High-Resolution Atomic Spectroscopy of Laser-Cooled Ions.

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1. – Introduction.

The experiments of Dehmelt and his collaborators in the 1960s[1] demonstrated that the stored-ion technique is useful for precise and accurate spectroscopy. The main reasons for this are: 1) Ions can be localized in space for long periods of time. This has the effects that the first-order Doppler shift averages to zero and that, in principle, very high resolution can be obtained because of the long resonance times. 2) When the ions are trapped in high vacuum, the perturbations to their internal structure (caused mainly by second-order Stark shifts[2]) are small. Historically, trapped-ion temperatures were typically above ambient temperatures so the uncertainty in the second-order Doppler shift limited the accuracy in very-high-resolution experiments. With laser cooling, this uncertainty can be significantly reduced.

One of the main interests of several laboratories is the development of frequency standards and clocks using stored ions. Although the examples discussed here are primarily taken from the work at NIST, the authors are aware of related work currently being pursued at (in alphabetical order) Communications Research Laboratory (CRL), Tokyo; Hamburg University; Hewlett Packard, San Jose; IBM, San Jose; Imperial College, London; Jet Propulsion Laboratory (JPL), Pasadena; Korea Standards Research Institute, Taejon, Korea; Laboratoire de l'Horloge Atomique (LHA), Orsay; Mainz University; Max Planck Institute, Garching; National Physical Laboratory (NPL), Teddington; National Research Council (NRC), Ottawa; National Research Laboratory of Metrology (NRLM), Tsukuba; Physikalisch-Technische Bundesanstalt (PTB),

Braunschweig; and the University of Washington, Seattle. This work has been summarized in ref. [3].

The high resolution obtained has also allowed searches for possible anomalous physical effects such as violations of local Lorentz invariance [4], nonlinear effects in quantum mechanics [5] and nonmagnetic spin-dependent forces [6]. The high detection sensitivity (discussed below) has allowed demonstrations of quantum properties of the electromagnetic field such as photon antibunching [7-10], and demonstrations of quantum measurements such as quantum jumps [8, 11-14] and the quantum Zeno effect [15]. In this lecture we do not discuss these experiments; rather, we concentrate on describing approaches to achieving very high accuracy and resolution in the spectroscopy of ions.

2. – Approach.

In attempting to achieve high resolution and accuracy in stored-ion experiments, we have made the following assumptions: 1) we would like a large number of ions in order to maximize signal-to-noise ratio, 2) for a «cloud» of many ions in the trap, the uncertainty in the measurement of the second-order Doppler shift will ultimately be the largest contribution to inaccuracy, and 3) for a cloud of ions, the magnitude of the second-order Doppler shift decreases as the number of ions in the trap decreases.

These assumptions are supported by experiment and theoretical analysis [16-21]. For a given number of ions, the second-order Doppler shift can be minimized when the temperature of the random or thermal degrees of freedom is reduced to a small value by some means (for example, by the use of buffer gas collisions [19, 20] or laser cooling [21, 22]). For a cloud of ions (which has three-dimensional extent), the motion in the nonthermal degrees of freedom dominates the second-order Doppler shift. For the Paul trap, the second-order Doppler shift is dominated by the velocity in the r.f. micromotion [16-20]. The basic idea is as follows: in the quadrupole Paul trap, the r.f. micromotion velocity increases with the distance of the ion from the center of the trap. As the number of trapped ions increases, space charge repulsion holds ions farther from the center of the trap thereby increasing the ions' micromotion speed and second-order Doppler shift. For a spherical cloud of ions, the number of ions N is proportional to the fractional second-order Doppler shift, averaged over the cloud, $\langle \Delta\nu_{D2}/\nu_0 \rangle$ [17-19]:

$$N = -2.16 \cdot 10^{16} r_{cl} M \langle \Delta\nu_{D2}/\nu_0 \rangle / Z^2 \qquad \text{(r.f. trap)}, \tag{1}$$

where r_{cl} is the cloud radius in centimeters, M is the ion mass in atomic mass units and Z is the ion charge in units of the proton charge. The number of ions for a given second-order Doppler shift can be increased by using a nonspherical ion cloud geometry in an elongated trap [20]. However,

the basic idea that as the number of ions in the cloud increases so does $|\langle \Delta\nu_{D2}/\nu_0 \rangle|$ still holds.

In the Penning trap, when the cyclotron and axial kinetic energies are reduced to small values, the second-order Doppler shift is dominated by the velocity of rotation of the ion cloud. In this case, N and $\langle \Delta\nu_{D2}/\nu_0 \rangle$ are related by [16-18]

$$N = 3.10 \cdot 10^{13} B \langle -\Delta\nu_{D2}/\nu_0 \rangle^{1/2} z_{cl} (r_{cl} - r_c')/Z \quad \text{(Penning trap)}, \tag{2}$$

where B is the trap magnetic-field strength in tesla, $2z_{cl}$ and r_{cl} are the cloud height and radius in centimeters, $r_c' \equiv (5\langle -\Delta\nu_{D2}/\nu_0 \rangle)^{1/2} c/\Omega_c$ (Ω_c is the ion's cyclotron frequency and c is the speed of light), and Z is the ion charge in units of proton charge. Equations (1) and (2) are valid for one species of ion in the trap.

Therefore, for a cloud of ions in either trap, $|\langle \Delta\nu_{D2}/\nu_0 \rangle|$ increases as N increases and we are faced with a compromise in design. For good signal-to-noise ratio and good frequency stability, we desire large N. However, this increases $|\langle \Delta\nu_{D2}/\nu_0 \rangle|$ and consequently decreases accuracy because of our inability to measure precisely the velocity distributions needed to determine $\langle \Delta\nu_{D2}/\nu_0 \rangle$. This trade-off between stability and accuracy has resulted in different approaches. In the work of one group [19,23], a stored-$^{199}Hg^+$-ion standard with excellent stability has been realized. In these experiments $N \simeq 2 \cdot 10^6$ and $\langle \Delta\nu_{D2}/\nu_0 \rangle \simeq -2 \cdot 10^{-12}$, so an accuracy of 10^{-14} would require a knowledge of $\langle v^2 \rangle$ to 0.5% precision. Inaccuracy due to second-order Doppler shifts can be reduced in the elongated trap geometry of ref. [20] but, independent of the trap geometry, the accuracy can generally be improved by using smaller numbers. (In the work of ref. [20], a short-term stability of $\sigma_y(\tau) \simeq 1.6 \cdot 10^{-13} \tau^{-1/2}$ was obtained.)

A special situation is realized for a single trapped ion or for a «string» of ions along the z-axis in the Penning or linear Paul trap (discussed below). In these cases, the second-order Doppler shift due to the nonthermal motion is approximately equal to that of the thermal motion, which can be very low.

At NIST, the primary goal of frequency standard work has been high accuracy. The preceding arguments have led us to use small ion clouds ($N \lesssim 10^4$) which are laser-cooled. The loss in short-term stability due to reduced numbers can be partially regained by going to very long interrogation times. These trade-offs are apparent from the expression for frequency stability if we assume the Ramsey method of interrogation and 100% detection efficiency [18]. For these conditions, the fractional frequency stability [24] can be expressed as [18]

$$\sigma_y(\tau) = (\tau N T_R \omega_0^2)^{-1/2} \qquad (\tau > T_R), \tag{3}$$

where τ is the averaging time, T_R is the Ramsey interrogation time, and ω_0 is the clock transition frequency (in radians per second). From this expression, we also see the importance of making ω_0 large.

Of course, to achieve high accuracy, we must also account for the perturbations due to static and time-varying multipole interactions for electric, magnetic and gravitational fields. These include atom-trap field interactions, collisions, shifts due to uncontrolled electric and magnetic fields, and gravitational red shifts. However, for a cloud of ions in the trap, the dominant uncertainty will probably be the uncertainty in the second-order Doppler shift. For a single ion or a string of ions, the limitation will probably depend on the experiment.

3. – $^9Be^+$ hyperfine clock.

In this experiment, an oscillator has been locked to the $(m_I = -1/2, m_J = 1/2) \leftrightarrow (-3/2, 1/2)$ nuclear spin-flip hyperfine «clock» transition ($\omega_0/2\pi \simeq 303$ MHz) in the ground state of $^9Be^+$ (fig. 1). The basic idea of this experiment has been described previously [5, 25-27]; the current experiment works as follows [5]: Between 5000 and 10000 $^9Be^+$ ions and 50000 and 150000 $^{26}Mg^+$ ions were simultaneously stored in a cylindrical Penning trap with $B \simeq 0.8194$ T under conditions of high vacuum ($\lesssim 10^{-8}$ Pa). At this magnetic field, the clock transition depends only quadratically on magnetic-field fluctuations, and, therefore, the accuracy is not limited by the small field fluctuations present in the experiment. To minimize second-order Doppler shifts of the clock transition, the $^9Be^+$ ions were cooled to less than 250 mK in the following manner: The $^{26}Mg^+$ ions were directly laser-cooled and compressed by a narrow-band ($\simeq 1$ MHz) laser radiation source at 280 nm. The $^9Be^+$ ions were then sympathetically cooled [28] by their Coulomb interaction with the cold Mg^+ ions. A

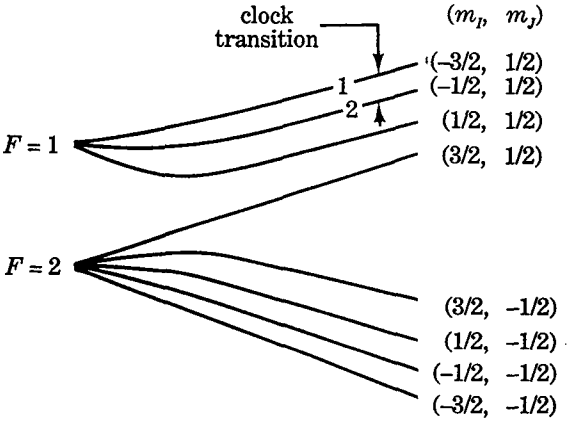

Fig. 1. – Hyperfine energy levels (not drawn to scale) of the $^9Be^+$ $2s\ ^2S_{1/2}$ ground state as a function of magnetic field. At $B = 0.8194$ T the 303 MHz clock transition is independent of magnetic field to first order.

narrow-band 313 nm radiation source was used to optically pump and detect the $^9Be^+$ ions [25-27]. With the 313 nm source tuned to the $2s^2S_{1/2}$ ($m_I = 3/2$, $m_J = 1/2$) to $2p\,^2P_{3/2}(3/2, 3/2)$ transition, 94% of the $^9Be^+$ ions were optically pumped into the $2s^2S_{1/2}(3/2, 1/2)$ ground state. The 313 nm source was then turned off to avoid optical pumping and a.c. Stark shifts while the clock transition was driven.

The clock transition was detected by the following method: After the 313 nm source was turned off, the ions in the $(3/2, 1/2)$ state were transferred to the $(1/2, 1/2)$ state and then to the $(-1/2, 1/2)$ state by two successive r.f. π pulses. Each pulse was 0.2 s long and was resonant with the appropriate transition frequency (around 321 MHz and 311 MHz, respectively). The clock transition was then driven by Ramsey's method of separated oscillatory fields (in the time domain) with r.f. pulses of about 1 s duration and a free precession time on the order of 100 s. This transferred some of the ions from the $(-1/2, 1/2)$ state to the $(-3/2, 1/2)$ state. Those ions remaining in the $(-1/2, 1/2)$ state were then transferred back to the $(3/2, 1/2)$ state by reversing the order of the two r.f. π pulses. The 313 nm source was then turned back on, and the population of ions in the $(-3/2, 1/2)$ state was registered as a decrease in the $^9Be^+$ fluorescence, relative to the steady-state fluorescence, during the first second that the 313 nm source was on. (The optical-repumping time of the ions from the $(-3/2, 1/2)$ state to the $(3/2, 1/2)$ state was about 10 s.) The sympathetic cooling of the $^9Be^+$ ions by the continuously cooled Mg^+ ions is necessary if long interrogation times are to be used, since otherwise the $^9Be^+$ ions would heat up while the 313 nm laser is off [27].

The Ramsey signal was used to steer the frequency of a synthesized r.f. source [5, 25-27]. Measurements were taken near the frequencies corresponding to the half-minimum points of the Ramsey signal on both sides of the center frequency. The difference in the measured signal strengths on either side of the line center was used to electronically steer the average frequency of the synthesizer to ω_0. Most runs were taken with a commercial cesium beam clock (fractional frequency stability $\sigma_y(\tau) \simeq 6 \cdot 10^{-12}\tau^{-1/2}$ for measurement time τ in seconds) as the reference oscillator, but a few runs were taken with a passive hydrogen maser ($\sigma_y(\tau) \simeq (2 \div 3) \cdot 10^{-12}\tau^{-1/2}$) as the reference oscillator. Stabilities of the $^9Be^+$ clock were measured to be better than $3 \cdot 10^{-12}\tau^{-1/2}$ for the number of ions used, which is within a factor of 4 of the theoretical maximum stability given by eq. (3). The systematic offset of our measurement due to the second-order Doppler frequency shift was dominated by the velocity in the rotation motion of the ions about the trap axis. This shift was measured to be $(-1.2 \pm 0.5) \cdot 10^{-14}$.

A pressure shift with an unexpectedly large value was discovered when the background gas pressure was increased. The background gas pressure could be increased by moving the magnet of the sputter ion pump which evacuated the trap region so that it overlapped fewer pumping cells and reduced the pumping

speed. (We checked to make sure the magnetic field at the site of the ions was not disturbed.) The composition of the gas was not known since the pressure was measured with a Bayard-Alpert gauge. The observed pressure shift is approximately 1000 times larger than those observed for inert gases on $^{137}Ba^+$[29] and $^{199}Hg^+$[23].

The large difference between our measured pressure shift and other measured pressure shifts is not understood at this time. One possible explanation is suggested by studies [30] of radiative association of C^+ with H_2 to form CH_2^+. In the models of this process, it is assumed that the H_2 can stick to the C^+ for a long enough time to allow the C^+-H_2 complex to radiatively stabilize. This sticking is possible because the collision energy can be taken up by the internal degrees of freedom in the H_2 molecule or the C^+-H_2 complex. The sticking time can be orders of magnitude longer than the interaction time during a simple elastic collision. If a similar sticking mechanism is active in Be^+ collisions with the background gas, it may account for the apparent large pressure shift.

The uncertainty in this pressure shift limits the accuracy of the current $^9Be^+$ clock measurements to around 1 part in 10^{13}. Eventually, it may be necessary to use liquid-He cryopumping to reduce the background pressure.

4. – $^{199}Hg^+$ optical clock.

The velocity in the micromotion for an ion in a quadrupole Paul trap is proportional to the distance of the ion from the center of the trap. For two or more laser-cooled ions in the trap, the Coulomb repulsion between ions holds them away from the trap center, and the second-order Doppler shift is dominated by the velocity of micromotion. However, a single ion can be held near the trap center if sufficiently cooled. In this case the kinetic energy in the micromotion can be about equal to that of the secular motion [17, 18, 31, 32]. If the ion is laser-cooled, resulting Doppler shifts can be extremely small; uncertainties can be less than 1 part in 10^{20} in some cases [32, 33]. However, with $N = 1$, stability is marginal unless we make ω_0 high enough (see eq. (3)). One way to accomplish this is to let ω_0 correspond to an optical transition. DEHMELT suggested this idea in 1973 [21]. The reasons that a clock based on an optical transition in an ion has not been realized yet are: 1) it took several years to isolate and manipulate single ions in the traps, 2) local oscillators (visible lasers) with linewidths of a few tens of hertz are only recently available and linewidths below 1 Hz are still desired, and 3) accurate comparison of laser and microwave frequencies is extremely difficult and remains an important problem. Nevertheless the potential accuracy ($\simeq 10^{-18}$) of single-ion optical frequency standards is extremely high [21, 34], so it is important to pursue this research.

At NIST we have investigated the use of the $5d^{10}6s\ ^2S_{1/2} \rightarrow 5d^9 6s^2\ ^2D_{5/2}$ electric-quadrupole transition ($\omega_0/2\pi \simeq 1.07 \cdot 10^{15}$ Hz) in $^{198}Hg^+$ and $^{199}Hg^+$ (see

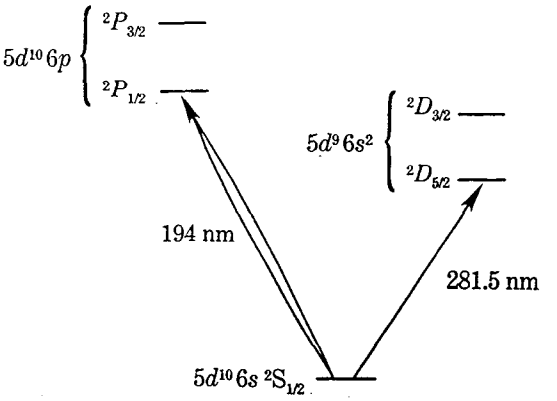

Fig. 2. – Simplified optical energy level diagram for Hg^+. The lifetime of the $^2D_{5/2}$ level is about 0.1 s which would give a natural linewidth of approximately 2 Hz on the electric-quadrupole $^2S_{1/2} \rightarrow {}^2D_{5/2}$ transition. By observing the presence or absence of fluorescence from the $^2S_{1/2} \rightarrow {}^2P_{1/2}$ transition, the quadrupole «clock» transition can be detected with 100% efficiency.

fig. 2) as an optical frequency standard [35]. The single mercury ion is confined in a miniature r.f. trap that has internal dimensions of $r_0 \simeq 466\,\mu m$ and $z_0 \simeq$ $\simeq 330\,\mu m$ [35, 36]. The amplitude of the trapping field (frequency $\Omega/2\pi \simeq$ $\simeq (21 \div 23)$ MHz) can be varied to a peak of 1.2 kV. The ion is laser-cooled to a few millikelvin by a few microwatt of c.w. laser radiation that is tuned below the $^2S_{1/2}$-$^2P_{1/2}$ first resonance line near 194 nm. In order to cool all motional degrees of freedom to near the Doppler-cooling limit [22] ($T = \hbar\gamma/2k_B \simeq 1.7$ mK), the 194 nm radiation irradiates the ion from 2 orthogonal directions, both of which are at an angle of 55° with respect to the symmetry (z) axis of the trap. The 282 nm radiation that drives the narrow $^2S_{1/2}$-$^2D_{5/2}$ transition is obtained by frequency-doubling the radiation from a narrow-band c.w. ring dye laser. The frequency of the laser is stabilized by locking it to a stable Fabry-Perot cavity. The frequency of the laser is scanned by an acousto-optic modulator that is driven by a computer-controlled synthesizer. Up to a few microwatt of 282 nm radiation could be focussed onto the ion in a direction counterpropagating with one of the 194 nm light beams.

Optical-optical double resonance was used to detect transitions driven by the 282 nm laser from the ion's $^2S_{1/2}$ ground state to the metastable $^2D_{5/2}$ state [35-37]. The 194 nm fluorescence rate from the laser-cooled ion is high when the ion is cycling between the $^2S_{1/2}$ and $^2P_{1/2}$ states (fig. 2) and essentially zero when the ion is in the metastable $^2D_{5/2}$ state. The $^2S_{1/2}$-$^2D_{5/2}$ resonance spectrum was obtained by probing the S-D transition at a particular frequency for the 282 nm radiation for 20 ms, then turning off the 282 nm radiation and turning on the 194 nm radiation to look for the presence or absence of scattered photons at 194 nm. The two radiation fields are alternately applied to avoid

light shifts and broadening of the narrow S-D transition by the 194 nm radiation. If there was no fluorescence at 194 nm, a transition into the metastable D state had occurred (that is the electron was «shelved» [38] into the D state); the presence of 194 nm fluorescence indicated that the ion was in the ground state and no transition was recorded for this frequency of the 282 nm laser. The frequency of the 282 nm radiation was then stepped and the measurement cycle repeated. As the frequency was swept back and forth each new result at a particular frequency of the 282 nm radiation was averaged with the previous measurements at that frequency. Normalization (or digitization) of the signal was obtained by assigning a 1 to each measurement of high fluorescence and a 0 to each measurement of no fluorescence. The high fluorescence made it possible to determine the state of the atom with almost no ambiguity in a few milliseconds.

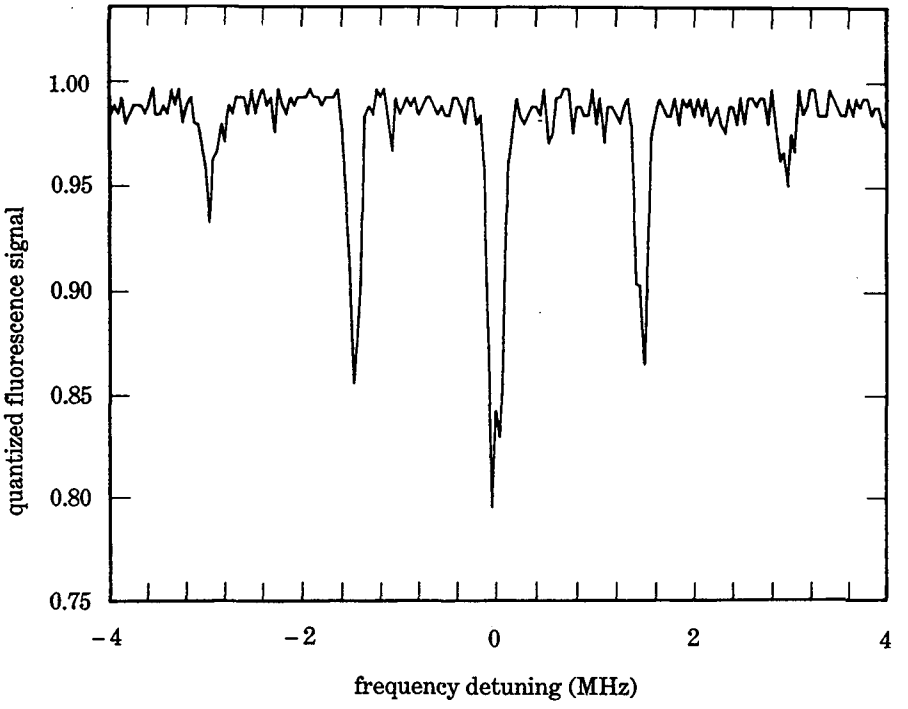

Fig. 3. – Quantized signal showing the electric-quadrupole-allowed $5d^{10}6s\ ^2S_{1/2}(m_J = -1/2)$-$5d^9 6s^2\ ^2D_{5/2}(m_J = 1/2)$ transition in a single, laser-cooled $^{198}Hg^+$ ion. On the horizontal axis is plotted the relative detuning from line center in frequency units at 282 nm. On the vertical axis is plotted the probability that the fluorescence from the $6s\ ^2S_{1/2}$-$6p\ ^2P_{1/2}$ first-resonance transition, excited by laser radiation at 194 nm, is on. The electric-quadrupole-allowed S-D transition and the first-resonance S-P transition are probed sequentially in order to avoid light shifts and broadening of the narrow S-D transition. Clearly resolved are the recoilless absorption resonance (carrier) and the Doppler sidebands due to the residual secular motion of the laser-cooled ion. Each point is the average of 230 measurement cycles (from ref. [36]).

Thus it is possible to reach the shot noise limit imposed by the single atomic absorber [36].

The quantized fluorescence signal obtained from an 8 MHz scan of the 282 nm laser through the $^2S_{1/2}(m_J = -1/2) \rightarrow {}^2D_{5/2}(m_J = 1/2)$ Zeeman component of the electric-quadrupole transition in $^{198}Hg^+$ is shown in fig. 3. The recoilless absorption resonance (carrier) and the motional sidebands due to the secular motion in the harmonic well of the r.f. trap are completely resolved [36].

To avoid broadening of the quadrupole transition due to magnetic-field fluctuations, we have more recently [35, 37] performed the same experiment on the $^2S_{1/2}(F = 0, m_F = 0) \rightarrow {}^2D_{5/2}(F = 2, m_F = 0)$ transition in $^{199}Hg^+$ which becomes field independent as $B \rightarrow 0$. The carrier is now observed with a linewidth $\Delta\nu \leqslant$ $\leqslant 100$ Hz (limited by laser spectral purity), which gives a line Q of about 10^{13}, the highest reported in atomic or molecular spectroscopy. Current efforts are devoted to improving the 282 nm laser spectral purity by locking it to a more stable reference cacity [35]. If the laser's spectral purity can be made high enough, then, when the laser is locked to the ion transition, stabilities are anticipated to be better than $10^{-15}\tau^{-1/2}$ and accuracies could be 1 part in 10^{18} or better.

5. – Future Penning trap experiments.

The $^9Be^+$ ion experiments have the primary disadvantage that ω_0 is relatively low and the resulting frequency stabilities are modest. We might hope to substitute $^{201}Hg^+$ ions in place of the $^9Be^+$ ions because $^{201}Hg^+$ has a higher-frequency clock transition ($\omega_0/2\pi \simeq 26$ GHz) which is field independent at $B = 0.534$ T [16]. However, two disadvantages compared to the $^9Be^+$ case arise: 1) If the Hg^+ is sympathetically cooled by lighter ions such as Mg^+ or Cd^+, it will reside in an annulus surrounding the lighter ions [28]; this makes the second-order Doppler shift larger for a given density and number of ions. 2) $^9Be^+$ and $^{25}Mg^+$ have simple optical-pumping schemes whereby a single laser frequency can be used to optically pump into nearly a single ground-state sublevel. For $^{201}Hg^+$ or $^{199}Hg^+$ in a strong field (required for a Penning trap) optical-pumping schemes would require auxiliary laser lines at 194 nm and microwave oscillators to manipulate the ground-state sublevels [16]; the simple optical-pumping schemes as in the case of $^9Be^+$ and $^{25}Mg^+$ do not appear possible. $^{199}Hg^+$ in a Penning trap would provide a very interesting system when magnets of high enough field strength become available. For example, the $(m_I = = 1/2, m_J = 1/2) \leftrightarrow (-1/2, 1/2)$ hyperfine transition in the ground state of $^{199}Hg^+$ ($\omega_0/2\pi \simeq 20.9$ GHz) is field independent at $B = 43.9$ T. At present, we must await the required magnet.

Within the limits imposed by today's techcnology, an experiment similar to

the $^9Be^+$ experiment but with better expected performance might be provided by $^{67}Zn^+$ ions [39]. The clock transition could be the $(m_I = 2/5, m_J = 1/2) \leftrightarrow$ $\leftrightarrow (3/2, 1/2)$ transition ($\omega_0/2\pi \simeq 1\,\mathrm{GHz}$) which is field independent at $B \simeq 8\,\mathrm{T}$. Some other examples are summarized in ref. [40].

6. – Future Paul trap experiments.

The advantage of the Paul trap is that a magnetic field is not required for trapping. This allows us to be rid of a cumbersome magnet and allows use of transitions which are field independent at $B = 0$. The primary disadvantage is that up to the present time it has been impossible to laser cool very many ions ($N > 100$). As discussed above, the use of small numbers may not be a limitation if ω_0 can be made big enough. This is the basic philosophy behind the single-ion optical frequency standards. Even for $\omega_0/2\pi = 40.5\,\mathrm{GHz}$ ($^{199}Hg^+$) and $N = 1$, from eq. (3) we can expect [18] $\sigma_y(\tau) = 3.9 \cdot 10^{-13}\tau^{-1/2}$ when $T_R = 100\,\mathrm{s}$. Because the second-order Doppler shift is expected to be so small for small numbers of ions, we examine this case more closely.

The main advantage of using a single ion in a quadrupole Paul trap is that the kinetic energy of micromotion can be on the order of the secular-motion energy. For a single $^{199}Hg^+$ ion cooled to the Doppler-cooling limit [36], the second-order Doppler shift would be [18, 32] $\langle \Delta\nu_{D2}/\nu_0 \rangle = -2.3 \cdot 10^{-18}$. In a quadrupole ion trap, two or more ions in the trap are pushed from the center of the trap by the mutual Coulomb repulsion and the second-order Doppler shift is higher. Consider the trap shown in fig. 4. In this trap, the r.f. electric fields are transverse to the trap axis for the entire z extent of the trap. If a single string of ions is trapped along the z-axis, then the kinetic energy of micromotion is about

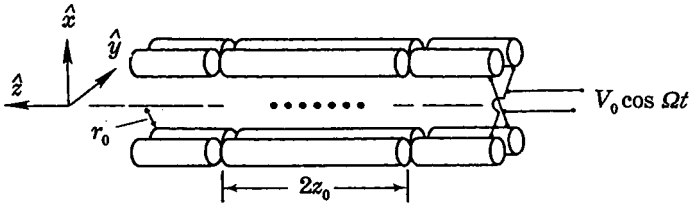

Fig. 4. – Linear trap configuration. The alternating r.f. voltage $V_0 \cos\Omega t$ is assumed to be applied to diagonally opposing electrodes as shown. We assume the end portions of the electrodes are long enough that the resulting r.f. potential at the position of the ions is independent of z, so that the r.f. electric fields are parallel to the (x, y)-plane. To trap ions along z, the center four electrodes are set to static ground potential and the two sets of four electrodes on either end are set at a static potential U_0 ($U_0 > 0$ to trap positive ions). The average position of the ions could be made to coincide with the r.f. electric-field null by applying different static potentials to the four central rods to correct for contact potential offsets etc. This geometry would allow laser beams to be directed along the z-axis. Such a trap might also be useful for studying fundamental atom-radiation interactions such as the statistics of fluorescence from or the interaction of a cavity with two or more atoms whose positions are localized to less than $\lambda/2\pi$.

equal to the kinetic energy in the secular motion. The fractional second-order Doppler shift could be as low as $5kT/2mc^2$. This is 5/6 of the value for a quadrupole r.f. trap [17] because of the absence of r.f. micromotion along the z direction. At the Doppler-cooling limit, this gives $\Delta\nu_{D2}/\nu_0 \simeq -2\cdot10^{-18}$ for Hg^+ ions in the string. This kind of trap was first demonstrated for atomic ions by DREES and PAUL [41] and CHURCH [42]. PRESTAGE *et al.* [20] have demonstrated a $^{199}Hg^+$ microwave clock with excellent short-term stability using a cloud of ions elongated in the z direction in a linear Paul trap. DEHMELT suggested using a thin string of ions to suppress the second-order Doppler shift in such a trap [38]. Strings of atomic ions have been observed at Garching [43] and NIST [44].

Use of the trap of fig. 4 would allow $N \gg 1$ giving good stability and still yield a small second-order Doppler shift. For the experimentally achievable conditions given in ref. [40], $N = 50$ ions could be stored along the z-axis like «beads on a string» with ion spacings of approximately 5 μm. With this spacing each ion could be independently detected by using an image detector [45, 46]. Therefore, each ion could be treated as an independent atomic clock where the clock transition could be detected with 100% efficiency [36]. From eq. (3), for $T_R = 100$ s and $\omega_0/2\pi = 40.5$ GHz ($^{199}Hg^+$) the frequency stability of this clock «ensemble» would be $\sigma_y(\tau) = 5.5\cdot10^{-14}\tau^{-1/2}$. For these long interrogation times, sympathetic cooling might be required to avoid heating while the Hg^+ optical-pumping laser was turned off to avoid light shifts during the Ramsey period. The ions used to cool the Hg^+ ions would also find a position in the string of ions along the z-axis. Arrays of traps have also been proposed previously [47]. These trap arrays would have the same advantages as above for increasing N and minimizing the second-order Doppler shift.

For optical spectroscopy, lasers with high spectral purity are required. This is the current limitation to reaching the natural linewidth resolution in the NIST Hg^+ optical experiments. Lasers which are locked to reference cavities have been shown to track the cavity frequency to precisions of much less than 1 Hz [44, 48]. The problem then remains that the length of the cavity and, therefore, the frequency of the laser are modulated by acoustic noise with deviations typically much greater than 1 Hz. It is desired to make this reference cavity stable enough so that the frequency deviations of the laser are less than the inverse of the attack time required to lock the laser to the ion resonance. The cavity-stabilized laser can be locked to the ion resonance line in a manner similar to that described for the $^9Be^+$ hyperfine transition. This has been demonstrated in ref. [37].

The main systematic error for the Hg^+ optical experiment may ultimately be the uncertainty in the shift of clock transition from static electric fields of quadrupole symmetry such as those caused by static potentials applied to the trap electrodes [21]. The basic idea is that the D state of the clock transition has a quadrupole moment that can interact with static fields of quadrupole symme-

try to give a shift which must then be measured. Although it is troublesome, we should be able to remove this offset from a clock calibration to about the same precision as the measurement precision [18]. DEHMELT has pointed out the advantage of the singly ionized group IIIA atoms in this respect [21]; the interesting clock transitions in these ions are the $^1S_0 \rightarrow {}^3P_0$ intercombination lines which are not shifted by static quadrupole electric fields. However, at low magnetic field these transitions have magnetic-field dependence on the order of the nuclear Zeeman effect. Therefore, careful control of the magnetic field would be required [21]. At higher fields, field-independent transitions could be used to advantage [49]. A linear trap or trap arrays could be used to increase signal-to-noise ratio as described above. For clock transitions involving a state with a quadrupole moment, such as for Hg^+, the mutual Coulomb interaction between ions would cause an additional quadrupole frequency shift which must be taken into account [2]. For the group IIIA ions, this shift would be absent.

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