Radiative Lifetime Measurement of the Ba^+ Metastable $D_{3/2}$ State

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The extremely long lifetimes of the metastable D states of the Ba^+ ion make it difficult to measure them accurately due to their high sensitivity to collisional effects. Thus a discrepancy exists between theories and experiments for the Ba^+ $D_{3/2}$ state. We have measured the $D_{3/2}$ lifetime under a nearly ideal condition, i.e., single trapped Ba^+ ions in ultrahigh vacuum. The obtained value of 79.8 ± 4.6 s resolves the existing discrepancy. The collisional quenching is found insignificant in our experiment, but there are indications of a non-negligible fine-structure mixing effect. [S0031-9007(97)03480-7]

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The natural radiative lifetimes of excited states in the Ba^+ ion have been measured by several groups recently [1–4]. Experimental data of such atomic properties are of great interest because they can be compared with *ab initio* theoretical calculations and thereby verify the validity of the theories. Ba^+ becomes especially relevant in this context since an atomic parity nonconservation experiment using the Ba^+ ion has been proposed [5]. A good theory is needed to determine the weak interaction coupling from the atomic measurements. A comparison between the predicted radiative lifetimes of the Ba^+ excited states from *ab initio* calculations [6] and the reported experimental data show that the theories and experiments agree within experimental uncertainties for three out of the first four excited states. There exists a large discrepancy for the metastable $D_{3/2}$ state. On the one hand, relativistic many-body calculations by Guet *et al.* [6] predict 83.7 s and multiconfiguration Hartree-Fock calculations by Glorieux *et al.* [7] yield 78.8 s. On the other hand, the most recent experimental result obtained by Werth *et al.* [4] is 48 ± 6 s. That experiment was done with Ba^+ ion clouds in an rf trap. In order to keep the ion storage time reasonably long in the experiment, a He buffer gas had to be used. The reported lifetime was then extrapolated to zero pressure. However, the pressure extrapolation does not always remove collisional effects. As the authors of Ref. [4] pointed out, collisions with residual gases may also induce fine-structure mixing of the $D_{3/2}$ state with the $D_{5/2}$ state. Since the $D_{5/2}$ state has a shorter lifetime of 32 s [2], their value could represent a lower limit for the $D_{3/2}$ state.

The extremely long natural lifetimes of the Ba^+ metastable states make experimental measurements very sensitive to effects of even infrequent collisions. Using ion clouds in a buffer gas is less than an ideal environment in which to carry out such measurements. Indeed, the lifetime of the $D_{5/2}$ state previously measured in clouds [8] is 47 s, while the measurements with single trapped ions [2] in ultrahigh vacuum (UHV) gives 32 s, with which the theoretical value agrees reasonably well. This suggests that the lifetime of the $D_{3/2}$ state should be measured in a similar environment as well to resolve the discrepancy. In this Letter we will present a measurement of the radiative lifetime of the metastable $D_{3/2}$ state in the Ba^+ ion using single trapped ions. The single ions are confined in an rf trap inside an UHV enclosure. We will show that the collisional effects are not negligible even in such a system.

To measure the $D_{3/2}$ state lifetime, we rely on the powerful technique of quantum shelving of single ions [2]. The relevant energy levels of Ba^+ are shown in Fig. 1. By turning on both the 493 nm (blue) and the 650 nm (red) lasers focused on the trapped single ion, many fluorescence photons are scattered per second when the ion is in the ground state. These photons can be easily detected with a photomultiplier. When the ion is excited to the long-lived metastable $D_{5/2}$ state ("shelved"), no photons are scattered at all. Therefore, the fluorescence signal off or on indicates whether the ion is in the $D_{5/2}$ state or the ground state. In fact, the $D_{5/2}$ lifetime was obtained by measuring the dwell time of the ion in the $D_{5/2}$ state, i.e, the fluorescence-off period [2]. Unlike the $D_{5/2}$ state, however, the $D_{3/2}$ state is connected to the $P_{1/2}$ state by an allowed electric dipole transition. An attempt to detect the fluorescence by turning on the two lasers would destroy the state of the freely decaying ion. So one needs to leave the ion in the $D_{3/2}$ state unperturbed

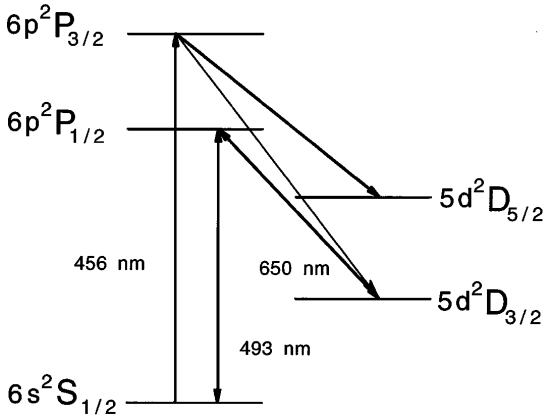

FIG. 1. The relevant Ba^+ ion energy level structure (not to scale).

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for a while and then determine whether a spontaneous decay has occurred.

Our experiment prepares the ion initially in the $D_{3/2}$ state. After a given wait time t which allows the ion to decay spontaneously without any perturbation, a shelving pulse is sent to excite the ion from the ground state to the $D_{5/2}$ state. The shelving can be achieved through the $P_{3/2}$ state by using a 456 nm discharge lamp as will be described later. If the ion has decayed to the ground state during the waiting period, then the shelving pulse excites the electron to $D_{5/2}$ with the probability P_{sh}. Since the probability of the ion having decayed from $D_{3/2}$ to the ground state after time t is $(1 - e^{-t/\tau})$, the probability of finding the ion in the shelving state is simply

$$P_{\mathrm{exp}} = P_{\mathrm{sh}}(1 - e^{-t/\tau}) .$$

By measuring the successful shelving rate P_{exp} at various wait times t, the $D_{3/2}$ state lifetime can be determined.

Single Ba^+ ions are trapped in a Paul-Straubel trap as described earlier [9]. The trap is housed in a vacuum enclosure continuously pumped by a 20 l triode ion pump. The background pressure is 4×10^{-11} Torr. The trapped single ions routinely stay in the trap for a couple of days without continuous laser cooling. Two laser beams of 493 and 650 nm are focused on the ion for both laser-cooling and ion signal detection. The fluorescence photons at 493 nm are collected and counted. Since the measurement process is expected to be slow, it is necessary to frequency-stabilize the lasers to prevent long-term drifts. With the 493 nm dye laser frequency locked to a Te_2 saturation line and the 650 nm diode laser frequency locked to a temperature-stabilized reference cavity, the system can run for hours without interruption.

The ion is first cooled and prepared in the $D_{3/2}$ state. The state preparation is accomplished by switching the blue laser off about 5 ms later than the red laser. This delay ensures that the electron gets pumped to the $D_{3/2}$ state if left in the ground state. Both the blue and red laser beams are switched on and off with mechanical choppers. The mechanical choppers, though rather crude, were necessary to achieve a light cutoff ratio of more than 100 db. This is easily understandable since the blue and red lasers drive the electron in and out of the ground state and the $D_{3/2}$ state at rates of the order of 10^8 s^{-1}. Any leakage of the laser light can induce transitions during the lifetime of the $D_{3/2}$ state, thus altering the measured lifetime. The same pulses of the visible lasers are used for both the initial state preparation and the state monitoring after shelving attempts at the end of wait periods. The pulse widths are set at 0.5 s.

To shelve the electron from the ground state to the $D_{5/2}$ state, we use a barium hollow cathode discharge lamp as used in the first demonstration of the "quantum jumps" [2]. A 456 nm interference filter is placed in front of the lamp. The 456 nm light excites the electron in the ground state to the $P_{3/2}$ state, which in turn spontaneously decays to the two D states or back to the ground state. If the lamp on-time is sufficiently long compared to the lamp excitation rate, the electron will end up in the D states with 100% probability. The chance of it being in $D_{5/2}$ is about 90%, determined by the branching ratio to the two D states. In the actual experiment, however, the lamp on time was set to be 0.5 s to reduce the timing uncertainty. The overall shelving probability P_{sh} from the ground state to $D_{5/2}$, which is a combination of the probabilities of the excitation from the ground state to $P_{3/2}$ and the spontaneous decay from $P_{3/2}$ to $D_{5/2}$, was measured separately by preparing the ion in the ground state instead of $D_{3/2}$. The measured shelving probability P_{sh} is 0.82 ± 0.01.

The overall experiment proceeded as follows. The ion is initially prepared in the $D_{3/2}$ state. Then the ion is left undisturbed for a time period t. At the end of the waiting period, the discharge lamp is switched on for 0.5 s. This is followed by the 0.5 s pulses of the blue and red lasers. If the fluorescence is detected, the shelving is unsuccessful and the above process repeats. Otherwise, we wait for the $D_{5/2}$ to decay spontaneously and record the dwell time rather than drive it down. This has the benefit of obtaining the lifetime of $D_{5/2}$ at the same time. The measured $D_{5/2}$ lifetime can be compared with the previous reported values.

Figure 2 shows the measured data points. The vertical axis is the measured probability P of the $D_{3/2}$ state having decayed after time t, $P = P_{\mathrm{exp}}/P_{\mathrm{sh}}$. The wait time t ranges from 4 to 150 s. Each data point represents at least 200 individual measurements. Therefore, the entire data collection took many hours. To effectively indicate the number of measurements, we put the expected error bar at each point. The errors at each wait time are calculated from the number of measurements assuming a binomial distribution; i.e., $\delta P_{\mathrm{exp}} = \sqrt{P_{\mathrm{exp}}(1 - P_{\mathrm{exp}})/N}$, where N is the total number of attempts. The total errors shown in Fig. 2 also include the P_{sh} measurement error which

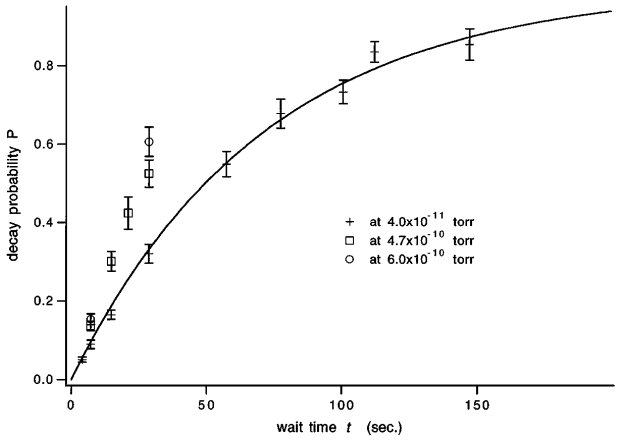

FIG. 2. Measured probabilities of single Ba^+ ions having decayed from the $D_{3/2}$ state after wait time t. The error bars are calculated from a binomial distribution (see text).

can be significant at longer t. To estimate the background pressure effects, we also took a few data points at higher pressures. By turning off the ion pump and keeping the ion gauge on, the background pressure was maintained around 5×10^{-10} Torr region. Slightly different steady-state pressures were reached on different days, enabling us to make measurements at two high pressure readings. These data are also shown in Fig. 2.

A straightforward least square fit of the data in Fig. 2 to the function $P = 1 - e^{-t/\tau}$ gives $\tau = 71$ s. However, as discussed previously, the collisional effects could be significant for the lifetime measurement of a long-lived metastable state even at the ultra low pressure of 10^{-11} Torr. Some collisional quenching effects have been studied before for the $D_{5/2}$ state [3,10]. At experimental conditions similar to the present one, the collisional quenching rate was found to be roughly 2.5×10^7 s^{-1} per Torr, resulting in a 2 s correction to the measured 30 s lifetime of the $D_{5/2}$ state [2].

In addition to the quenching effect, there is also the collisional fine-structure mixing as discussed in Ref. [4]. The fine-structure mixing results from collisions with neutral molecules which mix the two fine-structure D states [11]. The effective radiative decay rate with the collisional mixing can be expressed as

$$\frac{1}{\tau_{\text{eff}}} = (1 - \alpha)\frac{1}{\tau_{3/2}} + \alpha \frac{1}{\tau_{5/2}},$$

where $\alpha \leq 1$ is the mixing coefficient of the $D_{5/2}$ state. α in general is a function of the time and could reach a steady-state value quickly if the mixing rate is much faster than the decay rate. In the latter case, the effective lifetime is independent of pressure in the absence of the collisional quenching. So, unlike the collisional quenching, the fine-structure mixing effect in general cannot be removed by a simple pressure extrapolation. Under our experimental condition where the background pressure is extremely low, the collisional mixing is slow and weak. As an approximation, we can assume the mixing coefficient α is linear in time, i.e., $\alpha = r_m t$. r_m is some constant determined by the mixing cross section and the collisional rate. Then it can be shown that a Ba^+ ion initially in the $D_{3/2}$ state decays to the ground state after time t with the probability

$$P = 1 - \exp\left(-\frac{t}{\tau_{3/2}[1 - \frac{r_m}{2}(\frac{\tau_{3/2}}{\tau_{5/2}} - 1)t]}\right).$$

This shows that the measured lifetime τ for a given wait time t is a linear function of t itself. To estimate the fine-structure mixing effect in our measurement, instead of fitting the data in Fig. 2 directly, we evaluate τ at each wait time t, as shown in Fig. 3. A trend of shortened lifetimes with longer wait times is clearly seen. Again, the errors shown in the figure are calculated based on those in Fig. 2. With the weighted linear fit we find $\tau_{3/2} = 77.9 \pm 4.3$ s, 7 s longer than the straightforward fitted value. The fitted r_m value is 1.5×10^{-3} s^{-1}.

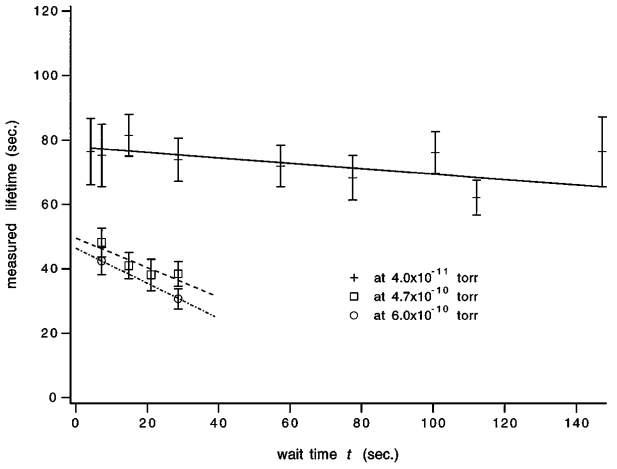

FIG. 3. Plots of the evaluated lifetime τ at each wait time t. The lines are the linear fit curves.

Indeed, the mixing is weak, but nevertheless should be considered significant in our measurement.

In the above discussion, we have ignored the collisional quenching. The quenching effect is expected to be small. It is also reasonable to assume that the quenching rates are similar for the two D states. Under this assumption, it is easy to show that the above result is still valid, but the value includes the quenching effect. To have a rough estimate of the quenching effect, we took measurements at two elevated pressures near 5×10^{-10} Torr. One has to be careful here, since the mixing will be a much larger effect at higher pressure. To minimize the uncertainty, we chose data points with the wait times $\leq$28.6 s. As before, the $t = 0$ values of the lifetime are extrapolated. With the three pressure points, as shown in Fig. 4, we extrapolate the lifetime to zero pressure. It yields 79.8 ± 4.6 s as the final value for the radiative lifetime of the $D_{3/2}$ state. The quenching correction resulted in an about 2 s longer lifetime, well within one standard deviation of the 4.6 s statistical uncertainty in the measurement.

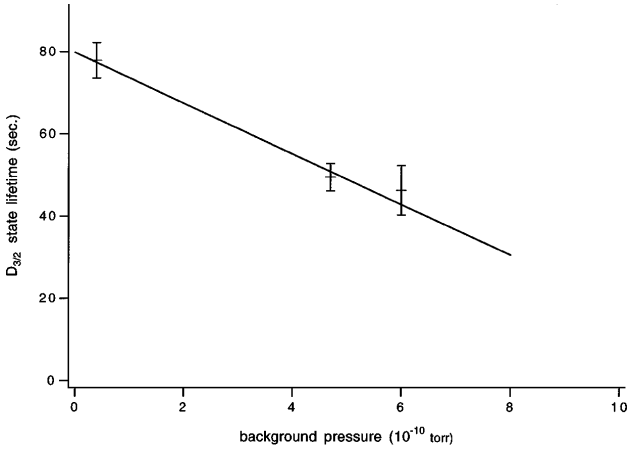

FIG. 4. Plot of the lifetimes extrapolated to $t = 0$ at different pressures.

Other measurement errors such as the timing errors are negligible.

As mentioned earlier, the present experiment also obtains the $D_{5/2}$ lifetime for comparison purposes. The direct measured value at 4×10^{-11} Torr pressure is 31.5 ± 2.6 s. The collisional quenching rate is measured to be 1.9×10^{7} s^{-1} per Torr. By excluding this quenching, the extrapolated value at zero pressure is 32.3 s. These results are quite consistent with the previous measurements [2,3].

In conclusion, we have measured the natural radiative lifetime of the $D_{3/2}$ state for the Ba^{+} ion. The experiment was done with a single trapped ion in UHV. We found that, under the UHV condition, the collisional quenching is relatively insignificant while the fine-structure mixing effect is still measurable. Our present measurement brings the theoretical values within one standard deviation of the experimental value of 79.8 ± 4.6 s, and thus has resolved the existing discrepancy.

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