*., 2006; Goldstein *et al*., 2009) and demonstrations of this increased sensitivity have been made (Meyer *et al.*, 2001; Leibfried *et al*., 2004, 2005; Roos *et al.*, 2006; Leroux, Schleier-Smith, and Vuletić, 2010; Monz *et al*., 2011). These demonstrations were made in the limit that noise was dominated by "projection noise," the fundamental noise arising from the fluctuations in which state the system is projected into upon measurement (Wineland *et al*., 1982; Itano *et al*., 1993). This might be the case in a spectroscopy experiment where the interrogation time is limited by a particular experimental constraint, like the duration of flight of atoms in a cesium fountain clock or by the desire to hold the temperature of ions below a certain value if they are heated during interrogation. However, if significant phase noise is present in either the atoms themselves (Huelga *et al*., 1997) or the interrogating radiation (Wineland *et al*., 1998; Buzek, Derka, and Massar, 1999; André, Sørensen, and Lukin, 2004; Rosenband, 2012), the gain from entanglement can be lost. This puts a premium on finding probe oscillators that are stable enough that the projection noise dominates for the desired probe duration.

Some ions of spectroscopic interest may be difficult to detect because they either don't have a cycling transition or lack a cycling transition at a convenient wavelength. In some cases, this limitation can be overcome by simultaneously storing the ion(s) of spectroscopic interest

with a "logic" ion or ions whose states can be more easily detected. Following the Cirac and Zoller scheme, we can use the internal-to-motion-state-transfer process described above. Here, the idea is to first transfer the two states of interest in the spectroscopy ion to the ground and first excited states of a mode of the ions' coupled motion. This is then followed by mapping the motional states onto the logic ion, which is subsequently measured (Wineland *et al*., 2002). In a project led by Till Rosenband at NIST, this technique has been used to detect optical transitions in $^{27}Al^{+}$ ions by transferring the relevant $^{27}Al^{+}$ states to a $^{9}Be^{+}$ or $^{25}Mg^{+}$ logic ion, which is then measured (Schmidt *et al*., 2005). It is now used routinely in an accurate optical clock based on $^{27}Al^{+}$ (Rosenband *et al*., 2008; Chou, Hume, Koelemeij *et al*., 2010) and might also be extended to molecular ions. Currently, the $^{27}Al^{+}$ single-ion optical clock has the smallest systematic error of any clock at somewhat below 1 part in 10^{17} (Chou, Hume, Koelemeij *et al*., 2010). This level of precision has enabled observations of the predictions of Einstein's general theory of relativity on a human scale, such as time dilation for bicycling speeds and the gravitational redshift for height changes of around 30 cm (Chou, Hume, Rosenband, and Wineland, 2010). Such clocks may become useful tools in geodesy.

The information transfer and readout process employed in the $^{27}Al^{+}/^{9}Be^{+}$ clock experiments typically had a fidelity of about 0.85, limited by errors caused by the ions' thermal motion in modes not used for information transfer [so-called "Debye-Waller" factors from Mössbauer spectroscopy (Lipkin, 1973; Wineland *et al*., 1998)]. However, the quantum logic detection process is a QND type of measurement in that it doesn't disturb the detected populations of the $^{27}Al^{+}$ ion. It can therefore be repeated to gain better information on the $^{27}Al^{+}$ ion's (projected) state. By use of real-time Bayesian analysis on successive detection cycles, the readout fidelity was improved from 0.85 to 0.9994 (Hume, Rosenband, and Wineland, 2007). This experiment shares similarities with those of the Paris cavity- QED group, where successive probe atoms are used to perform QND measurements of the photon number in a cavity (Deléglise *et al*., 2008). In Hume, Rosenband, and Wineland (2007), the same atom ($^{9}Be^{+}$) is reset after each detection cycle and used again. Also, because the detection was accomplished in real time, the procedure was adaptive,

requiring on each run a minimum number of detection cycles to reach a certain measurement fidelity.

VII. Summary

I have tried to give a brief account of some of the developments that have taken place in the area of quantum state manipulation of small numbers of trapped atomic ions. With apologies, I have omitted several aspects of this subject and for the topics discussed here, I primarily used examples from the NIST, Boulder group. Much of the other work has been discussed in various comprehensive articles and reviews; see, for example, Cirac *et al*. (1996), Wineland *et al*. (1998), Šašura and Bužek (2002), Leibfried, Blatt, Monroe, and Wineland (2003), Lee *et al*. (2005), Blatt and Wineland (2008), Duan and Monroe (2008, 2010), Häffner, Roos, and Blatt (2008), Kielpinski (2008), Monroe and Lukin (2008), Blatt and Roos (2012), Korenblit *et al*. (2012), Monroe *et al*. (2012), and Schneider, Porras, and Schaetz (2012). Reviews on advanced clocks including those based on ions are contained in Gill (2005, 2011), Maleki (2008), and Margolis (2009) [see also Madej *et al*. (2012) and references therein].

Acknowledgments

Certainly my role in this work is very small when compared to that of my colleagues both at NIST and around the world, who have made so many important contributions. Having been recognized by the Royal Swedish Academy of Sciences is really more recognition of our field rather than individual accomplishment; many others are at least as deserving. Just the work of the NIST group was due to the combined efforts of a very large number of people. I have been lucky to work with NIST permanent staff members Jim Bergquist, John Bollinger, Bob Drullinger, and Wayne Itano for my entire career, and we have been fortunate to be joined by Didi Leibfried and Till Rosenband in the last decade. Chris Monroe was a very important part of our group from 1992 to 2000 and now has his own group at the University of Maryland. Of course our successes would not have happened if not for the dedication of many students, postdocs, and visiting scientists to our group, numbering over 100 people. Having a group working directly together or on related problems has been a source of strength for us, and the congenial

atmosphere over the years has made our efforts so enjoyable. Throughout my career, our group has enjoyed the support and encouragement of NBS/NIST management. My direct supervisors over the years, Helmut Hellwig, Sam Stein, Don Sullivan, and Tom O'Brian, have always supported our goals and desires as much as possible. More recently, we have also enjoyed the support of Carl Williams, who heads NIST's quantum information program. We are all indebted to our laboratory director, Katharine Gebbie, for her support and encouragement. Perhaps one measure of her success is that I am the fourth person, after Bill Phillips, Eric Cornell, and Jan Hall, to receive a Nobel Prize during her tenure as lab director. We are also grateful for the support of agencies outside of NIST, such as AFOSR, ARO, DARPA, ONR, and various intelligence agencies who have supported our work on quantum information. I have great respect for the leaders of some of our group's strongest competition such as Rainer Blatt (Innsbruck) and Chris Monroe (University of Maryland) and have enjoyed their friendship for many years. It was also a great pleasure to share this recognition with Serge Haroche. I have known Serge for about 25 years and have enjoyed both his group's elegant science and also the mutual friendship that my wife and I have shared with him and his wife, Claudine. Most importantly, I have been very fortunate to have the support, understanding, and patience of my wife Sedna and sons Charles and Michael. I thank John Bollinger, Wayne Itano, Didi Leibfried, and Till Rosenband for helpful suggestions on the manuscript.

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American Association of Physics Teachers, Chesapeake Section	Frank R. Haig, S. J.
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American Institute of Aeronautics and Astronautics	David W. Brandt
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American Meteorological Society	Vacant
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