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Lifetime determination of the $5d^2\ ^3F_2$ state in barium using trapped atoms

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Magneto-optically trapped atoms enable the determination of lifetimes of metastable states and higher lying excited states like the $5d^2\ ^3F_2$ state in barium. The state is efficiently populated by driving strong transitions from metastable states within the cooling cycle of the barium magneto-optical trap (MOT). The lifetime is inferred from the increase of MOT fluorescence after the transfer of up to 30% of the trapped atoms to this state. The radiative decay of the $5d^2\ ^3F_2$ state cascades to the cooling cycle of the MOT with a probability of 96.0(7)% corresponding to a trap loss of 4.0(7)% and its lifetime is determined to 160(10) μs. This is in good agreement with the theoretically calculated lifetime of 190 μs [V. A. Dzuba and V. V. Flambaum, J. Phys. B **40**, 227 (2007)]. The determined loss of 4.0(7)% from the cooling cycle is compared with the theoretically calculated branching ratios. This measurement extends the efficacy of trapped atoms to measure lifetimes of higher, long-lived states and validate the atomic structure calculations of heavy multielectron systems.

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I. INTRODUCTION

Lifetime measurements of the atomic states provide an important information about the absolute transition probability. Such measurements test the wave functions of atomic theory calculations which have been extended to heavy alkaline-earth-metal elements [1]. They permit the examination of configuration mixing of even- and odd-parity states which is prominent due to relativistic effects in the case of heavy two-electron systems. The theoretical methods provide access to the calculation of dipole matrix elements and oscillator strengths. Wave functions and theoretical methods are required, e.g., for the interpretation of fundamental symmetry tests which are currently underway [2–4]. The heaviest alkaline-earth-metal radioactive radium (Ra) atom is of interest for experimental searches of permanent electric dipole moments (EDMs), which simultaneously violate parity and time reversal symmetry [5]. The symmetry violating effects are amplified in Ra isotopes due to their unique nuclear and atomic structure. However, the enhancements are estimated from different approaches to many-body calculation which have discrepancies [6–8]. Barium (Ba) has been used as a reference to check the consistency of the calculational approach. We investigate barium as a precursor to radium.

Various experimental techniques have been used to measure the lifetimes of the excited states utilizing magneto-optically trapped atoms. These are photoassociation spectroscopy [9–11], electron-shelving [12] delayed resonance fluorescence [13–17], and other methods [18–21]. Laser-induced fluorescence can be employed if the atoms remain in one of the states of the optical cooling cycle. This is exploited here to measure the lifetime of the $5d^2\ ^3F_2$ state in Ba.

Laser cooling on the strong $6s^2\ ^1S_0$-$6s6p\ ^1P_1$ transition of barium requires us to drive several repump transitions (Fig. 1) at the same time [22,23]. The $6s6p\ ^1P_1$ state branches mostly to the $6s^2\ ^1S_0$ ground state and to 0.3% to the metastable $6s5d\ ^1D_2$ and $6s5d\ ^3D_{2,1}$ states. The decay rates from the $6s6p\ ^1P_1$, $6s6p\ ^3P_{1,2}$, and $5d6p\ ^3D_1$ states to the metastable D states and other low-lying states are given in Table I. Although the main optical cooling force arises from the strong $6s^2\ ^1S_0$-$6s6p\ ^1P_1$ transition, about half of the atoms are in the metastable D states in steady state of this multicolor MOT [23].

The $5d^2\ ^3F_2$ state is populated by pumping the atoms from the cooling cycle to the $5d6p\ ^3D_1$ state which has a lifetime of 17.0(5) ns [24]. A fraction of 30% of the atoms is driven to the $5d^2\ ^3F_2$ level. The $5d6p\ ^3D_1$ state decays to the $6s5d\ ^3D_1$, $6s5d\ ^3D_2$, $6s^2\ ^1S_0$, and $5d^2\ ^3F_2$ states. Branching to the $6s5d\ ^1D_2$, $5d^2\ ^3P_{0,1,2}$, and $5d^2\ ^1D_2$ states given in Table I is negligible. Additionally, losses from the cooling cycle of the MOT are determined in order to estimate branching ratios, which are otherwise difficult to access. The obtained results are compared with the theoretical estimations of lifetimes and branching ratios in barium (Table II).

II. EXPERIMENTAL METHOD

A thermal barium atomic beam produced from a resistively heated oven operated at 800 K is slowed to capture velocities of the MOT using the slowing laser light at wavelength $\lambda_1 = 553.7$ nm and the repump laser light at $\lambda_{ir1} = 1500.4$ nm and $\lambda_{ir2} = 1130.4$ nm. These slowed atoms are captured by trapping laser beams at wavelength λ_1 and at λ_{ir1}, λ_{ir2}, and $\lambda_3 = 659.7$ nm, which are overlapped at the center of the trapping region. The MOT fluorescence at wavelength λ_1 is detected by a photomultiplier tube. The light for cooling and trapping is generated from a dye laser pumped by a Nd:YAG laser. The laser frequencies necessary for slowing and trapping are produced with acousto-optic modulators (AOMs). The laser light at λ_{ir1} is generated by a distributed feedback (DFB) semiconductor laser, and a fiber laser for light at λ_{ir2}. Two semiconductor diode lasers at wavelengths $\lambda_2 = 667.7$ nm and $\lambda_3 = 659.7$ nm

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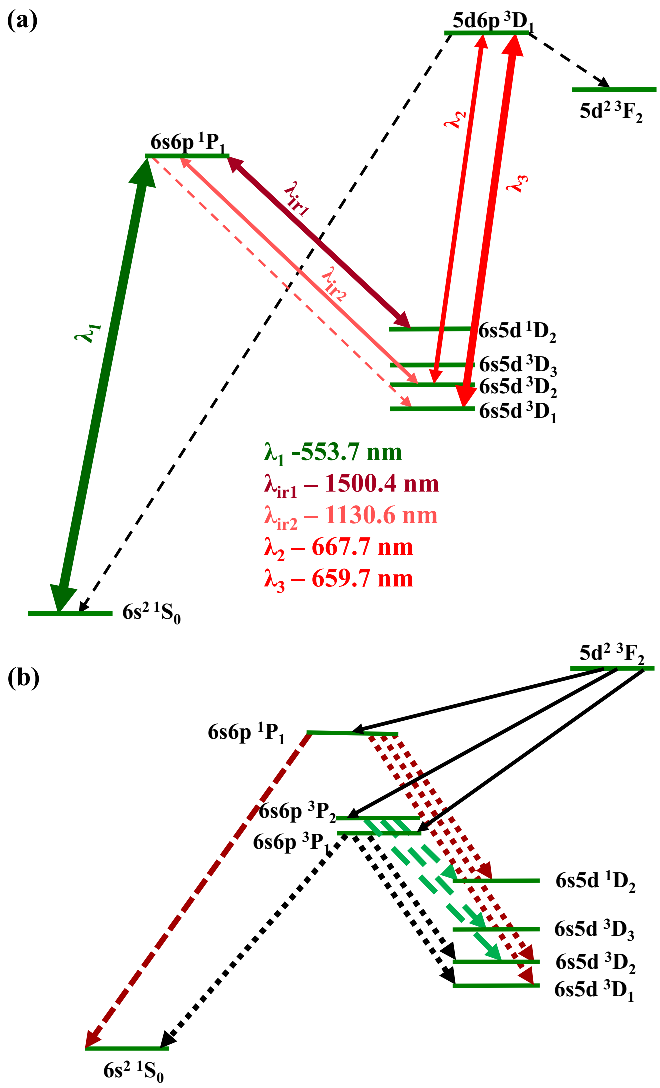

FIG. 1. (Color online) Energy levels of atomic barium. :(a) Energy levels used for laser cooling and trapping of barium. The solid arrows indicate laser-driven transitions and the dashed black arrows indicate the decay channels. The vacuum wavelengths of the laser-driven transitions are given in λ_i. (b) Decay cascade of the $5d^2\,{}^3F_2$ level to lower lying states is shown. The cascading to the $6s5d\,{}^3D_3$ state (red dashed arrows) constitutes the only loss channel from the cooling cycle of the MOT.

are used to drive the $6s5d\,{}^3D_{2,1}$-$5d6p\,{}^3D_1$ transitions. The repumping via the $6s5d\,{}^3D_{2,1}$-$5d6p\,{}^3D_1$ transitions avoids coherent Raman resonances. These resonances significantly influence the infrared transition repumping efficiency at λ_{ir2} and λ_{ir3} [23]. Further advantages are visible wavelength laser diodes with sufficient power and faster repumping due to the transition strength. Around 10 mW of light is available for the experiment allowing saturation of the $6s5d\,{}^3D_2$-$5d6p\,{}^3D_1(\lambda_2)$ transition.

In this work we focus on the lifetime measurement of the $5d^2\,{}^3F_2$ state. The barium MOT is operated in steady state by continuous multilaser repumping in the six-level system of the $6s^2\,{}^1S_0$, $6s6p\,{}^1P_1$, $6s5d\,{}^1D_2$, $6s5d\,{}^3D_2$, $6s5d\,{}^3D_1$, and $5d6p\,{}^3D_1$ states. The average population in the $5d6p\,{}^3D_1$ state is about 10^{-3} because of the small branching of the $6s6p\,{}^1P_1$ state to the $6s5d\,{}^3D_1$ state (Table I). Laser light at λ_2 is pulsed by passing through an AOM. The rise time of the diffracted beam is less than 50 ns which is more than two orders of

TABLE I. Optical transition, vacuum wavelengths, decay rates, and lifetimes in barium relevant to this work. The lifetimes and the decay rates are taken from.

Upper level	Lower level	λ (nm)	$10^7 A_{ik}$ (s^{-1})
$6s6p\,{}^1P_1$	$6s^2\,{}^1S_0$	553.7	11.9(1)[b]
8.0(5) ns[a]	$6s5d\,{}^1D_2$	1500.4	0.025(2)[b]
	$6s5d\,{}^3D_2$	1130.4	0.011(2)[b]
	$6s5d\,{}^3D_1$	1107.8	0.000 31(5)[b]
$6s6p\,{}^3P_1$	$6s5d\,{}^3D_2$	2923.0	0.0318(32)[d]
1345(14) ns[c]	$6s^2\,{}^1S_0$	791.32	0.0299(38)[d]
	$6s5d\,{}^3D_1$	2775.7	0.0123(12)[d]
	$6s5d\,{}^1D_2$	8056.5	0.000 06[e]
$6s6p\,{}^3P_2$	$6s5d\,{}^3D_3$	2552.2	0.048[e]
1.4 μs[f]	$6s5d\,{}^3D_2$	2326.0	0.01[e]
	$6s5d\,{}^3D_1$	2231.8	0.0009[e]
	$6s5d\,{}^1D_2$	4718.4	0.0001[e]
$5d6p\,{}^3D_1$	$6s5d\,{}^3D_1$	659.7	3.7(2)[h]
17.0(5) ns[g]	$6s5d\,{}^3D_2$	667.7	1.8(2)[h]
	$6s^2\,{}^1S_0$	413.4	0.15(2)[h]
	$5d^2\,{}^3F_2$	3068.2	0.063(38)[h]
	$6s5d\,{}^1D_2$	781.5	<0.0058(17)[h]
	$5d^2\,{}^3P_0$	101 70	
	$5d^2\,{}^3P_1$	140 40	
	$5d^2\,{}^3P_2$	366 04	
	$5d^2\,{}^1D_2$	8847.2	

[a]Reference [22].
[b]Reference [25].
[c]Reference [14].
[d]Reference [26].
[e]Reference [27].
[f]Reference [1].
[g]Reference [24].
[h]Reference [28].

TABLE II. Fractional branching for the decay cascade of the $5d^2\,{}^3F_2$ state. The decay from the $6s6p\,{}^3P_2$ state to the $6s5d\,{}^3D_3$ state results in loss of atoms from the cooling cycle. The last row is the fractional loss from the cooling cycle from two different calculations and this work.

Decay branching	Ref. [27]	Ref. [1]	Resultant of branching
$5d^2\,{}^3F_2\rightarrow$			
$6s6p\,{}^1P_1$	19%	2%	$6s^2\,{}^1S_0$
$6s6p\,{}^3P_1$	42%	89%	$6s^2\,{}^1S_0$
$6s6p\,{}^3P_2$	39%	9%	–
$6s6p\,{}^3P_2\rightarrow$			
$6s5d\,{}^1D_2$	≪1%	≪1%	$6s^2\,{}^1S_0$
$6s5d\,{}^3D_1$	2%	3%	$6s^2\,{}^1S_0$
$6s5d\,{}^3D_2$	17%	23%	$6s^2\,{}^1S_0$
$6s5d\,{}^3D_3$	81%	74%	Trap loss
$5d^2\,{}^3F_2\rightarrow$			This work
$6s5d\,{}^3D_3$	∼ 31.6%	∼ 6.7%	4.0(7)%

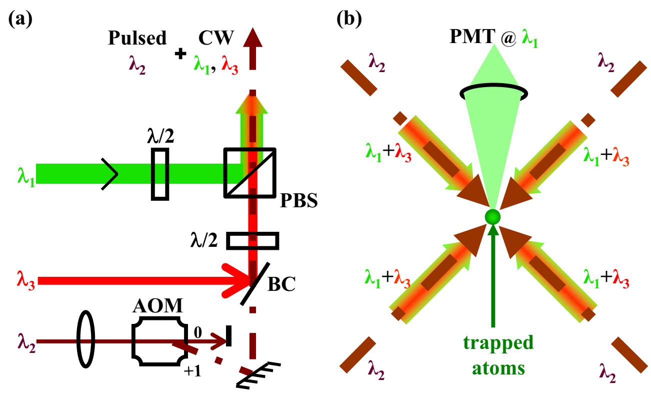

FIG. 2. (Color online) Schematic setup for the measurement of the $5d^2\,{}^3F_2$ state lifetime in a barium MOT. (a) The trapping light and the light at 667.7 nm (λ_2) and 659.7 nm (λ_3) are overlapped before they are sent to the MOT. The λ_2 laser light (dashed brown arrow) is pulsed with an acousto-optic modulator. (b) Configuration for the observation of the fluorescence from the trapped atoms (green sphere). The light at $\lambda_2 = 667.7$ nm and $\lambda_3 = 659.7$ nm optically pumps atoms to the $5d^2\,{}^3F_2$ state.

magnitude less than the typical pulse length T_p. The beam is overlapped on a beam combiner (BC) with cw laser light at λ_3. The two laser beams are further overlapped with λ_1 trapping laser beams using a combination of half-wave plates and a polarizing beam splitter (PBS) [see Fig. 2(a)]. The velocity of the trapped atoms is below 1 m/s and the typical MOT lifetime is about 1.5 s [29]. Under these conditions the atoms in the dark $5d^2\,{}^3F_2$ state travel an average distance of 0.2 mm before cascading back to the cooling cycle and remain in the detection volume of radius 3 mm for many lifetimes.

The experimental procedure is the following. The MOT is loaded for several seconds to achieve steady state in the trap. The light at wavelength λ_2 is absent during that period.

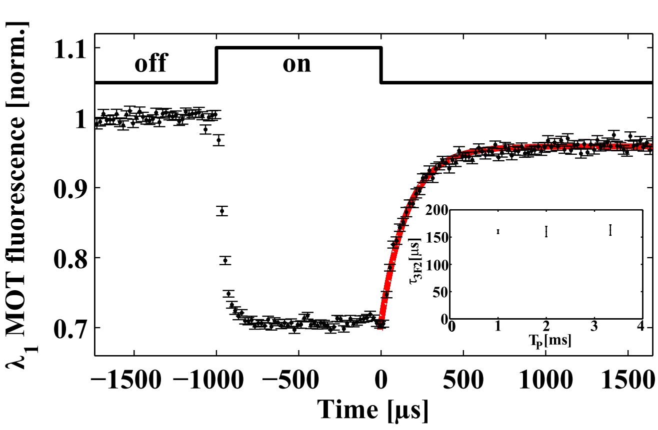

FIG. 3. (Color online) Fluorescence spectrum $s(t)$ from the trapped barium atoms detected at wavelength λ_1 from pulsed excitation of the $6s5d\,{}^3D_2$-$5d6p\,{}^3D_1$ transition at wavelength λ_2. The MOT fluorescence is normalized to the signal level before the light pulse at λ_2. The laser pulse at λ_2 is introduced from $t = -T_p \ldots 0$. The fluorescence signal is fitted for $t \geqslant 0$ to the function (1) in order to obtain the lifetime of the $5d^2\,{}^3F_2$ state. This yielded a lifetime $\tau = 160(10)$ μs and a loss $L = 4.2(2)\%$ of trapped atoms. Inset: Lifetime of the $5d^2\,{}^3F_2$ state at different λ_2 pulse lengths T_p.

The MOT population is monitored by the fluorescence at wavelength λ_1. The laser pulse at wavelength λ_2 of length $T_p =$ 1–3 ms repumps the ${}^3D_{1,2}$ population via the $5d6p\,{}^3D_1$ state of which about one-half decays directly to the $6s^2\,{}^1S_0$ ground state under emission of 413 nm photons. This fluorescence is an independent measurement of the MOT population [23]. The remaining atoms predominantly decay to the $5d^2\,{}^3F_2$ state. This removes a fraction from the cooling cycle and the λ_1 MOT fluorescence decreases until a steady state is reached. Up to 30% of the atoms can be accumulated in the $5d^2\,{}^3F_2$ state. After the λ_2 laser pulse is switched off, the decay of the $5d^2\,{}^3F_2$ state into several lower lying states [Fig. 1(b)] causes an increase in MOT fluorescence with a characteristic rise time. The time scale is dominated by the lifetime of the $5d^2\,{}^3F_2$ state because lifetimes of all intermediate states are shorter than 1.4 μs (Table I). In addition, the trap fluorescence does not reach the same level as before the laser pulse at wavelength λ_2. This missing fraction constitutes the loss L into the only untrapped $6s5d\,{}^3D_3$ state. The measurements are repeated for different λ_2 laser pulse lengths (see inset in Fig. 3). The contribution from loading of the atomic beam is estimated from the ratio of the length of the measurement cycle to the trap lifetime. This amounts to 10^{-3} for a pulse length of $T_p = 3$ ms.

III. RESULTS AND DISCUSSION

The normalized fluorescence spectrum $s(t)$ is shown in Fig. 3. For $t > 0$, $s(t)$ is described by an exponential decay function and a loss fraction (Fig. 3)

$$s(t) = 1 - \gamma \exp(-t/\tau_c) - L, \quad \text{for } t > 0, \tag{1}$$

where γ is the fraction of trapped atoms in the $5d^2\,{}^3F_2$ state, τ_c is the characteristic time scale of the decay back into the cooling cycle of the MOT and L is the fractional loss of trapped atoms. The decay time τ_c is dominated by τ of the $5d^2\,{}^3F_2$ state since all other decay times in the cascade are orders of magnitude faster than τ. The lifetime of the $5d^2\,{}^3F_2$ state determined by fitting to Eq. (1) is $\tau = 160(10)$ μs (Fig. 3) and the trap loss fraction, $L = 4.2(2)\%$.

The depletion of the MOT fluorescence due to the λ_2 laser pulse is a measure of the steady-state population in the $5d^2\,{}^3F_2$ state. The losses from the trap then depend on the branching to the only untrapped $6s5d\,{}^3D_3$ state and the probability P to pump into the $5d^2\,{}^3F_2$ state during the length T_p of the laser pulse, which is given by

$$P = \frac{\int_{-T_p}^{0} [1 - s(t)]\, dt}{\Delta t}, \tag{2}$$

where $s(t)$ is the normalized MOT fluorescence signal [Eq. (1)] and Δt is the average time required for cycling once through the $5d^2\,{}^3F_2$ state. The cycling time Δt is the sum of τ and the time required for pumping atoms into the $5d^2\,{}^3F_2$ state. The latter is estimated as 150(50) μs from the branching of the $6s6p\,{}^1P_1$ state to the $6s5d\,{}^3D$ states and from the parameters of the λ_1 trapping beams. In the six-level barium MOT system, the loss ℓ from the cooling cycle for cycling once through the $5d^2\,{}^3F_2$ state is

$$\ell = \frac{L}{P}. \tag{3}$$

The loss is determined to be $\ell = 4.0(7)\%$ from several measurements at different λ_2 pulse lengths, T_P. This cascading fraction from the $5d^2\ {}^3F_2$ state is the fractional loss to the $6s5d\ {}^3D_3$ state corresponding to the trap loss as all other states decay back to the cooling cycle. This is in agreement with the calculated branching fractions from the $5d^2\ {}^3F_2$ state to the $6s5d\ {}^3D_3$ state (Table II).

IV. CONCLUSIONS

To summarize, we have shown that cold atoms allow access to lifetime measurements of highly excited states, in particular, lifetimes of states which cannot be determined in atomic beams. The lifetime of the $5d^2\ {}^3F_2$ state in barium is measured using a magneto-optical trap. The measured value of 160(10) μs is in good agreement with the theoretically calculated value of 190 μs. Further, the fractional loss of the trapped atoms is measured as 4.0(7)% and agrees with an estimate based on calculated branching fractions of the $5d^2\ {}^3F_2$ state [1]. This test of the atomic theory gives confidence in the predictive power for heavy alkaline-earth elements, in particular radium, which is relevant for experimental searches for symmetry violation.

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