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54. We thank the referees for constructive comments on this manuscript. Recent work performed at the Geophysical Laboratory and described above was supported by grants from the following agencies and institutions: the National Science Foundation (grants EAR-83114064, EAR-8319209, EAR-8418706), the National Aeronautics and Space Administration (grant NAGW-214), the U.S. Department of Energy (grant DE-AS05-80ER10754), and the Carnegie Institution of Washington.

Laser Spectroscopy of Trapped Atomic Ions

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Recent developments in laser spectroscopy of atomic ions stored in electromagnetic traps are reviewed with emphasis on techniques that appear to hold the greatest promise of attaining extremely high resolution. Among these techniques are laser cooling and the use of single, isolated ions as experimental samples. Doppler shifts and other perturbing influences can be largely eliminated. Atomic resonances with line widths of a few parts in 10^{11} have been observed at frequencies ranging from the radio frequency to the ultraviolet. Experimental accuracies of one part in 10^{18} appear to be attainable.

ATOMIC SPECTROSCOPY DATES FROM THE 19TH CENTURY, when it was discovered that atomic vapors emitted and absorbed light at discrete resonance wavelengths, characteristic of each chemical element. When the quantum theory of atoms was developed in the 20th century by Niels Bohr and others, it was realized that these characteristic patterns of resonances, called spectra, were due to the quantum nature of the atom. The atom normally exists only in certain states of definite energy. Transitions between these allowed states are accompanied by the absorption or emission of quanta of light, called photons. The frequency ν of the light is related to the energy change ΔE of the atom by the formula $h\nu = |\Delta E|$, where h is Planck's constant. Accurate and detailed information about atomic spectra was crucial to the development of the modern theory of quantum mechanics.

Traditional (that is, nonlaser) optical spectroscopic methods, such as dispersing the light emitted from a gas with a diffraction grating, are limited in resolution by Doppler frequency shifts. Doppler shifts are the result of the motions of the atoms in a gas and cause the resonance absorption or emission lines to be much broader than the natural line widths (the line widths that would be observed if the atoms were motionless and isolated from perturbing influences such as collisions). Under typical laboratory conditions, the Doppler broadening results in a line width of about one millionth of the transition frequency, whereas the natural line widths are typically at least 100 times narrower. For example, the 280-nm first resonance line of Mg^+, which has a frequency of about 1.07×10^{15} Hz, has a Doppler-broadened line width at room temperature of about 3 GHz, whereas the natural line width is only 43 MHz. For a transition with a stable lower level, the natural line width (in hertz) is the inverse of the mean lifetime of the upper level (in seconds), divided by 2π.

The development of tunable lasers in the 1970s led to great advances in the resolution and accuracy with which optical atomic spectra could be observed. Laser light sources have high intensity and narrow line width. These properties make it possible to use various nonlinear spectroscopic techniques, such as saturated absorption or multiphoton absorption, that cancel the effects of first-order Doppler shifts, that is, Doppler shifts that are linear in the velocities of the atoms (*1*). The natural line width and the second-order Doppler shift, which is quadratic in the atomic velocities, still remain, however. The second-order Doppler shift is a result of relativistic time dilation. The atomic resonance is shifted like a clock, which runs at a rate that is slower for a moving atom than for an atom at rest. For typical laboratory conditions, this shift is very small, about one part in 10^{12}. Sometimes, even this shift can be troublesome. A good example is the work of Barger *et al.* on the 657-nm transition of calcium, which has a frequency of 4.57×10^{14}

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Hz and a natural line width of 410 Hz (*2*). Resonances with line widths as small as 2 kHz were observed, but they were severely shifted and distorted by the second-order Doppler shift.

In order to reduce both the first- and second-order Doppler shifts, some method of reducing the temperature of the atoms under study is required. An atomic vapor placed in a conventional refrigeration device would quickly condense on the walls of the container. But laser cooling, a method by which laser radiation pressure is used to reduce the velocities of atoms, achieves the cooling without contact with material objects (*3–5*). The technique was proposed independently by Hänsch and Schawlow (*6*) for free atoms and by Wineland and Dehmelt (*7*) for trapped ions. Laser cooling to temperatures on the order of 1 mK has been demonstrated with some kinds of atoms and atomic ions. This reduces the second-order Doppler shifts to the level where they are not a problem. An atom or atomic ion of mass 100 u (unified atomic mass units) cooled to 1 mK has a second-order Doppler shift of 1.4 parts in 10^{18}.

Transit-time broadening is another effect that limits the resolution with which spectra can be observed. The observed resonance line width cannot be much less than the inverse of the observation time. This is a consequence of Heisenberg's uncertainty relation applied to time and energy. In the case of the work on calcium, the observation time was limited to about 0.3 msec by the time it took the atoms to pass through the 21-cm-long resonance region (*2*). One way to increase the observation time is by trapping the atoms with electromagnetic fields. In some experiments, atoms are confined with specially coated walls or by buffer gases, but collisions with the wall or buffer gas molecules shift and broaden the observed resonances. Atomic ions can be trapped for long periods by fields that do not disturb their resonance frequencies by significant amounts. Neutral atoms have been trapped by static magnetic fields (*8*) and by optical fields (*9, 10*), but the trapping fields tend to strongly perturb the resonances.

The work described in this article has the goal of achieving greatly improved spectroscopic resolution and accuracy. The experimental methods involve the use of frequency-stabilized lasers to measure the spectra of atomic ions, sometimes single ions, that are laser cooled and electromagnetically trapped. The most obvious applications of such work are frequency standards and clocks of great accuracy; this is the primary goal of the work done in our laboratories at the National Bureau of Standards (NBS). These experiments have applications to other areas of physics, however, such as atomic physics and quantum optics. One example is the recent observation of quantum jumps (sudden changes of quantum state) of an individual atomic ion (*11–13*).

Ion Traps

Ion traps confine ions by means of electric and magnetic fields (*14, 15*). Two types of ion traps are commonly used for spectroscopic experiments, the Penning trap and the Paul or rf (radio frequency) trap.

The Penning trap is based on a combination of static electric and magnetic fields. The electric fields are produced by applying an electric potential between the ring and endcap electrodes, which are shown schematically in Fig. 1. The electric forces repel the ions from the endcaps and provide confinement along the axis of the trap. A strong, uniform magnetic field is applied along the trap axis to provide radial confinement. Full three-dimensional confinement is provided by this combination of electric and magnetic fields. For a trap with inside dimensions of about 1 cm, a typical value of the electric potential difference is 1 volt and of the magnetic field, 1 tesla. The potential energy of an ion is lowered as it moves out radially from the axis of the trap. Hence, even though an ion would remain trapped forever if undisturbed, collisions with neutral background gas molecules increase the radial extent of the orbit until eventually the ion collides with the ring. In practice, under conditions of high vacuum, it is not uncommon for an ion to remain confined for days.

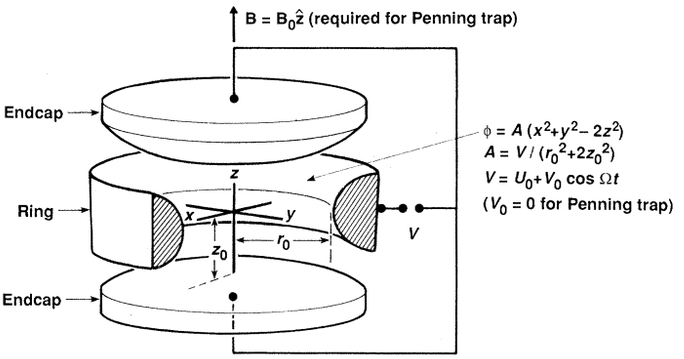

Fig. 1. Electrodes for a Penning or rf ion trap. The electric potential field ϕ is created by applying the voltage V between the endcap electrodes and the ring electrode. The uniform magnetic field **B** is required only for a Penning trap. [Adapted from (*42*) with permission from Plenum Press]

The rf trap uses oscillating electric fields and does not require a magnetic field. The electrode structure is the same as that of a Penning trap (see Fig. 1), but the applied electric potential varies sinusoidally in time. The electric field forces an ion to oscillate in position with an amplitude proportional to the field strength. The phase of the motion with respect to the applied field is such that the average force on the ion in the spatially nonuniform field is directed toward regions of weaker field (the center of the trap in this case). The trajectory of an ion can be separated into a part that oscillates at the frequency of the applied field, called the micromotion, and a part that varies more slowly, called the secular motion. The oscillating field creates an effective potential-energy well, sometimes called a pseudopotential, for the secular motion of an ion. For a trap with electrodes like those in Fig. 1, the pseudopotential is approximately a three-dimensional harmonic well, so the secular motion is characterized by well-defined frequencies. The electrodes of an rf trap identical to those used in the NBS experiments with Hg^+ ions (*12, 16, 17*) are shown in Fig. 2. The inside radius of the trap is slightly less than 1 mm. Under typical operating conditions, an electric potential of peak amplitude 570 volts and frequency 21 MHz is applied, creating a pseudopotential well about 15 eV deep. The frequency of the secular motion is about 1.5 MHz. In some cases, an ion can be held in the trap for several days before it is lost, possibly through a chemical reaction with a residual gas molecule.

Laser Cooling

It has been known for a long time that light can exert a force on a material body. In 1873, Maxwell showed that his theory of electromagnetic fields predicted that a beam of light would exert a force on a reflecting or absorbing body. This phenomenon is commonly known as radiation pressure. Light has momentum, the momentum density being numerically equal to the energy density divided by c, the speed of light. Radiation pressure is thus a consequence of conservation of momentum. In the early 1900s, radiation pressure was observed experimentally by Nichols and Hull in the United States (*18*) and by Lebedev in Russia (*19*), and shown to be in good agreement with theory. In 1933, Frisch first observed radiation pressure on the atomic scale (*20*). In his experiment, a beam of

sodium atoms traveling through an evacuated chamber was deflected by a lamp emitting sodium resonance light of wavelength 589 nm. The average deflection of an atom was that due to a transfer of momentum equal to that of a single photon, $h\nu/c$.

The force exerted by light on atoms is often divided into two parts. These are called the light-pressure or scattering force and the gradient or dipole force (*21*). In some simple cases, these two forces can be clearly distinguished, but in the general case, particularly for high light intensities, the simple descriptions of the forces break down, and a quantum-mechanical description is required.

The scattering force is simply radiation pressure at the atomic level, the force observed by Frisch (*20*). The average scattering force is in the direction of propagation of the light and is equal to the product of the momentum per photon and the photon scattering rate. The force reaches a maximum when the light is resonant with an atomic transition. Fluctuations in the scattering force arise because the photon scatterings take place at random times and because the direction of the reemitted photon, and hence the direction of the recoil momentum due to this reemission, is random.

The dipole force can be understood by considering the atom to be a polarizable body. The optical electric field induces an electric dipole moment in the atom; the induced dipole is acted on by the optical electric field. If the light intensity is spatially nonuniform, a force is induced parallel to the gradient of the intensity. The dipole force attracts an atom to a region of high light intensity if the frequency of the light is below the atomic resonance and repels it if it is above. Sodium atoms have been trapped, by means of the dipole force, near the focus of a laser beam tuned below the first resonance transition (*10*).

The laser cooling that has been demonstrated in ion traps is based on the use of the scattering force only. [It is also possible to cool by use of the dipole force (*22*), as has recently been demonstrated in an atomic beam experiment (*23*), but this method does not appear to have any advantage for trapped ions.] The theoretical description of the cooling depends on whether the natural line width γ of the resonance transition used for cooling is greater than or less than the frequencies of the oscillatory motion (ω_v) of the ion in the trap. The former case is called the heavy-particle or weak-binding limit; the latter is called the sideband-cooling or strong-binding limit (*3, 5, 24*). (Both γ and ω_v are expressed in angular frequency units.)

Consider first the heavy-particle limit ($\gamma >> \omega_v$, where ω_v is any of the motional frequencies). In this limit, each photon scattering event takes place in a time much less than a motional cycle, so that the scattering force can be considered to be made up of a series of impulses. Assume that a trapped ion is irradiated with a beam of light tuned lower than the resonance frequency. When the velocity of the ion is opposed to the direction of propagation of the laser beam, the frequency of the light in the frame of reference of the ion is Doppler-shifted closer to resonance than if it had zero velocity. Hence, the magnitude of the scattering force is higher when the atom moves against the laser beam, and its velocity is damped. When it moves in the same direction as the light, the light is Doppler-shifted away from resonance, and the magnitude of the scattering force is reduced. The net effect is to reduce the kinetic energy of the ion, that is, to cool it. (The ion is heated if the light is

Fig. 2. The electrodes of a small rf trap, shown on a coin for scale.

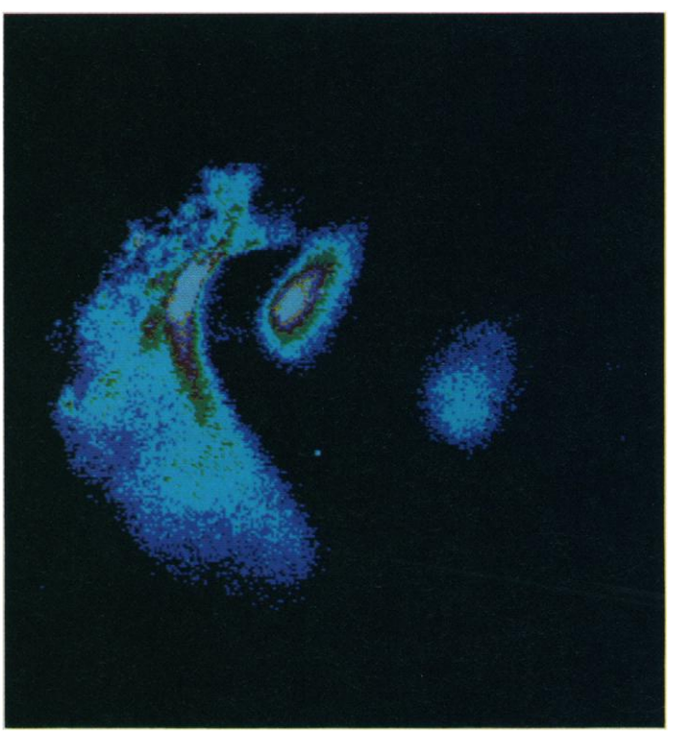

Fig. 3. False color image of a single Hg^+ ion (small isolated dot slightly below the center) stored in an rf trap. The other shapes are due to light reflected from the trap electrodes, whose orientation is the same as in Fig. 2. The largest reflection is from the front surface of the ring electrode. The two smaller reflections are from the edges of one of the endcap electrodes. Computerized imaging system developed by C. Manney and J. J. Bollinger (NBS). [Adapted from (*43*) with permission from the American Institute of Physics]

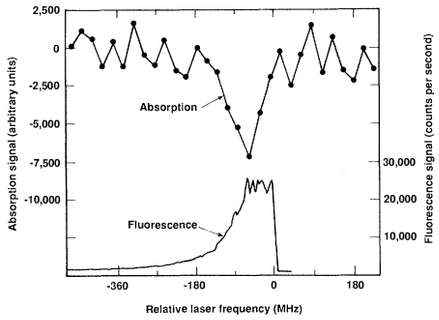

Fig. 4. The absorption signal of a single Hg^+ ion stored in an rf trap (upper trace). Radio-frequency modulation and demodulation techniques were used to reduce noise, resulting in a signal proportional to the derivative of the absorption with respect to laser frequency. The lower trace is the fluorescence signal, observed at the same time. The fluorescence drops off suddenly when the frequency is above resonance, due to laser heating. The flattened top of this curve is due to the frequency modulation. Integration times per point are 50 seconds and 10 seconds in the upper and lower traces, respectively. [Adapted from (*17*) with permission from the Optical Society of America]

higher in frequency than the resonance.) The minimum number of photon scatterings required to cool an ion substantially, starting from 300 K, is on the order of the ratio of the ion's momentum to a photon momentum, or about 10,000. Because of the inherent fluctuations of the scattering force, which cause heating, the kinetic energy is not damped to zero, but eventually reaches a steady value that depends on the degree of frequency detuning below the resonance. The detuning that gives the lowest temperature is equal to one half of the natural line width. This minimum temperature T_{min} is given by the relation $k_B T_{min} \simeq \hbar\gamma/2$, where k_B is Boltzmann's constant and $\hbar$ is Planck's constant divided by 2π. For example, T_{min} for Mg^+ ($\gamma/2\pi$ = 43 MHz) is 1 mK.

Now consider the sideband-cooling ($\gamma << \omega_v$) limit. The absorption spectrum of the ion consists of an unshifted resonance line at angular frequency ω_0, called the carrier, and a series of discrete lines on both sides of the carrier, each having (ideally) the natural line width, and separated by multiples and combinations of the motional frequencies. These extra lines, called motional sidebands, are the result of the periodic frequency modulation of the light frequency, arising from the Doppler effect, as observed by the moving ion. To cool an ion, the frequency of the laser beam is tuned to a sideband on the low-frequency side of the carrier, for example, to $\omega_0 - p\omega_v$, where p is a positive integer. The ion is induced to make transitions to the upper electronic state, with a decrease in the vibrational energy. When the ion makes a transition back to its ground electronic state, it may either increase or decrease its vibrational energy, but the average vibrational energy change is equal to $R = (\hbar\omega_0)^2/(2Mc^2)$, where M is the mass of the ion. The quantity R, sometimes called the recoil energy, is the kinetic energy that an initially motionless-free ion acquires after emitting a photon of energy $\hbar\omega_0$. If $R < p\hbar\omega_v$, the ion is cooled. If $R << \hbar\omega_v$, which is not hard to satisfy in practice, and if the frequency width of the light is much less than γ, then the ion can be cooled until it occupies the lowest quantum state of the trap potential most of the time. In this limit, the fraction of the time that the ion is not in the lowest state is on the order of $(\gamma/\omega_v)^2$ (*3, 5, 24, 25*). The sideband cooling discussion can also be applied to the $\gamma >> \omega_v$ case (*24, 26*). Recently, Lindberg, Javanainen, and Stenholm (*27*) have treated theoretically the general case of laser cooling of an ion trapped in a harmonic well, for an arbitrary ratio of γ to ω_v and also for arbitrary laser intensity. The lowest temperatures are obtained in the low-intensity limit.

The first experimental observations of laser cooling were made in 1978 at NBS by Wineland, Drullinger, and Walls (*28*) and at the University of Heidelberg by Neuhauser, Hohenstatt, Toschek, and Dehmelt (*26*). The NBS experiment used Mg^+ ions in a Penning trap, whereas the Heidelberg experiment used Ba^+ ions in an rf trap. In later experiments at these and other laboratories, cooling of trapped ions has been observed to temperatures in the range of a few millikelvin, close to the theoretical limit. So far, only strongly allowed transitions have been used for cooling, so the theory for the $\gamma >> \omega_v$ case applies.

Only a few kinds of ions have been laser-cooled. Laser cooling requires that the ion be cycled repeatedly between the ground state and an excited state. To be laser-cooled easily, an ion should have a strongly allowed resonance transition, at a wavelength where a tunable, continuous wave radiation source is available, and be free of intermediate metastable levels that would interrupt the cycling. Even Ba^+, one of the first ions to be laser-cooled, is not ideal, since it can decay into a metastable level from the upper level of the resonance transition. A second laser is required to drive the ion out of the metastable level. A way to cool a species of ion that cannot be laser-cooled directly is to store it in a trap together with a species that can be cooled directly. As one species is laser-cooled, the other species is cooled indirectly by Coulomb collisions. This technique, called sympathetic laser cooling, has been demonstrated in an experiment in which Hg^+ ions in a Penning trap were cooled to less than 1 K by Coulomb coupling with laser-cooled Be^+ ions (*29*).

Spectroscopy of Atomic Ions in Traps

A number of spectroscopic measurements have been made of non-laser-cooled ions in traps, particularly at microwave frequencies (*14, 15, 30, 31*). In the more accurate of these experiments, the uncertainty was due mainly to the second-order Doppler shift. For cases in which laser cooling can be applied, Doppler shifts are suppressed, and spectra can be observed with extremely high resolution, nearly free from perturbing influences.

Dicke showed in 1953 that the confinement of an atom to a

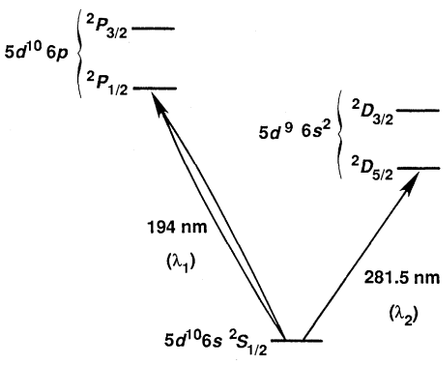

Fig. 5. Lowest energy levels of Hg^+. Absorption of a λ_2 photon is detected by the cessation of laser-induced fluorescence at λ_1. [Adapted from (*12*) with permission from the American Institute of Physics]

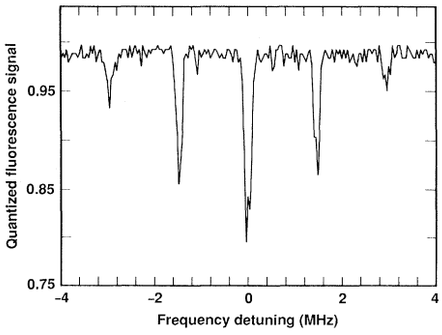

Fig. 6. The 281.5-nm transition observed in a single Hg^+ ion. The frequency detuning of the laser is plotted on the horizontal axis. The intensity ratios of the motional sidebands to the carrier (central line) indicate a temperature of about 6 mK. [Adapted from (*16*) with permission from the American Institute of Physics]

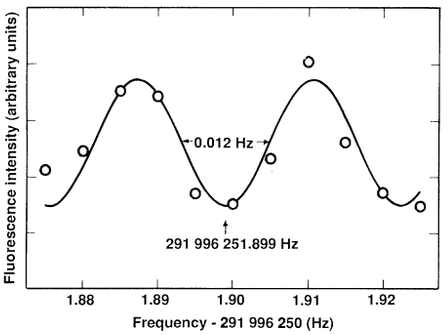

Fig. 7. Graph of a radio frequency hyperfine-structure resonance in $^{25}Mg^+$ ions stored in a Penning trap. Fluorescence intensity is plotted as a function of field frequency minus an offset of 291 996 250 Hz. Arrow indicates center of resonance. The oscillatory line shape results from the use of Ramsey's separated-oscillatory-field method. [Adapted from (*38*) with permission from the American Institute of Physics]

region of space smaller than the resonance-radiation wavelength leads to a suppression of the first-order Doppler shift broadening (*32*). It is easy to satisfy this condition, commonly called the Lamb-Dicke criterion, for a microwave resonance, since the wavelengths can be several centimeters long. Microwave frequency standards such as the atomic hydrogen maser make use of this fact. In the optical region, it can be satisfied for a single, laser-cooled ion confined in a trap. (Coulomb repulsion between ions makes it difficult to satisfy the Lamb-Dicke criterion when more than one ion is in the trap.)

The second-order Doppler shift of an ion that is laser-cooled to T_{min} is around one part in 10^{18} even for a strongly allowed transition (high γ). If more than one ion is present in the trap, then the kinetic energies per ion, and hence the second-order Doppler shifts, are necessarily higher. In the case of the rf trap, the Coulomb repulsion between ions leads to their being trapped away from the center of the trap, so the oscillating electric field, and hence the micromotion, is nonzero. In the case of the Penning trap, the presence of the other ions leads to increased velocities of rotation around the trap axis in the presence of crossed electric and magnetic fields. It appears that, for a single ion, shifts of resonance frequencies owing to electric and magnetic fields could be as small as a part in 10^{18}, since the ion is trapped in a region where the electric field approaches zero (*33, 34*).

Naturally, the signal-to-noise ratio suffers when the sample under observation consists of only a single ion. However, once an ion has been trapped and cooled, its presence can be detected easily by laser-induced fluorescence, since a strongly allowed transition can scatter as many as 10^8 photons per second. In fact, a single, trapped Ba^+ ion has been observed visually, through a microscope, by laser-induced fluorescence (*35*). Single ions have been observed in both Penning and rf traps (*11, 13, 15, 30, 35*). Figure 3 shows a false color image of the 194-nm laser-induced fluorescence of a single Hg^+ ion confined in an rf trap. This image was obtained at NBS with a position-sensing photomultiplier tube interfaced to a computer. It has even been possible to detect the resonance absorption due to a single Hg^+ ion by a decrease of the intensity of the 194-nm beam passing through the trap (*17*). The fraction of the light absorbed by the ion was around 10^{-5}. Figure 4 shows the absorption signal and the fluorescence signal, measured simultaneously as the laser was swept through the resonance.

A transition with a narrow natural line width would be difficult to detect directly by fluorescence, since the long lifetime of the upper state limits the rate at which photons are emitted. A way around this problem was suggested by Dehmelt (*36*). Consider an atom that has both a strongly allowed transition (at wavelength λ_1) and a weakly allowed transition (at wavelength λ_2) from the ground state. Figure 5 shows an example of such an atom (Hg^+). The narrow resonance at λ_2 is detected as follows: The atom is assumed to be initially in the ground state. Laser light at a wavelength near λ_2 is pulsed on, possibly driving the atom to the long-lived upper level of this transition. Then light at a wavelength near λ_1 is pulsed on. If the atom had made a transition in the previous step, no fluorescence would be observed; otherwise fluorescence will be observed at an easily detectable level. The detection of presence or absence of the λ_1 fluorescence is much easier than attempting to detect the one λ_2 photon that eventually is emitted when the long-lived state spontaneously decays. The method is called "electron shelving," since the optical electron is temporarily shelved in the long-lived level.

So far, only a few measurements of the optical spectra of trapped laser-cooled ions have been made. Some measurements have been made of strongly allowed transitions in Be^+ and Mg^+, with resolutions near the natural line widths of tens of megahertz. These measurements have yielded some values for energy level splittings, including fine-structure and hyperfine-structure splittings (*15, 30*). A two-photon transition from the ground $6^2S_{1/2}$ to the $5^2D_{3/2}$ state has been observed in a single Ba^+ ion, with a resolution, limited by the laser line width, of 3 MHz (*37*). Recently, at NBS, the $5d^{10}6s\ ^2S_{1/2}$ to $5d^96s^2\ ^2D_{5/2}$ 281.5-nm transition has been observed in a single, trapped Hg^+ ion (*16*). The ion was laser-cooled, by means of the 194-nm $6s\ ^2S_{1/2}$ to $6p\ ^2P_{1/2}$ transition, to near the theoretical limit of 1.7 mK. Figure 6 shows the spectrum obtained by tuning a laser across the narrow *S*-to-*D* transition. The electron shelving detection method was used. Immediately after the 281.5-nm radiation was shut off, the 194-nm fluorescence intensity was measured. If it was low enough to indicate that the ion had made a transition to the *D*-state, the signal was defined to be 0. Otherwise, it was defined to be 1. The average of these measurements is plotted along the vertical axis. The carrier and the two closest motional sidebands on each side are visible. The line widths were deliberately broadened to reduce the time required to sweep the resonance. On other sweeps, line widths of about 30 kHz were observed, a result mainly of laser frequency instability. The lifetime of the upper level is approximately 0.1 second, so the natural line width is about 1.6 Hz. The size of the sidebands indicates that the ion has been cooled approximately to the Lamb-Dicke regime. The root-mean-square amplitude of motion is estimated to be 50 nm.

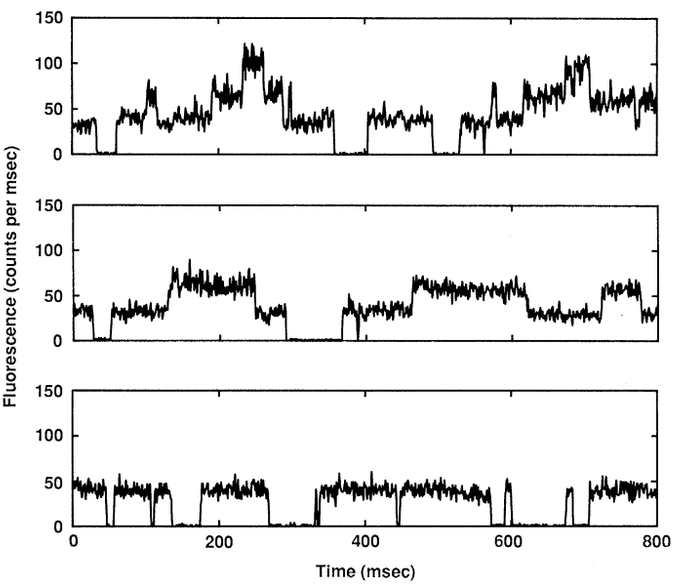

Fig. 8. Fluorescence intensity as a function of time for samples of three ions (top trace), two ions (middle trace), and one ion (bottom trace) in an rf trap. The fluorescence changes suddenly each time an Hg^+ ion either makes a transition to a metastable *D*-state (step down) or back to the ground *S*-state from a *D*-state (step up). The integration time per point is 1 msec.

Microwave spectra of trapped ions are usually observed by some form of microwave-optical double resonance. Hyperfine splittings and g-factors of the ground states of several atomic ions have been measured (*15, 30, 31*). So far, all of these experiments involve samples of more than one ion. Figure 7 shows an example of the kind of resolution that can be obtained with trapped ions (*38*). The line width of the resonance (a hyperfine transition in $^{25}Mg^+$) is only 12 mHz. The fractional resolution is about the same as for the Hg^+ S-to-D optical transition. In another experiment at NBS, the frequency of a hyperfine transition in $^9Be^+$ has been measured with an accuracy of about one part in 10^{13}, which is comparable to the most accurate frequency standards in existence (*39*). The accuracy was limited to this value by the second-order Doppler shift, because the laser cooling had to be shut off while the microwave transition was being driven, in order not to perturb the resonance frequency, and the ions heated up during this period. Sympathetic laser cooling using Mg^+ might provide a solution to this problem.

Quantum Jumps

Recently, Cook and Kimble (*40*) theoretically investigated a three-level atom like the one considered by Dehmelt in his "shelved electron" proposal. Light at wavelengths λ_1 and λ_2 was assumed to be present at the same time. They predicted that one would observe the λ_1 fluorescence to turn off and on abruptly as the atom made transitions (quantum jumps) to and from the long-lived upper level. This paper generated a great deal of theoretical interest, and the problem was approached from various viewpoints (*41*). The novelty of this problem is that theorists are accustomed to calculating, and experimenters are accustomed to measuring, ensemble averages rather than following the development in time of a single quantum system.

The experiment that is the most similar to the one treated by Cook and Kimble was performed at NBS on a single, trapped Hg^+ ion (*12*). Light at 194 nm (λ_1) and 281.5 nm (λ_2) was present at the same time (see Fig. 5). The 194-nm fluorescence level was observed to be bistable, switching suddenly between a steady level and zero. The rate at which the switches occurred increased as the intensity of the 281.5-nm light was increased. The statistical properties of the quantum jumps were consistent with theory and with previous measurements of the lifetime of the $^2D_{5/2}$ state. If the intensity of the 194-nm light is increased, then quantum jumps are observed even without the 281.5-nm light present. [These events occurred at a low rate in the data of (*12*).] The reason for these events is that the upper level of the 194-nm transition has a small probability (about 1 in 10^7) of decaying to the $^2D_{3/2}$ level rather than back to the ground state. The $^2D_{3/2}$ state has a lifetime of about 10 msec and about an equal probability of decaying either to the $^2D_{5/2}$ level or to the ground state. The bottom trace of Fig. 8 shows the 194-nm fluorescence intensity of a single ion as a function of time, clearly showing the quantum jumps. The middle and top traces in Fig. 8 show the fluorescence when two or three ions are in the trap. When more than one ion is present in the trap, the fluorescence level jumps between several discrete levels, depending upon how many ions are in one of the D-states. Quantum jumps have been observed by other groups, using trapped Ba^+ ions (*11, 13*).

Conclusions

Spectroscopy of trapped atomic ions has already achieved impressive levels of accuracy and resolution. The ultimate limits have certainly not been reached, and no fundamental (as opposed to technical) obstacles preventing the achievement of measurement accuracies (in special cases) of one part in 10^{18} are foreseen (*33, 34*). Interesting spin-offs from the quest for improved spectroscopic methods, such as the observation of quantum jumps, will no doubt continue to appear.

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44. We gratefully acknowledge the support of the Air Force Office of Scientific Research and the Office of Naval Research. This article is not subject to copyright in the United States.