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LASER COOLED, STORED ION EXPERIMENTS AT NBS AND POSSIBLE APPLICATIONS TO MICROWAVE AND OPTICAL FREQUENCY STANDARDS

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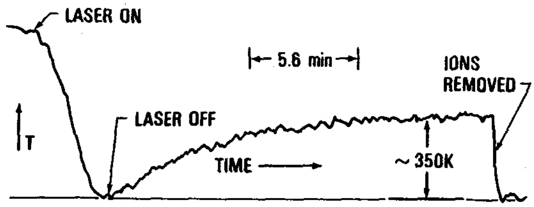
Abstract. - Research on stored ion frequency standards at the United States National Bureau of Standards is briefly discussed. We summarize past work and indicate directions of future research.

Introduction. - The purpose of this paper is to briefly summarize the work at the National Bureau of Standards (NBS) which has been directed towards realizing a frequency standard based on stored ions. The similar work of other groups [1] is not discussed here. This summary briefly describes past work and indicates future directions of research at NBS.

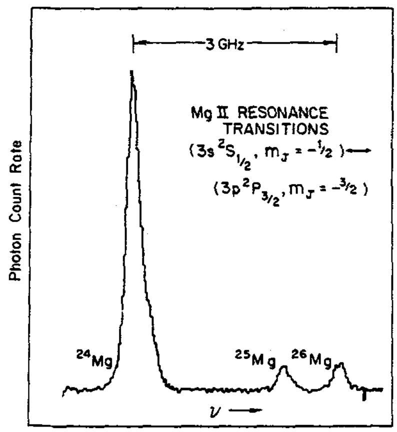
The stored ion work at NBS was initiated in the Fall of 1977. The goal of this work has been to devise a technique for realizing a frequency standard whose accuracy would be significantly better than that of the Cs atomic beam frequency standard. With this in mind, the ion storage method [2] was pursued because it can provide long confinement times without the usual perturbations associated with confinement (e.g., the wall shift in the H-maser).

The initial work was directed toward realizing laser cooling of ions [3], since this would provide a way to substantially suppress the frequency shift due to the second-order Doppler or time-dilation effect. For these initial experiments, Mg^+ ions were stored in a Penning trap. Mg^+ ions were chosen, since the electronic structure is fairly simple (i.e., like that of neutral alkali atoms) and required a laser source at 280 nm. The Penning trap was chosen because the residual heating mechanisms (at least for a cloud of ions) are less than for the rf trap.

In the first experiments [4], the temperature of the ions was monitored directly by the bolometric technique [5]. Figure 1 shows the Mg^+ ion temperature vs. time for a fixed laser detuning of about -2 GHz from the 3s $^2S_{1/2}$ ($M_J = -1/2$) → 3p $^2P_{3/2}$ ($M_J = -3/2$) transition frequency. The sensitivity of these temperature measurments was limited by noise in the electronic detection. Therefore, subsequent measurements detected the temperature by observing the fluorescence light scattered by the ions and measuring the Doppler widths of the optical lines [6].



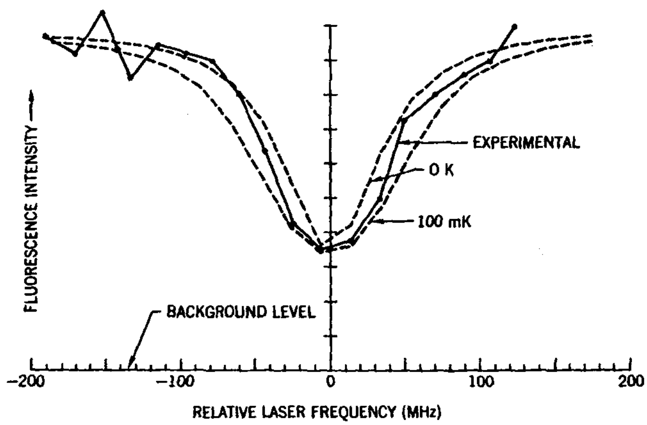
1. Ion temperature vs. time when laser cooling is applied for fixed laser detuning of -2 GHz. The ions were initially heated above equilibrium temperature with the laser. Laser cooling was then applied on the 3s $^2S_{\frac{1}{2}}$ $(M_J = -\frac{1}{2})$ $\rightarrow$ 3p $^2P_{3/2}$ $(M_J = -^3/_2)$ transition for a fixed time until a temperature approaching 0 K (< 40 K) was achieved. After the laser is turned off, the ions rethermalize to the ambient temperature.



2. Spectra of one Zeeman component of laser cooled $^{24,25,26}Mg^+$. The room temperature Doppler width of these lines is about 3 GHz. Only the $^{24}Mg^+$ is directly laser cooled. The $^{25}Mg^+$ hyperfine structure has been optically pumped resulting in the observation of only the $(M_J = -\frac{1}{2}, M_I = -^5/_2) \leftrightarrow (M_J = -\,^3/_2, M_I = -^5/_2)$ component.

Figure 2 shows the fluorescence light from a "low" power laser which is swept in frequency across the 3s $^2S_{\frac{1}{2}}$ $(M_J = -\frac{1}{2}) \rightarrow$ 3p $^2P_{3/2}$ $(M_J = -^3/_2)$ optical transitions of the three naturally occuring Mg^+ isotopes. At the same time, a higher power (approximately 10 times higher power) fixed-frequency laser is tuned to the low frequency side of the $^{24}Mg^+$ transition to keep the sample cold. (Note that the $^{25}Mg^+$ and $^{26}Mg^+$ isotopes are not directly laser cooled, but are cooled by Coulomb collisions with the cooled $^{24}Mg^+$.)

Lowest temperatures have so far been obtained for single ions [7] where cyclotron-magnetron "temperatures" of approximately 0.05 K have been obtained for a single $^{24}Mg^+$ ion. Figure 3 shows the double-resonance detection of the $^2S_{1/2}$ $(M_J = -1/2) \rightarrow {}^2P_{3/2}$ $(M_J = -1/2)$ optical transition in a single $^{24}Mg^+$ ion. Our theore-

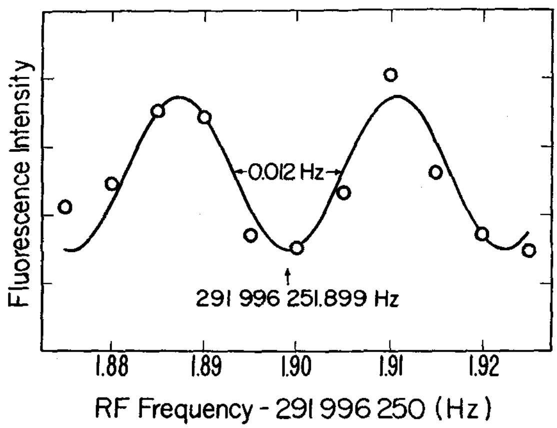


3. Double-resonance curve of a single $^{24}Mg^+$ ion. On the vertical axis is the fluorescence from a fixed-frequency laser (power approximately 5 μW) tuned to the $(^2P_{3/2}, M_J = -3/2) \leftarrow (^2S_{1/2}, M_J = -1/2)$ transition. Each point represents a 10 s integration; the connecting lines are only for clarity. The horizontal axis is the frequency of the low-power (<< 5 μW) laser which is continuously scanned across the $(^2P_{3/2}, M_J = -1/2) \leftarrow (^2S_{1/2}, M_J = -1/2)$ transition. The dashed curves are simulations of fluorescence at T = 0 K and 100 mK (no added noise). The solid curve is experimental data. From these data, we conclude T = 50 ± 30 mK.

tical predictions [8, also Itano, Wayne M., and Wineland, D. J., to be published] indicated that lower temperatures should be obtained; possible limitations may be caused by the presence of impurity ions in the trap. Future experimental investigations are planned; a new apparatus with improved fluorescence collection efficiency is being constructed.

In the Penning trap, ions are unstable with respect to collisions with background gas; that is, collisions cause the magnetron orbits to increase in size and this leads to ion loss. This process can be reversed by a technique which is formally equivalent to the usual laser cooling [4,6, also Itano, Wayne M., and Wineland, D. J., to be published]; specifically, the magnetron energy is <u>increased</u> and the orbit size reduced by laser scattering by spatially tailoring the laser beam. The result is that infinite confinement times are in principle possible in the Penning trap.

The ground-state structure of Mg^+ ions has been measured by double-resonance schemes where changes in the laser fluorescence can be used to monitor ground-state rf and microwave transitions [9,10]. This has led to a measurement of the ground-state hyperfine constant (A = -596.254 376(54) MHz) and g_I/g_J (= 9.299 484(75) x 10^{-5}) in $^{25}Mg^+$. For this ion, the derivative of the ground-state $(M_I, M_J) = (-^3/_2, \frac{1}{2})$ to $(-\frac{1}{2}, \frac{1}{2})$ transition with respect to magnetic field B_0 goes to zero at $B_0 \cong 1.24$ T. The corresponding resonance ($\nu \cong 292$ MHz) was observed at this field with linewidths as small as 0.012 Hz ($Q \cong 2.4 \times 10^{10}$) by implementing the Ramsey interference method with two coherent rf pulses separated in time by 41.4 s (see figure 4). We expect that such narrow resonance lines (and even narrower lines) can be observed in other ions (e.g., Hg^+) with higher ground-state transition frequencies. This could then yield extremely high Q (> 10^{12}) in microwave transitions which would be valuable for frequency standard applications.

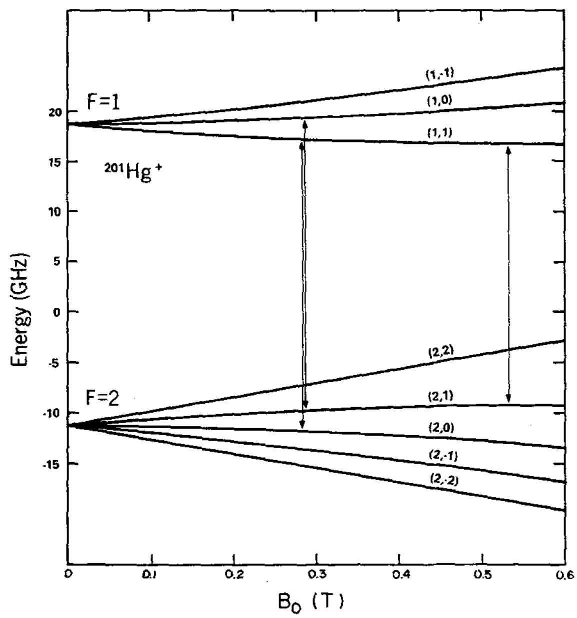


4. Rf resonance curve for the $(m_I, m_J) = (-^3/_2, \frac{1}{2})$ to $(-\frac{1}{2}, \frac{1}{2})$ ground-state hyperfine transition in $^{25}Mg^+$. Each circle represents the average of four measurements (total detection fluorescence integration time of 16 s). The oscillatory lineshape results from the use of the Ramsey method to drive the transition. Two coherent rf pulses of duration $\tau = 1.02$ s separated by T = 41.4 s were applied. The vertical arrow marks the central minimum, which corresponds to the resonance frequency.

In some double-resonance schemes, it is possible to scatter many photons for each microwave or optical "clock" photon absorbed. (In Ref. 10, a factor of about 10^6 was achieved.) This "quantum multiplication" should allow the signal-to-noise ratio in double-resonance detection schemes to be limited only by the statistical noise in the number of ions that have made the "clock" transition [9,11]. This will be extremely important for frequency standards based on ions where the number of ions is necessarily rather small.

Because of the above results, we have initiated work at NBS to realize microwave [11] and optical [11,12] frequency standards based on Hg^+ ions stored in a Penning trap.

The proposed microwave frequency standard is based on the $(F, M_F) = (1,1) \leftrightarrow (2,1)$ ground-state hyperfine transition in $^{201}Hg^+$, which is field-independent to first order at $B_0 \cong 0.534$ T, with frequency $\cong 25.9$ GHz (see figure 5). If B_0 can be controlled to slightly better than 0.1 ppm over the ion cloud, the fractional frequency shift can be kept below 10^{-15}. (At the "field-independent" point $\Delta\nu/\nu_0 = (\Delta B/B_0)^2/6$). The velocity in the magnetron motion will give a second-order Doppler shift; it should be controllable to 10^{-15} (see below). All other systematic shifts, such as those due to collisions, the trap electric fields, or thermal radiation appear to be less than 10^{-15} [11]. It should be possible to observe the transition with a Q of 2.6×10^{12} or better, by using optical pumping and detection techniques similar to those demonstrated with $^{25}Mg^+$. The accuracy of this standard could be better than 10^{-15}.



5. Ground-state hyperfine energy levels of $^{201}Hg^+$ vs. magnetic field. States are designated by the (F, M_F) representation. Three transitions are indicated at the fields where the transition frequencies are independent of magnetic field to first order.

The proposed optical frequency standard [11,12] is based on the two-photon-allowed $5d^{10}\,6s\ ^2S_{1/2} \rightarrow 5d^9\,6s^2\ ^2D_{5/2}$ Hg^+ transition, which has a natural Q of 7.4×10^{14}. The first-order Doppler effect can be elimated by driving the transition with counter-propagating 563.2 nm laser beams. Hyperfine-Zeeman components, whose magnetic field derivatives vanish at particular values of B_0, exist in $^{199}Hg^+$ and $^{201}Hg^+$. The two-photon transition can be detected with high efficiency by using the 194.2 nm fluorescence intensity as a probe of the ground-state population. Taking full advantage of the high Q transition would require a laser with linewidth less than 1 Hz, which does not exist at present. However, linewidths $\lesssim$ 100 Hz appear feasible and could be used for initial experiments. If the laser linewidth is less than the natural linewidth, then the ac Stark shift is about 2×10^{-15} near saturation. All other systematic shifts appear to be less than 10^{-15}. We note that the ac Stark shift can be made negligibly small by driving the single photon $^2S_{1/2} \rightarrow 2D_{5/2}$ quadrupole transition. In this case, it will be desirable to use a single ion that can be confined to approximately realize the Dicke criterion in order to suppress first-order Doppler effects.

The method currently being investigated for generating the required 194.2 nm radiation for laser cooling and optical detection is sum-frequency mixing in a KB5 crystal of the output of a 792 nm single-mode cw ring dye laser and the second harmonic, generated in an ADP crystal, of the output of a 514 nm stabilized, single-mode cw Ar^+ laser.

For the microwave frequency standard, it is especially desirable to use the largest possible number of ions in order to increase signal to noise. Unfortunately, as the number of ions is increased, the second-order Doppler shift due to magnetron rotation also increases, which will, therefore, limit the number of ions to about 10^5 or less for 10^{-15} accuracy [11]. (A similar problem exists for the rf trap due to the kinetic energy in the micromotion [13]). Therefore, studies will also be devoted to methods of controlling the density and shape of the ion clouds.

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