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Measurement of the lifetimes of the lowest $^3P_1$ state of neutral Ba and Ra

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The lifetimes of the lowest $^3P_1$ states of Ba and Ra were determined to be 1345±14 ns and 422±20 ns, respectively, by measuring the exponential decay of fluorescence after illuminating a thermal atomic beam with pulses of laser light. In addition, the $^1S_0(F=1/2)$-$^3P_1(F=3/2)$ transition frequency in ^{225}Ra was measured to be 13 999.269±0.001 cm^{-1} by referencing a nearby I_2 transition.

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Radium has recently attracted significant interest as an excellent atomic system for studying parity and time-reversal violation [1]. In isotopes such as ^{225}Ra($t_{1/2}$=14.9 days) that have an octupole-deformed nucleus, the signature of an electric dipole moment is expected to be amplified by over two orders of magnitude relative to previously studied systems [2–5]. In order to measure these effects, detailed knowledge of the atomic properties of radium is needed. Until now, the best measurements of the energy of the atomic levels were spectroscopic studies of the longest-lived radium isotope (^{226}Ra, $t_{1/2}$=1600 years) performed in 1934 [6,7]. More recently, radium isotopes in the mass range A=208–232 were produced at the ISOLDE facility and the isotope shifts and hyperfine splittings of a handful of low-lying levels were determined [8,9]. Many atomic properties, such as the lifetime of any excited state, remain unmeasured.

Calculations of many atomic lifetimes, branching ratios, and transition rates have been performed [10–13]. However, experimental data is lacking to constrain such calculations for radium. Spin-changing transitions, such as between the $7s^2\ ^1S_0$ ground state and the $7s7p\ ^3P_1$ state, are particularly challenging to calculate because of the sensitivity to relativistic effects. Predictions of the lifetime of this state vary from 250 to 505 ns [10–13]. Experimental results for low-lying states are needed to constrain the atomic wave functions.

For neutral barium, with a valence-shell electronic structure similar to that of radium, a large collection of experimental data exists [14]. Barium provides a convenient system to verify the accuracy of calculational techniques applied to radium because of the many experimental constraints. Barium also proved useful because statistics could be easily and rapidly accumulated. It was of interest to precisely measure the $6s6p\ ^3P_1$ lifetime to test the experimental technique and improve upon the precision of previous measurements [15,16].

In this paper, we report measurements of the frequency of the $^1S_0$-$^3P_1$ transition of ^{225}Ra and the lifetime of the $^3P_1$ state in both neutral barium and radium. For radium, the lifetime determines the transition probability because this state decays predominantly back to the $^1S_0$ ground state (Fig. 1).

The experimental layout is shown in Fig. 1. We chemically separated ^{225}Ra from a 250-μCi source of the long-lived parent nuclide ^{229}Th($t_{1/2}$=7300 years). The ^{225}Ra solution was loaded into a titanium crucible and dried. We added 100 mg of barium metal not only as a source of atoms for study but also to aid in the reduction of the radium and passivation of the crucible surfaces. At temperatures above 650 °C, neutral atomic beams of barium (>10^9 atoms/s) and radium (>10^6 atoms/s) emerged from the aperture of the crucible exit channel with a divergence angle of less than 50 mr. Although operation at a constant temperature resulted in a slow decline in output, steady atomic fluxes were maintained over several days of measurement by slowly increasing the oven temperature. The pressure in the vacuum chamber was maintained in the 10^{-7} torr range.

The intercombination transitions of radium and barium at 714.3 and 791.1 nm, respectively, were excited using light

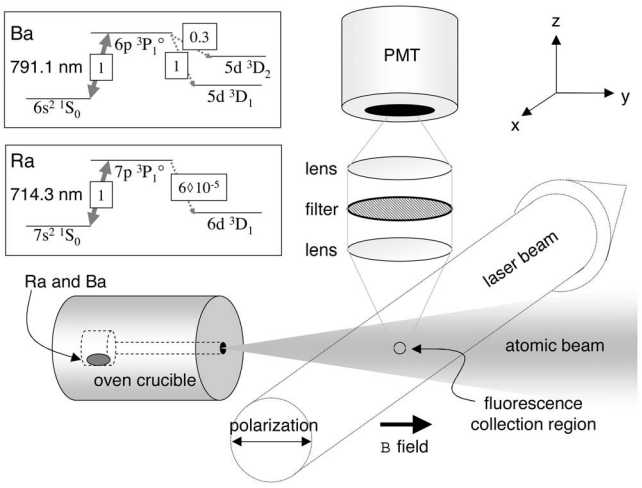

FIG. 1. Experimental layout geometry (not to scale). The laser beam is shown passing transversely across the atomic beam with linear polarization and the applied magnetic field, B, in the $+y$ direction. The laser beam diameter (1.0–4.5 cm) was larger than the diameter of the fluorescence collection region (0.3 cm). Additional measurements were made on barium with the laser beam directed in the $-y$ direction and linearly polarized in the x direction. In the atomic energy diagrams, the relative branching ratios shown in the rectangles [10] for the dominant decays to dark states (dotted arrows) and the transition of interest (solid arrows).

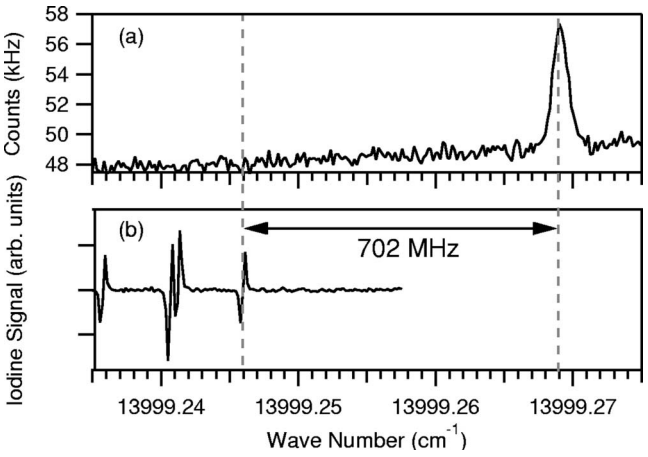

FIG. 2. Fluorescence detected from the $^1S_0(F=1/2)$-$^3P_1(F=3/2)$ transition in ^{225}Ra is shown in (a). The frequency of nearby I_2 transitions are shown in (b). During the experiments, the indicated I_2 transition was used as a frequency reference for the probe beam.

from a Ti:sapphire ring laser pumped by a frequency-doubled, diode-pumped Nd:YVO_4 laser. The linearly polarized laser beam transversed perpendicularly across the atomic beam 4 cm downstream from the oven aperture as shown in Fig. 1. The laser beam was expanded to diameters as large as 4.5 cm to minimize effects due to the atomic beam propagation. Its linewidth was broadened to 20 MHz by double passing through an acousto-optic modulator (AOM) operated with a radio-frequency (rf) carrier mixed with white noise. The increased linewidth matched the transverse Doppler spread due to the divergence of the atomic beam. The laser light was shuttered with an extinction ratio of better than 1000:1 after 100 ns by switching off the rf signal applied to this AOM. In addition, the laser beam could be aligned to counterpropagate against the atomic beam. By illuminating the atoms in these two different ways, systematic effects associated with the lifetime measurement were checked.

A simple lens system focused 5% of the scattered light from atoms in a roughly 15 mm^3 volume onto a photomultiplier tube (PMT) where photons were detected with a 10% quantum efficiency. Signal amplification and discrimination were handled by a commercial PMT package. PMT dark counts were typically 100 Hz. Optical bandpass filters with bandwidths of 4 nm (for radium) and 10 nm (for barium) were used to reduce backgrounds from the crucible and heating elements to several kHz.

For the measurements on ^{225}Ra, the laser frequency was locked to a hyperfine transition of molecular I_2 at 13 999.2459 cm^{-1} (see Fig. 2), which is part of the Doppler-broadened absorption peak at 13 999.2363 cm^{-1} [17], using optical heterodyne saturation spectroscopy [18]. Using a double-passed AOM, the I_2 spectroscopy beam frequency was shifted so that the I_2 and ^{225}Ra transitions of interest were excited simultaneously. The barium fluorescence signals were large and measurement times sufficiently short that special care maintaining the laser frequency was not required.

We determined the $^1S_0(F=1/2)$-$^3P_1(F=3/2)$ transition frequency in ^{225}Ra to be 13 999.269±0.001 cm^{-1} by scanning the laser over the transition and locating the peak of the fluorescence relative to the I_2 transition used to control the laser frequency. Figure 2 shows the radium fluorescence signal relative to nearby I_2 transitions. The absolute frequency of the I_2 transition was determined with a precision of ±0.0005 cm^{-1} by commercial software based on the results of [19]. A comparable uncertainty arose from the difficulty in assuring the laser beam and atomic beam were perfectly perpendicular. We also observed fluorescence from the $^1S_0(F=1/2)$-$^3P_1(F=1/2)$ transition at 13 999.76 cm^{-1}, as expected from previous measurements of the hyperfine structure [9]. From these results, the $^1S_0$-$^3P_1$ transition frequencies for ^{212}Ra and $^{222-226}$Ra were also determined to the same precision after correcting for the measured isotope shifts [9]. We infer a value of 13 999.357±0.001 cm^{-1} for this transition in ^{226}Ra that is 700 MHz lower than the previous absolute measurement [6].

The $^3P_1$ lifetimes were determined from the exponential decay of fluorescence detected at the PMT after shuttering the laser beam. For barium, measurement cycles consisted of pulsing the laser beam on for 1 μs followed by shuttering the laser for 9 μs. For radium, the cycle was similar with the laser on for 0.5 μs and off for 2.9 μs. These cycles were repeated until sufficient statistics were accumulated. Upon detection of a photon, the timing system would count cycles of an 80-MHz clock until stopped by the beginning of the next laser pulse.

To eliminate the effects of Zeeman quantum beats, a magnetic field $B=8.5\pm0.5$ G was applied along the same direction as the linear polarization of the laser beam to split the Zeeman sublevels by ≈20 MHz. Any residual effect due to imperfect polarization would lead to a modulation of the fluorescence emission with a period of 60 ns. No such modulation was observed in the data. Lowering the magnetic field to 4 G had no observable effect on the extracted lifetime. The alternative approach of nulling out any residual magnetic fields would have required the field be less than a few milliGauss because of the relatively long lifetimes studied here.

Figure 3 shows the signal to background and the fit to the data for typical ^{138}Ba and ^{225}Ra measurements. Backgrounds were measured by repeating the measurement with the laser light detuned from the atomic transition by several GHz. The only background to persist beyond 100 ns was blackbody radiation from the heated oven that was distributed uniformly in time. The data acquisition could only be started once per measurement cycle so an exact analytic correction for the loss of subsequent events was applied to the data. We subtracted the background from the data before performing a three parameter least-squares fit to a single exponential and uniform offset. We obtained values of the χ^2 per degree of freedom in the range 0.9–1.1, showing that the data are well represented by this functional form. The measured lifetimes were consistent within statistical uncertainty for various timing cuts, even when the analysis was extended to times earlier than 100 ns.

Systematic effects were thoroughly investigated using barium, for which data could be rapidly accumulated. The excitation laser beam was directed either transverse to or

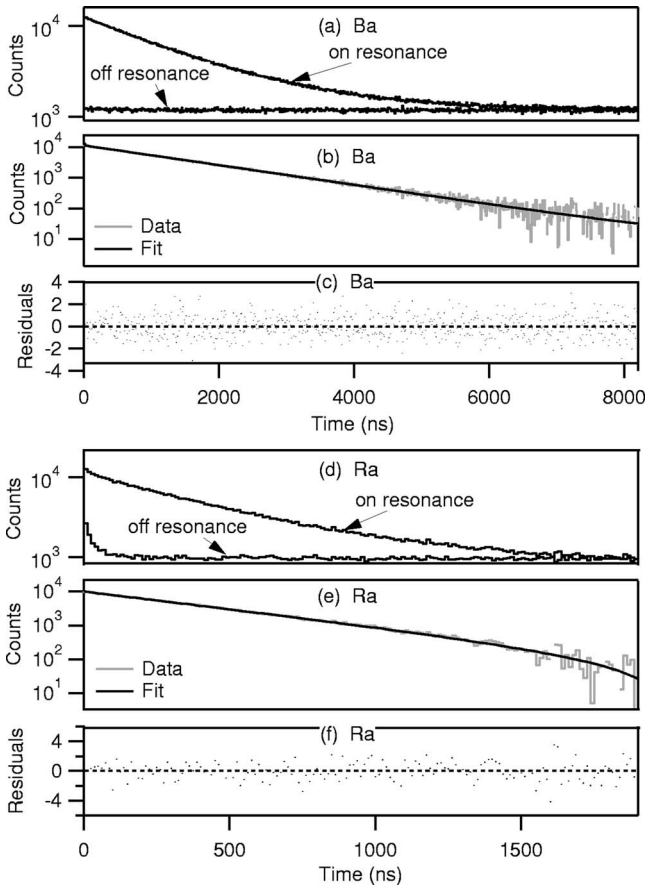

FIG. 3. Typical distribution of photon arrival times after shuttering the laser are shown in (a) for barium and in (d) for radium. In the "off resonance" data, the laser detuning was ≈3 GHz from the atomic transitions of interest. The background-subtracted data with fits to an exponential decay with a time-independent background are shown in (b) for barium and in (e) for radium. The fits give 1344±3 ns and 422±3 ns (statistical uncertainty only) for the lifetimes of the displayed barium and radium data, respectively. The residuals for the fit, normalized by the square root of the number of counts in each bin, are shown in (c) for barium and (f) for radium.

antiparallel to the atomic beam. These two measurement geometries have different sets of systematic effects and obtaining agreement between these measurements was an important cross-check of the technique.

Measurements in the transverse geometry are susceptible to effects due to the atomic motion—excited barium (radium) atoms traveled on average 0.6 mm (0.15 mm) before decaying. We limited this effect by using a laser excitation region (4.5 cm diam) that was significantly larger than the fluorescence collection region (0.3 cm diam). We estimated that the laser beam was centered relative to the collection optics to better than 0.2 cm. Therefore, the flux of excited atoms drifting into and out of the collection region should be nearly identical. The laser intensity used was 0.9 μW/cm^2. The size of motion-dependent effects were investigated by trying to upset this balance by decreasing the laser beam diameter and displacing the laser beam path along the direction of the atomic motion. No statistically significant shift was seen when the beam diameter was decreased to 2.0 cm.

TABLE I. Summary of $^3P_1$ lifetime results for atomic barium and radium.

Atom	Expt.	Calc.	Ref.
Ba	1345±14 ns		this work
	1351±55 ns		[15]
	1200±100 ns		[16]
		1370 ns	[10]
		1520 ns	[11]
		1230 ns	[13]
		994–1120 ns	[20]
Ra	422±20 ns		this work
		505 ns	[10]
		420 ns	[11]
		250 ns	[12]
		362 ns	[13]

All these measurements yielded lifetime values in the range 1342–1346 ns with statistical uncertainties of ≈3 ns.

The use of a 0.6-cm-diam counterpropagating laser beam to illuminate all atoms that enter the fluorescence region essentially eliminated the atomic motion effects described previously. This technique was used only with barium because of difficulties in collecting sufficient statistics, as only ≈10% of the atoms were in resonance due to the greatly increased Doppler spread. Pumping atoms to a dark state (most atoms excited to the $^3P_1$ state decay to the $5d^3D_2$ or $5d^3D_1$ metastable states as can be seen in Fig. 1) as they propagate would diminish the fluorescence signal from downstream atoms and lead to a systematic increase in the lifetime. By extending the time between laser pulses to 200 μs, atoms have enough time to drift past the fluorescence collection region and are probed at most once. No change in the measured lifetime was observed when this period was varied between 9–200 μs. No velocity dependent effects were observed when the laser frequency was detuned by ±150 MHz, corresponding to a change in average velocity of probed atoms by 200 m/s. In addition, there was no significant change in the lifetime when the laser beam path was off axis by several millimeters, the laser power was varied from 30–300 μW/cm^2, or the laser beam area was decreased by a factor of 5. These results agree with the transverse excitation measurements; the lifetimes are in the range 1332–1359 ns with typical statistical uncertainties of 5 ns.

As consistent results were obtained for the barium $^3P_1$ lifetime using both techniques, we take this to indicate that systematic effects were not significant. In particular, the agreement indicates that the laser beam alignment for the transverse measurements for barium did not introduce a systematic shift larger than 10 ns.

For radium, only measurements in the transverse geometry were possible because of weak fluorescence signals. The laser beam alignment was identical during the radium and barium measurements. Any effect due to the transit of atoms before decaying would be approximately five times smaller for radium than for barium because of the shorter lifetime

and smaller velocities due to increased mass. In the majority of these measurements, the laser intensity was $I=270\ \mu\mathrm{W/cm^2}$ with a beam diameter of 1.0 cm. There was no significant change in the measured lifetime when the laser beam diameter was increased to 1.8 cm and the intersection region between the laser and atomic beams was shifted along the atomic beam direction by ±2.5 mm.

No statistically significant shift in the data was uncovered by varying the experimental conditions. Overall, we find the individual results are slightly overscattered (χ^2 per degree-of-freedom of 1.8 for both the Ba and Ra lifetime weighted averages) relative to the small statistical uncertainties (typically $\approx$0.4% for Ba and $\approx$2% for Ra). This most likely results from small, unresolved shifts in the measurements. The twelve ^{138}Ba measurements and six ^{225}Ra measurements, each conducted under a different set of experimental conditions, fall in the ranges 1345 ± 14 ns and 422 ± 20 ns, respectively. We conservatively take these ranges as the 1σ total uncertainty in our measurements. A summary of results for the lifetimes of the $^3P_1$ states in barium and radium is presented in Table I. The results of previous measurements and several calculations are also included for comparison. For radium, we find remarkable agreement with the calculated results of [11] and the results of [10] fall within the 20–30 % uncertainty estimated by the authors.

Development of a magneto-optical trap for ^{225}Ra based on this transition is currently underway. With trapped samples, it should be possible to improve the precision of this measurement and perform detailed spectroscopy of the radium atom.

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[1] J. S. M. Ginges and V. V. Flambaum, Phys. Rep. **397**, 63 (2004).
[2] V. Spevak and N. Auerbach, Phys. Lett. B **359**, 254 (1995).
[3] V. Spevak, N. Auerbach, and V. V. Flambaum, Phys. Rev. C **56**, 1357 (1997).
[4] J. Engel, J. L. Friar, and A. C. Hayes, Phys. Rev. C **61**, 035502 (2000).
[5] J. Dobaczewski and J. Engel, Phys. Rev. Lett. **94**, 232502 (2005).
[6] E. Rasmussen, Z. Phys. **86**, 1934 (1934).
[7] H. N. Russell, Phys. Rev. **46**, 989 (1934).
[8] S. A. Ahmad, W. Klempt, R. Neugart, E. W. Otten, K. Wendt, C. Ekstrom, and T. I. Collaboration, Phys. Lett. **133B**, 47 (1983).
[9] K. Wendt, S. A. Ahmad, W. Klempt, R. Neugart, E. W. Otten, and H. H. Stroke, Z. Phys. D: At., Mol. Clusters **4**, 227 (1987).
[10] V. A. Dzuba, V. V. Flambaum, and J. S. M. Ginges, Phys. Rev. A **61**, 062509 (2000).
[11] P. Hafner and W. H. E. Schwarz, J. Phys. B **11**, 2975 (1978).
[12] J. Bruneau, J. Phys. B **17**, 3009 (1984).
[13] V. A. Dzuba and J. S. M. Ginges, e-print physics/0511199.
[14] J. J. Curry, J. Phys. Chem. Ref. Data **33**, 725 (2004).
[15] J. Brust and A. C. Gallagher, Phys. Rev. A **52**, 2120 (1995).
[16] H. Bucka and H. H. Nagel, Ann. Phys. **8**, 329 (1961).
[17] S. Gerstenkorn, J. Verges, and J. Chevillard, *Altas Du Spectre D'Absorption De la Molecule D'Iode* (Laboratoire Aime Cotton, Orsay, France, 1982). We locked to the highest wave number transition of line number 692.
[18] J. L. Hall, L. Hollberg, T. Baer, and H. G. Robinson, Appl. Phys. Lett. **39**, 680 (1981).
[19] H. Knockel, B. Bodermann, and E. Tiemann, Eur. Phys. J. D **28**, 199 (2004).
[20] D. Kulaga, J. Migdalek, and O. Bar, J. Phys. B **34**, 4775 (2001).