Lifetime of the $7s6d\ ^1D_2$ atomic state of radium

W. L. Trimble,[1] I. A. Sulai,[1,2,3] I. Ahmad,[1] K. Bailey,[1] B. Graner,[1,3] J. P. Greene,[1] R. J. Holt,[1] W. Korsch,[4] Z.-T. Lu,[1,2,3] P. Mueller,[1] and T. P. O'Connor[1]

[1]*Physics Division, Argonne National Laboratory, Argonne, Illinois 60439, USA*
[2]*Department of Physics, The University of Chicago, Chicago, Illinois 60637, USA*
[3]*The Enrico Fermi Institute, The University of Chicago, Chicago, Illinois 60637, USA*
[4]*Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506, USA*

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The lifetime of the $7s6d\ ^1D_2$ state of atomic radium is determined to be 385(45) μs using cold ^{226}Ra atoms prepared in a magneto-optical trap. The $^1D_2$ state is populated from the decay of the $^1P_1$ state which is excited by a pulse of 483 nm light. The decay of the $^1D_2$ state is observed by detecting delayed fluorescence at 714 nm from the last step in the decay sequence $^1P_1$-$^1D_2$-$^3P_1$-$^1S_0$. The measured lifetime is compared to a number of theoretical calculations. An improved value of the $7s7p\ ^1P_1$ level of 20 715.598(6) cm^{-1} is obtained.

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I. INTRODUCTION

Radium is the highest-Z neutral atom for which laser cooling and trapping have been demonstrated [1]. It is an attractive candidate for tests of fundamental symmetries, since relativistic and finite-size-nuclear effects are enhanced in heavy atoms and since laser techniques permit long observation times and potentially precise measurements with small samples.

There are two transitions from the ground state that can be used for laser trapping and cooling of atomic radium (Fig. 1). The intercombination transition $^1S_0$-$^3P_1$ ($\Gamma = 2\pi \times 380$ kHz) at 714 nm can be repeatedly excited to provide a maximum acceleration of 3×10^3 m s^{-2}. This transition is quasicycling since $^3P_1$ has an estimated branching ratio to $^3D_1$ of 5×10^{-5}, but a single repump laser tuned to the $^3D_1$-$^1P_1$ transition at 1428 nm is sufficient to bring the atoms back into the cooling cycle. Compared to the intercombination transition, the singlet-to-singlet transition $^1S_0$-$^1P_1$ ($\Gamma \sim 2\pi \times 30$ MHz) at 483 nm has a much stronger transition strength and it can be used to achieve an acceleration as large as 3×10^5 m s^{-2}, 100 times larger than the first scheme. However, the $^1P_1$ state has much stronger leak channels. Its total branching ratio to $^1D_2$ and ^3D_J states is estimated to be 2×10^{-3}. Consequently, a radium atom can scatter on average only 500 photons of 483 nm before going dark. Recent work demonstrating the trapping of neutral barium has shown that manipulation of such "leaky" atomic systems is possible with a sufficient number of repump lasers [3]. The work presented here will help evaluate the feasibility of using the $^1S_0$-$^1P_1$ transition for more efficient slowing and trapping of radium by measuring previously unknown lifetimes and branching ratios of excited states in radium.

TABLE I. Values of the $^1D_2$ lifetime, theory, and experiment.

Work	Theory	Experiment
[6]	710 μs	
[7]	129 μs	
[2]	1.37 ms	
This work		385(45) μs

The lifetime of the $^1D_2$ state has been calculated a number of times and the results range from 129 μs to 1.37 ms (see Table I for references). The difficulty in atomic theory arises from a near cancellation of two large transition amplitudes [4]. Previous determination of the $^3P_1$ lifetime was performed using a thermal beam of radium atoms [5]. The long lifetime of $^1D_2$ makes that approach difficult. Instead, the measurement presented here is performed on cold ^{226}Ra atoms prepared in a magneto-optical trap (MOT). This work represents only the second experimental determination of an excited-state lifetime in atomic radium. It will help anchor atomic theory calculations and better model the interaction between the radium atom and lasers used to manipulate it.

II. EXPERIMENTAL SETUP

The apparatus for laser cooling and trapping of radium atoms is described in detail in [1]. In short, neutral ^{226}Ra

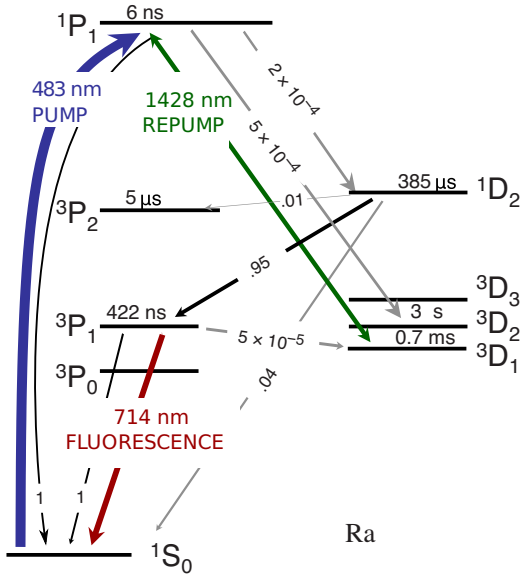

FIG. 1. (Color online) Level diagram for the lowest eight electronic states of neutral radium. The lifetimes of $^1D_2$ and $^3P_1$ are experimentally determined; other lifetimes and branching ratios are based on theoretical calculations [2,6].

($t_{1/2}$=1600 yr) atoms emerging from a thermal atomic beam source at 500 °C are transversely cooled, slowed in a Zeeman slower, and captured in a MOT, all by exciting the intercombination transition, $^1S_0$-$^3P_1$, at 714 nm using light from a Ti:sapphire ring laser. A diode laser at 1428 nm is tuned to the $^3D_1$-$^1P_1$ transition and overlapped with one of the MOT beams to repump atoms from $^3D_1$. Typically 500–1000 cold ^{226}Ra atoms are captured in the MOT with a trap lifetime of 2 s.

Typically, 1.5 mW of 483 nm light is produced by frequency doubling a 100 mW diode laser at 966 nm in a periodically poled lithium niobate waveguide. 483 nm light with an intensity of 100 mW cm^{-2} is directed into the MOT at a 5° angle to one of the MOT beams and is retroreflected using a lens and mirror to adjust the intensity of the reflected beam. When tuned to the $^1S_0$-$^1P_1$ resonance, the 483 nm laser pumps atoms to the metastable states, causing loss from the trap. The 483 nm laser frequency and beam pointing is adjusted for maximal trap loss at low intensity. The $^1S_0$-$^1P_1$ resonance frequency in ^{226}Ra is measured to be 20 715.598(6) cm^{-1} using a laser wavelength meter (Bristol Instruments 621). This value is 0.1 cm^{-1} below the value determined from grating spectrometer measurements in the 1930s [8,9] and places the $^3D_1$ level at 13 715.76 cm^{-1} based on the measurement of the $^3D_1$-$^1P_1$ wavelength [1].

For the lifetime measurement, the trapping light at 714 nm and the repump light at 1428 nm are kept on for 990 ms in the atom capture phase and are then blocked by mechanical shutters for 10 ms in the measurement phase [Fig. 2(c)]. The two phases cycle at 1 Hz rate. To keep the detector dark counts low, the detector is opened only after the MOT light is shuttered. During the first 5 ms of the 10 ms measurement phase, the cold atoms fall while the detector shutter opens. They are then excited by a 100 μs pulse of 483 nm light. Fast amplitude modulation of the 483 nm light is achieved with an acousto-optical modulator. Each excited atom emits either a 483 nm photon or decays to one of the excited metastable states. Those metastable states then eventually decay to the ground state. Theoretical branching ratios predict that most of the atoms pumped to $^1D_2$ will decay by emitting a photon at 714 nm while the atoms pumped to other states will not decay rapidly via $^3P_1$. The atomic fluorescence at 714 and 483 nm is separated by color with a dichroic mirror and optical band-pass filters and detected by two separate photomultiplier tubes. A computer-based data-acquisition system records the arrival times of the detected photons.

III. RESULTS

Figure 2(a) shows the 714 nm fluorescence collected from the MOT when the 483 nm pulse is applied without shuttering the MOT beams. During each 483 nm flash, the MOT fluorescence promptly decreases, declining to nearly background levels by the end of the pulse. Between 5% and 20% of the MOT, fluorescence returns within 1 ms of the end of the 483 nm flash. The relative intensities of the incoming beam and its reflection are adjusted to maximize the fraction of atoms which return following the flash.

Figure 2(b) shows the 714 nm fluorescence collected from the atoms when the MOT light is blocked. 714 nm fluorescence rises during the 483 nm exposure and falls following it. The delayed fluorescence is observed only when the 483 nm laser is tuned to the $^1S_0$-$^1P_1$ resonance.

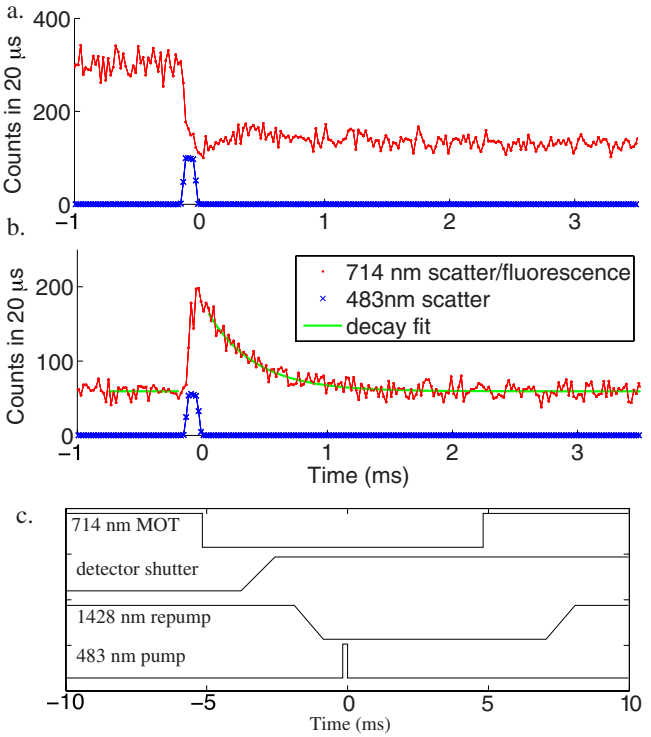

FIG. 2. (Color online) (a) Suppression of MOT fluorescence at 714 nm caused by 483 nm light pumping radium to metastable states. This panel represents data accumulated for 11 min with the MOT beams kept on. (b) 714 nm flash following 483 nm excitation of radium atoms shows the decay of $^1D_2$. The MOT beams are off for the collection of these data. This panel represents data accumulated for 10 h with the timing structure in (c).

The arrival-time data are fitted using maximum likelihood and weighted least squares. 714 nm fluorescence data are used excluding those taken during the 483 nm flash. Since the count rates in the detector are low, maximum likelihood is the more appropriate algorithm, though the two techniques agreed within their uncertainties. Data taken under the same experimental conditions are summed; three separate experimental conditions are fitted separately for comparison. Data taken with imbalanced 483 nm beams and with the repump on throughout the experiment had longer but insignificantly different fitted lifetimes. Data taken with imbalanced beams are discarded and the remainder are summed. This set, comprising 6 h of integration, gives a lifetime of 385(26) μs.

The observed delayed 714 nm fluorescence could be influenced by factors other than the $^1D_2$ lifetime. In a previous work, it was observed that room-temperature blackbody radiation pumps atoms from $^3P_0$ to $^3D_1$ at a rate of 200 Hz [1]. This radiation should also pump atoms from $^3D_1$ to $^3P_1$ at approximately 40 Hz. Though the lifetime of the $^3D_1$ is predicted to be on the same scale as the state of interest, the weak strength of the blackbody coupling between the states ensures that a small fraction of the $^3D_1$ can contaminate the 714 nm signal. The lifetime could be biased as much as 5% toward longer lifetimes if the population in the $^3D_1$ state is as large as 75%.

TABLE II. Error budget.

Source	Error
Motion from recoils	31
Black-body repumping from $^3D_1$	19
Motion from gravity	5
Statistical	26
Total	45 μs

The scattering of 483 nm light pushes the atoms and the motion of the atoms out of the viewing area of the detector could bias the resulting lifetime measurement. The atoms can scatter an average of 500 photons of 483 nm before decaying to the $^1D_2$ metastable state; this many single-photon recoils in the same direction would confer an average velocity of 2 m s^{-1} to the atoms. The 483 nm beams are balanced to reduce this effect. Assuming a 10% residual imbalance, the atoms will move 1 mm in 5 ms, less than the 2 mm radius viewing area of the detector. To test whether the atoms are leaving the viewing area before the decay is finished, some of the data were taken with the retroreflection blocked, providing a maximum imbalance in the momentum imparted by the 483 nm pump beam. The lifetime measured with a maximally imbalanced 483 nm excitation is 8% longer than the lifetimes with balanced beams. This is less than the 13% statistical uncertainty. This failure of an imbalanced beam to shorten the measured lifetime is evidence that the motion of the atoms out of the field of view of the detector does not influence the lifetime measurement at a level comparable to the statistical uncertainty in the measurement.

The radium atoms can also move away from the viewing region due to their initial velocity ($\sim$3 cm s^{-1}, 150 μm in 5 ms) and to falling under gravity (125 μm in 5 ms). Both effects are small compared to the effect of pushing by the 483 nm light. Quantum interference that might modulate the observed fluorescence is suppressed since the decay that is observed is one step in a three-photon cascade [10]. Interference effects caused by a nonzero magnetic field would be further washed out by the 1.5 G/cm axial field gradient over the 1 mm size of the atomic cloud. Table II lists estimates of the contributions to the total uncertainty.

IV. DISCUSSION

The lifetime of $^1D_2$ is expected to be dominated by the decay branch to $^3P_1$. The presence of the former state complicates cooling using the 483 nm transition by sequestering atoms and preventing them from participating in cooling for fractions of a millisecond. This measurement permits more accurate modeling of the atomic populations in the presence of 483 nm and repumping light.

This measurement of the radium $7s6d\ ^1D_2$ lifetime is limited by statistics; larger numbers of trapped atoms would permit more thorough investigation of systematic shifts to the measured decay, particularly experimental limits on the shifts caused by imbalanced forces and the viewing area of the detector.

In addition to excited-state lifetimes, experiments with cold radium atoms can also probe hyperfine splittings in some excited states and atomic polarizabilities to provide experimental anchors for the models of the atomic wave functions. It is interesting to note that the metastable states with the longest radiative lifetimes, $^3P_0$ and $^3D_3$, will absorb room-temperature blackbody photons in 5 and 200 ms, respectively, limiting the prospects for room-temperature experiments in metastable states of radium. The $^3D_2$ state, only 5 cm^{-1} below $^3P_1$, is only slightly affected by blackbody radiation.

In summary, this lifetime measurement for $^1D_2$ and refinement of the energies of the $7s7d\ ^1P_1$ and $7s6d\ ^3D_1$ levels in atomic radium should permit the construction of more accurate models of the atom, both for predicting the distribution of the atoms in their internal states for experiment and for interpreting the observable signatures of interesting physics arising from interactions with the nucleus.

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