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## Stark shift of a single barium ion and potential application to zero-point confinement in a rf trap

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(Received 20 April 1994)

Recently we have observed the Stark shift in an optical transition of a single trapped Ba<sup>+</sup> ion. To enhance the shift in the  $6S_{1/2}$ - $5D_{5/2}$  clock transition, the ion is displaced from the saddle point of the trapping potential. The Stark shift constants,  $\sigma_s$ , for the three pairs of Zeeman components are found to be the following: 6.1(7), 7.5(8), and 11.7(12) mHz/(V/cm)<sup>2</sup> for  $|M_S| = \frac{1}{2}$  to  $|M_D| = \frac{1}{2}$ ,  $\frac{3}{2}$ , and  $\frac{5}{2}$  transitions, respectively. A scheme is also proposed to use the Stark shift to identify the zero-point confinement state of a single trapped Ba<sup>+</sup> ion.

PACS number(s): 32.60. + i, 32.80. Pj, 35.80. + s

A single ion in a rf trap is well known to be ideally suited for high-resolution optical spectroscopy because of its considerable isolation and localization in a perturbationfree environment. Future atomic clocks with stability approaching one part in 10<sup>18</sup> have been proposed based on such single trapped ions [1,2]. Other fundamental physics experiments may also benefit from them [3]. One of the remaining factors limiting the ultimate stability and accuracy in these experiments is the second-order Stark shift due to the finite rf field that confines the ion [1]. This effect is very small for a single laser-cooled ion confined at the exact rf saddle point of the trap, i.e., when no additional dc fields exist. At the current stage of laser frequency stability, such Stark shifts cannot be directly detected. In this paper, we present a Stark shift measurement of the  $6S_{1/2}$ - $5D_{5/2}$  clock transition of a single Ba<sup>+</sup> in a rf trap. In our experiment, the electric field causing the Stark shift is the trapping rf field itself, greatly enhanced by displacing the ion from the center of the trap with an applied external dc field. Our experiment is a measurement of the second-order Stark effect in a single trapped Ba<sup>+</sup>. Experiments on other species of single ions can be done in the same fashion. In addition, we propose to use the Stark shift to identify the vibrational level that the ion occupies near the zero level.

The experiment was done using a single  $^{138}\text{Ba}^+$  confined in a Paul-Straubel trap [4]. The trap and the remainder of the system have previously been described in detail [4,5]. A brief description will be given here in connection with this experiment. The trap is made of a slightly elliptic wire loop with a 0.3-mm inside diameter. Three pairs of compensation plates are placed in the

planes of a cube with the trap at its center and the zplates parallel to the loop plane (the z axis coincides with the trap axis). When an  $\Omega/2\pi = 26$  MHz rf voltage is applied to the loop electrode, as described in Ref. [4], the center region of the trap has a quadrupole potential distribution,  $\Phi = A (x^2 + y^2 - 2z^2) \sin \Omega t$ . An ion in such a rapid oscillatory field moves on average as if in the pseudopotential  $\overline{\Phi} = \frac{1}{2}m(\omega_x^2 \overline{x}^2 + \omega_y^2 \overline{y}^2 + \omega_z^2 \overline{z}^2)$ , where the  $\omega$ 's are the secular frequencies and m is the mass of the ion. The motional amplitude of the ion is reduced (cooled) by the laser radiation at 493 and 650 nm, usually to a temperature of mK. At such a temperature, the orbit size  $z_0$ of the ion is very small, on the order of tens of nanometers, well within the Lamb-Dicke regime of the Ba<sup>+</sup> 1.76- $\mu$ m clock transition ( $z_0 < \lambda/2\pi$ ). Therefore, the first-order Doppler shift is completely suppressed and high-resolution spectroscopy can be performed. The fluorescence from the allowed  $6^{2}S_{1/2} - 6^{2}P_{1/2}$  transition at 493 nm serves as the ion detection signal. The  $6^{2}S_{1/2} - 5^{2}D_{5/2}$  clock transition at 1.76  $\mu$ m is driven by a frequency stabilized laser locked to a Zerodur cavity. The laser has a linewidth about 600 Hz and an average long-term drift rate of 4 Hz/s. The quantum shelving scheme is used to detect the weak clock transition [6]. The entire trap tube assembly is housed in a magnetic shield that reduces both 60-Hz laboratory fields and external dc fields. A pair of magnet coils is placed inside the magnetic shield to generate a homogeneous 5-G field along the trap z axis at the center region of the trap. This applied field is necessary to prevent optical pumping in the  $5^2 D_{3/2} - 6^2 P_{1/2}$  "clean-out" transition at 650 nm.

2738

The ion spectrum thus obtained typically has a linewidth of about 1 to 2 kHz, a combination of the laser linewidth and the residual 60-Hz "hum" magnetic fields. The laser frequency scan is accomplished by using a guided-wave electro-optic (EO) modulator.

Even for this highly confined ion at the center of the trap, a small 26-MHz field of strength  $4Az_0$  is present, which keeps the ion in place. This residual rf field causes the second-order Stark shift, which may limit the ultimate precision achievable with such trapped ions. The Stark shift is quadratic in the electric-field strength and can be expressed as  $\delta_s = \frac{1}{2}\sigma_s E_{\Omega}^2$ , where  $E_{\Omega}$  is the electric-field amplitude at the ion site and  $\sigma_s$  is the Stark shift constant. The  $\frac{1}{2}$  factor comes from the ac average. At the ion temperature T, it can be shown that the residual ac Stark shift is  $\delta_s = \sigma_s m \Omega^2 k_B T / e^2$ , where  $k_B$  is the Boltzmann constant and e is the ionic charge. The shift depends only on the temperature of the ion for a given trapping frequency  $\Omega$  regardless of the trap well depth. When there exists an additional dc electric field  $E_d$  (for example, in the z direction), the ion will be pushed away from the saddle point of the pseudopotential to a point  $z_c$ , where the pseudotrapping force balances the dc field. The averaged motion of the ion is still equivalent to that in a harmonic potential well. But the ion now sees a larger rf field,  $4Az_c$ . Hence a larger Stark shift results. It is easy to show that the amplitude ratio of the rf field to the dc fields is  $\sqrt{2}\Omega/\omega_z$ . This factor is on the order of 10 in our experiment, making the rf field still the primary cause of the shift. The total Stark shift is  $\delta_s = \sigma_s (\Omega/\omega_z)^2 E_d^2$  $+\sigma_s m \Omega^2 k_B T/e^2$ . Often, the first term is larger if an appreciable dc field is present.

In the absence of any dc fields, the estimated sub-hertz Stark shift of a single trapped and cooled Ba<sup>+</sup> cannot be detected with our 600-Hz clock laser. In our experiment, a relatively large external dc field is applied along the zaxis. The field is generated by a voltage applied to the pair of z-compensation plates. The dc force pushes the ion off the trap center far enough so that the Stark shift can be observed. One side effect associated with the large rf field at the ion position is the enhanced micromotion [4,7]. The excessively large micromotion reduces the ion fluorescence rate due to Doppler broadening. The ion can even be moved out of the tightly focused cooling laser (and its image moved out of the small fluorescence detection pinhole). Therefore, it was necessary to pulse the external voltage so that it is switched on only during the clock excitation cycle and off during the fluorescence detection cycle. No appreciable ion heating results when the ion is being moved back and forth between the center and a displaced point so long as the pulse rise time is not too short and no spurious spikes are present. We found a 0.5-ms pulse rise time convenient in our experiment. In the meantime, the external field is switched on every other clock transition cycle. In this way, one obtains the shifted and unshifted resonances in the same frequency sweep (Fig. 1), reducing the effect of potential frequency drifts between sweeps. In a typical sweep, the averaging time for each point is about 2 s with 50% duty cycle. Both the clock transition and the fluorescence detection



FIG. 1. A typical Stark shift spectrum of the  $Ba^+$  clock transition. The number of the transitions is plotted against the EO modulator frequency increment. The left peak is the Stark shifted resonance with 158 V applied to the z-compensation plates. The open dots are experimental data and the solid line is a fitted Gaussian curve with full width at half maximum (FWHM) of 1.6 kHz. The shift is 7.1 kHz towards higher frequency since the lower EO modulator sideband is used. The inset shows a narrower line of 600-Hz FWHM width obtainable in the most favorable experimental conditions.

cycles are about 10 ms. The frequency steps are 500 Hz. The Stark shift in Fig. 1 is for the  $M_S = +\frac{1}{2}$  to  $M_D = +\frac{1}{2}$ Zeeman transition with 158 V applied to the z compensation plates. The shift is 7.1 kHz towards higher frequency since the lower EO modulator sideband is used. The linewidths in the spectrum are about 1.6 kHz. Narrower 600-Hz linewidths can be obtained in the most favorable operating conditions as shown in the inset of Fig. 1. To further minimize the frequency drift when the sweep time between two resonances is long, the simultaneous frequency sweep technique is used. In this scheme, the scanning laser frequency is alternately shifted back and forth by an amount approximately equal to the Stark shift. Then the shifted and unshifted transitions come into resonances at the same time.

There is no direct way to determine the rf field strength  $E_{\Omega}$  that the ion sees in the trap. One could in principle estimate the field strength from the trapping parameters and the trap geometry. But the latter is difficult to measure. We rely on the fact that the oscillatory micromotion amplitude  $\zeta$  of the ion is determined by the rf field strength  $E_{\Omega}$ . Neglecting the weak damping (due to laser cooling), we have  $m \Omega^2 \zeta = e E_{\Omega}$ . The oscillatory motion of the ion in turn modulates the laser frequency with the modulation index  $\beta = 2\pi \zeta \cos \alpha / \lambda$ , where  $\alpha$  is the angle between the rf field and laser propagation direction and  $\boldsymbol{\lambda}$ is the transition wavelength. The modulation produces discrete micomotion sidebands. By measuring the distribution of the first four micromotion sidebands and comparing them with the frequency modulation expansion in Bessel functions, we were able to determine the modulation index  $\beta$  for a given applied voltage. Figure 2 shows the plot of the measured modulation index  $\beta$  and the corresponding rf field strength versus the applied voltage. The angle  $\alpha$  is estimated to be 45°. As shown in the plot,



FIG. 2. The measured micromotion modulation index versus applied dc voltage. The right axis gives the corresponding rf field strength assuming a  $45^{\circ}$  angle between the rf field and the laser propagation direction.

the rf field strength at the ion site is quite linear with the applied voltage within the entire experimental range. Unfortunately, we cannot determine the angle to better than a few degrees. Thus, an upper limit of 4% systematic error is assigned to the rf field strength measurement.

The Stark shift in the Ba<sup>+</sup>  $6S_{1/2}$ - $5D_{5/2}$  transition has a scalar part and a tensor part [8]. The tensor part results from the *M*-dependent shift of the  $D_{5/2}$  state. Since the shifts depend only on |M|, the symmetric pairs of the Zeeman lines from the center of the transition have the same shifts. The shifts of the different Zeeman pairs are well resolved. In the actual experiment, we measured the Stark shifts of three pairs out of the ten Zeeman lines,  $M = \pm \frac{1}{2}$  to  $\pm \frac{1}{2}$ ,  $M = \pm \frac{1}{2}$  to  $\pm \frac{3}{2}$ , and  $M = \pm \frac{1}{2}$  to  $\pm \frac{5}{2}$ . The measured values in each pair agree well within the measurement error. This indicates that non-Stark shifts such as Zeeman shifts due to the spatial inhomogeneity of the magnetic field and possible Bloch-Seigert shifts [9] are relatively small. We take the averaged Stark shifts of the Zeeman pairs as the measured shifts. Figure 3 shows the



FIG. 3. The Stark shifts of the three pairs of the Zeeman lines are plotted against the squares of the applied dc voltages. The error bars reflect the statistic fluctuations of the measurement for each Zeeman line.

averaged Stark shifts of the three pairs of the Zeeman transitions. The error bars in the figure merely indicate the size of the fluctuations in each Zeeman line measurement. The larger error in the outer Zeeman lines reflects the higher sensitivity to fluctuations of the magnetic field. Since the Stark shift is quadratic in the rf strength field, and therefore the applied voltage, the shifts are plotted against the applied voltage squared in Fig. 3. By fitting the data with straight lines through the origin, we obtain the following Stark shift constants  $\sigma_s$  for the three Zeeman transitions: 6.1(2)(5) mHz/(V/cm)<sup>2</sup> ( $|M_S| = \frac{1}{2}$ to  $|M_D| = \frac{1}{2}$ , 7.5(2)(6) mHz/(V/cm)<sup>2</sup> ( $|M_S| = \frac{1}{2}$  to  $|M_D| = \frac{3}{2}$ , and 11.7(3)(9) mHz/(V/cm)<sup>2</sup> ( $|M_S| = \frac{1}{2}$  to  $|M_D| = \frac{5}{2}$ ). The first errors are the fitting errors and the second errors are the estimated systematic errors. We also calculated the same Stark shift constants using the usual second-order perturbation method. The calculation only uses a few Ba<sup>+</sup> transition strengths published in the literature [10] and neglects all other state contributions. We find the corresponding theoretical  $\sigma_s$ 's to be 8.2, 10, and 14 mHz/ $(V/cm)^2$ , respectively. These values are about 30% higher than our measured ones. We attribute the discrepancies to the lack of accurate transition strengths for the  $5D_{5/2}$  state in the theoretical estimate and the systematic errors in our experiment.

The calculated ion temperature of single Ba<sup>+</sup> in the Doppler cooling limit is 1 mK. With  $\Omega/2\pi=26$  MHz and secular frequency  $\omega_z = 2.6$  MHz in our experiment, the orbit size  $z_0$  is 15 nm. According to the above Stark shift constants, the average shift of the clock transition will be 2 mHz,  $10^{-17}$  of the transition frequency. However, if there is a stray dc field present (due, for instance, to the patch effect of electrode surface potential), the Stark effect will be increased. A mere 5-mV potential difference across a 0.3-mm diameter trap will produce a Stark shift of 80 mHz, already much larger than the natural linewidth of the Ba<sup>+</sup> transition (5 mHz). Furthermore, the micromotion energy excited is equivalent to that of the vibrational level  $v \approx 500$ . Therefore, one cannot overlook the importance of reducing the stray dc fields [4].

It is perhaps interesting to compare the quadratic Stark shift with the quadrupole shift of the  $D_{5/2}$  state due to electric-field gradients, another potential factor limiting the frequency stability of trapped single ions. The inherent rf trapping field gradient causes a second-order ac quadrupole shift. When the magnetic field is perfectly aligned with the z axis of the quadrupole field and the latter has radial symmetry, it can be shown that the quadrupole shift has a minimum of much less than 1 mHz. For our experimental setup, where the alignment is not perfect and the trap has no radial degeneracy, this shift is estimated to be about a few tenths of a hertz due to the nonzero quadrupole interactions among the Zeeman multiplets of the D state. However, the quadrupole shift is totally absent in the clock transitions of the group III ions, which are well recognized as the better candidates for atomic clocks [1,11].

Ultimately, one would like to confine the ion in the ground vibrational state—the zero-point confinement [12,13]. The Stark effect we discussed above provides a

practical way to identify individual vibrational levels v in the trap via the associated Stark shift. The zero-point wave-packet spread is  $z_0^2 = \hbar/m\omega$ . Independent of trap geometry, the trapping field strength at  $z_0$  causes a Stark shift  $\delta_s = \sigma_s (\hbar m/e^2) \omega \Omega^2 = 0.34$  mHz. Increasing  $\omega_z$  and  $\Omega$  each by a factor of 4, as proposed in a zero-point confinement experiment [13], appears feasible, yielding  $\delta_s(v=0) \approx 22$  mHz and spacings of 44 mHz between v=0,1,2,3... components with linewidths approaching the 5-mHz natural width in the most favorable case. Choosing the v=0 component effectively realizes zeropoint confinement, as the line then has the frequency it

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has in the zero vibrational state.

In conclusion, we have measured the Stark shifts of single trapped  $Ba^+$  in a rf trap. The experimental values of the Stark shifts are found to be in fair agreement with the theoretical estimates within their accuracies. In addition, we have shown the feasibility of using the Stark shift to identify the zero-point confinement state of an ion in a rf trap.

Our research is supported by the National Science Foundation under the Single Atomic Particle at Rest in Free Space project.

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