

# Laser Cooling and Double Resonance Spectroscopy of Stored Ions

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## 1. Introduction

The use of ion storage techniques for spectroscopy is motivated by the fact that ions can be confined by electric and magnetic fields for long periods of time without suffering the large perturbations which usually accompany other methods of confinement, such as those due to collisions with buffer gas molecules. Linewidths as small as a few Hz and Q's as high as  $10^{10}$  have been observed on ground-state hyperfine transitions of atomic ions stored in rf quadrupole traps [1-3]. The accuracy of these measurements has been limited largely by the second-order Doppler shift. The signal-to-noise ratios have been limited by the small number of ions that can be stored (about  $10^5$ - $10^6$ ) and by the difficulty of detecting transitions. Hyperfine and Zeeman transitions have been detected by charge exchange [1], fluorescence [2,3], photodetachment [4], and photodissociation [5].

In this paper we discuss recent work at the National Bureau of Standards (NBS) in this area. The second-order Doppler shift can be reduced by laser (resonant light pressure) cooling. This technique has been demonstrated in experiments on stored ions by our group at NBS [6-8] and also by a group at Heidelberg [9]. Other work at NBS has been directed toward the realization of high-efficiency laser-optical-pumping, double-resonance detection techniques [10]. These techniques have been used to make the first highprecision hyperfine structure measurements of  $^{25}\text{Mg}^+$  [11]. We anticipate that the laser cooling and double resonance techniques will find practical application in the development of frequency standards based on stored ions [12].

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## 2. Apparatus

A block diagram of the apparatus is shown in Fig.1. The  $\text{Mg}^+$  ions are confined in a Penning-style ion trap. They are irradiated by light resonant with the  $3s\ ^2S_{1/2} \rightarrow 3p\ ^2P_{3/2}$  280 nm transition and also by rf and microwave radiation resonant with various transitions between ground-state sublevels. Scattered 280 nm photons are collected by a mirror and counted by a photomultiplier tube, with a net detection efficiency of about  $10^{-5}$ .

A Penning trap confines ions by a combination of a uniform static magnetic field,  $\vec{B} = B_0 \hat{z}$ , and a quadrupolar electrostatic potential. The magnetic field causes the ions to move in circular "cyclotron" orbits in the x-y plane. The electric fields provide a harmonic restoring force for the axial (z) motion and cause the centers of the cyclotron orbits to move in the x-y plane in circular "magnetron" orbits around the trap symmetry axis. The angular frequencies corresponding to the axial, cyclotron, and magnetron motions are denoted by  $\omega_z$ ,  $\omega_c'$ , and  $\omega_m$  respectively. For typical operating conditions,  $\omega_z \cong 2\pi \cdot 200$  kHz,  $\omega_c' \cong 2\pi \cdot 800$  kHz, and  $\omega_m \cong 2\pi \cdot 25$  kHz. Typically,  $B_0 \cong 1$  T, and a potential  $V_0 \cong 7$  V is applied across the trap electrodes, which have inside dimensions of about 1 cm. The axial and cyclotron motions are thermal and stable. The magnetron motion is nonthermal and unstable, since an increase in the size of the magnetron orbit leads to a decrease in the total (kinetic plus potential) energy. However, storage times of about 1 day have been observed, even without laser cooling. The background pressure is typically  $\leq 10^{-8}$  Pa.

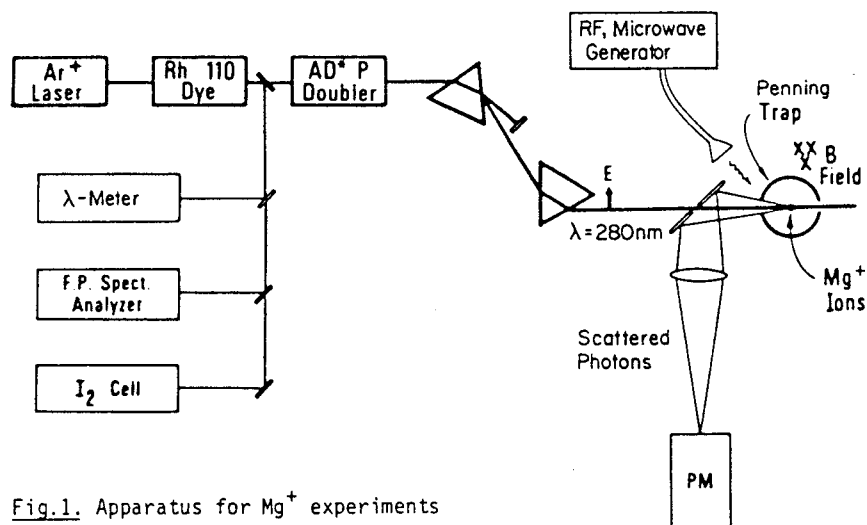


Fig.1. Apparatus for  $\text{Mg}^+$  experiments

The 280 nm radiation was produced by frequency doubling the output of a single-mode cw Rhodamine 110 dye laser in a 90° phase-matched AD\*P crystal. Between 5 and 30  $\mu\text{W}$  of UV radiation was generated in a bandwidth of about 1 MHz. The dye laser could be long-term stabilized to discrete frequencies to less than 1 MHz by locking it to saturated absorption features in  $\text{I}_2$ . Fine tuning could be done by varying the trap magnetic field to Zeeman shift the  $\text{Mg}^+$  levels. For efficient laser cooling, the UV linewidth must be less than the Doppler width of the transition (3 GHz at 300K), but need not be much less than the natural linewidth (43 MHz).

### 3. Laser Cooling in a Penning Trap

The basic principle of laser cooling is that light pressure can be used to damp the velocity of an atom or ion if the light frequency is tuned slightly below that of a strong optical transition. The process is easily described for the case of free or harmonically bound atoms [9,13-16]. Atoms whose velocities are directed toward the light source see a frequency Doppler shifted closer to resonance; if their velocities are directed away from the source, the frequency shift is away from resonance. Therefore, the atoms tend to absorb photons when their velocity is directed toward the source. This cools them, since the momentum of the absorbed photons reduces the atomic momentum. (The photons are re-emitted in random directions.) Three-dimensional cooling can be obtained with six laser beams directed along the  $\pm x$ ,  $\pm y$ , and  $\pm z$  directions. A single beam suffices to cool all oscillational modes of an atom bound in a three-dimensional harmonic potential, if the modes are nondegenerate and if the beam is not directed along one of the principal axes.

Cooling of the axial and cyclotron modes of an ion in a Penning trap takes place by the process just described for free or harmonically bound atoms, if the light is tuned below resonance [17]. Cooling of the magnetron mode can be accomplished by focusing the light beam so that it is more intense on the side of the trap axis on which the magnetron motion recedes from the light source [6-8,17]. By "cooling" of a mode, we mean the reduction of the average kinetic energy in that mode, regardless of what happens to the potential energy. In order to efficiently cool all modes, the angle between the beam and the z axis should be about 45°; the optimum angle depends on the angular distribution of the scattered photons [17]. We have derived rate equations for the laser cooling of an ion in a Penning trap by

a single nonuniform light beam [17]. Solving these equations, we find that it should be possible to cool an ion to a "temperature"  $T$  given by  $k_B T \approx \hbar \gamma / 2$ , where  $k_B$  is Boltzmann's constant and  $\gamma$  is the natural linewidth of the upper state. The "temperature" is defined in terms of the mean kinetic energy and may be different for the different modes. For  $Mg^+$ , this minimum temperature is about 1 mK.

#### 4. Optical Pumping and Double Resonance

The extremely long ground-state relaxation times possible for stored ions make it possible to observe very narrow transition linewidths and to observe very weak optical pumping processes. An ion must scatter about  $10^4$  optical photons in order to be cooled significantly below room temperature. Therefore, even weak depopulation pumping processes would have an adverse effect on the cooling. Cooling, in turn, influences the optical pumping by reducing the Doppler broadening until it is less than the natural linewidth. This makes it possible for a monochromatic laser to interact strongly with all of the ions and not just with a small velocity class.

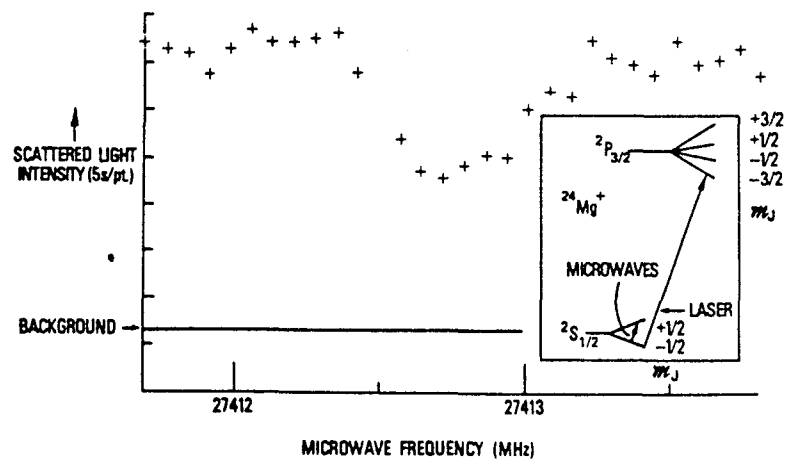


Fig.2. Optical pumping and double resonance of  $^{24}Mg^+$

Generally, the  $(^2S_{1/2}, M_J = -1/2) \rightarrow (^2P_{3/2}, M_J = -3/2)$  Zeeman component was used for laser cooling of  $^{24}Mg^+$ , which has  $I=0$  (see Fig.2). This transition does not lead to depopulation pumping of the ground state, since the  $M_J = -3/2$  excited state sublevel must decay to the  $M_J = -1/2$  ground state

sublevel. When this transition is driven near resonance with light polarized perpendicular to the magnetic field, the  $(^2S_{1/2}, M_J = -1/2) + (^2P_{3/2}, M_J = +1/2)$  and  $(^2S_{1/2}, M_J = +1/2) + (^2P_{3/2}, M_J = -1/2)$  transitions are driven weakly in their Lorentzian wings (the Zeeman splitting is much greater than the Doppler broadening). In the steady state, which is achieved in  $\leq 1$  s, 16/17  $\approx$  94% of the population is in the  $M_J = -1/2$  ground-state sublevel [10].

The ground-state Zeeman transition  $(^2S_{1/2}, M_J = -1/2 \rightarrow +1/2)$  can be detected by a decrease in the fluorescence intensity. This "flop-out" detection method can be very efficient, since a transition due to a single microwave photon interrupts the flow of scattered optical photons until the ion is pumped back to the  $M_J = -1/2$  sublevel by weak, off-resonance scattering. Since the decrease in the number of scattered photons per ion can be very large, it is possible to make up for poor light collection and detector quantum efficiency, so that the transition can be detected with nearly unit efficiency. Similar detection methods have been proposed previously [18].

The  $(^2S_{1/2}, M_I = -5/2, M_J = -1/2) \rightarrow (^2P_{3/2}, M_I = -5/2, M_J = -3/2)$  hyperfine-Zeeman component was generally used for laser cooling of  $^{25}\text{Mg}^+$  ( $I = 5/2$ ). In the steady state, about 16/17 of the population is pumped into the  $(M_I = -5/2, M_J = -1/2)$  ground-state sublevel. Pumping into the  $M_J = -1/2$  manifold takes place by the same mechanism as in  $^{24}\text{Mg}^+$ . Pumping into the  $M_I = -5/2$  sublevels takes place because of hyperfine coupling in the excited state [10]. Any transition which decreases the population in the  $(M_I = -5/2, M_J = -1/2)$  ground-state sublevel can be detected by a decrease in the fluorescence intensity.

## 5. Results

In the Penning trap, low temperatures become easier to achieve as the ion density is reduced. This is because the radial electric field due to space charge increases the magnetron velocity. This problem does not exist for a single, isolated ion, and the lowest temperatures were observed for this case [8].

Figure 3 shows the fluorescence from a small number of  $^{24}\text{Mg}^+$  ions as a function of time. After the ions were cooled and localized at trap center, an oven containing  $^{25}\text{Mg}$  (98% isotopic purity) was heated in order to induce the resonant charge exchange reaction  $(^{24}\text{Mg}^+ + ^{25}\text{Mg} \rightarrow ^{24}\text{Mg} + ^{25}\text{Mg}^+)$ . The

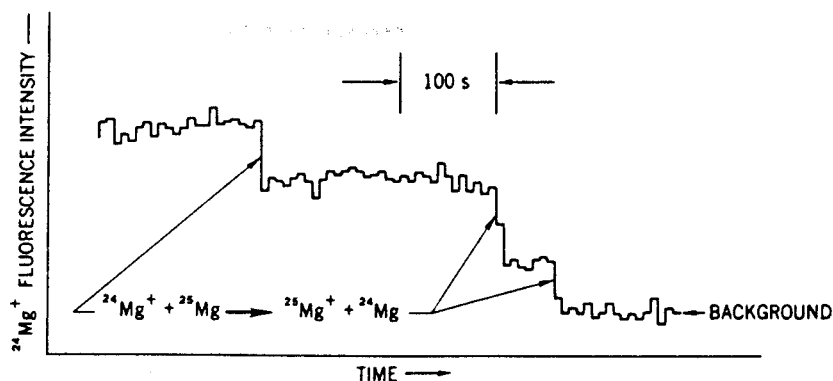


Fig.3. Fluorescence from 3, 2, and 1  $^{24}\text{Mg}^+$  ions

resulting  $^{25}\text{Mg}^+$  ions were ejected from the trap by resonant cyclotronmagneton rf excitation. The three large step decreases in fluorescence are due to loss of the  $^{24}\text{Mg}^+$  ions, one at a time, and the last plateau above background is the fluorescence from a single ion. The signal was about 50 photons/s per ion.

The "temperature" of a single  $^{24}\text{Mg}^+$  ion was determined from the Doppler width by optical-optical double resonance (see Fig.4). One laser was tuned slightly below the  $(^2S_{1/2}, M_J = -1/2) \rightarrow (^2P_{3/2}, M_J = -3/2)$  transition, to provide cooling and fluorescence detection. A low power laser was swept continuously across the  $(^2S_{1/2}, M_J = -1/2) \rightarrow (^2P_{3/2}, M_J = -1/2)$  transition, which was detected by a decrease in the fluorescence. The resulting lineshape reflects both the natural and Doppler broadenings. The data points represent 10s integrations; the connecting lines are only for clarity. Simulated curves are shown for temperatures of 0 K and 1000 mK. We estimate that  $T = 50 \pm 30$  mK. Since the light was incident at  $82^\circ$  with respect to the z axis, this is essentially a measurement of the cyclotron-magneton (x-y) temperature. The axial (z) temperature was estimated to be about 600 mK by probing the axial excursions with a focused laser beam. According to our calculations, it should be possible to obtain a cyclotron-magneton temperature of about 1 mK and an axial temperature of about 11 mK for these conditions [17]. At present, the discrepancy is not understood, but may be due to the presence of impurity ions in the trap.

Ground-state Zeeman and hyperfine transitions were detected by the optical-pumping, double-resonance methods outlined previously. The only transition in  $^{24}\text{Mg}^+$  is the electronic spin flip transition (see Fig.2). Several

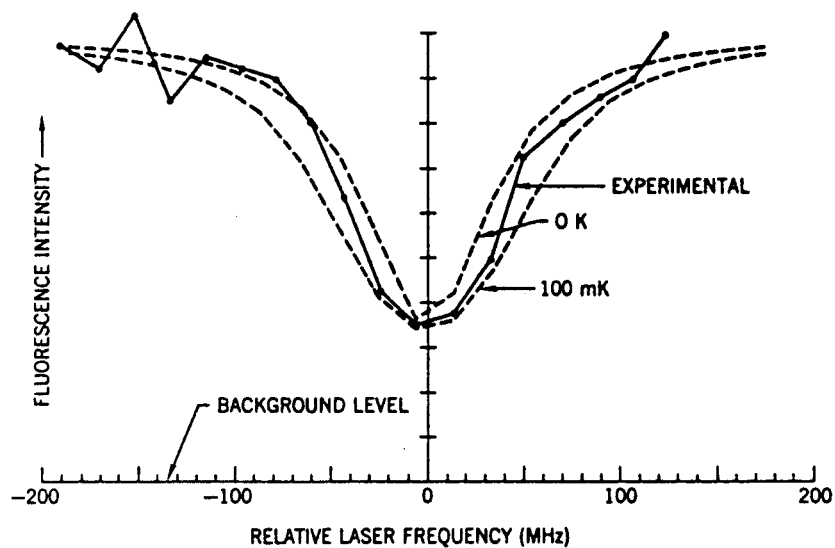


Fig.4. Optical-optical double resonance of a single  $^{24}\text{Mg}^+$  ion

transitions in  $^{25}\text{Mg}^+$ , corresponding to both nuclear and electronic spin flip transitions, were observed. Most transitions were broadened by the instability of  $B_0$ , which was, at best, about 1 ppm in a few seconds. The magnetic field derivative of the  $(M_I, M_J) = (-3/2, +1/2) \leftrightarrow (-1/2, +1/2)$  transition goes to zero at  $B_0 \approx 1.2398$  T. Near this field, the transition was observed with linewidths as narrow as 0.012 Hz (see Fig.5). The oscil-

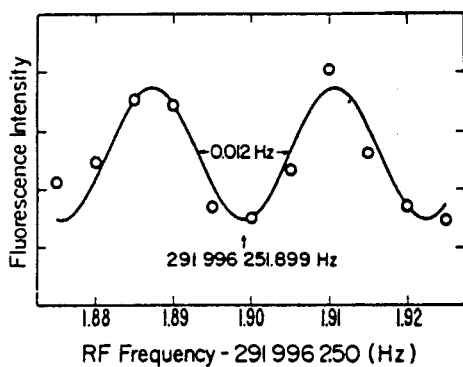


Fig.5.  $^{25}\text{Mg}^+$   $(M_I, M_J) = (-3/2, +1/2) \leftrightarrow (-1/2, +1/2)$  ground-state hyperfine resonance

latory lineshape results from the use of the Ramsey interference method [19], which was implemented by driving the transition with two coherent rf pulses of 1.02 s duration separated by 41.4 s. The center of the resonance can be determined with an uncertainty of about  $10^{-11}$ . Several different transition frequencies were measured in order to obtain separately the hyperfine constant ( $A = -596.254\ 376\ (54)\ \text{MHz}$ ) and the nuclear-to-electronic g factor ratio ( $g_I/g_J = 9.299\ 484\ (75) \times 10^{-5}$ ) [11].

## 6. Future Possibilities

Work is being initiated at NBS on the development of a microwave frequency and time standard based on a hyperfine transition in  $^{201}\text{Hg}^+$  and of an optical frequency standard based on a two-photon transition to a metastable state in  $^{199}\text{Hg}^+$  or  $^{201}\text{Hg}^+$ . Both proposed standards are based on a cloud of  $\text{Hg}^+$  ions stored in a Penning trap and use the  $6s\ ^2S_{1/2} \rightarrow 6p\ ^2P_{1/2}$  194 nm transition for laser cooling.

The proposed microwave frequency standard is based on the  $(F, M_F) = (1,1) \leftrightarrow (2,1)$  transition, which is field-independent to first order at  $B_0 \approx 0.534\ \text{T}$ , with frequency  $\approx 25.9\ \text{GHz}$ . 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  $10^{-15}$ . All other systematic shifts, such as those due to the second-order Doppler effect, collisions, the trap electric fields, or thermal radiation, appear to be less than  $10^{-15}$ . It should be possible to observe the transition with a Q of  $2.6 \times 10^{12}$  or better, by using optical pumping and detection techniques similar to those demonstrated with  $^{25}\text{Mg}^+$ . The accuracy of this standard could be as good as  $10^{-15}$ , which is about 100 times better than that of the best frequency standards now available.

The proposed optical frequency standard is based on the two-photon-allowed  $5d^{10}\ 6s\ ^2S_{1/2} \rightarrow 5d^9\ 6s^2\ ^2D_{5/2}$   $\text{Hg}^+$  transition, which has a natural Q of  $7.4 \times 10^{14}$  [12]. The first-order Doppler effect can be eliminated 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  $^{199}\text{Hg}^+$  and  $^{201}\text{Hg}^+$ . 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  $\leq 100\ \text{Hz}$  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  $2 \times 10^{-15}$  near saturation. All other systematic shifts appear to be less than  $10^{-15}$ .

The method currently being investigated for generating the required 194.2 nm radiation is sum-frequency mixing in a K85 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<sup>+</sup> laser. The method has been demonstrated previously with pulsed lasers [21]. Further details of these proposed Hg<sup>+</sup> frequency standards are published elsewhere [12].

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