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High-Resolution Optical Spectra of Laser Cooled Ions

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Abstract. We obtain essentially Doppler free spectra of the naturally occuring isotopes of Mg^+, which are bound in a Penning trap, by using a frequency stabilized laser to continuously cool the ions, while the scatter rate from a second, frequency swept laser is monitored. We show that the magnetron motion as well as the cyclotron and axial motion can be minimized. Line position measurements yielding resonance transition energy, isotope and hyperfine shifts are reported.

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Radiation-pressure cooling to substantially reduce Doppler broadening, shifts, and residual velocity effects to all orders [1, 2] represents an important step in the study of ultra-narrow, lifetime limited transitions. The method reported here investigates the full Doppler width of the quantum absorber studied; but, because the absorbers are substantially cooled by radiation pressure, the line width can be dominated by the natural resonance width. Furthermore, in contrast to conventional sub-Doppler techniques, this method directly reduces all velocity effects.

Recently, cooling of a sample of bound resonant absorbers by radiation pressure has been demonstrated [1, 2]. In [1] Mg^+ ions were confined to a room temperature Penning trap and were irradiated near the $^2S_{1/2}-{}^2P_{3/2}$ resonance transition (280 nm) with the output of a frequency doubled cw dye laser. In (such) a two-level system, the process of absorption followed by re-emission returns the atom to the original internal state; but, in general, a different kinetic energy state. After many such scattering events, the collective momentum of the absorbed photons can be made to cancel the thermal momentum of the absorber, whereas the recoil momenta from the randomly re-emitted photons cancel to first order [3]. In this paper, we describe some of the interesting features of electromagnetically trapped ions which are cooled by this technique and also report the first high-resolution spectra obtained on a laser cooled species. Preliminary results of this work are reported in [4].

Briefly, the eigenmotions of ions in a Penning trap [5] are a simple harmonic oscillation in the electrostatic well along the trap axis, a cyclotron orbit in the plane perpendicular to the trap axis, and a magnetron drift orbit also in that plane. The axial vibration and the cyclotron orbital motions are thermal and coupled through the long range coulomb collisions between ions in the trap, whereas the magnetron motion is nonthermal and not strongly coupled to the other motions [1, 5]. In fact, the magnetron motion is in an unstable equilibrium in the trap, but the confinement time of unperturbed ions is observed to be days.

Experiment

In our present experiments, the laser beam (diameter $\approx 100\,\mu m$) crosses the trap normal to the horizontal trap axis (**B** field axis) and is also linearly polarized perpendicular to the trap axis. With this configuration, the laser directly interacts with both the cyclotron and magnetron motion. These motions can be represented as [6]

$$\mathbf{r} = x + iy = \mathbf{r}_c + \mathbf{r}_m = \mathbf{r}_{c_0} e^{-i\omega_c t} + \mathbf{r}_{m_0} e^{-i\omega_m t}, \quad (1)$$

where $\mathbf{r}_c$ and $\mathbf{r}_m$ are the cyclotron and magnetron radius vectors, respectively, and ω_c, ω_m the associated angular frequencies. If the ions undergo an incremental change in velocity, $\Delta\mathbf{v}$, per scattering event, we find

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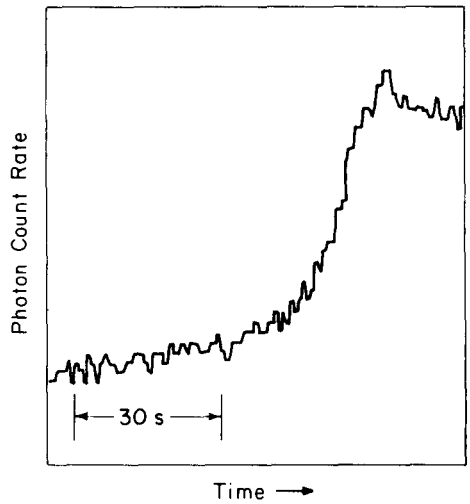

Fig. 1. Scatter rate vs. time after the laser has been blocked for 5 min. On unblocking the laser, the ion cloud which has begun to spread and heat, yields a low scatter rate at first, but returns to its previous value in approximately 1 min

that there is a corresponding incremental change in the radius vector given by

$$\Delta \mathbf{r}_c = \frac{\mathrm{i}}{\omega_c - \omega_m}\Delta \mathbf{v} \quad \text{and} \quad \Delta \mathbf{r}_m = \frac{-\mathrm{i}}{\omega_c - \omega_m}\Delta \mathbf{v}. \tag{2}$$

By irradiating the side of the ion cloud which recedes from the laser with photons of frequency $\omega_L < \omega_0$, both $\mathbf{r}_c$ and $\mathbf{r}_m$ can be made smaller. This effect is demonstrated below.

A cloud of $\lesssim 10^2$ Mg^+ ions is loaded into the trap. The static magnetic field is 0.976 tesla and the electric potential is ≈ 7 V. The laser beam is initially positioned about 600 μm below the trap center; that is, where the magnetron motion carries the ions in the direction away from the laser. The laser frequency is swept up to and held at approximately the half-intensity point of the resonance transition which is Doppler shifted by the magnetron motion. Photons which are backscattered into an $f/4$ cone are collected and counted. The overall detection efficiency is $\approx 3 \times 10^{-5}$. The scatter rate is observed to decrease in time indicating that the ions move closer to trap center. This process is repeated by moving the laser beam toward trap center in 100 μm steps. When the beam reaches a position of ≈ 100 μm below trap center, the scatter rate remains constant, which indicates a stable cloud configuration (dia: $\leqq 200$ μm) has been obtained.

Additionally, if the laser radiation is blocked, the ions begin to heat and spatially spread; however, the low entropy ion cloud can be reconstituted at trap center by simply unblocking the laser. This is indicated in Fig. 1, where a plot of scatter rate vs. time is shown, after the laser had been blocked for 5 min. The signal rises and then stabilizes at the previous scatter rate, indicating that the ions have returned to the preblocked configuration at trap center. If the laser beam is positioned above trap center, the cloud is "blown-out" to a large radius, but still remains confined to the trap. Once again, we can reform a small, cold ion cloud by spatially stepping the laser below trap center and repeating the above procedure.

If the ions are localized and cooled and the laser beam maintained about 50 μm below trap center, an estimate of ion temperature can be obtained by rapidly scanning the laser through the optical transition and recording the scatter rate as a function of laser frequency. The broken curve (circles) in Fig. 2 shows a typical plot of scatter rate vs. laser frequency. As the frequency is swept into resonance, the ions are maintained cold and a nearly Lorentzian line shape is produced. However, even at our low laser power (≈ 8 μW) and rapid sweep (700 MHz/s), the scattering process at laser frequencies blue of resonance significantly heats the ions which results in the observed line asymmetry. The half-width at half-maximum (HWHM) of the cooled side of the line is ≈ 40 MHz corresponding to a temperature of ≈ 0.5 K. It is to be noted however, that the laser beam, which has non-zero extent, interacts with a spread of magnetron orbits and velocities. The magnitude of the resulting Doppler broadening is a function of the cloud size and charge density, but it is expected to result in the major portion of the observed 40 MHz half width. This indicates that the cyclotron and axial motions may be substantially colder than 0.5 K. This residual Doppler effect due to magnetron motion could be avoided by interrogating the ions with the laser directed along the trap axis.

To help overcome the effects of heating as the laser is scanned through the line, a second, frequency-doubled, ring dye laser is introduced into the trap. The ring laser is frequency-stabilized by locking to a saturated absorption hyperfine feature in iodine whose doubled frequency is nearly coincidental with but to the low frequency side of $^{24}Mg^+$. The stabilized laser serves to keep the sample cold as the first laser is scanned at reduced power. For reasons not fully understood at this time, the half line-widths obtained in this fashion are a factor of two broader than those obtained with one laser if all other parameters are unchanged. If the static magnetic field is increased from the previous ≈ 1.0 to ≈ 1.4 Tesla, the two laser scheme also produces narrow lines corresponding to temperatures ≈ 0.5 K. In Fig. 2, we show the line profile of the two laser generated signal (solid curve).

Equally important, the second fixed frequency laser allows us to do high-resolution spectroscopy of like or

dissimilar ionic species. The magnesium used in these experiments had a natural isotopic mixture containing approximately 80% $^{24}Mg^+$ and 10% each $^{25,26}Mg^+$. The $^{25,26}Mg^+$ lines are positioned in the high frequency wings of $^{24}Mg^+$. Thus, scanning these lines heats $^{24}Mg^+$, which in turn heats $^{25}Mg^+$ and $^{26}Mg^+$ by Coulomb collisions. This is in addition to the heating caused by scanning to the high frequency side of $^{25}Mg^+$ and $^{26}Mg^+$. These heating effects are largely eliminated by the frequency stabilized laser. Whereas $^{24}Mg^+$ is cooled directly by the fixed frequency laser, $^{25}Mg^+$ and $^{26}Mg^+$ are cooled and maintained cold by Coulomb interactions with the cooled $^{24}Mg^+$. This confirms an earlier conjecture [1] that one ion (in this case, $^{24}Mg^+$) can be used to cool other ions of spectroscopic interest.

Monitoring the scatter rate of the low power, frequency swept laser results in the spectrum shown in Fig. 3. $^{24}Mg^+$ and $^{26}Mg^+$ have zero nuclear spin and yield single lines, while $^{25}Mg^+$ has nuclear spin $I=5/2$ and would ordinarily exhibit six lines. However, the nuclear hyperfine components are optically pumped such that nearly all of the ^{25}Mg ions are in the $M_I=-5/2$ state. Hence, $^{25}Mg^+$ is also observed as a single line. The relative positions of the observed lines depend upon the isotope shifts and (in the case of $^{25}Mg^+$) the hyperfine coupling constants. The isotope shift of the $^2S_{1/2}-{}^2P_{3/2}$ line in MgII is measured to be

$$\Delta\nu(^{24}Mg-{}^{26}Mg)=3.050\pm0.1\,\text{GHz}. \quad (3)$$

Combining this result with the mass shift formula, which is a good approximation for low Z elements such as Mg^+, we calculate that the isotope shift for $^{25}Mg^+$ is

$$\Delta\nu(^{26}Mg-{}^{25}Mg)=1.461\pm0.1\,\text{GHz}. \quad (4)$$

The frequency difference between $^{25}Mg(m_I=-5/2, m_J=-3/2\leftrightarrow m_J=-5/2, m_J=-1/2)$ and $^{26}Mg(m_J=-3/2\leftrightarrow m_J=-1/2)$ optical transitions is

$$\Delta f=772.1\pm60\,\text{MHz}. \quad (5)$$

From this result, we obtain the hyperfine coupling constant, a_{3S}, for the $^2S_{1/2}$ state of $^{25}Mg^+$

$$a_{3S}=-607.8\pm50\,\text{MHz}. \quad (6)$$

This is in fair agreement with earlier works of Crawford [7] and Weber [8]. For this calculation, we first estimated the $^2P_{3/2}$ coupling constant to be $a_{3P}=-19.1\pm5$ MHz using the Goudsmit formula and the known fine structure separation [9]. The uncertainty in this value of ±5 MHz contributes an uncertainty ±15 MHz in the value calculated for a_{3S}. We expect to substantially improve the accuracy in the a value for both states with a double resonance technique presently being pursued.

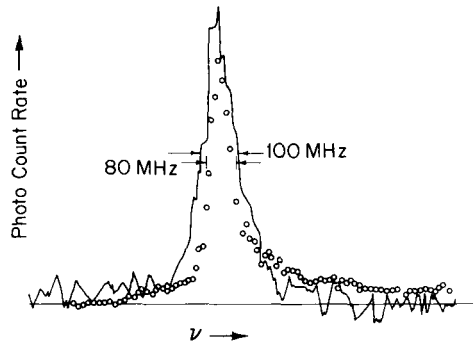

Fig. 2. Scatter rate vs. laser frequency. The curve shown in circles is with one laser only. The solid curve uses one laser to produce cooling while a second scans the line profile

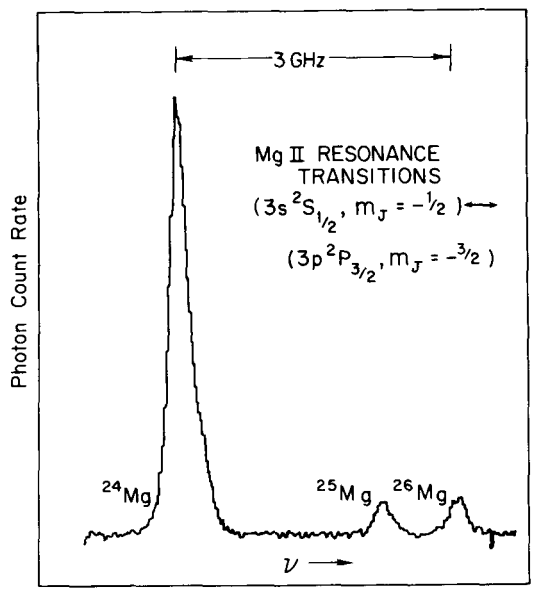

Fig. 3. Spectra of one Zeeman component of laser cooled $^{24,25,26}Mg^+$. The room temperature Doppler width of these lines is about 3 GHz. Only the $^{24}Mg^+$ is directly laser cooled. The $^{25}Mg^+$ hyperfine structure has been optically pumped resulting in the observation of a $(m_J=-1/2, m_I=-5/2)\leftrightarrow(m_J=-3/2,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

By measuring the frequency difference between the center of the Doppler broadened I_2 line and the particular hyperfine component to which the stabilized ring laser is locked, we are able to reference the $^{24}Mg^+$ $3^2S_{1/2}\leftrightarrow3^2P_{3/2}$ resonance transition to the known iodine spectra [10]. Subtracting out the linear Zeeman shift, we determine the $3^2S_{1/2}\leftrightarrow3^2P_{3/2}$ resonance transition to be $35{,}760.834\pm0.004\,\text{cm}^{-1}$. This compares favorably to the accepted value of $35{,}760.88\,\text{cm}^{-1}$ [11].

Conclusion

In conclusion, we have presented high-resolution spectroscopy of laser cooled Mg^+ ions electromagnetically confined to a Penning trap. We have demonstrated a

general two laser scheme which provides both radiation pressure cooling and high-resolution spectroscopy on the cooled species. Additionally, we have demonstrated sympathetic cooling of a second species by long range Coulomb cooling (i.e. $^{25}Mg^+$ and $^{26}Mg^+$ by $^{24}Mg^+$). We have minimized the motions of the trapped ions yielding a temperature <0.5 K; but, most likely, substantially colder for the axial and cyclotron motions. We make the observation that the present experiments are limited in accuracy and resolution by the broadening and shift of the optical resonances due to residual magnetron motion. This limitation can be removed by probing the cooled ions with a laser directed along the trap axis.

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References

1. D.J.Wineland, R.E.Drullinger, F.L.Walls: Phys. Rev. Lett. **40**, 1639–1642 (1978)
2. W.Neuhauser, M.Hohenstatt, P.Toschek, H.Dehmelt: Phys. Rev. Lett. **41**, 233–236 (1978)
3. D.J.Wineland, W.M.Itano: Phys. Rev. A **20**, 1521–1540 (1979)
4. R.E.Drullinger, D.J.Wineland: In *Laser Spectroscopy IV*, ed. by H.Walther and K.W.Rothe, Springer Series Opt. Sci. **21** (Springer, Berlin, Heidelberg, New York 1979)
5. H.G.Dehmelt, F.L.Walls: Phys. Rev. Lett. **21**, 127–130 (1968); H.Dehmelt: In *Advances in Atomic and Molecular Physics*, ed. by D.R.Bates and I.Esterman (Academic Press, New York 1967, 1969), Vols. **3**, 53–72 and **5**, 109–154; D.J.Wineland, H.G.Dehmelt: J. Appl. Phys. **46**, 919–930 (1975)
6. J.Byrne, P.S.Farago: Proc. Phys. Soc. (London **86**, 801–815 (1965)
7. M.F.Crawford, F.M.Kelly, A.L.Schawlow, W.M.Gray: Phys. Rev. **76**, 1527–1528 (1949)
8. E.Weber: University of Heidelberg, private communication
9. Average of values obtained from Eqs. (III.84) and (III.86) of N.F.Ramsey: *Molecular Beams* (Oxford University Press 1956) p. 75
10. S.Gerstenkorn, P.Luc: Rev. Phys. Appl. **14**, 791–794 (1979)
11. P.Risberg: Ark. Fys. **9**, 483 (1955)