Measurement of the lifetimes of *S* and *D* states below $n=31$ using cold Rydberg gas

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In this paper we have extended our previous work [de Oliveira *et al.*, Phys. Rev. A **65**, 031401 (2002)] by measuring the *S* and *D* Rydberg lifetimes for ^{85}Rb for principal quantum number between $n=26$ and 31 using a sample of cold Rydberg atoms. This range is limited by the amplitude of the electric field that we can generate in our present setup.

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Spectroscopy can be performed in either the frequency or time domain, and both can provide crucial tests for the quality of computed wave functions and can be used to optimize models of the electron distribution in complex atoms or molecules. Alkali-metal atoms have been used both theoretically and experimentally as prototypes for accurate measurements of Rydberg state lifetimes, tests for calculations of dipole matrix elements, oscillator strengths, core polarizabilities, and influence of blackbody radiation [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 work done in this field has 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 $n>20$, superradiance occurs too 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 by as much as 40%. Recently, we demonstrated the possibility of obtaining a high-precision measurement of the Rydberg state lifetimes by using pulsed field ionization detection on a sample of cold trapped atoms [8,9]. In this work we measured the lifetime of *S* and *D* states of ^{85}Rb as a function of the principal quantum number *n* using a sample of cold atoms for $n<31$, and therefore extending our previous work. We start by presenting a brief description of the experimental setup, followed by the results and discussion in which our results are compared with existing theory.

Our setup is as described in Ref. [9], but now a dispenser is used to provide the Rb vapor, instead of a reservoir. This setup is shown schematically in Fig. 1. The ^{85}Rb trapping laser beam is provided by a Ti:sapphire laser (Coherent 899) operating at 780 nm and tuned 5 MHz to the red of the atomic $5S_{1/2}(F=3)\rightarrow 5P_{3/2}(F'=4)$ transition. A repumping frequency is generated by an electro-optical modulator, in which one sideband is tuned to the $5S_{1/2}(F=2)\rightarrow 5P_{3/2}(F'=3)$ transition. The total power at the trapping transition is about 300 mW, and the light beams present a Gaussian profile with a waist of 1 cm full width at half maximum. The coils, in an anti-Helmholtz configuration, generate the magnetic field gradient for trapping the atomic sample with an axial field gradient of 10 G/cm. The number of trapped atoms was determined by imaging their fluorescence onto a calibrated photomultiplier tube (PMT), while their dimensions were measured with a calibrated charge-coupled device camera (CCD). The atomic densities were obtained using these quantities. In typical conditions of operation the trap is loaded with 10^8 atoms in almost Gaussian spatial distribution with a waist leading to maximum peak densities of $n\sim 3\times 10^{10}$ atoms/cm^3. The details about our experimental setup and detection technique are describe elsewhere [9]. Briefly, the cold Rb atoms are excited from the $5P_{3/2}$ to the Rydberg levels by a pulsed dye laser (Jaguar Continuum, 5 ns pulse duration, running at a 20 Hz repetition rate, and 1.5 mJ/pulse) pumped by the third harmonic of a neodymium-doped yttrium aluminum garnet (Nd:YAG) laser, operating at the wavelength of 480 nm. A cloud of atoms is formed between two metal grids, separated from each other by 1.2 cm, these grids consisting of nickel meshes with 95% transparency through which the magneto-optical trap (MOT) beams pass almost unperturbed. One of the grids is grounded while a high-voltage (HV) pulse (rise time 1 μs, peak voltage $\sim$2 kV) is applied in the other to ionize the

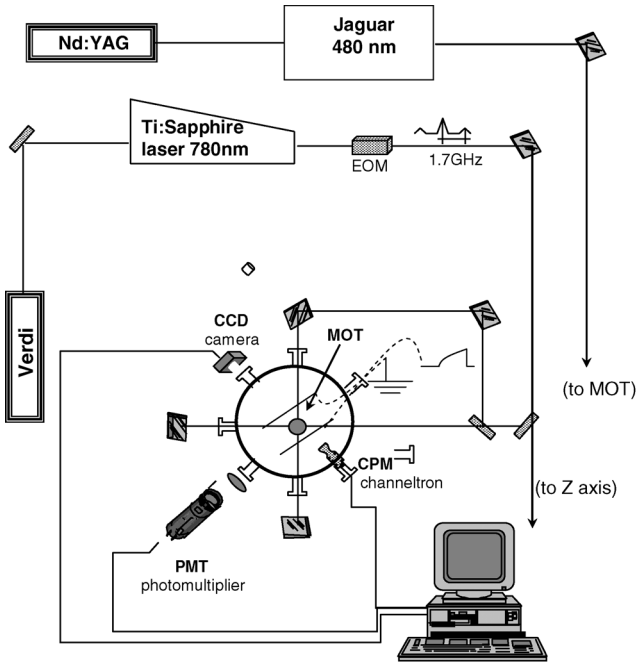

FIG. 1. Schematic experimental setup.

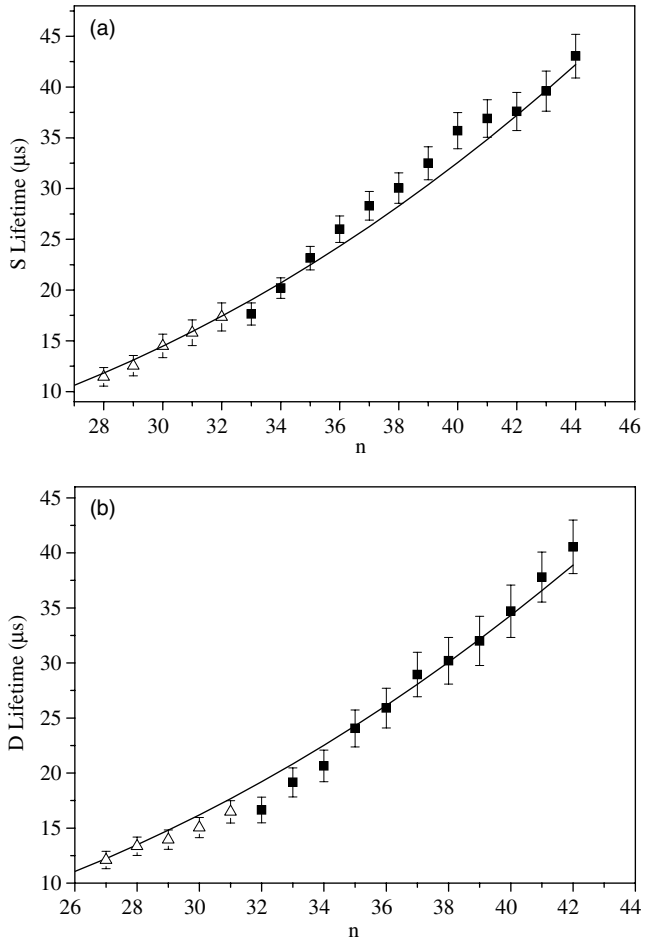

FIG. 2. (a) *S*- and (b) *D*-state lifetimes. The empty triangles are the current data and the squares are the data from our previous work [9]. The solid line represents the theoretical calculations by using Eq. (4).

Rydberg atoms. The HV pulse stays on over 2 μs, and the electrons produced are detected by a channel particle multiplier placed behind the second grid, and the electron signal is integrated by a boxcar integrator. By varying the delay between the optical excitation and the high-voltage pulse which ionize the Rydberg states, we are allowed to observe the time evolution of the Rydberg state population, and thus measure the lifetime.

The experiment runs according to the following time sequence: an external pulse delay generator (SRS, DG 535) provides the master clock and controls the switching on and off of the laser beams. The first step is to turn on a pulse with a controllable delay, which follows the Nd:YAG laser, triggering the excitation laser shot. A fast photodiode detects the ionizing laser pulse for deriving the needed trigger for the electron signal measurement. The boxcar signal is collected using a computer. We have used the same procedure described in our previous work [9] in order to avoid effects like collisions and superradiance, which impose serious limitations on the accuracy of lifetime measurements.

Figures 2(a) and 2(b) present the measured lifetimes for *S* and *D* states as a function of the principal quantum number *n*. The open up triangles are the current data and the squares are the data from our previous work [9]. The error bars were taken as the variance of several obtained measurements. The solid line represents the theoretical calculations, which we shall discuss in more details. We should point out that because of the linewidth of our pulsed laser, the lifetime for *D* states is due to both fine structure levels ($D_{3/2,5/2}$), and because of the amplitude limit of the electrical field we cannot detect principal quantum numbers lower than $n=26$.

To the best of our knowledge, there is still no published theoretical calculations for the Rb Rydberg state lifetimes at 300 K [10]. Therefore, we will compare our results with the calculations by Gounand for Rb; besides, this comparison is consistent with our previous work [11]. The lifetime of such states can be calculated using the lifetime at 0 K and also calculating the blackbody radiation, and the total lifetime will be given by [1]

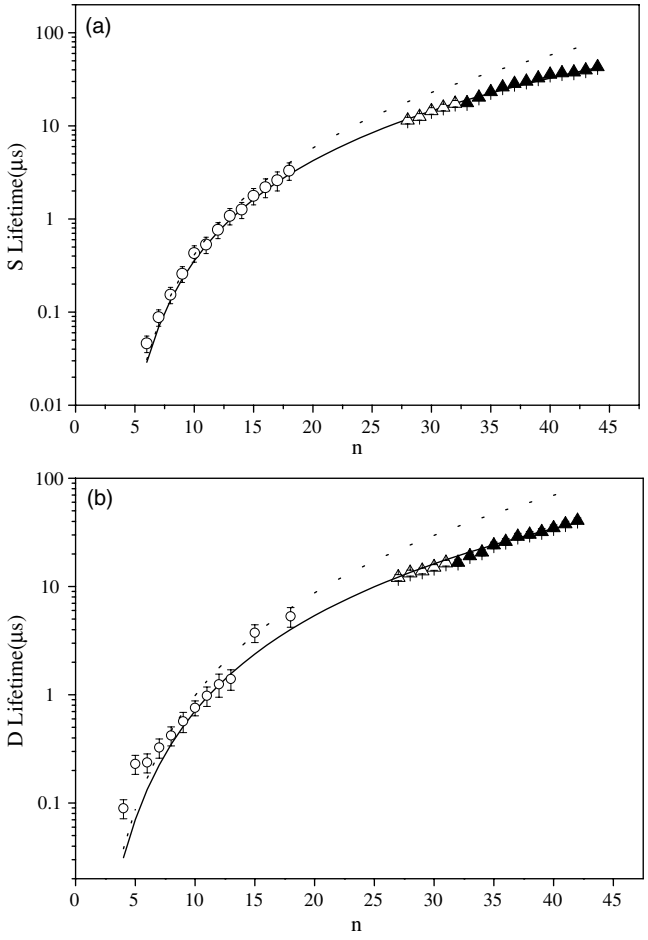

FIG. 3. Lifetime measurements of (a) *S* and (b) *D* states. In both figures are shown the measurements by using conventional techniques ([7], shown as open circles) and the cold atom technique (open up triangles for the current data and full up triangles for the old data [9], respectively). The solid line represents the theoretical calculations by using Eq. (4) for $T=300$ K and the dotted line represents the theoretical calculations of Ref. [5] for $T=0$ K.

$$\frac{1}{\tau} = \frac{1}{\tau_0} + \frac{1}{\tau_{bb}}, \tag{1}$$

where the total lifetime is a composition of the atomic lifetime τ_0 and the blackbody radiation contribution τ_{bb}, which has a strong dependence on the environmental temperature; in our case this temperature is about 300 K. For the evaluation of τ_0 we followed the same procedure as in our previous work (Refs. [5,9]), i.e., calculating from

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

where τ' and γ depend on the atomic species and state. δ is the quantum defect for a given state. τ_{bb} is calculated considering the transition rates for absorption and stimulated emission induced by the blackbody radiation. According to this theory, τ_{bb} can be written as

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

where α is the fine structure constant. Combining Eqs. (1)–(3), one can obtain the total lifetime, which is given by

$$\tau = \frac{3(n-\delta)^2}{4\alpha^3 K_B T} \frac{\tau'(n-\delta)^{\gamma}}{[\tau'(n-\delta)^{\gamma} + 3(n-\delta)^2/4\alpha^3 K_B T]}, \tag{4}$$

Using Eq. (4) we have fitted the data shown in Figs. 2(a) and 2(b). We can observe that there is a good agreement between experimental and theoretical data. From the fitting procedure, we obtain for the S state $\tau' = 1.43 \pm 0.05$ and $\gamma = 2.94 \pm 0.03$, and for the D state $\tau' = 1.9 \pm 0.07$ and $\gamma = 2.83 \pm 0.04$; which are consistent with our previous results [9].

In Fig. 3, we show the lifetimes obtained by using the conventional techniques (Ref. [7], shown as open circles) and by using the cold atom technique (open up triangles for the current data and full up triangles for the old data [9], respectively) for S [Fig. 3(a)] and D [Fig. 3(b)] states. We also show the theoretical curves obtained using the calculations of Ref. [5] (dotted line) for $T=0$ K, and Eq. (4) (solid line), which takes the effect of blackbody radiation into account for $T=300$ K. As one can see, the experiment using cold atoms allows us to perform high-precision lifetime measurements in a range inaccessible by conventional techniques. We should point out that for small principal quantum number the D states do not present a good quantitative agreement with the presented theory; which is not surprising due to its simplicity. This behavior can be reproduced by a more elaborate theory [10].

To summarize, we have measured the Rydberg state lifetimes for S and D states in the range of the principal quantum number between $n=26$ and 31, expanding on our previous work. These results are in good agreement with the results of our previous work [9], and also there is good agreement between the experimental data and the theoretical curves. The use of cold atoms in the low-density regime has proved once again to be a reliable technique to obtain accurate measurements of Rydberg lifetimes, and may be used to measure the lifetimes of others alkali-metal atoms. We hope that this will motivate future theoretical work in this field.

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[11] In our previous work, our lifetime definition is a factor of 2 different from the standard definition in this field [5]. But a mistake of a factor of 2 in the blackbody contribution canceled this factor, and the comparison was not compromised in our previous work. To avoid further confusion, we will use the standard definition from now on.