Precision Measurement of Light Shifts in a Single Trapped Ba^+ Ion

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Using a single trapped barium ion we have developed an rf spectroscopy technique to measure the ratio of the off-resonant vector ac Stark effect (or light shift) in the $6S_{1/2}$ and $5D_{5/2}$ states to 0.1% precision. We find $R = \Delta_S/\Delta_D = -11.494(13)$ at 514.531 nm where $\Delta_{S,D}$ are the light shifts of the $m = \pm 1/2$ splittings due to circularly polarized light. Comparison of this result with an *ab initio* calculation of R would yield a new test of atomic theory and improve the understanding of atomic structure needed to interpret a proposed atomic parity violation experiment. By appropriately choosing an off-resonant light shift wavelength one can emphasize the contribution of one or a few dipole matrix elements and precisely determine their values.

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Experimental tests of atomic theory often involve the measurement of atomic state lifetimes, oscillator strengths, polarizabilities [1], and other properties which depend directly on atomic dipole matrix elements. Absolute measurements of these quantities can be difficult. Another approach [2] is to make high precision measurements of properties which can be directly calculated using modern atomic theory techniques and depend on ratios of atomic matrix elements. Here we report a 0.1% measurement of the ratio R of the ac Stark effect [or light shift (LS)] in the $6S_{1/2}$ and $5D_{3/2}$ states of a singly ionized barium ion, isoelectronic to the well-studied alkali atom Cs. Comparison of this result with an *ab initio* calculation of R would yield a new test of atomic theory.

Since R is expressible as ratios of matrix elements (shown below), this measurement also establishes a sum rule relating the barium matrix elements known to ~1% or better (i.e., $\langle 6S_{1/2}||r||6P_{1/2,3/2}\rangle$) to matrix elements with about 10 times worse experimental precision [3–6]. One of the latter, $\langle 5D_{3/2}||r||6P_{1/2}\rangle$, is crucial for a proposed atomic parity violation experiment [7–9]. In our scheme, a particular matrix element can be studied by choosing a light shift wavelength close to the corresponding dipole transition so that other contributions remain small. The technique is generalizable to other trapped ion species with metastable states.

To second order in perturbation theory an off-resonant light beam of intensity I, frequency ω, and polarization $\hat{\boldsymbol{\epsilon}}$ causes a frequency shift $\Delta_{k,m}$ in atomic level $|k, m\rangle$ of

$$\Delta_{k,m} = 2\pi\alpha I \sum_{k',m',\pm\omega} \frac{|\langle k, m|\hat{\boldsymbol{\epsilon}} \cdot \mathbf{r}|k', m'\rangle|^2}{W_{k'} - W_k \pm \hbar\omega}, \qquad (1)$$

where α is the fine-structure constant, k stands for atomic quantum numbers nlj, and W_k is the unperturbed energy of level k. The sums extend over continuum states. Though it is difficult to measure the light intensity at the site of a trapped ion, I cancels in a measured ratio of light shifts. The light shift due to circularly polarized off-resonant light acts effectively as a magnetic field pointing along the direction of light propagation [10,11]. In our case, a laboratory magnetic field ($B_0 \sim 2.5$ G) is aligned with the light shift beam (see Fig. 1). Therefore, if the same magnetic sublevels in two atomic states are used, there is only second order dependence of the ratio on polarization errors and misalignment of the light shift beam with respect to the magnetic field.

By employing an rf spectroscopy technique [12] that features the metastable $5D_{5/2}$ "shelving" state as a means of detecting spin flips, we make measurements of the $m = -1/2 \leftrightarrow +1/2$ splittings ω_S and ω_D in the $6S_{1/2}$ and $5D_{3/2}$ states of a single $^{138}Ba^+$ ion without any light shift, which we interleave with measurements of the splittings, ω_S^{LS} and ω_D^{LS}, made with application of the light shift beam. The measured ratio of light shifts R is then

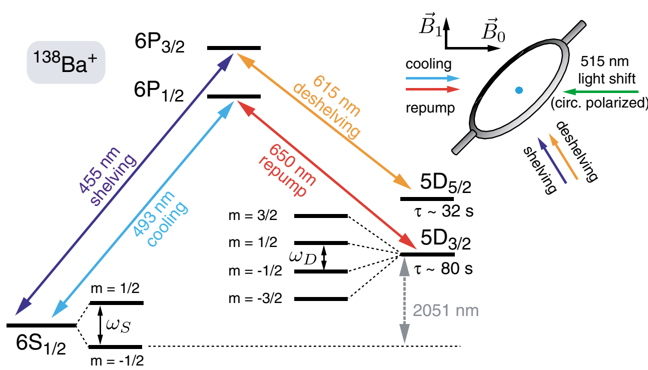

FIG. 1 (color online). This energy level diagram of $^{138}Ba^+$ shows the optical transitions and magnetic spin resonances ω_S and ω_D of interest in this work as well as the electric-dipole forbidden 2051 nm transition important to a proposed atomic parity violation experiment. Note the several second lifetimes [19,20] of the metastable $5D_{3/2}$ and $5D_{5/2}$ states. The inset cartoon of a Paul-Straubel [13] trap and ion shows the alignment of the lasers and rf spin-flip field $\vec{B}_1$ with respect to the static magnetic field $\vec{B}_0$. See text for description of the ω_S and ω_D measurement technique using electron shelving.

$$R \equiv \frac{\Delta_S}{\Delta_D} = \frac{\omega_S^{\mathrm{LS}} - \omega_S}{\omega_D^{\mathrm{LS}} - \omega_D}, \tag{2}$$

which is expressed using Eq. (1) as

$$R = \frac{\sum_{k',\pm\omega} f(6S_{1/2}, k') \frac{|\langle 6S_{1/2}||r||k'\rangle|^2}{(W_{k'} - W_{6S_{1/2}} \pm \hbar\omega)}}{\sum_{k',\pm\omega} f(5D_{3/2}, k') \frac{|\langle 5D_{3/2}||r||k'\rangle|^2}{(W_{k'} - W_{5D_{3/2}} \pm \hbar\omega)}} \tag{3}$$

Here $\langle k||r||k'\rangle$ are reduced matrix elements and $f(k, k')$ are angular factors obtained from Clebsch-Gordan coefficients. A precise atomic calculation would have to include effects of the core and higher-order terms in order to achieve an accuracy better than 1%.

Because the light shift of level k due to level k' depends inversely on the detuning between the applied light frequency and the splitting between k and k', choosing a light shift frequency ω close to an allowed dipole resonance will make one or a few terms of the sum in Eq. (3) dominate. For instance, the contributions of the $\langle 6S_{1/2}||r||6P_{1/2,3/2}\rangle$ and $\langle 5D_{3/2}||r||6P_{1/2,3/2}\rangle$ terms dominate the shifts Δ_S and Δ_D for most visible wavelengths as shown in Fig. 2.

We trap a single $^{138}\mathrm{Ba}^+$ ion by applying several hundred volts of 10 MHz rf to a twisted wire Paul-Straubel [13] trap. We cool the ion to the Lamb-Dicke regime on the 493 nm $6S_{1/2} \leftrightarrow 6P_{1/2}$ transition using a frequency doubled 986 nm diode laser while also applying a 650 nm beam which prevents pumping to the long-lived $5D_{3/2}$ state. This repump laser is frequency modulated at 2 kHz to a depth of $\sim$100 MHz to frequency lock it optogalvanically to a barium hollow cathode lamp and to eliminate coherent effects which otherwise appear on the cooling transition [14]. A static magnetic field aligned along both lasers splits the magnetic sublevels by a few MHz and eliminates dark states [15]. High intensity filtered light-emitting diodes perform shelving to and deshelving from the $5D_{5/2}$ state with rates $\gtrsim$ 10 Hz.

Because earlier reports [9,12] gave details of the rf spectroscopy measurement in the $6S_{1/2}$ state and since an important systematic effect arises from the $5D_{3/2}$ resonance line shape, we describe the measurement in the $5D_{3/2}$ state in detail here. The red repumping beam is aligned with the magnetic field. Making it left circular polarized (σ^-) with an electro-optic modulator causes optical pumping into a state in the lower manifold ($m = -1/2, -3/2$) of $5D_{3/2}$. With the cooling and repumping beams turned off, a properly timed rf pulse, resonant with the $m = -1/2 \leftrightarrow +1/2$ splitting, moves the ion to the upper ($m = +1/2, +3/2$) manifold with moderate probability. We must now readout the occurrence of this rf spin flip. Applying a pulse of σ^- repumping light to cause a decay to $6S_{1/2}$ followed by a pulse of shelving (455 nm, $6S_{1/2} \rightarrow 6P_{3/2}$), light transfers an ion in the upper $5D_{3/2}$ manifold to the $5D_{5/2}$ shelving state while leaving the lower manifold population untouched. Now detection of 493 nm fluorescence upon reapplication of the cooling and repumping beams is correlated to whether the ion is in the $5D_{5/2}$ shelved state and, hence, whether the rf pulse was on resonance. By repeating the measurement to gather statistics at several spin-flip radio frequencies, one obtains resonance line shapes as shown in Fig. 3.

The size and shape of the coherent features (e.g., sidebands) of the line depend primarily on the initial condition of the ion; that is, with what probability a it is prepared via optical pumping into the $m = -1/2$ state instead of $m = -3/2$. In principle $a \leq 0.8$ because of differential pumping rates to $6P_{1/2}$; lower values will result from laser/

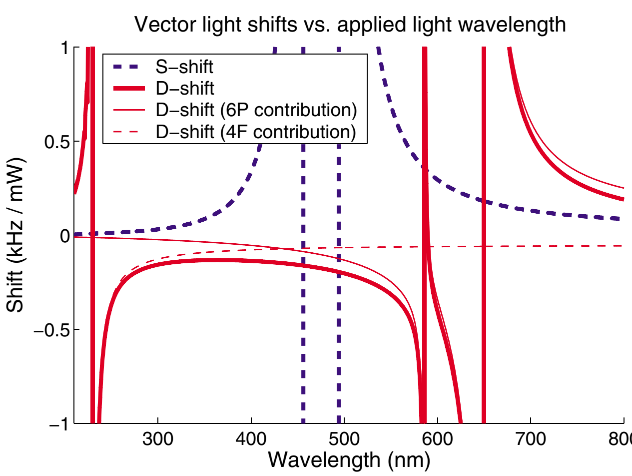

FIG. 2 (color online). Estimated light shifts of the $6S_{1/2}$ (thick dashed) and $5D_{3/2}$ (thick solid) $m = -1/2 \leftrightarrow 1/2$ Zeeman resonances as a function of applied light shift wavelength assuming a 20 μm spot. Note the divergences near atomic resonances. Separately plotted are the $\langle 5D_{3/2}||r||6P_{1/2,3/2}\rangle$ contributions to the shift in the $5D_{3/2}$ state (thin solid) which dominates in the visible spectrum and the $\langle 5D_{3/2}||r||4F_{5/2}\rangle$ contribution (thin dashed). The calculation includes electric-dipole coupling with the $6P_{1/2}$, $6P_{3/2}$, $7P_{1/2}$, $7P_{3/2}$ [3–6], and $4F_{5/2}$ [21] states.

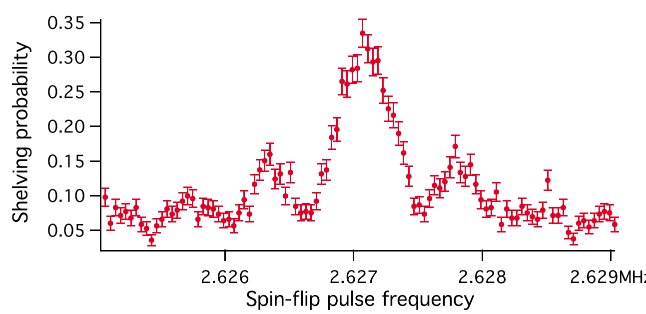

FIG. 3 (color online). Example of a $5D_{3/2}$ resonance scan. The probability that the ion is shelved in the metastable $5D_{5/2}$ state is correlated to whether a rf π pulse was on resonance with the $5D_{3/2}$ $m = -1/2 \leftrightarrow +1/2$ splitting. Here, homogenous broadening dominates the linewidth and coherent features unique to this four state system are visible.

magnetic field misalignment and poor red laser polarization quality. By fitting resonances using the optical Bloch equations, we have determined that $0.5 \leq a \leq 0.78$ are typically observed values.

We chose the convenient 514.531 nm (air wavelength) line of an argon-ion laser as the off-resonant light shifting beam. Approximately 1 W of laser light travels through an acousto-optic modulator (AOM) used for intensity stabilization and couples into a single-mode polarization-maintaining fiber after which are circular-polarizing optics and the intensity stabilization photodiode. The use of the AOM, fiber, polarization optics, and sensor in this order translates laser pointing and polarization noise into intensity noise which is nulled by the AOM servo to much better than 0.1% over a dc-10 kHz bandwidth.

A single data run consists of four interleaved measurements: $6S_{1/2}$ and $5D_{3/2}$ Zeeman resonances with and without the application of a light shift beam. The four resonances are typically limited by broadening due to small fluctuations in the magnetic field or light shift beam intensity, pointing, and polarization. They are therefore fit to Gaussians to determine the resonance centers and uncertainties. We assign statistical errors to each light shift ratio measurement by propagating these uncertainties through Eq. (2). About two hours is required to reach a statistical precision of 1% per measurement.

Systematic effects common to shifted and unshifted resonance measurements completely cancel. Many other systematic effects are eliminated because we interleave resonance measurements in both the $6S_{1/2}$ and $5D_{3/2}$ states and are only concerned with the ratio of their shifts. Effects that remain are discussed below; they fall into the categories of line shape effects, alignment and polarization errors, ion displacements, magnetic field variations, coupling to the trapping rf field, and spectral purity of the light shift laser.

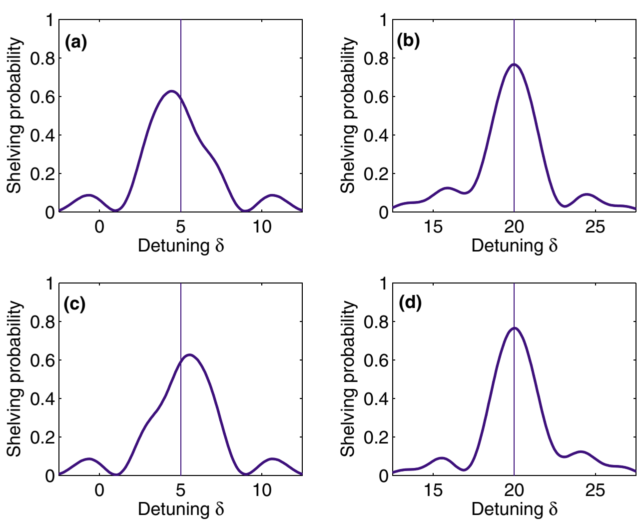

FIG. 4 (color online). The most important systematic effect arises from distortions in the $5D_{3/2}$ resonance line shape caused by the tensor piece of the light shift. The vertical lines indicate the simulated light shift values Δ_D in units of Ω_D. Shown in (a) and (c) are simulated line shapes for $\Delta_D/\Omega_D = 5$ while (b) and (d) show $\Delta_D/\Omega_D = 20$. The initial conditions for (a) and (b) were $a = 0.78$ into the lower spin state manifold of $5D_{3/2}$ while (c) and (d) were prepared in the upper spin state manifold.

The most significant systematic effect is caused by the line shape of the light shifted $5D_{3/2}$ resonance. $J > 1/2$ in this state, so a tensor component to the light shift makes the outer splittings $m = -3/2 \leftrightarrow -1/2$ and $m = +1/2 \leftrightarrow +3/2$ different from the splitting of interest $m = -1/2 \leftrightarrow +1/2$. This has been observed to cause distortions in the $5D_{3/2}$ resonance line shapes when the light shifts Δ_D are comparable to the spin-flip Rabi oscillation frequency Ω_D. The distortion near the center of the line decreases markedly with increasing Δ_D/Ω_D as illustrated in Fig. 4. The distortion is worse at higher a values and at light shift wavelengths with significant $\langle 5D_{3/2}||r||4F_{5/2}\rangle$ contribution. The sign of the line-center error changes with reversing the light shift beam polarization relative to the magnetic field, or with initially preparing the ion in the opposite $5D_{3/2}$ manifold. We confirmed these reversals, which explain the increased scatter seen in data with low Δ_D/Ω_D (see Fig. 5) since the sign of the effect was not controlled. We minimize this systematic by only including data in the final analysis for which $\Delta_D \gg \Omega_D$. By choosing the cutoff $\Delta_D/\Omega_D > 20$, our models suggest the average remaining effect is less than 0.05%.

A misalignment α of the light shift beam from the magnetic field decreases observed light shifts and alters the measured R. Throughout the experiment the light shifts Δ_S and Δ_D are kept small with respect to the splittings ω_S and ω_D so that these errors are quadratic in α and multiplied by factors Δ_S/ω_S or Δ_D/ω_D. For a typical data run, $\Delta_S \sim 100$ kHz at $\omega_S \sim 6.8$ MHz, we estimate an $\alpha = 10°$

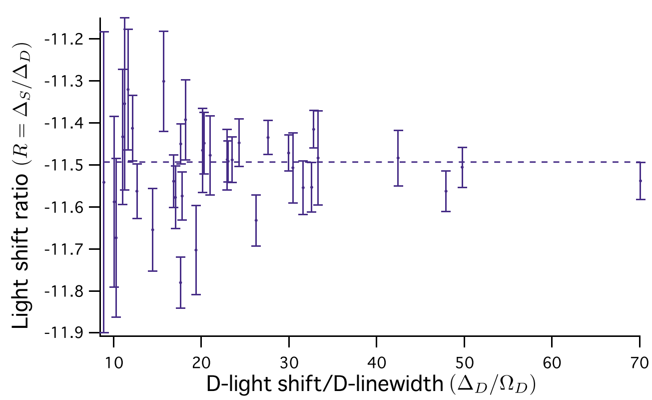

FIG. 5 (color online). Observed light shift ratios R defined in Eq. (2) are shown against the most significant systematic parameter: the ratio of measured light shift in the $5D_{3/2}$ state and the spin-flip Rabi-frequency Δ_D/Ω_D. For low values of this ratio, the tensor light shift distorts the D-resonance line shape leading to systematic shifts of either sign and variable size. If only data with $\Delta_D/\Omega_D > 20$ are considered (putting any potential systematic below 0.1% in our models), the straight line fit $R = -11.494(13)$ explains the data with $\chi_r = 1.0$.

misalignment causes $<0.1\%$ change in R while independently we have constrained $\alpha < 5°$. Errors in the light shift laser polarization couple to misalignments to produce systematic shifts in the $5D_{3/2}$ splitting due to the tensor light shift. These are kept well below 0.1% by maintaining a measured polarization quality of $\sigma > 0.95$ in addition to the above misalignment bounds.

The pseudopotential due to the rf trap completely overpowers the expected dipole force experienced by an ion in a gradient of the light shift beam ($P < 100$ mW, with a ~ 20 μm spot). We have ruled out any effects which might displace the ion in a beam gradient by taking some data with the ion near the edge of the beam to set bounds and taking the majority of data with the ion centered in the beam spot to $\pm 5\%$. We detected no dependence of Δ_S on the Rabi spin-flip frequency Ω_S which would imply displacement of the ion caused by the spin-flip rf. Changes in the magnetic field at the site of the ion accompanying the application of the light shift laser, for instance due to the activation of a shutter, have been ruled out at the 1 Hz level by running the experiment with the light shift laser turned off and seeing no anomalous shift.

Capacitive pickup of the trap rf creates magnetic fields that shift (like the Bloch-Siegert effect) the $6S_{1/2}$ and $5D_{3/2}$ resonances up to a few kHz. This effect is calibrated for each run by comparing the measured ratio of g factors ω_S/ω_D against its well measured value [16]. Most of this effect is common to both shifted and unshifted resonances since $\Delta_S \ll \omega_S$ and $\Delta_D \ll \omega_D$ by design; we apply corrections, no higher than 0.06% in any single run, to account for the net effect on R.

Commercial wave meters confirmed no significant wavelength drift with occasional small changes in argon-ion laser tube pressure. Background argon-ion fluorescence coupled via the optical fiber to the ion was measured below 20 μW which can only account for sub-Hz light shifts.

All data are shown in Fig. 5. By cutting data with $\Delta_D/\Omega_D < 20$ to minimize systematics as discussed above we obtain the result $R = -11.494(13)$, where the quoted uncertainty includes a statistical contribution of $\sigma_{\text{stat}} = 0.008$ and total systematic uncertainty $\sigma_{\text{syst}} = 0.010$ added in quadrature. This agrees with a theoretical estimate of $R_{\text{est}} = 11(1)$ obtained using published matrix elements and uncertainties (see Fig. 2 note). An *ab initio* calculation of R would provide an exacting test of atomic theory.

In conclusion, we have demonstrated that the measurement of off-resonant light shifts in single trapped ions via electron shelving is a viable technique for precisely testing atomic theory. Future plans include performing this measurement again at a different off-resonant wavelength in order to untangle $\langle 5D_{3/2}||r||6P_{1/2,3/2}\rangle$ and $\langle 5D_{3/2}||r||4F_{5/2}\rangle$ contributions in Eq. (3). The apparatus might also be used to measure the $5D_{3/2}$ electronic quadrupole moment via a strong dc potential applied to the trap ring [17]. Such quadrupole Stark shifts impact single ion frequency standards [18] and merit further study in a calculable system such as $^{138}Ba^+$.

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