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Recoilless optical absorption and Doppler sidebands of a single trapped ion

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Spectroscopic measurements of the electric-quadrupole-allowed $5d^{10}6s\,^2S_{1/2}$ to $5d^96s^2\,^2D_{5/2}$ transition near 282 nm on a single, laser-cooled Hg^+ ion give a recoil-free absorption line (carrier) and well-resolved motional sidebands. From the intensity ratio of the sidebands to the carrier, the effective temperature of the Hg^+ ion was determined to be near the theoretical minimum of 1.7 mK. A fractional resolution of better than 3×10^{-11} for this ultraviolet transition is achieved.

Following the success of recent experiments on single, laser-cooled ions, most notably those experiments showing quantum jumps,[1-3] photon antibunching,[2,4] and absorption by a single ion,[5] several groups are now at the threshold of atomic spectroscopy that promises fractional resolution and accuracy exceeding 1 part in 10^{15}. In the work reported here, we experimentally demonstrate the spectroscopic resolution and signal-to-noise ratio that are attainable with a single atom nearly at rest in space. We have resolved the recoilless optical resonance[6,7] and motional sidebands of the $5d^{10}6s\,^2S_{1/2}$ to $5d^96s^2\,^2D_{5/2}$ transition ($\lambda=282$ nm) on a laser cooled $^{198}Hg^+$ ion confined in an rf trap. Since we can detect each transition to the metastable $^2D_{5/2}$ state with nearly 100% efficiency,[2] there is essentially no instrumental noise. The noise results only from the quantum statistical fluctuations in the transition probability of the single ion.[8] The unshifted resonance and sidebands shown in Fig. 1 are a convincing demonstration in the optical region of Dicke's original prediction of the spectral features of an atom whose spatial excursions are constrained to the order of λ.[6] From the sideband structure we are able to make a determination of the amplitude of the motion of the ion and its effective temperature.[9,10] We anticipate that by further cooling with narrowband laser radiation that is tuned to the first lower sideband of the $^2S_{1/2}$-$^2D_{5/2}$ transition, the Hg^+ ion can be made to reside at the zero-point energy of the trap's harmonic well most of the time.[9,11,12]

The experimental setup is essentially the same as that described in our earlier studies on the two-photon spectroscopy of Hg^+,[13] quantum jumps,[2] and single-ion absorption.[5] A mercury atom that is ionized by a weak electron beam is captured in a miniature rf (Paul) trap that has internal dimensions of $r_0\simeq455$ μm and $z_0\simeq320$ μm. The rf trapping frequency was 21.07 MHz with a peak voltage amplitude of about 730 V. The ion is laser cooled by a few microwatts of cw laser radiation that is frequency tuned below the $6s\,^2S_{1/2}$-$6p\,^2P_{1/2}$ first resonance line near 194 nm.[2,14] When the Hg^+ ion is cold and the 194-nm radiation has sufficient intensity to saturate the strongly allowed S-P transition, 2×10^8 photons/s are scattered. With our collection efficiency, this corresponds to an observed peak count rate of about 10^5 s^{-1} against a background of less than 50 s^{-1}.

The 282-nm radiation that drives the $^2S_{1/2}$-$^2D_{5/2}$ transition is obtained by frequency doubling the radiation from a cw ring dye laser that has been spectrally narrowed and stabilized in long term to less than 15 kHz. For this experiment, the dye laser is stabilized to a high-finesse optical cavity[15] to give good short-term stability. In long term, the laser is stabilized by FM optical heterodyne spectroscopy to a saturated-absorption hyperfine component in $^{129}I_2$.[16] The pressure in the iodine cell is not controlled against temperature variations, and this gave rise to pressure-induced frequency fluctuations that dominated the stability of the laser for times greater than about 10 s. The frequency of the laser is scanned by tuning the frequency applied to an acousto-optic modulator through which the laser beam is passed. We eliminated angle variation in the beam that was frequency shifted

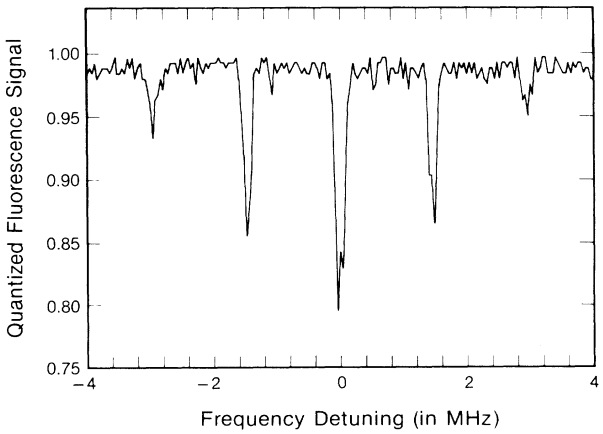

FIG. 1. Quantized signal showing the electric-quadrupole-allowed $5d^{10}6s\,^2S_{1/2}(m_J=-\frac{1}{2})$-$5d^96s^2\,^2D_{5/2}(m_J=\frac{1}{2})$ transition in a single, laser-cooled $^{198}Hg^+$ ion. On the horizontal axis is plotted the relative detuning from line center in frequency units at 282 nm. 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. The integration time per point is about 16 s (230 measurement cycles).

and scanned by using the twice-frequency-shifted beam obtained by retroreflecting the first-order frequency-shifted beam back through the crystal. A computer controls the frequency and amplitude of the synthesizer that drives the acousto-optic modulator. Up to a few microwatts of 282-nm radiation could be focused onto the ion in a direction either orthogonal to, or counterpropagating with the 194-nm light beam. A magnetic field of approximately 1.2 mT (12 G) was applied parallel to the electric field vector of the linearly polarized 282-nm radiation to give well-resolved Zeeman components. This configuration gives a selection rule of $|\Delta m_J| = 1$ for the electric-quadrupole-allowed transitions to the various Zeeman states.

Optical-optical double-resonance utilizing quantum amplification[13,17-19] was used to detect transitions driven by the 282-nm laser to the metastable $^2D_{5/2}$ state. Our version of this method makes use of the fact that the 194-nm fluorescence intensity level is bistable; high when the ion is cycling between the S and P states (the "on" state) and nearly zero when it is in a metastable D state (the "off" state).[1-3] The fluorescence intensity in the on state is high enough that the state of the atom can be determined in a few milliseconds with nearly 100% efficiency. The full measurement cycle was as follows: A series of measurements of the 194-nm fluorescence was made, using a counter with a 10-ms integration period. As soon as the counter reading per measurement period was high enough to indicate that the ion was in the on state, the 194-nm radiation was shut off and the 282-nm radiation was pulsed on for 20 ms. Then, the 194-nm radiation was turned on again, and the counter was read. If the reading was low enough to indicate that the ion had made a transition to the $^2D_{5/2}$ state (the off state), the signal was defined to be 0. Otherwise, it was defined to be 1. The 282-nm laser frequency was then stepped, and the cycle was repeated. As the laser frequency is swept back and forth through the resonance, the quantized measurement of the fluorescence signal at each frequency step is averaged with the previous measurements made at that same frequency. Since we could detect the state of the ion with nearly 100% efficiency, there was essentially no instrumental noise in the measurement process. Occasionally, while the 194-nm radiation was on, the ion decayed from the $^2P_{1/2}$ state to the metastable $^2D_{3/2}$ state rather than directly to the ground state.[2] This process led to a background rate of false transitions which was minimized by the quantized data-collecting method described above and by decreasing the 194-nm fluorescence level (thereby decreasing the $^2P_{1/2}$-$^2D_{3/2}$ decay rate) until it was just high enough for the quantized detection method to work. The quantized measurement scheme also removes any contribution to the signal base line due to intensity variations in the 194-nm source. The 282- and 194-nm radiation were chopped so that they were never on at the same time. This eliminated shifts and broadening of the narrow 282-nm resonance due to the 194-nm radiation.[18,20,21]

Figure 1 shows the fluorescence signal obtained from an 8-MHz scan of the 282-nm laser through the

$$5d^{10}6s\,^2S_{1/2}(m_J = -\tfrac{1}{2}) \rightarrow 5d^9 6s^2\,^2D_{5/2}(m_J = \tfrac{1}{2})$$

Zeeman component of the laser-cooled Hg^+ ion. The recoilless absorption resonance (carrier)[6,7] and the motional sidebands due to the secular motion of the ion in the harmonic well of the rf trap are completely resolved. The inhomogeneous rf electric field produces a pseudopotential harmonic well with a radial frequency of approximately 1.46 MHz and an axial frequency of approximately twice the radial frequency, or about 2.9 MHz. An ion trapped in this pseudopotential well will execute nearly independent harmonic motions in the radial and axial directions. These harmonic motions frequency modulate the laser radiation, as seen by the trapped ion and produce sidebands on the carrier frequency at the radial and axial frequencies and their harmonics.[9] Thus, the number and amplitude of sidebands are a direct measure of the amplitude of the ion's motion and its effective temperature. In Fig. 1, only two pairs of sidebands are obtained. In this figure the 282-nm intensity is adjusted to be close to saturation on the carrier in order to enhance the relative strength of the sidebands to the carrier. Also, the laser linewidth was broadened to about 80 kHz (at 563 nm) by modulating the frequency of the acousto-optic modulator in order to reduce the number of data points required for the sweep. A careful comparison of the sideband intensities [Ref. 9, Eq. (44)], including the effects of saturation, gives an ion temperature for the secular motion of about 5.8 mK. This is near the theoretical laser-cooling limit for $^{198}Hg^+$ on the first resonance line at 194 nm given by[9] $T_{\min} = \hbar\gamma/2k_B \simeq 1.7$ mK.

The spectrum in Fig. 1 was taken with the 194-nm beam counterpropagating to the 282-nm beam. This arrangement gives optimum cooling to the ion motion in the direction that is probed by the 282-nm beam. We have also probed in a direction perpendicular to the propagation direction of the 194-nm beam. In this case, if there is perfect symmetry in the radial plane of the trap, then the ion will heat without bound in the radial direction perpendicular to the cooling beam.[9,11] Trap asymmetry in the radial plane permits cooling in all directions provided the $\mathbf{k}$ vector of the cooling beam is not in the radial plane nor collinear with the trap axis.[9,11,18] Even though cooling in all directions is possible, the limiting temperature will be higher in the direction perpendicular to the cooling beam due to recoil heating in that direction. From our data, we find this temperature to be only a few mK higher, indicating a fairly rapid energy transfer between the ion's motional degrees of freedom.

Stray static electric fields in the trap can produce residual micromotion at the rf drive frequency of 21.07 MHz. This happens because the static field shifts the ion's equilibrium point to a region where the force from the static field is compensated for by the force from the pseudopotential. In this case, there is residual micromotion at the bottom of the well even for a cold ion. The micromotion will produce sidebands on the carrier at harmonics of the micromotion frequency analogous to the secular motion sidebands. The appearance of rf sidebands in our initial scans indicated the presence of a small stray static field perhaps caused by contact potential variations on one or more of the electrodes. It was possible to null out the rf sidebands by applying a compensating dc voltage of about

0.3 V to one of the end caps. To null the rf sidebands in all directions would likely require extra compensation electrodes.

In Fig. 2, we show high-resolution scans through the Doppler-free resonance at line center and across the first upper sideband. The average integration time per point is about 10 s (160 measurement cycles). The linewidth at half maximum is approximately 30 kHz at 282 nm (or 15 kHz at 563 nm), giving a fractional resolution of about 3×10^{-11}. If we can obtain a resolution that approaches the natural linewidth limit of 1.6 Hz, the required integration time per point for the same signal-to-noise ratio of Fig. 2 will increase by a factor of less than 2. For the data of Fig. 2, an effective temperature of 1.6 ± 0.5 mK and an rms amplitude of 28 ± 5 nm for the secular motion was obtained. However, over several sets of data similar to that of Fig. 2, we obtained temperatures that ranged from 1.6 to 6.7 mK. The discrepancies between the measured temperatures and the theoretical minimum might be due to the uncertainty in the tuning of the 194-nm cooling laser to the half-power point on the S-P transition. If the frequency of the laser were detuned to a longer wavelength by a natural linewidth, the temperature obtained would be about twice the minimum. Also, some heating may have been caused by collisions with the background gas. Finally, if the frequency of the 282-nm laser is tuned to the first lower secular sideband, it would be possible to cool the ion until it occupied the zero point or $\langle n_i\rangle=0$ ($i=x,y,z$) energy state of the harmonic well most of the time.[9,11,12,18]

In summary, we have laser cooled a single ion and probed with high resolution the $^2S_{1/2}$-$^2D_{5/2}$ quadrupole resonance at 282 nm. The ability to detect each transition with almost no ambiguity eliminated nearly all instrumental noise. We have resolved the recoilless resonance line and the Doppler-generated motional sidebands. From the relative strength of the sidebands, a direct measurement of the ion temperature and its spatial excursion was made.

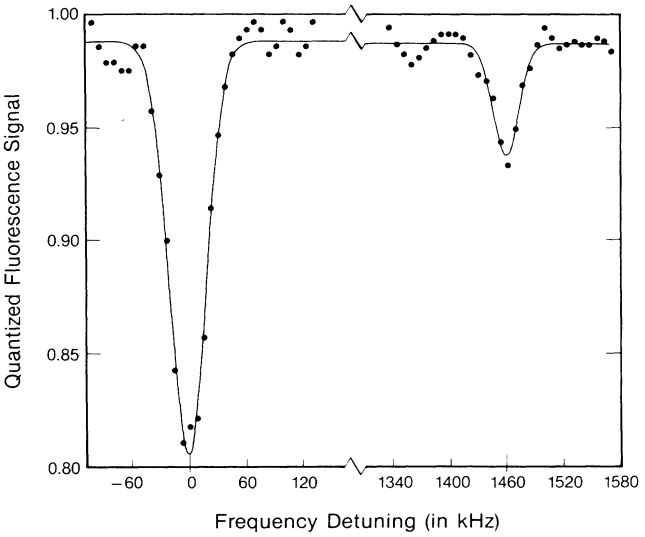

FIG. 2. High-resolution scan through the recoilless absorption resonance and first (upper) sideband of the $^2S_{1/2}$-$^2D_{5/2}$ ($m_J=-\frac{1}{2}$-$m_J=\frac{1}{2}$) transition in a single laser-cooled $^{198}Hg^+$ ion. The full width at half maximum of the recoilless absorption resonance and of the upper sideband is about 30 kHz at $\lambda=282$ nm (15 kHz at $\lambda=563$ nm). A comparison of the carrier-to-sideband intensity gives a temperature of 1.6 ± 0.5 mK, in agreement with the theoretical minimum of 1.7 mK. The integration time per point is about 10 s (160 measurement cycles).

We anticipate improved spectral purity of the probe laser and better magnetic shielding of the Hg^+ ion in the future to give increased spectral resolution.

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