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Lifetime determination of the 5d\(^2\) 3\(^F_2\) 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\(^2\) 3\(^F_2\) 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\(^2\) 3\(^F_2\) 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) \(\mu\)s. This is in good agreement with the theoretically calculated lifetime of 190 \(\mu\)s [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\(^2\) 3\(^F_2\) state in Ba.

Laser cooling on the strong 6s\(^2\) 1\(^S_0\)-6s6p 1\(^P_1\) transition of barium requires us to drive several repump transitions (Fig. 1) at the same time [22,23]. The 6s6p 1\(^P_1\) state branches mostly to the 6s\(^2\) 1\(^S_0\) ground state and to 0.3% to the metastable 6s5d 3\(^D_2\) and 6s5d 3\(^D_2\) states. The decay rates from the 6s6p 1\(^P_1\), 6s6p 3\(^P_{1,2}\), and 5d6p 3\(^D_1\) 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\(^2\) 1\(^S_0\)-6s6p 1\(^P_1\) transition, about half of the atoms are in the metastable D states in steady state of this multicolor MOT [23]. The 5d\(^2\) 3\(^F_2\) state is populated by pumping the atoms from the cooling cycle to the 5d6p 3\(^D_1\) state which has a lifetime of 17.0(5) ns [24]. A fraction of 30% of the atoms is driven to the 5d\(^2\) 3\(^F_2\) level. The 5d6p 3\(^D_1\) state decays to the 6s5d 3\(^D_1\), 6s5d 3\(^D_2\), 6s\(^2\) 1\(^S_0\), and 5d\(^2\) 3\(^F_2\) states. Branching to the 6s5d 3\(^D_2\), 5d2 3\(^P_{0,1,2}\), and 5d\(^2\) 3\(^D_2\) 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 \(\lambda_{r1} = 1500.4\) nm and \(\lambda_{r2} = 1130.4\) nm. These slowed atoms are captured by trapping laser beams at wavelength \(\lambda_1\) and at \(\lambda_{r1}, \lambda_{r2}\), and \(\lambda_3 = 659.7\) nm, which are overlapped at the center of the trapping region. The MOT fluorescence at wavelength \(\lambda_1\) 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 \(\lambda_{r1}\) is generated by a distributed feedback (DFB) semiconductor laser, and a fiber laser for light at \(\lambda_{r2}\). Two semiconductor diode lasers at wavelengths \(\lambda_2 = 667.7\) nm and \(\lambda_3 = 659.7\) nm

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are used to drive the 6s5d 3D2-5d6p 3D1 transitions. The repumping via the 6s5d 3D2-5d6p 3D1 transitions avoids coherent Raman resonances. These resonances significantly influence the infrared transition repumping efficiency at λ1 and λ3 [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 3D2-5d6p 3D1(λ2) transition.

In this work we focus on the lifetime measurement of the 5d 2 F2 state. The barium MOT is operated in steady state by continuous multilaser repumping in the six-level system of the 6s2 1S0, 6s6p 1P1, 6s5d 1D2, 6s5d 3D1, and 5d6p 3D1 states. The average population in the 5d6p 3D1 state is about 10⁻³ because of the small branching of the 6s6p 1P1 state to the 6s5d 3D1 state (Table I). Laser light at λ3 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 magnitude.

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The MOT population is monitored by the fluorescence at wavelength $\lambda_1$. The laser pulse at wavelength $\lambda_2$ of length $T_p = 1-3$ ms repumps the $^3D_{1,2}$ population via the $5d^2\,^3F_2$ 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 $\lambda_1$ 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 $\lambda_2$ 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 $\mu$s (Table I). In addition, the trap fluorescence does not reach the same level as before the laser pulse at wavelength $\lambda_2$. This missing fraction constitutes the loss $L$ into the only untrapped $6s5d\,^3D_3$ state. The measurements are repeated for different $\lambda_2$ 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)

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

where $\gamma$ is the fraction of trapped atoms in the $5d^2\,^3F_2$ state, $\tau_c$ 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 $\tau_c$ is dominated by $\tau$ of the $5d^2\,^3F_2$ state since all other decay times in the cascade are orders of magnitude faster than $\tau$. The lifetime of the $5d^2\,^3F_2$ state determined by fitting to Eq. (1) is $\tau = 160(10)$ $\mu$s (Fig. 3) and the trap loss fraction, $L = 4.2(2)\%$.

The depletion of the MOT fluorescence due to the $\lambda_2$ 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

$$P = \frac{\int_0^{T_p} [1 - s(t)] \, dt}{\Delta t},$$

where $s(t)$ is the normalized MOT fluorescence signal [Eq. (1)] and $\Delta t$ is the average time required for cycling once through the $5d^2\,^3F_2$ state. The cycling time $\Delta t$ is the sum of $\tau$ and the time required for pumping atoms into the $5d^2\,^3F_2$ state. The latter is estimated as 150(50) $\mu$s from the branching of the $6s6p\,^3P_1$ state to the $6s5d\,^3D$ states and from the parameters of the $\lambda_1$ trapping beams. In the six-level barium MOT system, the loss $\ell$ from the cooling cycle for cycling once through the $5d^2\,^3F_2$ state is

$$\ell = \frac{L}{P}.$$
The loss is determined to be \( \ell = 4.0(7)\% \) from several measurements at different \( \lambda_2 \) pulse lengths, \( T_\ell \). This cascading fraction from the \( 5d^2 F_2 \) state is the fractional loss to the \( 6s5d^3 D_3 \) 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 \( 5d^2 F_2 \) state to the \( 6s5d^3 D_3 \) 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 \( 5d^2 F_2 \) state in barium is measured using a magneto-optical trap. The measured value of \( 160(10) \mu s \) is in good agreement with the theoretically calculated value of \( 190 \mu s \). 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 \( 5d^2 F_2 \) 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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