Measurement of the lifetime of the metastable $5d_{3/2}26d_{3/2}$, $J=0$ autoionizing state of barium

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We report a measurement of the lifetime of the metastable $5d_{3/2}26d_{3/2}$, $J=0$ autoionizing state of barium. We have determined the lifetime to be 190(10) ns.

The recent study of inhibited autoionization in barium[1] demonstrated that the internal interferences between autoionizing states can lead to a dramatic suppression of the autoionization process. Several states of the $5d_{3/2}nd$ series were observed to have very narrow spectral widths. The data showed a variation in the scaled autoionization rate (spectral width) of these states of at least 3 orders of magnitude. One of these states, the $5d_{3/2}26d_{3/2}$, $J=0$, had a measured width of no more than 8.5 MHz, corresponding to a lifetime of $\sim$20 ns. We have directly measured the lifetime of this state by exciting atoms from the metastable state to another autoionizing state with an independently pumped dye laser and then monitoring the decay products as a function of the time at which the excitation took place.

Ideally, one would measure the lifetime of a state by directly observing the decay products (photons or electrons, typically) as a function of time. This requires a temporal resolution much smaller than the lifetime being measured. In the case of the metastable state, the fluorescent rate is expected to be very small, thus leaving only electrons or ions to be monitored. For a lifetime on the order of 100 ns, the experiment must be able to collect charged particles in $\sim$10 ns for a reasonable resolution. It is difficult to move heavy ions or low-energy electrons (0.58 eV energy) on these time scales without affecting the parent state.

A second, indirect technique is to measure the spectral linewidth of a state and infer a lifetime.[1] For long lifetimes this requires an excitation laser with a very narrow linewidth. Furthermore, various other effects, such as Doppler broadening or Stark shifts, can change the linewidth without affecting the state's lifetime. Consequently, the lifetime deduced from a linewidth measurement gives a lower limit, which is usually accurate only for short-lived states.

We have utilized, therefore, a pump laser—delayed probe laser combination to measure the lifetime of the $5d_{3/2}26d_{3/2}$ $J=0$ metastable state. Figure 1 shows the relevant energy levels. Two tunable, pulsed dye lasers, pumped by the third harmonic of a Nd:YAG (YAG denotes yttrium aluminum garnet) oscillator, excited barium atoms in an effusive atomic beam to the metastable state. The probe laser, also a tunable dye laser pumped by a second Nd:YAG oscillator, excited the atoms from the $5d_{3/2}26d_{3/2}$, $J=0$ metastable state to the $6p_{3/2}26d_{3/2}$, $J=1$ autoionizing state. This laser system was triggered by a computer-driven, voltage-controlled, time-delay generator. This enabled us to continuously vary the time between the pump laser and the probe laser to map out the temporal behavior of the state. All three lasers had the following characteristics: linewidth ~ 0.3 cm^{-1}; pulse duration $\sim$5 ns; energy per pulse ~ 100 μJ.

Since the metastable state decays primarily by autoionization producing 0.58 eV energy electrons and ground-state ions [$Ba^+(6S_{1/2})$], measuring the total ion yield alone would not give a measure of the relative population in the metastable state. However, the further excited $6p_{3/2}26d_{3/2}$ state autoionizes to produce $Ba^+(6P_{1/2})$ and 0.18 eV electrons *and* $Ba^+(6S_{1/2})$ and 2.70 eV electrons. We have monitored, therefore, only the "hot" electrons (2.7 eV) by applying a retarding voltage to the electrons.

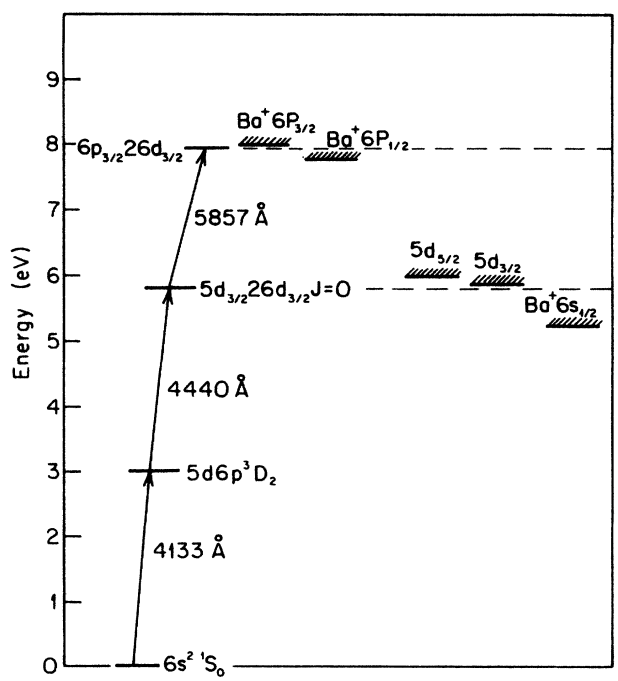

FIG. 1. Barium energy-level diagram showing the excitation scheme to populate the $5d_{3/2}26d_{3/2}$, $J=0$ state. The third laser monitors the metastable population by further exciting the atoms to the autoionizing $6p_{3/2}26d_{3/2}$, $J-1$ state.

We actually excited the atoms in a field-free region between two grounded plates, but the top plate had a small hole in it to pass electrons into a 15-cm drift region where the retarding voltage was applied. The hot electrons were then detected by a channel electron multiplier, and integrated by a boxcar averager.

The experiment was performed as follows. All three lasers were tuned to maximize the hot electron signal with the delay time set at zero (all lasers arriving at about the same time). The computer then swept the delay time between the first two lasers and the probe laser. Figure 2 shows typical data. The vertical axis is the normalized hot-electron signal; the horizontal axis is the time delay in nanoseconds; and the points at negative times indicate the noise level. Each point on the graph represents 30 laser shots averaged together. In order to avoid possible drifts while collecting the data, these 30 shots were taken in three sets of 10 shots each and subsequently added together. We collected three such sets of data for each delay time. The spread in the data points is indicative of the statistical errors. To check for systematic errors, we applied small dc electric fields (~ 1 V/cm), but observed no change in the lifetime, although under high signal conditions we did observe an apparent shortening of the lifetime. We believe that this was either due to the space-charge effects on the last electrons out, or to microfields in the plasma shortening the lifetime of the metastable state. The solid line is a linear least-squares fit using all of the data, corresponding to a lifetime of 190(10) ns, where the uncertainty only includes statistical variations.

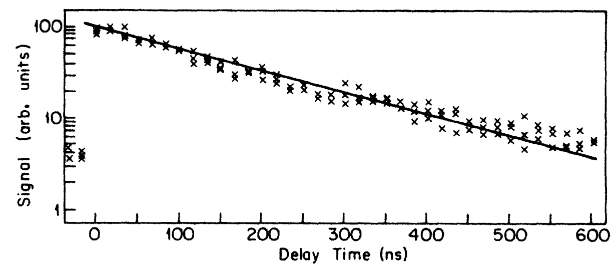

FIG. 2. The hot-electron signal vs delay time. Each point represents 30 laser shots. The solid line is a least-squares fit yielding a lifetime of 190(10) ns.

In conclusion, we have measured the lifetime of the $5d_{3/2}26d_{3/2}$, $J=0$ metastable state of barium. This lifetime implies an autoionization induced linewidth of 0.84 MHz. With this measurement, the variation in autoionization rates as described by Neukammer *et al.* was actually over 5 orders of magnitude instead of 3.

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[1]J. Neukammer, H. Rinneberg, G. Jonsson, W. E Cooke, H. Hieronymus, A Konig, K. Vietzke, and H. Springer-Bolk, Phys. Rev. Lett. 55, 1979 (1985).