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Measurement of Rydberg-state lifetimes using cold trapped atoms

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In this paper, we demonstrate the use of a cold trapped sample of neutral atoms to perform high-precision measurement of the lifetime for a sequence of Rydberg states of ^{85}Rb. States of principal quantum number lying between $n=31$ and $n=45$ are produced and their lifetimes measured using field ionization. Experimental results are compared with existing theories. This is an important demonstration concerning the possibilities of high-precision spectroscopy in the time domain using cold atomic samples.

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Accurate measurements of the lifetimes of Rydberg states can provide powerful tests for theoretical calculations of dipole matrix elements, oscillator strengths, core polarizabilities, and influence of blackbody radiation on radiative lifetimes. Alkali-metal atoms have been used both theoretically and experimentally as prototypes for the study of these problems [1]. Although the alkali-metal atoms are among the easiest to treat theoretically, the available predictions for the Rydberg-state lifetimes present variations from 5% to 15% [2–6]. The error bars of the experimental results are even worse, for $n>15$ they can be larger than 25% [7]. This large uncertainty comes mainly from the fact that most of the works done in this field have used conventional techniques to measure lifetimes, relying on the observation of atomic fluorescence decay of thermal atoms either in cells or in atomic beams. For levels where $n>20$, super-radiance occurs rapidly, making it impossible to perform a fair lifetime measurement. Another important effect present in these experiments is the blackbody radiation, which can appreciably alter the observed decay rates. At room temperature, the blackbody radiation can decrease the lifetime for as much as 40%. Spencer and co-workers [8] were the first to eliminate this influence measuring the lifetime of sodium Rydberg states using an atomic sodium beam and a liquid-helium-cooled environment. They were able to measure lifetimes smaller than 25 μs at a temperature of 30 K. However, after this pioneerins work, no more studies have been done in order to either eliminate or decrease the effect of collisions and super-radiance on lifetime measurements using thermal atoms. Recently, we had demonstrated the possibility to obtain a high-precision measurement of the lifetime of the ^{85}Rb $27D$ Rydberg state by using pulsed field ionization on a sample of cold trapped atoms [9]. In this work, we measured the lifetime of S, P, and D states of ^{85}Rb as a function of the principal quantum number n using a sample of cold atoms. Due to the ultralow velocities of the atoms and the low collision rate, we were able to measure longer lifetimes (>100 μs). This report begins with a description of the experimental setup, followed by the results and the discussion, in which we present a comparison of our measurement with existing theory.

Our experiment is composed of a magneto-optical trap (MOT) operating in a closed stainless steel vapor cell. The Rb vapor from a reservoir kept at $-20\,^{\circ}$C effuses through a valve into the main chamber maintained at a background pressure of 10^{-10} torr. The trapping laser beam is provided by an injection-locked diode laser tuned -5 MHz of the red of the atomic $5S_{1/2}(F=3)\rightarrow 5P_{3/2}(F'=4)$ transition (ω_1). This laser beam is divided into three equally intense MOT beams, which are mutually orthogonal and retroreflected, intersect at the center of the quadrupole magnetic field. The magnetic field is generated by a pair of anti-Helmholtz coils located outside the chamber and its axial field gradient is about 10 Gauss/cm near the center. Another laser beam, provided by a second diode laser, passes through an acousto-optical modulator (AOM), generating an extra laser beam tuned to the red of the $5S_{1/2}(F=2)\rightarrow 5P_{3/2}(F'=3)$ transition ω_2, which works as a repumper. A radio frequency (rf) switch connected to this AOM allows the trap to operate in such way that first there is a trapping phase (on the order of 50 ms) with ω_1 and ω_2 present, and then a probe phase (on the order of 1 ms), where only ω_1 is present. During the probe phase, ω_2 is turned off and all the atoms are optically pumped to the ground state $5S_{1/2}(F=2)$ by ω_1 in about 20 μs. After the optical pumping, the trapping force does not act on the atoms anymore, and the atomic sample is allowed to expand for about 1 ms before the excitation to the Rydberg states takes place. This procedure decreases the atomic density, avoiding collisional and super-radiance effects on the radiative lifetime [9]. After the expansion an extra laser frequency ω_3, nearly resonant to the $5P_{3/2}\rightarrow nl$ transition, is provided by a pulsed dye laser (1 mJ/pulse, 4 ns, repetition rate 20 Hz, $\lambda\sim 482$ nm) pumped by the third harmonic of a Nd:YAG (yttrium aluminum garnet) laser. The frequency of ω_3 is measured by a high resolution-pulsed wavemeter (Burleigh WA4500), which presents a resolution of 0.01 cm^{-1}. According to Fig. 1, during the laser pulse duration the atoms are coupled from the $5S_{1/2}$ ground-state laser to a virtual level (dashed line in Fig. 1) by ω_1 and from there to the Rydberg state by ω_3. Therefore, the transition $5S_{1/2}\rightarrow nl$ takes place via a two-photon process, nearly resonant with the intermediate level $5P_{3/2}$, which results in a considerable enhancement of the absorption cross section. The total trap intensity used in our study was about 50 mW/cm^2. The number of trapped atoms is measured by

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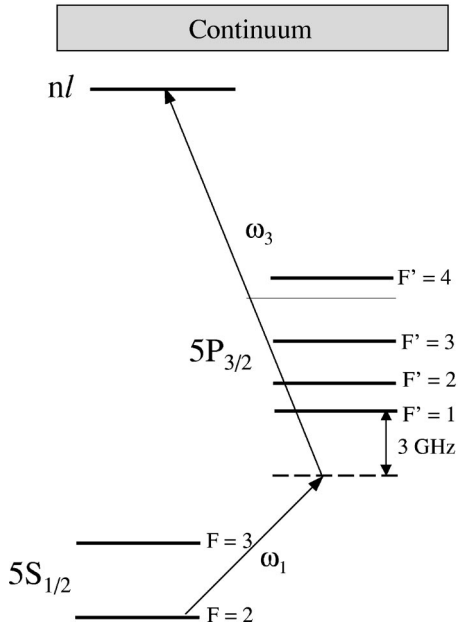

FIG. 1. Energy level diagram of rubidium, showing the relevant transitions for the lifetime measurement. In the probe cycle the atoms are optically pumped to the hyperfine ground state $5S_{1/2}(F=2)$. The frequency ω_1 is tuned to the red of the $5S_{1/2}(F=3) \rightarrow 5P_{3/2}(F'=4)$ transition and the frequency ω_3 is nearly resonant to the $5P_{3/2} \rightarrow nl$ transition.

imaging their fluorescence onto a calibrated photomultiplier and the volume of the cloud (before and after expansion) can be derived from pictures taken with a camera (charge-coupled device). These two quantities are used to calculate the atomic density. With our trap operating under optimal conditions, the total number of atoms before expansion was $N_{\mathrm{Rb}} \sim 5 \times 10^7$ and the density was about 2×10^{10} atoms/cm^3. The cold cloud of atoms is formed between two metal grids 3.2 cm apart from each other. One of the grids is grounded while a high-voltage (HV) pulse (rise-time 1 μs, peak voltage $\sim$1.2 kV) is applied in the other one to ionize the Rydberg atoms. The HV pulse stays on for over 2 μs, and the ions produced are detected by a channeltron particle multiplier placed behind the second grid, and are analyzed by a boxcar integrator. The acoustic-optical modulator, the pulsed laser, and the boxcar integrator are synchronized by a pulse delay generator (local oscillator).

The time sequence of the events is presented in Fig. 2. The sequence begins with the turning off of the repumper light, then the sample is allowed to expand for a time of the order of 1 ms before the laser pulse excites the Rydberg state. After a given delay time Δt from the pulsed excitation, the HV pulse is turned on and the boxcar integrator is gated to compensate for the time of flight of the ions. The signals are obtained by varying the delay time Δt and recording the ionized atoms by the HV pulse, which enables one to measure the fraction of Rydberg atoms that survive spontaneous decay between the excitation (laser pulse) and the ionization.

At high atomic density ($>10^{10}$ atoms/cm^3) in the trap (i.e., before expansion), collisions and super-radiance effects may influence the exponential decay. However, for our experiment we always operate the trap at low Rydberg atomic density regime ($n \sim 5 \times 10^6$ atoms/cm^3) obtained by controlling the expansion and the pulsed laser intensity. In this regime, the average time between collisions is much higher than the lifetime and the atomic density is smaller than required for superradiance effect [9]. Therefore, the atoms are basically individualized and the lifetime of the excited level is only influenced by the blackbody radiation.

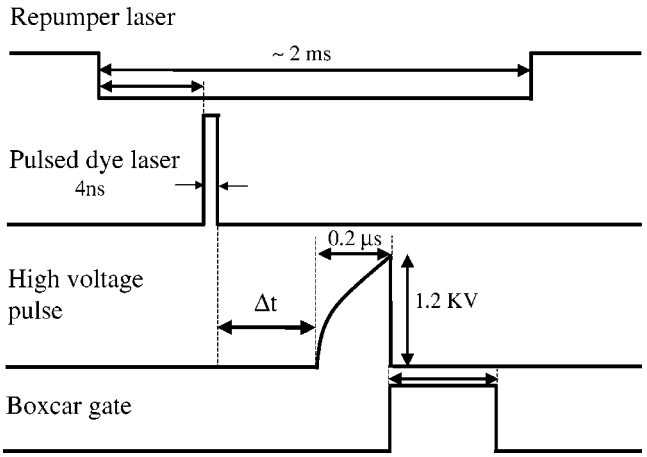

FIG. 2. Event sequence in the experiment. At $t=0$ the repumper is turned off and the sample is allowed to expand for $\sim$1 ms. The Rydberg state is excited by the pulsed laser after the expansion. After a given delay time Δt from the pulsed excitation, the HV pulse is turned on and the boxcar integrator is gated to compensate for the time of flight of the ions.

The observation of the Rydberg population as a function of the time allows one to obtain the lifetime for a given state, as shown in our previous work [9]. Figures 3(a)–3(b) present the measured lifetimes for *S*, *P*, and *D* states as a function of the principal quantum number *n*. The error bars were taken as the variance of several obtained measurements. Due to the linewidth of our pulsed laser, the lifetimes presented here for the nP and nD states are an average for both fine structure levels ($P_{1/2,3/2}$ and $D_{3/2,5/2}$). From Figs. 3(a) and 3(b), we can observe that nP states have longer lifetimes than nS and nD states. This fact is due to their small oscillator strengths connecting to the lower states, which leads to a smaller Einstein *A* coefficient when compared to nS and nD states; and therefore to longer lifetimes [1]. On the other hand, due to their long lifetimes, the nP states are the most affected states by blackbody radiation. One can write

$$\frac{1}{\tau} = \frac{1}{\tau_o} + \frac{1}{\tau_{bb}}, \tag{1}$$

where the total lifetime τ is now a composition of the atomic lifetime τ_o and the effect caused by blackbody radiation τ_{bb}, which is highly dependent on the environmental temperature felt by the atoms; in our case the vacuum chamber is at 300 K. Evaluation of τ_o requires knowledge of the dipole matrix elements between the level under study and all the other radiatively connected levels. To the best of our knowledge, no theoretical work has considered the Rb states at 300 K. In order to compare with the experimental data, we shall use a

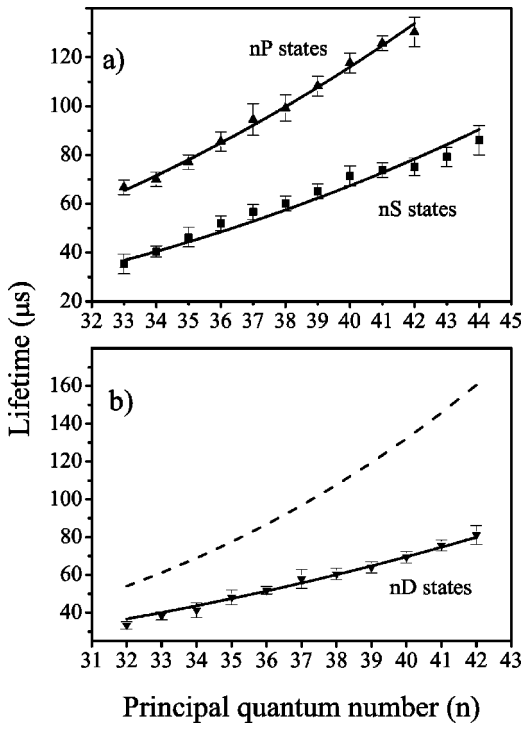

FIG. 3. Measured lifetime as a function of the principal quantum number for (a) *S* and *P* states; (b) *D* states. The solid lines are obtained with a fitting using Refs. [5,10]. The dashed line in (b) was obtained with Eq. (4) from our previous work [9].

simpler theory, based on Coulomb approximation, to calculate the lifetimes for Rb at 0 K. According to Gounand [5], τ_o can be expressed in a compact and consistent way as

$$\tau_0 = \tau'(n-\delta)^{\gamma}, \tag{2}$$

where τ' and γ depend on the atomic species and state. The parameter δ is the quantum defect for a given state. In order to calculate τ_{bb}, we have considered a theory proposed by Cooke and Gallagher [10] to calculate the transition rates for absorption and stimulated emission induced by blackbody radiation. Within this theory, τ_{bb} can be written as

$$\frac{1}{\tau_{bb}} = \frac{4\alpha^3 K_B T}{3n^2}, \tag{3}$$

where α is the fine structure constant. Equation (3) is expressed in atomic units; and its accuracy increases as the principal quantum number increases. The solid lines in Figs. 2(a) and 2(b) are obtained with a fit using Eqs. (1)–(3) and combining Refs. [5] and [10] and considering τ' and γ as free parameters for each state. From these fittings, we extract τ' and γ, as well as their error bars. The obtained values are resumed in Table I. The theoretical values, obtained from Ref. [5], for τ' and γ are also shown in Table I. The agreement is quite good between the fitting and the theory. We should point out that the weak dependence of the quantum defect on the principal quantum number [11] was not considered here.

TABLE I. Comparison between the parameters τ'_{Th} and γ_{Th} obtained from Ref. [5] and the ones obtained from the fitting of Fig. 3 (τ'_{Ex} and γ_{Ex}).

States	τ'_{Th} (ns) [5]	γ_{Th} [5]	τ'_{Ex} (ns)	γ_{Ex}
S	1.43	2.94	1.45 ± 0.03	3.02 ± 0.02
P	2.76	3.02	2.80 ± 0.03	3.01 ± 0.03
D	2.09	2.85	2.10 ± 0.02	2.89 ± 0.02

Another possible approach is to base our discussion on the calculation made by Theodosiou [6], which produced a vast table of the lifetimes of Rydberg states in alkali metal at different temperatures (0 K, 350 K, and 460 K) for the principal quantum number from $n=4$ to $n=18$. Using an interpolation procedure, as in our previous work [9], between $T=0$ K and $T=350$ K, we have calculated these lifetimes at 300 K. The lifetimes for the nD states from $n=4$ to $n=18$ obey a power-law expression type

$$\tau(n) = 294.242 + 0.096(n-1.347)^{3.867}, \tag{4}$$

where τ is given in units of 10^{-9} s and the value 1.347 is the quantum defect for the *D* states. In Fig. 2(b) the dashed line is obtained using Eq. (4) and it is clearly in disagreement with the experimental data. From this comparison we can conclude that Eq. (4) does not reproduce the experimental results. For a fair comparison, it would be necessary to calculate the lifetime at 300 K following the procedure described by Theodosiou [6], which is a complex calculation and is not the scope of this paper.

In conclusion, in this paper we have measured high-precision values of lifetime for a sequence of Rydberg levels of Rb atom using a sample of cold atoms. In these measurements collisional and super-radiance effects were suppressed without compromising the signal to noise ratio. We have fitted our experimental results using a combination of two simple theories; one which calculates the lifetimes of Rydberg states τ_0 at $T=0$ K and another which takes in account the blackbody radiation effect τ_{bb}. Our results are a good demonstration of the use of cold atoms to perform high-resolution time-domain spectroscopy, which can be explored to reopen old issues related to many-body theories to calculate lifetimes. The technique of using cold atoms can also be applied to cavity quantum electrodynamics where suppression or enhancement of lifetime are possible [12].

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