Observation of a midinfrared fine-structure transition for the single trapped barium ion

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We report observation of laser excitation of a narrow, magnetic-dipole-allowed, infrared transition in a single trapped ion. Quantum jumps of a Ba^+ atom were observed using a diode laser tuned in resonance with the excited-state $5d\,^2D_{3/2}-5d\,^2D_{5/2}$ transition at 12.48 μm. Single-ion resonance line shapes were obtained and a transition frequency of $\nu=24\,012\,033\pm11$ MHz was determined. Calculated quantum jump rates of the *D-D* transition for the applied laser spectral brightness yield agreement with experimentally observed values.

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The unique environment provided by the confinement of ions in an electromagnetic trap at high vacuum has opened new opportunities in atomic spectroscopy at very high resolution and for a possible new generation of frequency standards [1–5]. The long interrogation times and small confinement volume for the ion provide the possibility to examine transitions with very narrow linewidths without transit time limitations. In addition, for a trapped ion collisional perturbations are small and using the techniques of laser cooling, the perturbing effect of Doppler broadening and shifts can be minimal. To date, the microwave and near visible spectral regions have been used in the study of narrow linewidth transitions of trapped ions. Clouds of trapped ions have been used in the study of hyperfine transitions in the microwave region [2,3]. The frequency resolution obtained in such work was comparable to that provided by Cs beam frequency standards. Recently, single-ion studies using ground to excited electronic state, electric quadrupole transitions in the region of $10^{14}-10^{15}$ Hz have begun [1,4,5]. In the case of the work dealing with the single Hg^+ ion [5], fractional frequency resolutions of better than 2×10^{-13} have been obtained.

Dehmelt and co-workers [6], Werth [7], and our group [8] have proposed the use of narrow linewidth, fine-structure transitions in trapped ion samples as interesting candidates for ultra-high-resolution spectroscopy. Some of the important advantages of these transitions include their very high transition frequency to natural linewidth ratio (Q up to 10^{15}) allowing the possibility for extreme precision. Also, the transition frequencies of these systems lie principally in the far- to midinfrared region of the spectrum where phase-locked frequency chains exist [9] for the measurement of the transition frequency relative to a Cs time standard. In the present work, we report the observation of optical excitation of a midinfrared, fine-structure transition in a single, trapped ion.

The fine-structure transition studied in this work is the $5d\,^2D_{3/2}-5d\,^2D_{5/2}$ transition in Ba^+ at a wavelength of 12.5 μm (see Fig. 1). The lifetimes of the metastable ^2D levels are very long since dipole transitions to the ground state are forbidden. Experiments measuring the natural lifetime of these levels have yielded $t=34.5\pm3.5$ s for the $5d\,^2D_{5/2}$ level [10,11] and $t=17.5\pm4$ s for the $5d\,^2D_{3/2}$ level [12] allowing a possible resolution of better than 0.02 Hz at 24.0 THz ($Q>10^{15}$). Calculation of the transition moment of the $^2D_{3/2}-{}^2D_{5/2}$ magnetic-dipole transition (assuming negligible change in the radial wave function) yields a reduced matrix element for the transition of $\langle\frac{3}{2}|M|\frac{5}{2}\rangle=(\frac{12}{5})^{1/2}\mu_B$, where μ_B is the Bohr magneton and a spontaneous emission rate of $A_{D\text{-}D}=0.006$ s^{-1}. These values indicated that effective saturation of the magnetic-dipole transition could be achieved using a standard lead-salt diode laser source operating with a typical output power (0.1 mW) and linewidth (10^8 Hz). Since the accuracy of the previous best determination of the *D-D* transition frequency was quite low [13] ($\sigma=800.97\pm0.01$ cm^{-1}), the goal of the present work was to perform a preliminary study of the transition and

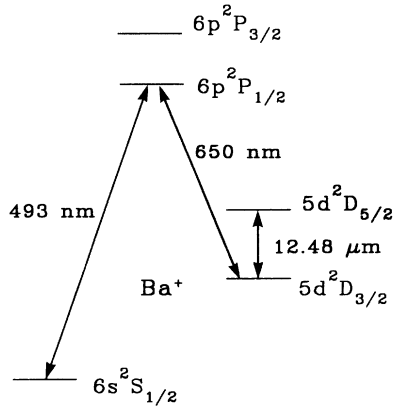

FIG. 1. Energy-level diagram for the lowest electronic states of Ba^+ showing the transitions used for detection and cooling (493 and 650 nm) and for excitation of the magnetic-dipole transition at 12.5 μm.

determine such parameters as transition frequency and excitation rate using a tunable laser diode source with moderate resolution.

The experimental arrangement used in trapping, observing, and laser cooling the Ba^+ ions was essentially the same as that employed in previous studies and is described in detail elsewhere [11]. The trapping of the Ba^+ atoms is provided by a radio-frequency trap with an effective pseudopotential well depth for $^{138}Ba^+$ of $D_z=16$ V and $D_r=8.0$ V. Laser cooling and fluorescence excitation of the barium ion was provided by excitation on the $6p\ ^2P_{1/2}-6s\ ^2S_{1/2}$ ($\lambda=493.5$ nm) and the $6p\ ^2P_{1/2}-5d\ ^2D_{3/2}$ ($\lambda=650$ nm) transitions (see Fig. 1). The laser light was provided from commercial ring dye lasers of 1-MHz linewidths. In most experiments, the 493-nm laser was tuned approximately 50 MHz below line center for effective laser cooling while the 650-nm laser was maintained about 10–40 MHz below line center. The radiation from both lasers was focused to a spot size $\omega=25\pm10$ μm at the trap center with incident laser powers of 30 μW at 493.5 nm and approximately 25–100 μW at 650 nm. Detection of the Ba^+ was achieved by observing the 493 nm S-P fluorescence using a photon counting photomultiplier system and a digital rate meter, recorded by a computer for storage and analysis.

Radiation at 12.5 μm was generated by a cryogenic, tunable lead-salt diode laser (Laser Photonics Analytics L5615-0790). Initial tests using a monochromator for mode selection showed that single mode operation of the laser was possible in the wavelength region of interest, with 50 ± 10 μW of power. Thereafter, the monochromator was not used. The short-term linewidth of the laser light was determined to be 40 ± 10 MHz. A portion of the output radiation was sent via a beam splitter to two diagnostic systems for determination of the output frequency and frequency tuning rate of the laser. One portion of the beam was sent through a 50-cm cell filled with $^{15}NH_3$ at 13 Pa and then focused on a Hg-Cd-Te detector. The observed absorption lines as the laser was scanned were then compared with the known absorption spectrum [14] of $^{15}NH_3$ to determine the absolute output frequency of the diode laser. The $sP(8,7)$ transition of the ν_2 fundamental band in $^{15}NH_3$ was known to be in close proximity to the desired Ba^+ D-D transition frequency and once identified, $sP(8,7)$ was used as the absolute frequency reference for the diode laser. A second portion of the output diode laser power was sent to a $\delta\nu=45.9$ MHz free spectral range confocal étalon with a finesse better than 20. The transmitted light through the étalon served to provide a frequency scale as the laser was scanned and were used to calibrate observed diode laser tuning rate as a function of changes in diode laser current. The diode laser output power sent directly into the ion trap was focused using an $f=0.27$ m NaCl lens. In order for the 12.5-μm radiation to enter the vacuum enclosure, a BaF_2 window system using indium seals was mounted on the vacuum chamber where the visible laser beams emerge from the trap. As a result, the infrared light direction was counterpropagating to the incident light at 493 and 650 nm. Despite the low bakeout temperature set by the use of indium seals, a good vacuum was eventually obtained of better than 0.1 μPa, consisting mostly of H_2. Based on our previous collisional quenching studies [11], the calculated collisional perturbation of the ion by the background gas reduced the $5d\ ^2D_{5/2}$ lifetime by no more than 10%. The infrared power at trap center was $P_0=5.9\pm2.3$ μW with the measured beam spot being elliptical with parameters $\omega_x=233\pm70$ μm and $\omega_y=165\pm50$ μm.

In initial experiments, a search for the D-D transition was undertaken by loading a cloud of Ba^+ atoms in the trap. Typically 40–50 ions were loaded and the fluorescence at 493.5 nm was quite strong (10^5 detected counts s^{-1}). Excitation by the infrared light on the D-D transition resulted in ions being shelved in the metastable $5d\ ^2D_{5/2}$ level and a decrease in the total fluorescence at 493 nm. Figure 2 shows the observed D-D transition signal in the S-P fluorescence as the laser was scanned in the vicinity of the $^{15}NH_3$ $sP(8,7)$ transition frequency. In order that a derivative signal be obtained from the $^{15}NH_3$ reference cell, a small frequency dither on the diode laser frequency (<30 MHz) was applied. The frequency scan in Fig. 2 is from lower to higher frequencies and one can observe the rapid drop in fluorescence as the 12.5-μm light is swept into resonance. The slow decay back to maximum fluorescence is due to the laser quickly sweeping through the transition and ions shelved in the $^2D_{5/2}$ metastable level then slowly decay with essentially the natural decay rate of 0.03 s^{-1}. Determinations of the D-D transition frequency were made by comparing the Ba^+ transition signal with the $^{15}NH_3$ $sP(8,7)$ reference absorption and using the étalon fringe scale to measure the frequency displacement. Both upward and downward frequency scans were performed to eliminate asymmetry due to the slow natural decay of atoms out of the upper metastable level. The mean value of the data yielded the D-D transition frequency at a frequency displacement of $+0.3\pm7$ MHz from the $sP(8,7)$ reference transition. The indicated error is only the statistical error and other error sources such as those due to Doppler shifts may contribute uncertainty to the same order as the quoted statistical uncertainty.

The next stage of experiments involved the use of a single, trapped, laser cooled Ba^+ atom. As in other spectroscopic studies of dipole-forbidden single-ion transitions [4,5], the excitation into and out of the metastable level was monitored via quantum jumps in the strong fluorescence. The sequence for the study of excitation at different detunings proceeded in the following manner. The diode laser frequency was initially steered to the reference $sP(8,7)$ absorption frequency. A known frequency detuning (by using a small offset current) was applied to the diode laser. The quantum jump bright and dark rates were than recorded by computer for a fixed period of time (typically 200 s). No modulation was applied to the diode laser during this interrogation period. In addition, the ion was alternately illuminated by the 650-nm and 12.5-μm light during the interrogation period in order to minimize power broadening of the lower $5d\ ^2D_{3/2}$ level. The number of stimulated events was determined by counting each change in fluorescence bright and dark periods during the 650-nm illumination.

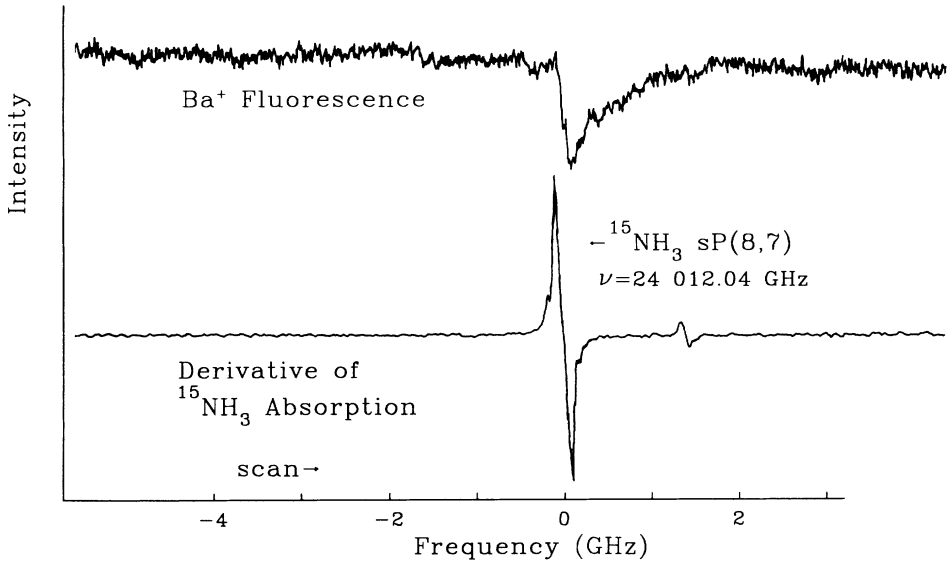

FIG. 2. Observed decrease in 493-nm fluorescence from a cloud of 50 Ba^+ atoms due to excitation on *D-D* fine-structure transition at 12.5 μm. Superimposed below the Ba^+ fluorescence signal is the reference derivative absorption spectrum of $^{15}NH_3$ showing the absorption resonance of the $sP(8,7)$ transition.

The alternation between the 650-nm and infrared radiation occurred at a rate of about 6 Hz. The intensity of the infrared radiation was sufficiently low that on average, less than one *D-D* transition occurred during a full cycle of alternate illumination at 650 nm and 12.5 μm. At the end of the 200-s measurement period, laser detuning was then checked by relocking the diode laser frequency to the $sP(8,7)$ reference line and noting any change in current offset. By this technique, the quantum jump excitation rate as a function of detuning was obtained with minimum drift of the diode laser frequency during the experiment and minimal power broadening of the *D-D* transition. The single trapped ion temperature was estimated to be $T < 10$ K from observation of the *S-P* and *P-D* line shapes. The Doppler broadening for the *D-D* transition is thus estimated to be less than 4 MHz, which is small compared to the diode laser linewidth. In the single-ion quantum jump studies, the ion in the *S-P-D* superposition (2000 fluorescent and background counts per second) was unambiguously observed relative to the periods when the ion resided in the $5d\,^2D_{5/2}$ state (600 background counts per second). Figure 3 shows the observed quantum jump mean bright rate R_+ $(=1/\langle T_{on}\rangle$ where $\langle T_{on}\rangle$ is the mean length of the bright periods) as a function of the various detunings applied to the infrared laser. The line shape observed shows a width of $\delta\nu = 80 \pm 10$ MHz, which is larger than that expected from the measured short-term linewidth of the diode laser. The broadening is believed to arise from the frequency drift of the unstabilized diode laser during the 200-s interrogation periods. The center frequency of the transition was determined to be $\delta\nu = -10 \pm 10$ MHz relative to the $sP(8,7)$ center frequency, in agreement with the ion cloud measurements. D'Cunha and co-workers [14] have determined the $^{15}NH_3$ $sP(8,7)$ center frequency to be $\sigma = 800.955\,67 \pm 0.000\,20$ cm^{-1} and recently Johns [15] has obtained an improved value of $\sigma = 800.955\,524 \pm 0.000\,070$ cm^{-1}. Using the latter value and our single-ion measurement, we have determined the absolute frequency of the *D-D* transition to be $\sigma = 800.955\,19 \pm 0.000\,40$ cm^{-1} or $\nu = 24\,012\,033 \pm 11$

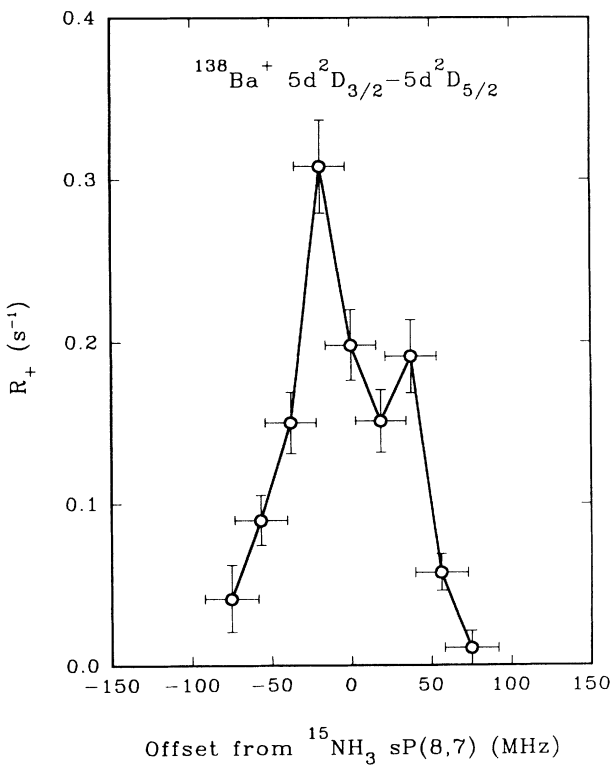

FIG. 3. Observed single-ion $D_{3/2}-D_{5/2}$ transition quantum jump rate as a function of diode laser detuning. The zero of the frequency scale is referenced to the center frequency of the $sP(8,7)$ transition of the ν_2 band in $^{15}NH_3$ located at $\nu = 24\,012.04$ GHz.

MHz. We can compare the mean observed quantum jump dark and bright periods with the calculated transition periods. Studies of the mean bright period $\langle T_{on} \rangle$ and dark period $\langle T_{off} \rangle$ were made with simultaneous excitation on the visible and infrared transition. Nienhuis [16] has presented the theoretical bright and dark rates for broadband excitation on a weak transition as

$$1/\langle T_{on} \rangle = R_+ = R_{12} N_1 , \quad (1)$$

$$1/\langle T_{off} \rangle = R_- = A_2 + R_{21} , \quad (2)$$

where N_1 is the population probability in the coupled lower state of the weak transition, A_2 is the spontaneous decay rate of the upper state, and R_{12} and R_{21} are, respectively, the stimulated absorption and emission rates on the weak transition. In our case, the lower state of the weak transition also has a spontaneous decay rate (0.06 s^{-1}), however, the excitation of the strong *S-P-D* system is rapid (10^6 s^{-1}) and the quantum jump periods are essentially unperturbed. From our calculated transition moments for the magnetic-dipole transition, we have calculated the Einstein B coefficients for the weak transition to be $B_{12} = 9.7 \times 10^{14}$ $J^{-1} s^{-2} m^3$ and $B_{21} = 6.5 \times 10^{14}$ $J^{-1} s^{-2} m^3$. Given the estimated intensity at trap center and spectral width observed in the single-ion line-shape scans together with the estimated steady-state population fraction in the $5d\,^2D_{3/2}$ level and the known spontaneous decay rate out the $5d\,^2D_{5/2}$ level, the mean bright and dark periods are calculated as $\langle T_{on} \rangle = 0.91 \pm 0.6$ s and $\langle T_{off} \rangle = 0.40 \pm 0.3$ s. These results compare favorably with those observed under optimum detuning and laser powers $\langle T_{on} \rangle = 1.6 \pm 0.1$ s and $\langle T_{off} \rangle = 0.78 \pm 0.07$ s. We note that the observed excitation rate into and out of the metastable level (1 s^{-1}) is much more rapid than the natural decay rate (0.03 s^{-1}) showing that we have in our system sufficient spectral brightness to drive the *D-D* transition well into the saturation.

In summary, this investigation has demonstrated the excitation and observation of an electric-dipole-forbidden, midinfrared transition in a single trapped ion. The absolute transition frequency has been determined and represents a significant improvement in accuracy over the previous known value. Calculations of the expected quantum jump periods due to such excitation are in reasonable agreement with the experimentally observed values. Experiments are now underway to use an optically pumped NH_3 laser as the midinfrared probe laser. Such NH_3 lasers have been previously shown to provide linewidth in the subkilohertz regime [17], which should allow a significant increase in resolution.

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