Two-Photon Optical Spectroscopy of Trapped Hg II

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Traditionally, precision frequency measurements and lifetime measurements have provided numerous and stringent tests of physical principles. Two of the important merits of trapped ions for precision frequency spectroscopy and lifetime measurements are long confinement times and a gentle, highly non-perturbative environment. Present experiments give the first glimpse of the deep reservoir of precision spectroscopic investigations possible with stored ions. Examples include the g-2 experiment that yields the highest precision yet obtained in fundamental tests of QED [1],and the measurement of the electron to proton mass ratio to an accuracy of 0.04 ppm [2]. In other experiments, trapped ions are radiatively cooled to temperatures below 0.1K [3-5]. This directly reduces Doppler effects to all orders and thereby enhances resolution and precision in the method of trapped ion spectroscopy. Furthermore, because the radiation of the laser used for cooling is usually tuned to near resonance with a highly allowed transition in order to facilitate efficient cooling, many photons per second per ion are scattered; the high rate of scattered photons provides a powerful scheme not only to detect ions but also to detect atomic transitions to metastable states by the method of double resonance [6,7]. This method can give unit detection efficiency or, equivalently, a signal-to-noise (S/N) ratio limited only by the quantum-statistical fluctuation in the number of ions that make the transition and not by detection solid angle, detector quantum efficiency, etc. [8]. This is the maximum S/N ratio possible. We further note that this highly efficient detector of an atomic transition works even if the cycling/cooling transition and the transition to the metastable state share no common level [9]. With the continual and rapid maturation of laser cooled, stored ion spectroscopy, there has come a flurry of activity. Examples include a 300 fold improvement in the limits for spatial anisotropy by using laser cooled, electromagnetically trapped $^{9}Be^{+}$ ions [10], a laser cooled atomic frequency standard with an accuracy comparable to the best cesium beam atomic frequency standards [11], a demonstration of strong coupling in a small cloud of laser compressed and laser cooled ions [12], and a precise measurement of the electron-to-proton mass ratio [13]. In this paper we describe some of the first results obtained in our study of the Doppler-free, two-photon $5d^{10}\ 6s\ ^{2}S_{1/2} - 5d^{9}\ 6s^{2}\ ^{2}D_{5/2}$ transition in singly ionized mercury stored in a miniature rf trap [14].

A cross-sectional view of the trap electrodes is shown in Fig. 1. In contrast to the customary hyperbolic surfaces which produce a harmonic potential nearly everywhere in the trap volume, our trap was

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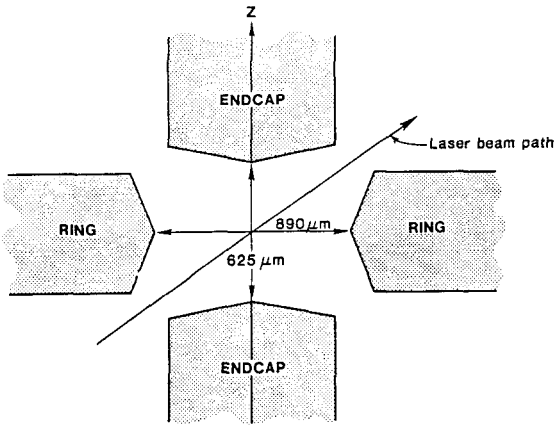

Fig. 1. Schematic showing cross-section view of trap electrodes. The electrodes are figures of revolution about the z-axis and are made from molybdenum.

machined with simple conical cuts. However, the trap dimensions and angles were chosen to make the fourth and sixth order anharmonic terms in the trapping potential vanish [15]. For practical purposes, then, the potential was harmonic in the central volume of the trap. Trapping was obtained by driving the electrodes with an oscillating electric field with a voltage amplitude $V_o \leq 1$ kV and a frequency near 21 MHz. The trap was housed in a vacuum vessel which was baked and pumped out to a partial pressure of background gas $\leq 10^{-7}$ Pa. A small amount of ^{198}Hg was admitted into the vacuum vessel and crossed by a low current electron beam near trap center in order to produce the trapped mercury ions. After loading 50-200 ions, the mercury vapor was frozen out in a liquid nitrogen cold trap and the system was back-filled with about 10^{-2} Pa of dry helium. The helium buffer gas was used to collisionally cool the mercury ions to near room temperature.

The $5d^{10}\ 6s\ ^2S_{1/2} - 5d^9\ 6s^2\ ^2D_{5/2}$ transition, driven by two photons with a wavelength near 563nm, was detected by the very sensitive optical double resonance scheme noted above. About 5 μW of narrowband, cw, sum-frequency-generated radiation near 194 nm [16] was focused through the trap between the ring electrodes and the endcaps. The beam waist, w_o, was approximately 25 μm and located near trap center. The frequency of the 194 nm radiation was tuned into coincidence with the $6s\ ^2S_{1/2} - 6p\ ^2P_{1/2}$ first resonance transition. The photons scattered by the ions into a small solid angle perpendicular to the 194 nm beam were detected and recorded. Rejection of stray photons scattered elsewhere in the system was achieved by means of a single spatial filter placed in the collection channel at the image of the ion cloud. The overall detection efficiency including solid angle, collection optics, filter and photomultiplier sensitivity was about 10^{-4}. The signal level was typically 2-10 x 10^3 counts/s and the signal to background level was better than 10/1. When the ions were driven out of the $^2S_{1/2}$ ground state into the metastable $^2D_{5/2}$ state, there was a decrease in the 194 nm fluorescence corresponding to the number of ions in the D state.

About 100 mW of the output from a cw ring dye laser oscillating near 563 nm was coupled through a single mode fiber and mode matched into a near concentric cavity placed around the rf trap. The relative position of the cavity and the trap was adjusted so that the cavity beam waist ($w_o \cong 30\mu m$) was located near the center of the cloud of trapped ions. The axes of the 563 nm beam, the 194 nm beam and the collection optics were mutually perpendicular. The cavity enhanced the one way power of the 563 nm radiation by about a factor of fifty, to give nearly 5 W of circulating power. Additionally, the high finesse of the cavity better ensured nearly equal intensity counter-propagating beams, which are necessary for high-

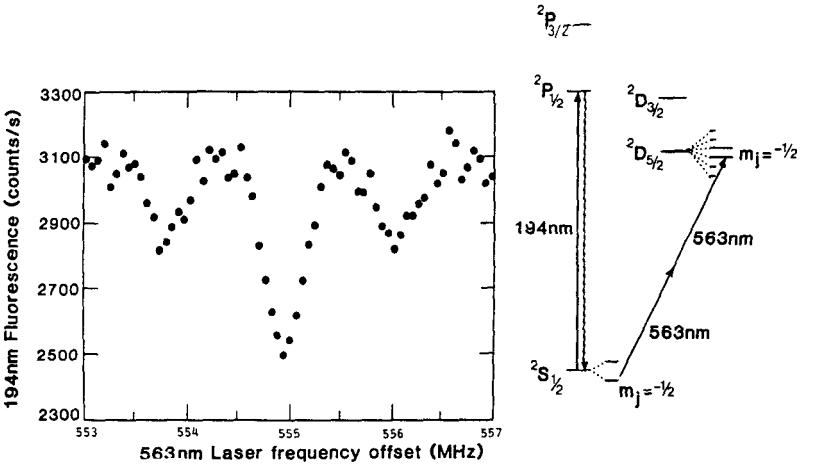

Fig. 2. Two-photon $^2S_{1/2}$-$^2D_{5/2}$ transition in $^{198}Hg^+$. AM sidebands caused by the harmonic secular motion of the ions are visible in this scan. The frequency scan is 4 MHz at the fundamental laser frequency ($\lambda \cong 563$ nm). The depth of the central component is about 25% of full scale. The integration time is 2 s/point. In the inset is a simplified energy-level diagram of $^{198}Hg^+$, depicting the levels of interest.

resolution Doppler-free two-photon spectroscopy. The linewidth of the ring dye laser in these preliminary experiments was of the order of 300 kHz. The frequency of the laser was offset locked and precisely scanned with respect to a second stabilized ring dye laser that was locked to a hyperfine component in the Doppler-free saturated absorption spectrum of $^{127}I_2$.

A 4 MHz scan of the frequency of the dye laser through the two-photon resonance is shown in Fig. 2. Also included in Fig. 2 is a simplified energy-level diagram showing the pertinent optical levels in Hg^+. A small magnetic field of approximately 11.6×10^{-4} T(11.6 G) which differentially Zeeman splits the ground and excited states, was applied by means of a pair of Helmholtz coils placed around the trap. The electric field vector of the linearly polarized 563 nm laser is oriented parallel to this field. The selection rule for the two-photon transition for this polarization is $\Delta m_J = 0$, and, thus, only two Zeeman components are expected, separated by approximately 13 MHz (approximately 6.5 MHz at the dye laser frequency). In Fig. 2, we scanned over only one of these components ($m_J = -1/2 \Leftrightarrow m_J = -1/2$) but saw sideband structure. This structure is due to amplitude modulation (AM) of the 563 nm laser intensity due to the harmonic secular motion of the ions in the rf trap,which carries the ions back and forth through the laser beam. Recall that the 563 nm waist is about 30 μm whereas the ion cloud volume is characterized by a linear extent of about 60μm. We note that any frequency modulation (FM) of the laser caused by the motion of the ions [17] is strongly suppressed if the cavity is well aligned,so that the k-vectors of the counter running light beams are anti-parallel. By changing the well depth of the trap, the harmonic frequency of the secular motion was changed,thereby shifting the sideband components on the two-photon signal. To our knowledge, this is the first observation of secular motion sidebands at optical frequencies. The depth of the central feature in Fig. 2 is nearly 25% of full scale,implying that we have nearly saturated the strongly forbidden two-photon transition. For the data of Fig. 2, the 194 nm laser irradiated the ions continuously. The linewidth of the two-photon resonance is about 420 kHz, and is determined in nearly equal parts by the 563 nm laser linewidth of about

320 kHz and by the nearly 270 kHz excitation rate of the $^2S_{1/2}$ state by the 194 nm radiation. When the 194 nm radiation is chopped, the two-photon linewidth drops to approximately 320 kHz.

In the near future we anticipate narrowing the 563 nm laser linewidth to the order of a few kHz and studying various systematic effects including pressure broadening and shifts, power broadening, and light shifts. Ultimately, we would like to narrow the laser linewidth to a value near that imposed by the 0.1 s natural lifetime of the D state, and to drive the two-photon (or single photon, electric quadrupole [8]) transition on a single, laser-cooled ion.

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1. P. B. Schwinberg, R. S. Van Dyck, Jr., and H. G. Dehmelt, Phys. Rev. Lett. 47, 1679 (1981).
2. R. S. Van Dyck, Jr. and P. B. Schwinberg, Phys. Rev. Lett. 47, 395 (1981)
3. D. J. Wineland, R. E. Drullinger, and F. L. Walls, Phys. Rev. Lett. 40, 1639 (1978).
4. W. Neuhauser, M. Hohenstatt, P. Toschek, and H. G. Dehmelt, Phys. Rev. Lett. 41, 233 (1978).
5. W. Nagourney, G. Janik, and H. G. Dehmelt, Proc. Natl. Acad. Sci. USA, 80, 643 (1983).
6. D. J. Wineland, W. M. Itano, J. C. Bergquist, J. J. Bollinger, and J. D. Prestage, in "Atomic Physics 9", R. S. Van Dyck, Jr. and E. N. Fortson, eds. (World Scientific Publ. Co., Singapore, 1984) p. 3.
7. H. G. Dehmelt, in "Advances in Laser Spectroscopy," F. T. Arecchi, F. Strumia, and H. Walther, eds. (Plenum, New York, 1983) p. 153.
8. D. J. Wineland, J. C. Bergquist, R. E. Drullinger, H. Hemmati, W. M. Itano, and F. L. Walls, J. Phys. (Orsay, Fr.) 42, C8-307 (1981); D. J. Wineland, W. M. Itano, J. C. Bergquist, and F. L. Walls, Proc. of 35th Annu. Symp. on Freq. Control, (1981) p. 602 (copies available from Electronic Industries Assoc., 2001 Eye St., NW, Washington, DC 200006).
9. W. M. Itano and D. J. Wineland, Phys. Rev. A 24, 1364 (1981).
10. J. D. Prestage, J. J. Bollinger, W. M. Itano, and D. J. Wineland, Phys. Rev. Let. 54, 2387 (1985).
11. J. J. Bollinger, J. D. Prestage, W. M. Itano, and D. J. Wineland, Phys. Rev. Lett. 54, 1000 (1985).
12. J. J. Bollinger and D. J. Wineland, Phys. Rev. Lett. 53, 348 (1984).
13. D. J. Wineland, J. J. Bollinger, and W. M. Itano, Phys. Rev. Lett. 50, 628; erratum: 50, 1333 (1983).
14. J. C. Bergquist, D. J. Wineland, W. M. Itano, H. Hemmati, H.-U. Daniel, and G. Leuchs, submitted for publication.
15. E. C. Beaty, to be published.
16. H. Hemmati, J. C. Bergquist, and W. M. Itano, Opt. Lett. 8, 73 (1983).
17. H. A. Schuessler, Appl. Phys. Lett. 18, 117 (1971); F. G. Major and J. L. Duchene, J. Phys. (Orsay) 36, 953 (1975); H. S. Lakkaraju and H. A. Schuessler, J. Appl. Phys. 53, 3967 (1982); M. Jardino, F. Plumelle, and M. Desaintfuscien, in "Laser Spectroscopy VI", H., P. Weber and W. Lüthy, eds. (Springer-Verlag, NY, 1983) p. 173; L. S. Cutler, R. P. Giffard and M. D. McGuire, Appl. Phys. B36, 137 (1985)