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Radiative lifetime measurement of ultracold cesium Rydberg states by a simplified optical pumping method

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We demonstrate one simplified all-optical method to measure the radiative lifetime of ultracold cesium (Cs) Rydberg atoms. This method is based on photodetection of one ground state atomic absorption in a ladder-type electromagnetic induced transparency (EIT), which contains one ground state, one intermediate state, and one Rydberg state. In the presence and absence of optical pumping between the Rydberg state and the intermediate state, the absorption difference with varying delay time can reconstruct the population evolution of target Rydberg atoms. By using this method, the radiative lifetimes of 50 $S_{1/2}$ and 50 $D_{5/2}$ of Cs atoms are measured to be 53 (9) μs and 42 (9) μs, respectively, which are consistent with theoretical calculations. The agreements show the reliability of our presented method, which can provide a route for researching light–matter interaction behavior without the need to quantify absorption characteristic.

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1. INTRODUCTION

Rydberg atoms have some distinctive properties, such as a large radius, strong coupling with external fields, small energy separation, large dipole–dipole interaction, and long radiative lifetimes. Benefiting from these merits, they have great applications in quantum sensing [1–3], quantum information [4,5], and quantum simulation [6–8]. The lifetime measurements of Rydberg atoms are not only of great importance for the theoretical calculation of dipole matrix elements [9], oscillator strengths [10], and polarizability [11], but also for the accuracy test of wave functions [12–14], blackbody radiation [15–17], and photoionization rates [18]. To our knowledge, there mainly exist three experimental methods for measuring Rydberg atomic lifetime: fluorescence detection [19,20], pulsed field ionization [21–29], and all-optical detection [30].

The fluorescence detection monitors the fluorescence from the Rydberg state to another specific state by using a photomultiplier or a monochromator. The evolution of Rydberg atomic population with the delay time is recorded, and then the lifetime of the Rydberg atoms is obtained. Marek and Niemax utilized a pulsed dye laser and delayed coincidence method to measure cesium (Cs) lifetime of the 7, 8, 9 $^2P_{1/2}$, and $^2P_{3/2}$ levels in pure Cs vapor and in a mixture of Cs and Xe [19]. Gallagher *et al.* have determined the radiative lifetimes of the $7s-11s$ and $5d-9d$ states of K with a time and wavelength resolved laser-induced-fluorescence approach [20]. The fluorescence detection is mainly applicable to Rydberg states with principle number $n<15$ because the collisions and super-radiance effects will decrease the accuracy of measurement for the higher-lying Rydberg states [21].

Pulse field ionization ionizes Rydberg atoms by means of a high-voltage pulse, and the resulting ions are detected through some special detection devices such as the micro-channel plate or single-photon counting module. Using this method, Spencer *et al.* [22] and Hansen [23] reported radiative lifetimes of Na and Li, respectively. In Ref. [24], Magalhães *et al.* observed the radiative lifetime of the $27D$ state of ^{85}Rb, which was a high-precision measurement of the radiative lifetime through a sample of cold atoms. Marcassa's group successively determined the lifetime of ^{85}Rb for principal quantum number between $n=26$ and 45 [25,26]. Feng *et al.* studied the lifetime of Cs Rydberg atoms, and they found that collisions and super-radiance effects have a great impact on the accuracy of lifetime measurement at high densities of Rydberg atoms [21]. It is universally acknowledged that blackbody radiation will greatly affect the lifetime of Rydberg atoms. To suppress the effect of

blackbody radiation, Magnani *et al.* introduced a finite size metal vacuum chamber to measure the lifetimes of nS states of Rb Rydberg atoms in the range of $40 \leq n \leq 70$ [27]. Although pulsed field ionization is more extensive for higher Rydberg states, it becomes difficult to measure lifetime of the target state with principal quantum numbers exceeding 60 because the ionization thresholds of adjacent Rydberg states become indistinguishable [28]. In high Rydberg atoms, the influence from blackbody radiation becomes observable by inducing transfer between selection-ruler allowed states. To exclude this influence, Branden *et al.* [29] utilized a millimeter wave to transfer the chosen target Rydberg state to another monitor state for measuring the lifetime of the target Rydberg state. It is worth noting that another disadvantage of this method is that it is only appropriate when there is an electron or ion detector in chamber.

In recent years, many experiments of Rydberg atoms rely on all-optical operation and detection, in which electromagnetic induced transparency (EIT) is the most used method [31–33]. In this kind of system, Mack *et al.* introduced a widely suitable, all-optical method for measuring the lifetime of the Rydberg state [30]. In their method, it is required to quantify the population of the target Rb Rydberg state by calibrating the number of ground state atoms with optical density. In this paper, we do not need to calibrate the number of the initial ground state atoms, rather, we only need to directly evaluate the Rydberg atom population by monitoring the changes of probe laser intensity in the presence and absence of optical pumping. We then apply this optical pumping method to measure the radiative lifetimes of ultracold Cs Rydberg atoms in $50S_{1/2}$ and $50D_{5/2}$ states and compare those with the theoretical calculations.

2. EXPERIMENTAL SETUP

The schematic of our experimental setup and relevant Rydberg energy level diagram of Cs atoms is shown in Fig. 1(a). The experiment operates in a closed stainless-steel vapor cell, and Cs vapor from a reservoir enters the main chamber, which lies in air with room temperature of around 300 K. Vacuum background pressure is about 3.0×10^{-6} Pa, and the axial field gradient of the quadrupole magnetic field is approximately 15 G/cm. Two Littrow external-cavity diode lasers (Toptica

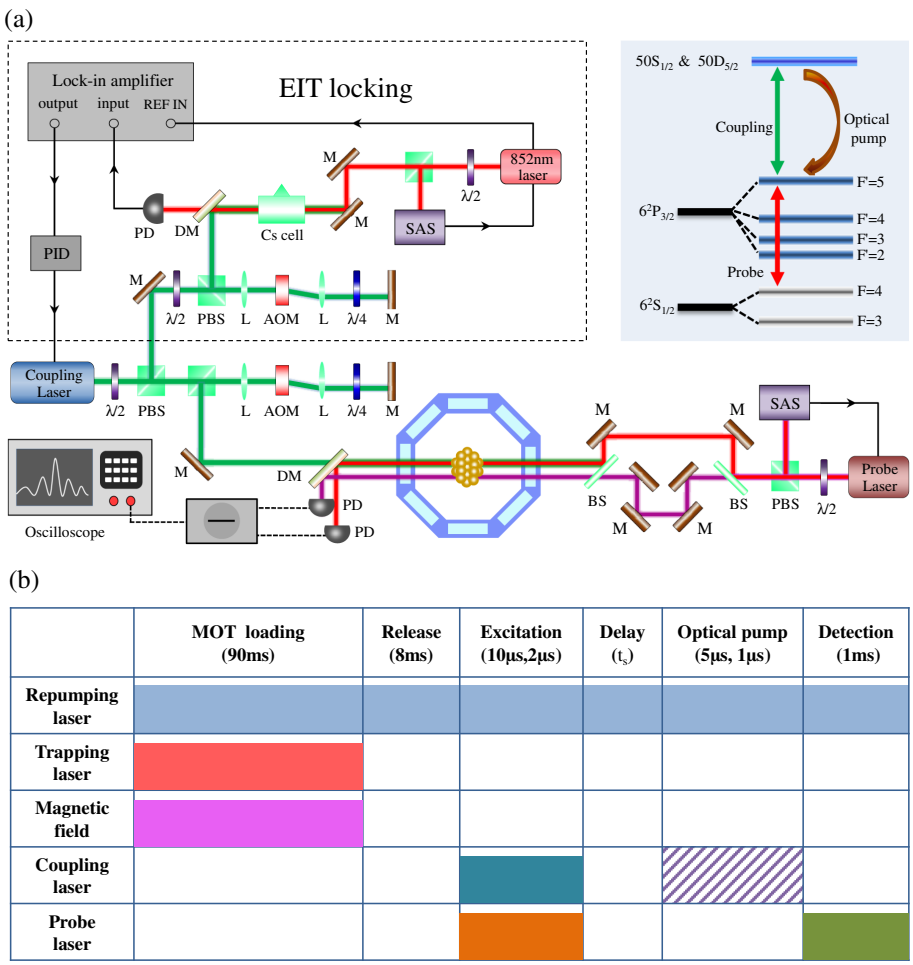

Fig. 1. (a) Schematic diagram of the experimental setup. $\lambda/2$, half-wave plate; PBS, polarization beam splitter; L, lens; AOM, acousto-optic modulator; $\lambda/4$, quarter-wave plate; M, mirror; PD, photodetector; DM, dichroic mirror; SAS, saturated absorption spectroscopy; BS, beam splitter. Inset, energy level diagram. The probe laser is fixed at the resonance transition $6^2S_{1/2}(F=4) \rightarrow 6^2P_{3/2}(F'=5)$, while the coupling laser is locked to the $6^2P_{3/2}(F'=5) \rightarrow 50S_{1/2}/50D_{5/2}$. (b) Time sequence in our experiment. The shadow with different colors means "on," the blank means "off," and the rectangle filled with diagonal lines means "selectively on or off."

DL100) separately generate trapping and repumping lasers. The acousto-optic modulator (AOM) is applied to adjust the trapping laser to red detuned about two natural linewidths (10 MHz) from the $6^2S_{1/2}(F=4) \rightarrow 6^2P_{3/2}(F'=5)$ atomic transition. The repumping laser is locked to the $6^2S_{1/2}(F=3) \rightarrow 6^2P_{3/2}(F'=4)$ atomic transition. For trapping lasers, the diameters are 15 mm, and the total power is 9 mW. The repumping beams are also 15 mm in diameter with a total power of approximately 5 mW. The translational temperature of cold atomic sample is approximately 100 μK measured by the time-of-flight imaging method.

The Cs ground state $6^2S_{1/2}$ $(F=4)$, excited state $6^2P_{3/2}$ $(F'=5)$, and Rydberg states $50S_{1/2}/50D_{5/2}$ consist of a three-level system. We obtain ultracold Rydberg atoms via a two-photon transition. The first excitation laser is fixed at the resonance transition $6^2S_{1/2}$ $(F=4) \rightarrow 6^2P_{3/2}$ $(F'=5)$ as a probe laser. While the second laser is locked at the $6^2P_{3/2}$ $(F'=5) \rightarrow 50S_{1/2}/50D_{5/2}$ as a coupling laser. For the probe laser, the Gaussian radius and power are around 40 μm and 2 μW, respectively, while for the coupling laser, the Gaussian radius and power are around 42 μm and 80 mW, respectively. The coupling laser is divided into two beams through a polarization beam splitter (PBS). One beam is used for frequency locking by a cascade EIT involving target Rydberg states in a Cs vapor cell at room temperature [34], in which a 852 nm laser is locked by saturated absorption spectroscopy (SAS) technique. The other beam acts as the coupling beam. For the purpose of reducing the fluctuation of the probe signal, we divide the probe laser into two beams using a beam splitter (BS). One is regarded as the probe beam actually overlapping with the coupling beam, while the other is regarded as a reference beam without overlapping with the coupling beam. We use two photodetectors (PDs) to observe the reference beam and probe beam behind a dichroic mirror (DM). The desirable signal is the subtraction of these signals detected by the two PDs. It is proved that the signal-to-noise ratio is improved by this way.

The time sequence is presented in Fig. 1(b), which is divided into six stages. (1) Loading: ultracold atoms are loaded into a standard MOT, where the trapping laser and the magnetic field are kept on around 90 ms. (2) Release: the obtained cold atoms freely release for 8 ms to ensure a low initial atom density where collisions and super-radiance effects become weak for the Rydberg atoms. (3) Excitation: the probe laser and coupling laser are turned on for producing Rydberg atoms. (4) Delay: the probe laser and coupling laser are turned off. The evolution curve of Rydberg atomic population is obtained by changing the delay time. (5) Optical pump (OP): in this procedure, the coupling laser can be selected to turn on or off. The laser is kept on in order to depump the target Rydberg atoms to the intermediate state, and they soon decay to the ground state, while the laser is turned off in order to make the target Rydberg atoms naturally decay to ground state. (6) Detection: in this period, only the probe laser is turned on. A repumping laser was always turned on to keep the ground state atoms from accumulating in the $F=3$ state. The repetition time is 100 ms in the experimental condition.

3. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 2(a) shows the evolution curves of probe intensity for the $50S_{1/2}$ state with the delay time $t_s = 0$. We turn off the coupling laser after excitation with an excitation time of 10 μs, then the atoms in the $50S_{1/2}$ state will naturally decay to a number of intermediate states. As the lifetimes of these intermediate states are short, they will decay toward the ground state quickly. With the accumulation of the ground state atoms, the intensity of probe laser after the atoms will decrease. When the atoms of the $50S_{1/2}$ state all fall to the ground state, the intensity of the probe laser will decrease to a constant value in the short time. The green curve of Fig. 2(a) represents our measured probe intensity. We begin to record experimental data after an OP time, which is chosen to be 5 μs for $50S_{1/2}$ state. Hereafter, we call this process as OP off process. We record the intensities of probe and reference beams as $I_{\text{probe off}}$ and $I_{\text{ref off}}$, respectively, at this moment. Thus, the initial transmission intensity can be derived as $I_{\text{OP off}} = I_{\text{probe off}} - I_{\text{ref off}}$.

If the coupling laser is turned on after excitation, it will act as a depumping laser. It depumps a fraction of atoms in the $50S_{1/2}$ state toward the specific state $6^2P_{3/2}(F'=5)$, then atoms soon reach the ground state. During the process, the probe laser is turned off to avoid reproduction of Rydberg atoms. The pink curve of Fig. 2(a) represents our measured probe intensity under this case. Similarly, we call this process as OP on process. This time the intensities of the probe and reference beams are labeled as $I_{\text{probe on}}$ and $I_{\text{ref on}}$, respectively. Now the transmission intensity is labeled as $I_{\text{OP on}} = I_{\text{probe on}} - I_{\text{ref on}}$. During the OP on process, the number of ground state atoms is larger owing to the action of the depumping laser, so the intensity of the probe laser is weaker. We need to notice that there is still a fraction of atoms without dumping during the OP on process, and they will decay naturally as described above. But the difference of probe laser intensity between the OP off and OP on process can directly reflect the depumped population of $50S_{1/2}$ state atoms, which can be described as $\alpha P(t_s) = I_{\text{OP off}} - I_{\text{OP on}}$, where α is depumping rate and t_s is delay time. The blue curve of Fig. 2(a) represents the obtained $\alpha P(t_s)$.

Figure 2(b) shows the depumped Rydberg population $\alpha P(t_s)$ as a function of time for different delay times. The blue, pink, and orange curves, respectively, represent the cases of $t_s = 0$ μs, 16 μs, and 32 μs. Solid lines represent the exponential fitting with the formula $y = y_0 + Ae^{-(t-t_s)/t_1}$. The fitted values are 1.041 (14), 0.614 (13), and 0.342 (14), corresponding to $t_s = 0$ μs, 16 μs, and 32 μs.

Figure 3(a) shows the depumped Rydberg population $\alpha P(t_s)$ as a function of delay time t_s for Rydberg state $50S_{1/2}$. Each data point is the average value of multiple sets of measurements with standard deviation, which is different from the fitting error in Fig. 2(b). For different t_s, α is a constant due to the same time and power of the optical pumping laser we used. Hence, $\alpha P(t_s)$ is proportional to the population of Rydberg atoms. We fit the experimental data with the formula $y = y_0 + Ae^{-t_s/\tau}$ and obtain the radiative lifetime of the $50S_{1/2}$ state with the value of 53 (9) μs. The offset y_0 is included because it is difficult and also unnecessary to make all the parameters of probe and reference beams exactly the same. Theoretically, the radiative lifetimes of Rydberg atoms with $15 < n < 80$ can be calculated

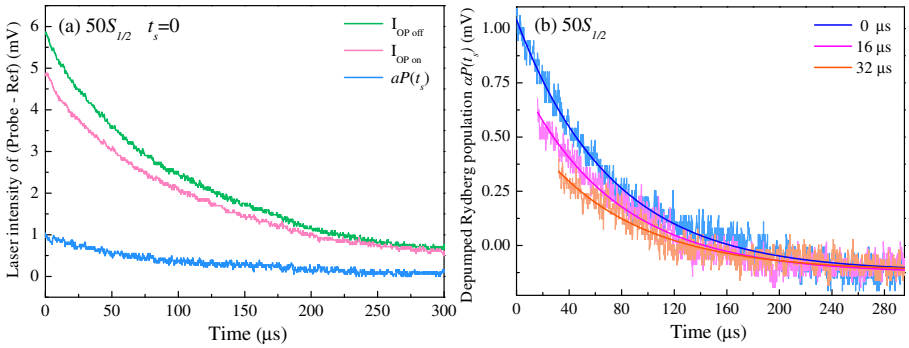

Fig. 2. (a) Evolution curves of probe intensity for the $50S_{1/2}$ state with delay time $t_s = 0$. (b) Measured $\alpha P(t_s)$ of the $50S_{1/2}$ state for different delay times. Solid lines represent exponential fitting.

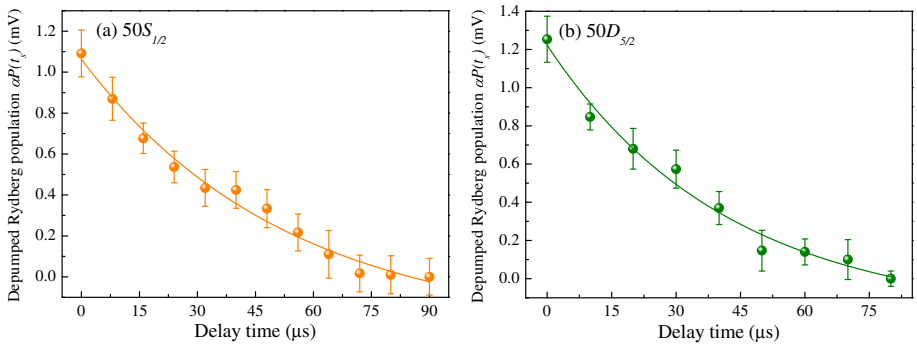

Fig. 3. Measured $\alpha P(t_s)$ of (a) the $50S_{1/2}$ state and (b) the $50D_{5/2}$ state with different t_s. The solid lines represent exponential fitting.

by the model developed by Beterov [35] with the accuracy better than 5%. The theoretical lifetime of Cs atoms in the $50S_{1/2}$ is 60.4 (3.0) μs at 300 K. Even our measurement is consistent with the theoretical calculation in view of the uncertainty, but it still shows as systematically lower. We think this deviation mainly arises from collisions and super-radiance effects, which are difficult to quantify. Besides of these two systematic errors, other random errors include instability of cold atom samples, intensity fluctuation of probe laser, noise and response of PD, etc.

In order to further check the applicability of this method, we measured the lifetime of the $50D_{5/2}$ in Fig. 3(b). The excitation and optical pumping times are chosen to be 2 μs and 1 μs, respectively, due to the larger transition electric dipole moment between $50D_{5/2}$ and $6^2P_{3/2}$ ($F' = 5$). The radiative lifetime of the $50D_{5/2}$ is fitted to be 42 (9) μs, and the calculated value is 46.6 (2.3) μs from the theory above. In Ref. [18], Tallant *et al.* have reported the radiative lifetime of $50D_{5/2}$ is 46 (3) μs via an electric field ramp. Thus, our measurement on this state shows similarity as the case of $50S_{1/2}$, supporting that the demonstrated method is reliable.

4. CONCLUSION

We demonstrate a simpler and operable all-optical method for measuring the lifetime of Rydberg atoms. This method is different from the traditional fluorescence and selective field ionization methods. It is also applicable to some high Rydberg states, does not need to accurately consider the ionization threshold of the target Rydberg state, and is without the need of the specific detector in experimental systems. Compared with the previous all-optical method in Ref. [30], we do not need to calibrate the number of the initial ground state atoms. Thence, the method provides a route for researching light–matter interaction behavior without the need to quantify absorption characteristic. With the help of this method, we measured the radiative lifetimes of $50S_{1/2}$ and $50D_{5/2}$ of Cs atoms, which are in agreement with existing theoretical and experimental values in view of the uncertainty. The main factors limiting the precision of lifetime measurement include stability of atom density, power stability of probe and reference beams, and response characteristics of two PDs. If these factors can be improved, e.g., a laser feedback system to servo the number of atoms, a power stabilization system to stabilize laser beams, or a balanced homodyne detector to unify the response characteristics, the measurement accuracy may be greatly improved.

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