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Hg^+ Single Ion Spectroscopy*

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Abstract

A single Hg^+ ion that is confined in an rf (Paul) trap can be laser cooled so that the amplitude of its motion is much less than a wavelength (the Dicke limit) for optical transitions. Recently, we have used the technique of optical sideband cooling to reach the zero point of motion. This realizes for the first time the fundamental limit of laser cooling for a bound atom and the ideal of an isolated atomic particle at rest in space to within the quantum mechanical limits imposed by the surrounding apparatus. In both limits, Doppler effects become negligible to all orders, the interrogation time is long and the fundamental shot noise detection limit of a single atom is readily attained.

Introduction

Laser cooling of a single atom has led to a number of important experiments in fundamental physics [1-5] and may allow optical spectroscopy and metrology with a resolution and accuracy that approach 1×10^{-18} [6-9]. One of the candidates for an optical frequency standard that approaches this accuracy is a single, laser-cooled $^{199}Hg^+$ ion confined in a miniature rf trap [8]. At NIST we have begun with an investigation of a single $^{198}Hg^+$ ion, laser cooled into the Dicke limit for optical transitions [8] and, in a second experiment, laser cooled in the sideband limit to the zero-point energy of motion [10,12]. The $^{198}Hg^+$ ion offers the experimental advantage of zero nuclear spin; hence, no hyperfine structure and a simpler optical configuration for cooling. However, all the narrow optical transitions have a magnetic field sensitivity of the order of 10 MHz/mT (1 MHz/G), whereas the $^{199}Hg^+$ ion has several first-order magnetic field-independent transitions.

In the work summarized here we begin to show the spectroscopic resolution and signal-to-noise ratio that are possible with a single atom nearly at rest in space. We have resolved the recoilless optical resonance and the motional sidebands of the $5d^{10}6s\ ^2S_{\frac{1}{2}}(m_J=-\frac{1}{2})$ - $5d^96s^2\ ^2D_{5/2}(m_J=\frac{1}{2})$ electric quadrupole transition ($\lambda\approx282$ nm) on a laser-cooled $^{198}Hg^+$ ion [8]. Since we can detect each transition to the

metastable $^2D_{5/2}$ state ($\gamma/2\pi \approx 1.7$ Hz) with nearly 100% efficiency, there is essentially no technical noise. The noise results only from the quantum statistical fluctuations in the transition probability of the single ion [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 [10,11,13]. The sidebands at $\omega_0 \pm n\omega_v$ (n an integer) are generated by the oscillatory motion at frequency ω_v of the ion in its confining harmonic well. If the ion is driven with narrowband radiation tuned to the first lower sideband, the atom will absorb photons of energy $\hbar(\omega_0-\omega_v)$ and in most cases re-emit photons of energy $\hbar\omega_0$. This reduces the atom's vibrational quantum number by 1 for each scattered photon at frequency ω_0. In this way we can obtain $\langle n_v \rangle \ll 1$, which means that the atom spends most of its time in the ground state level of its confining potential [10,12-14]. To the extent that the atom is in the ground state of its confining potential the fundamental limit of laser cooling for a confined particle has been reached [12].

Experiment

The mercury ion is confined in a miniature rf trap that has internal dimensions of $r_0 \approx 466$ μm and $z_0 \approx 330$ μm [8,15]. The amplitude of the trapping field (frequency $\Omega/2\pi \approx 21$-23 MHz) could be varied to a peak of 1.2 kV. The ion is laser cooled to a few millikelvins by a few microwatts of cw laser radiation [16] that is frequency tuned below the $^2S_{\frac{1}{2}}$ - $^2P_{\frac{1}{2}}$ first resonance line near 194 nm [2,8]. In order to cool all motional degrees of freedom to near the Doppler cooling limit ($T = \hbar\gamma/2k_B \approx 1.7$ mK) the 194 nm radiation irradiates the ion from 2 orthogonal directions, both of which are at an angle of 55° with respect to the symmetry (z) axis of the trap. The 282 nm radiation that drives the narrow $^2S_{\frac{1}{2}}$ - $^2D_{5/2}$ transition is obtained by frequency-doubling the radiation from a narrowband cw ring dye laser. In the long term, the laser is stabilized by FM optical heterodyne spectroscopy to a saturated absorption hyperfine component in $^{129}I_2$ [17]. The frequency of the laser is scanned by an acousto-optic modulator that is driven by a computer controlled synthesizer. Up to a few microwatts of 282 nm radiation could be focussed onto the ion in a direction counterpropagating with one of the 194 nm light beams.

Optical-optical double resonance (electron shelving) [1-3,8,15,18-20] with a net quantum amplification (including detection efficiency) in excess of 100 at 10 ms was used to detect transitions driven by the 282 nm laser to the metastable $^2D_{5/2}$ state. The fluorescence rate from the laser-cooled ion is high when the ion is cycling between the $^2S_{\frac{1}{2}}$ and $^2P_{\frac{1}{2}}$ states and nearly zero when it is in the metastable $^2D_{5/2}$ state [1-3,8]. Thus the $^2S_{\frac{1}{2}}$ - $^2D_{5/2}$ resonance spectrum was obtained by probing the S-D transition at a particular frequency for the 282 nm radiation for 20 ms, then turning off the 282 nm radiation and turning on the 194 nm radiation to look for the presence or absence of scattered photons at 194 nm (the two radiation fields are alternately applied to avoid light shifts and broadening of the narrow

S-D transition). If there was no fluorescence at 194 nm, a transition into the metastable D state had occurred; the presence of 194 nm fluorescence indicated that the ion was in the ground state and no transition was recorded for this frequency of the 282 nm laser. The frequency was then stepped and the measurement cycle repeated. As the frequency was stepped back and forth each new result at a particular frequency of the 282 nm radiation was averaged with the previous measurements at that frequency. Normalization (or quantization) of the signal was obtained by assigning a 1 to each measurement of high fluorescence and a 0 to each measurement of no fluorescence. The high fluorescence level made it possible to determine the state of the atom with almost no ambiguity in a few milliseconds. Thus, it is easy to reach the shot noise limit imposed by the single atomic absorber [8].

The quantized fluorescence signal obtained from an 8-MHz scan of the 282 nm laser through the $^2S_{\frac{1}{2}}(m_J=-\frac{1}{2}) \rightarrow {}^2D_{5/2}(m_J=\frac{1}{2})$ Zeeman component of the electric quadrupole transition is shown in Fig. 1 [8]. The recoilless-absorption resonance (carrier) [21,22] and the motional sidebands due to the secular motion in the harmonic well of the rf trap [10] are completely resolved. The number and amplitudes of the sidebands are a direct measure of the ion's motion and its effective temperature. A careful comparison of the intensities of the sidebands to that of the carrier [9] for several sets of data similar to that shown in Fig. 1, including the effects of saturation, gave temperatures that ranged from 1.6 to 6.7 mK. This is consistent with the theoretical laser-cooling limit for $^{198}Hg^+$ on the first resonance line at 194 nm given by $T_{min} = \hbar\gamma/2k_B \approx 1.7$ mK [10].

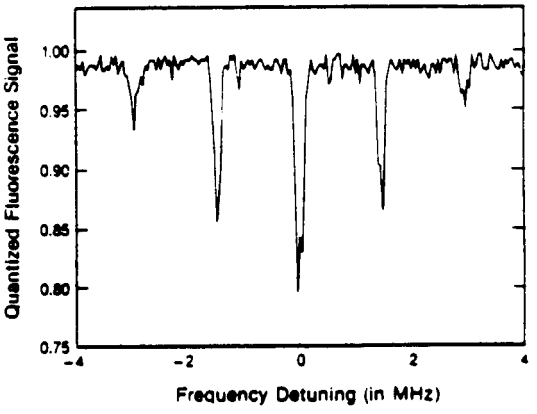

Fig. 1. Quantized signal showing the electric-quadrupole-allowed $5d^{10}6s\ ^2S_{\frac{1}{2}}(m_J=-\frac{1}{2})$-$5d^96s^2\ ^2D_{5/2}(m_J=\frac{1}{2})$ transition in a single, laser-cooled $^{198}Hg^+$ ion. On the horizontal axis is plotted the relative detuning from line center in frequency units at 282 nm. On the vertical axis is plotted the probability that the fluorescence from the $6s\ ^2S_{\frac{1}{2}}$-$6p\ ^2P_{\frac{1}{2}}$ 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).

In Fig. 2 we show a high resolution scan through the Doppler-free (recoilless) resonance at line center The full width at half maximum (FWHM) is approximately 1.7 kHz at 563 nm (3.4 kHz at 282 nm). This corresponds to a fractional resolution of about 3.4×10^{-12} ($Q\approx3\times10^{11}$). For this trace the laser was spectrally narrowed by locking to a mechanically, acoustically and thermally quiet reference cavity that had a finesse of about 60,000. The frequency of the laser fluctuates less than 1 Hz relative to this reference cavity. This is determined from the measurement of the residual noise in the error signal; the actual frequency fluctuations of the laser are then governed by the stability of the well isolated cavity [23]. The natural linewidth for the $^2S_{\frac{1}{2}}$ - $^2D_{5/2}$ transition is approximately 1.7 Hz [15]. The measured linewidth in Fig. 2 is dominated by fluctuations in the first-order magnetically-sensitive electric quadrupole transition in the $^{198}Hg^+$ ion. The field sensitivity of the $^2S_{\frac{1}{2}}(m_J=-\frac{1}{2})$ to $^2D_{5/2}(m_J=\frac{1}{2})$ transition is on the order of 22.4 MHz/mT (2.24 MHz/G). In our laboratory there are field fluctuations of a few milligausses with a frequency-noise spectrum that is dominated by 60 Hz and its harmonics. Shielding would help but is difficult in the present experiment. The better approach is to switch to the $^{199}Hg^+$ isotope. This isotope has first-order, field-independent transitions near zero magnetic field that should permit us to reach the 1.7 Hz natural linewidth. In this case, the ion will provide a good test of the spectral purity of the 282-nm laser.

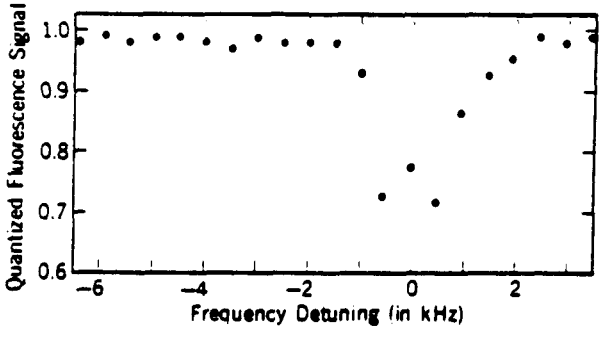

Fig. 2 High resolution scan through the recoilles absorption resonance of the $^2S_{\frac{1}{2}}(m_J=-\frac{1}{2})$-$^2D_{5/2}(m_J=1)$ transition in a single laser cooled $^{198}Hg^+$ ion. The full width at half maximum is about 1.7 kHz at λ=563 nm (3.4 kHz at λ=282 nm).

The possibility of cooling the Hg^+ ion further, to the zero point energy of motion, is intriguing for several reasons. First, cooling so that the average occupational number $\langle n_v \rangle$ for the motional energy of the bound atom is zero is a fundamental limit to laser-cooling for any bound particle [8]. This limit is imposed by the position-momentum Heisenberg uncertainty principle where the spatial confinement (Δx) is provided by the physical surroundings. Driving a single atom in a macroscopic trap to the zero point energy of motion also exploits the benign environment near the center of an rf trap - certainly a system worthy of consideration for the high resolution and accuracy in optical spectroscopy, metrology and frequency standards. Second, with an ion prepared in the lowest

vibrational state (most of the time), experiments such as squeezing the atom's position and momentum can be demonstrated.

Cooling to the zero point energy of motion is possible for a bound atom in the resolved sideband limit where the linewidth of the cooling transition ($^2S_{\frac{1}{2}}$ - $^2D_{5/2}$ in our case) is less than motional oscillation frequency of the atom [10,12,13]. When the ion is in the lowest or ground vibrational state (n_v=0) it can no longer resonantly absorb a photon at ω_0-ω_v; at this point the resonant feature at ω_0-ω_v disappears. Experimentally, a measurement of the amplitude of the resonant feature at ω_0-ω_v (for saturating intensity) is a direct measurement of $\langle n_v \rangle$ [13]. The result from ref. 12 is shown in Fig. 3.

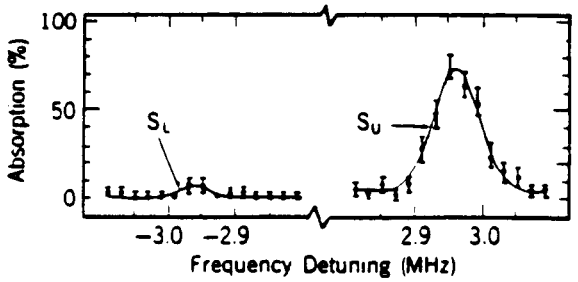

Fig. 3 First lower (S_L) and upper (S_u) motional sidebands of the $^2S_{\frac{1}{2}}$-$^2D_{5/2}$ electric quadrupole transition of a laser-sideband-cooled $^{198}Hg^+$ ion measured 10 ms after cooling. Data points are averages of 41 sweeps.

From these data we deduce that the y and z degrees of freedom are in the zero point energy state (n_v=0) 95% of the time. Because the 282 nm probe beam is nearly orthogonal to the x-axis, we were not sensitive to the x-motion. The uncertainty in the second-order Doppler shift in this example is dominated by the uncertainty in $\langle n_v \rangle$ and amounts to $\Delta\nu/\nu \approx 10^{-20}$ [13]. It could be made even lower by adiabatically lowering the potential well depth (thereby lowering ω_v) after the ion is cooled into the ground state.

With this system, the absorption of a <u>single</u> quantum of energy at a frequency in the megahertz range would raise n_v by one unit and this could be detected with an efficiency of nearly 100%. Also, it should be possible to produce squeezed states [24] of the atom's motion from the zero point energy state by a sudden, non-adiabatic weakening (and, in general, shifting) of the trap potential or by driving the atomic motion parametrically at 2 ω_v. If after some time the atom could then be returned to the n_v=0 state by reversing the above procedures, the zero point energy state could be unambiguously detected by the absence of the lower sideband. This would convincingly demonstrate the squeezed state of the single atom in the rf trap.

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