

rf Spectroscopy with a Single Ba⁺ Ion

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We have employed the method of shelving to measure Zeeman resonances of a single trapped Ba⁺ ion. We use optical pumping to place the ion in a selected magnetic sublevel of either the $6S_{1/2}$ ground state or the $5D_{3/2}$ metastable state. The ion is exposed to an rf field, and a probing/shelving mechanism detects whether spin-flip transitions have taken place. We have observed rf transitions with linewidths of 15 Hz, limited by magnetic-field noise. We have also observed the shift in the Zeeman frequency when the ion is illuminated by off-resonant light. A simultaneous measurement of such light shifts in two atomic states of Ba⁺ will permit a precise determination of the ratio of transition matrix elements.

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Introduction.—After the pioneering experiments on single atomic ions trapped in vacuum [1,2], single ions have been used for accurate atomic clocks [3,4], precise measurements of atomic masses [5,6] and of metastable atomic lifetimes [7], and studies of entangled states and quantum computing [8]. Thus far, studies involving the spin states of single ions have relied on large magnetic fields to optically resolve the Zeeman splitting [2].

In this Letter we report on a new technique for detecting the spin direction of a single ion at low (or zero) magnetic field. We have used this technique to observe rf Zeeman resonances in the $6S_{1/2}$ ground state and the $5D_{3/2}$ metastable state of Ba⁺ with linewidths down to 15 Hz, limited by magnetic-field fluctuations in the current apparatus.

Our technique is based on observing the fluorescence emitted by a single ion when it is continuously excited on an allowed optical transition. If the ion is placed in a metastable electronic level outside this absorption/emission cycle, the fluorescence stops, and the ion is said to be *shelved* in the metastable level [9–12]. We use a spin-sensitive extension to the method of shelving. Briefly, we prepare the ion in a spin state by optical pumping, then drive a spin-flip transition, and finally detect the spin state by placing the ion in a shelved state with a spin-dependent probability.

As an initial application of this technique we have measured the ac Stark shift, or light shift, of the $5D_{3/2}$ Zeeman resonance frequency that occurs when the ion is illuminated by an off-resonant laser beam. This measurement opens up the precise study of transition matrix elements in Ba⁺ by comparing the light shifts in two states (viz., $6S_{1/2}$ and $5D_{3/2}$) caused by the same light beam. Uncertainties in the intensity and polarization cancel in the ratio, enabling a precise comparison with atomic theory [13]. Our work also is a step toward a possible measurement of atomic parity nonconservation in Ba⁺; a similar light shift technique is the basis for such a measurement [14].

Apparatus.—Figure 1 shows a diagram of the experimental apparatus. A single ion is confined in a Paul-Straubel trap [15], consisting of a stainless-steel ring (1 mm in diameter) supported by thin leads of the same mate-

rial. The trap is connected to an rf oscillator that delivers a 10-MHz, 700-V (peak to peak) voltage to the ring. This produces a stably trapped ion with a secular frequency [15] of about 1 MHz. The ion is cooled using the $6S_{1/2} \rightarrow 6P_{1/2}$ transition at 493 nm (see Fig. 2). A photomultiplier collects about 1000 photons/sec of scattered light from the ion, thereby detecting its presence. From the $6P_{1/2}$ state, the ion can decay into the metastable $5D_{3/2}$ state, and a cleanup beam at 650 nm is necessary to drive the ion back into the absorption/emission cycle.

The cooling light is produced with a 20-mW 986-nm external-cavity diode laser that is frequency doubled within a KNbO₃ crystal in an enhancement cavity to produce ~ 0.5 – 1 mW of 493-nm light. To keep the laser frequency fixed to about 20 MHz below the $6S_{1/2} \rightarrow 6P_{1/2}$ transition, we lock it to the side of the optogalvanic resonance of a barium discharge lamp [16]. Since this resonance is Doppler broadened to about 1 GHz, the laser can be more conveniently locked when first shifted by a few hundred MHz. Therefore part of the beam is sent through two consecutive acousto-optical modulators (AOM) to the barium

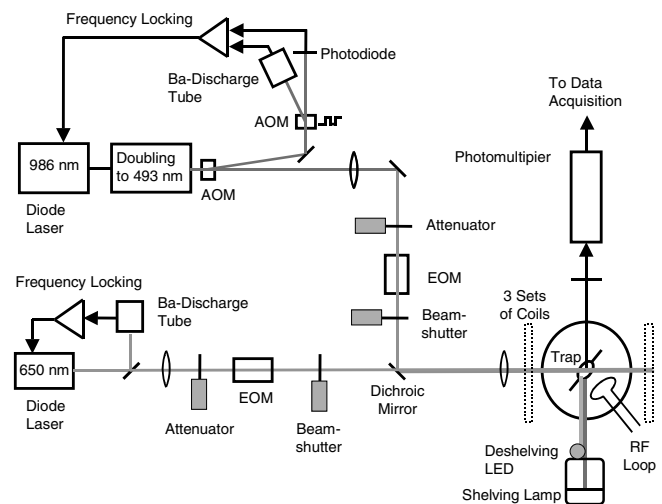


FIG. 1. Schematic of the apparatus used to perform rf spectroscopy with a single Ba⁺ ion.

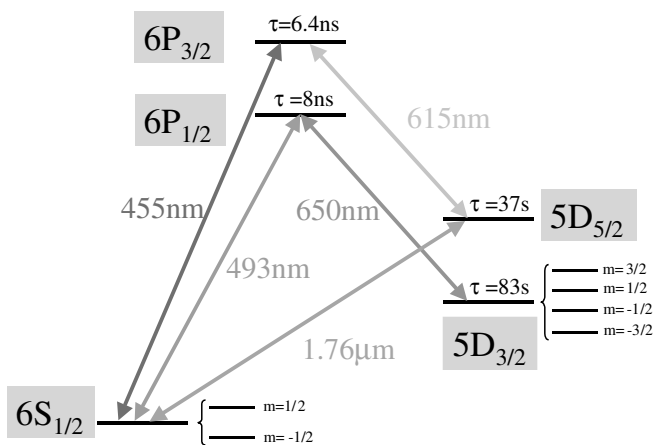


FIG. 2. Level diagram of Ba⁺ with transition wavelengths, natural lifetimes, and the relevant Zeeman structure. Level energies are not to scale.

discharge lamp, where it produces the optogalvanic signal. The second AOM is chopped to allow for phase-sensitive detection of this signal. The portion of the beam undeflected by the second AOM is detected by a photodiode and serves as a reference to eliminate laser-power dependence in this locking scheme.

The cleanup laser is a 1-mW external-cavity diode laser that is locked to a second barium discharge lamp. In this case, the laser is frequency modulated at 1 kHz with an amplitude of about 150 MHz, and the optogalvanic signal is phase-sensitively detected. With this large frequency modulation, all coherent effects involving the two lasers are washed out.

The beams are combined with a dichroic mirror and focused onto the ion after passing through mechanical shutters, switchable neutral density filters, and electro-optical modulators (EOM) for polarization control. Mechanical shutters are necessary to provide the high cutoff ratios needed (>100 dB) to prevent lifetime shortening of the state whose spin structure is under study.

Zeeman transitions are induced by an rf magnetic field produced by a wire loop with a diameter of 10 mm mounted 5 mm away from the trap. The system is housed in an ultrahigh vacuum enclosure with background pressures of 5×10^{-11} torr. Three coils provide the static magnetic field at the site of the ion. The chamber is surrounded by two layers of Mumetal to provide magnetic shielding.

The shelving transition at 455 nm is driven with light selected by an interference filter from a barium discharge tube. The ion can be “deshelved” (pumped out of the shelved state) using a bright orange light-emitting diode (LED) that covers the $5D_{5/2} \rightarrow 6P_{3/2}$ transition.

Method.—rf spectroscopy of Zeeman levels in both the $6S_{1/2}$ and the $5D_{3/2}$ states is possible using these techniques. The procedure for the $6S_{1/2}$ state will be described first in some detail.

First we set the cooling beam to be σ^- polarized along **B** while leaving the cleanup beam coupled to all $5D_{3/2}$

sublevels. As a result, the $6S_{1/2,-1/2}$ state will be left uncoupled, and after a few absorption/emission cycles the ion will be stuck there. Evidence of this optical pumping is immediately available from the fluorescence signal, which drops to a fraction of its full value.

With the ion prepared in the $6S_{1/2,-1/2}$ state, we turn off the cleanup laser and reduce the intensity of the cooling laser to act as a weak probe beam, causing a few transitions per second. At the same time we apply an rf field with a frequency close to the Zeeman splitting of the two sublevels. If the rf field is on resonance, spin-flip transitions between the two substates take place, and the probe beam can excite the ion to the $6P_{1/2,+1/2}$ state from where it will eventually decay into the $5D_{3/2}$ state. If the rf field is not on resonance, the ion will remain in the ground state.

Next we turn off the rf field and the probe beam and instead apply 455-nm light. This light excites the ion, provided it is still in the ground state, to the $6P_{3/2}$ state, from where it will ultimately decay to $5D_{5/2}$, the shelved state.

Finally we can turn on full-intensity, linearly polarized cooling and cleanup beams and observe either the absence of fluorescence if the ion has been shelved, or the presence thereof if the ion is in $5D_{3/2}$. We can make this observation during the first 1% of the lifetime of $5D_{5/2}$ and thus detect a shelved ion with close to 100% probability.

By repeating this process of optical pumping, spin-flip transition, and detection, the shelving probability for a particular rf frequency is determined. This shelving probability is correlated with the transition probability due to the applied rf field. Then by varying the rf frequencies around the Zeeman resonance, the transition resonance can be mapped out.

The correlation is not perfect because of two factors. First, the circular polarization of the cooling laser is imperfect, and as a result the $6S_{1/2,-1/2}$ state is not completely uncoupled. Thus, the ion can end up in the $5D_{3/2}$ state even when the rf frequency is not on resonance. This effect is minimized by low intensities and short exposure times. On the other hand, the ion needs several excitations into the $6P_{1/2}$ state because in only 22% of all cases will the ion decay into the desired $5D_{3/2}$ state. Second, an ion in $6P_{3/2}$ decays to $5D_{5/2}$ in only 90% of all D -state decays; in 10% it decays to the $5D_{3/2}$ state, which leads to the opposite shelving result.

The method described above works similarly for rf spectroscopy in the $5D_{3/2}$ state. Here the cleanup laser assumes the role of the cooling laser in the above description. When using a σ^- polarized cleanup laser, the ion is prepared in either the $5D_{3/2,-1/2}$ or the $5D_{3/2,-3/2}$ state. Alternatively, π^0 light will pump the ion to the $5D_{3/2,\pm 3/2}$ states. In either case, a transition to the other two available spin states is then induced by a suitable rf field, and a weak circularly polarized cleanup beam acts to transfer the ion to the $6S_{1/2}$ state if the rf field is on resonance. Then the same method of shelving is used to ultimately determine whether rf transitions take place.

Data.—To demonstrate the techniques described above we used it to measure Zeeman resonance frequencies in the $6S_{1/2}$ and $5D_{3/2}$ states. The external magnetic field was set to 2.5 G. For the $5D_{3/2}$ state the shelving probability was determined for ten rf frequencies around the estimated resonance in 30-Hz intervals. The upper graph of Fig. 3 shows the resulting distribution of shelving probabilities along with their statistical errors $\delta p_i = \sqrt{p_i(1-p_i)/N}$ for the $N = 600$ trials we performed for each rf frequency.

The measurement time for this scan was about 6 h. During this time the magnetic field at the site of the ion varies due to imperfectly shielded external magnetic fluctuations, domain changes of magnetic material, and current noise in the field-producing coils. The fluctuations likely follow a Gaussian distribution, which is reflected in the Gaussian fit to the data. This magnetic noise broadens the resonance to 50 Hz in the above example; linewidths down to 15 Hz have been observed with shorter integration times. These broadening effects overwhelm the 0.3-Hz linewidth of the transition caused by the finite interrogation time of the rf magnetic field (0.5 sec). However, with sufficient magnetic-field stabilization, it is expected that this very small linewidth will be realized. The ultimate limit on the

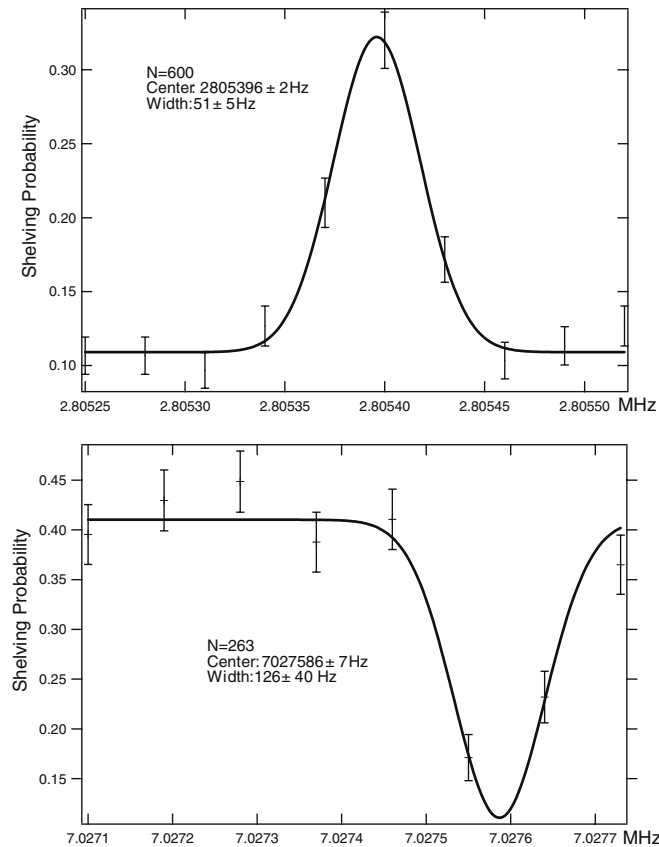


FIG. 3. The upper graph shows the shelving probability as a function of the applied rf frequency when probing the Zeeman splitting of the $5D_{3/2}$ state. Also shown is the statistical error associated with 600 trials for each data point, and a Gaussian fit to the data. The lower graph shows the same information for the $6S_{1/2}$ state and 263 trials.

linewidth would be given by the natural lifetime of the atomic state, ≈ 70 sec for $5D_{3/2}$.

For the $6S_{1/2}$ state the magnetic field was approximately the same, and the resonance was mapped out with eight shelving probabilities around the resonance in 75-Hz intervals. Here, as expected, one observes a minimum shelving probability on resonance, as shown in the lower graph of Fig. 3.

The method used to obtain rf resonances can be modified so that the rf field and the probe beam are turned on sequentially rather than simultaneously. This has the advantage that possible light shifts from the probe laser as well as small systematic effects from the magnetic field of the beam shutters are eliminated; on the other hand it yields a smaller change in shelving probability for the resonance condition. Data have also been taken using this alternative method.

Light shift.—With the tool of rf spectroscopy in place, ac Stark shifts of Zeeman sublevels, as defined in Eq. (1) below, can now be measured. In a first attempt to observe the effect, off-resonant π^0 light at 653 nm (close to the $6P_{1/2} \rightarrow 5D_{3/2}$ transition) was used. The light was produced by a dye laser with an 80-mW output and focused onto the ion. The off-resonant interaction with the $6P_{1/2}$ state shifts the $5D_{3/2, \pm 1/2}$ sublevels downwards, while the other two sublevels are not coupled and therefore are not affected.

The ion was first prepared in the $5D_{3/2, \pm 3/2}$ state. Without light shifts, one would expect a single resonance peak corresponding to the Zeeman splitting from both $m = \pm 3/2 \rightarrow m = \pm 1/2$ transitions. With the off-resonant laser turned on, the $m = +3/2 \rightarrow m = +1/2$ transition frequency is increased by the magnitude of the ac Stark shift while the $m = -3/2 \rightarrow m = -1/2$ frequency is reduced by the same amount. As a result, there should be two peaks symmetrically displaced from the original resonance peak. Furthermore, the two peaks should have half the height of the original peak, since for any given trial only one of the two transitions can take place.

The measurement sequence consisted of two alternating spin-resonance trials where the shelving probability is measured for a number of applied rf frequencies. The trials were identical in state preparation, interaction, and probing and differed only in the application of the off-resonant light during alternate trials. The result shown in Fig. 4 was obtained with a different, magnetically unshielded apparatus, which explains the 6-kHz linewidth of the central peak. The light-shifted peaks are even broader because of the intensity fluctuations of the unstabilized dye laser over the course of several hours.

A light field \mathbf{E} will cause ac Stark shifts in all atomic states given by

$$\Delta_{k,m} = \frac{e^2}{4\hbar} \sum_{k',m',\pm\omega} \frac{|\langle k,m|\vec{\mathbf{E}} \cdot \vec{\mathbf{r}}|k',m'\rangle|^2}{W_{k'} - W_k \pm \hbar\omega}, \quad (1)$$

where k stands for the quantum numbers nlj , W_k is the

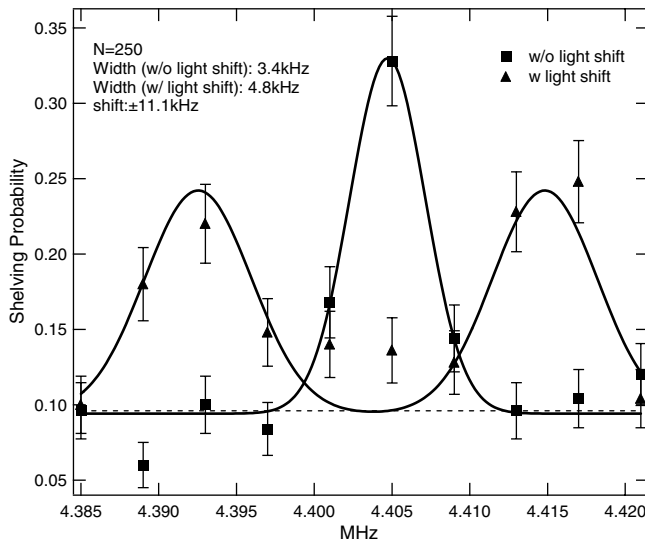


FIG. 4. Shelving probability as a function of the applied rf frequency when probing the Zeeman splitting of the $5D_{3/2}$ state, with and without the presence of off-resonant light.

energy of each level, and ω is the frequency of the light. The observed shift of 10 kHz is consistent with the off-resonant light focused to a $150\text{-}\mu\text{m}$ spot size.

If we measure the shifts in the $6S_{1/2}$ and $5D_{3/2}$ states using the same off-resonant light beam and compare the light shifts for the same value of m , then the field strength $|\mathbf{E}|$ as well as the angular factors cancel. The resulting ratio of reduced matrix elements is independent of the characteristics of the off-resonant beam, except for its frequency, which is easily determined. Misalignments between the applied magnetic field and the propagation direction of the off-resonant light should be negligible.

In particular, measuring the ratio of light shifts of the $5D_{3/2,\pm 1/2}$ and $6S_{1/2,\pm 1/2}$ states would determine the ratio of the matrix elements $\langle 6S_{1/2} || r || 6P \rangle$ and $\langle 5D_{3/2} || r || 6P \rangle$ which dominate the shifts for visible light. This would provide a precise test of atomic calculations on alkali-like ions.

The ratio could be measured at various wavelengths to enhance sensitivity to particular matrix elements. For instance, a wavelength close to 650 nm would be more sensitive to $\langle 5D_{3/2} || r || 6P_{1/2} \rangle$ than to $\langle 5D_{3/2} || r || 6P_{3/2} \rangle$. The precision of this ratio measurement is then limited by the relatively smaller shift in the $6S_{1/2}$ state due to larger mistuning. Nevertheless, 100 mW of power focused to a $50\text{-}\mu\text{m}$ spot size would yield a light shift of 2.5 kHz in the $6S_{1/2}$ state, which can be measured to better than 1% with the current apparatus and an intensity-stabilized dye laser.

In principle, the size of the achievable light shift in a state $|k, m\rangle$ would be limited by the degree to which

off-resonant light causes real transitions into other states. In general, the induced loss rate is

$$\Gamma_{k,m}^{\text{loss}} = \frac{e^2}{4\hbar} \sum_{k',m',\pm\omega} \Gamma_{k',m'} \frac{|\langle k, m | \vec{\mathbf{E}} \cdot \vec{\mathbf{r}} | k', m' \rangle|^2}{(W_{k'} - W_k \pm \hbar\omega)^2}, \quad (2)$$

where $\Gamma_{k',m'}$ is the spontaneous transition rate out of $|k', m'\rangle$. However, because $\Gamma_{k,m}^{\text{loss}}$ falls off with mistuning more rapidly than $\Delta_{k,m}$, real transitions will rarely be a limiting factor in practice.

Being able to measure light shifts in a single ion is also a proof of principle for a proposed experiment to measure parity nonconservation in a single barium ion [14]. The observable in such an experiment is a light shift in the Zeeman sublevels of the $6S_{1/2}$ state that is induced by coupling to the $5D_{3/2}$ state and depends on the amount of parity nonconservation.

In conclusion, we have demonstrated the measurement of Zeeman energy splittings in a single barium ion using a new, spin-sensitive shelving technique. We have observed Zeeman resonances of 15-Hz width limited by magnetic-field noise in our current apparatus. Using this technique, precise measurements of light shifts in Zeeman sublevels will be possible, and we have shown an initial observation of such shifts in the $5D_{3/2}$ state. Measuring light shifts leads to a ratio of matrix elements that can be used to improve atomic theory and is an essential step toward a parity nonconservation measurement in Ba^+ .

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