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Abstract

We report a precision measurement of the cesium $6P_{3/2}$ excited state lifetime. Two collimated, counter-propagating thermal Cs beams cross perpendicularly to femtosecond pulsed laser beams. High timing accuracy is achieved from having excitation and ionization laser pulses which originate from the same mode-locked laser. Using pulse selection we vary the separation in time between excitation and ionization laser pulses while counting the ions produced. We obtain a Cs $6P_{3/2}$ lifetime of 30.460(38) ns, which is a factor of two improvement from previous measurements and with an uncertainty of 0.12%, is one of the most accurate lifetime measurements on record.

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Atomic state lifetimes are useful to many areas of physics as one of the basic parameters of atomic structure. Precision lifetime measurements can be compared to predictions from *ab initio* atomic structure calculations, providing a direct test of the accuracy of the underlying theory. This is particularly important in cesium, where atomic parity nonconservation (PNC) has been measured on the parity-forbidden electric dipole $6S_{1/2} \rightarrow 7S_{1/2}$ transition [1, 2]. Atomic PNC measurements are of fundamental interest since they test the weak interaction at low energies, providing contraints on potential new physics beyond the standard model such as the masses of extra Z bosons [3]. The low-lying $6P$ atomic lifetimes are important since these states couple to the $6S$ and $7S$ states through parity-odd interactions, and with a decay channel only to the $6S$ ground state they directly provide the corresponding dipole matrix elements which are used in determining the PNC amplitude.

Previous measurements of excited state lifetimes have utilized atomic samples in vapor cells [4], thermal [5] and fast atomic beams [6], as well as cold trapped atoms [4, 7] and single trapped ions [8]. Two direct, high precision measurements of the Cs $6P_{3/2}$ state lifetime achieved uncertainties of 0.33% using time-correlated single-photon counting from ultrafast excitation of a thermal atomic Cs beam [5], and 0.23% from a Cs fast-beam position-correlated photon counting experiment [6]. Other methods to measure atomic lifetimes have included line-width measurements [9], beam-foil experiments [10], and photoassociative spectroscopy [11] among others.

In this Rapid Communication we present measurements of the Cs $6P_{3/2}$ atomic state lifetime using a pump-probe technique [12] based on the ultrafast pulses from a mode-locked femtosecond laser. Such lasers have broad applications from both the short optical pulses they generate along with the precise timing (and frequency spacing) of the mode-locked pulse train [13]. We take advantage of this temporal stability by using a mode-locked pulse to first excite Cs atoms to the $6P_{3/2}$ state (see Fig. 1a), with a subsequent frequency-doubled pulse ionizing atoms which remain in the excited state. Figure 1b illustrates the pulse selection scheme, whereby varying the excitation pulse selected with respect to the ionization pulse in time, the population of the $6P_{3/2}$ state is probed at integer multiples, $N\Delta t$, of the mode-locked laser pulse repetition rate ($\Delta t \sim 13.2$ ns). An excitation/ionization pulse pair occurs every $T = 4$ μs. The excitation and ionization laser beams are collinear and cross perpendicularly to counter-propagating Cs atomic beams as shown in Fig. 1c. The ions produced are counted with a Channeltron detector, providing near unity ion detection.

The first excited ^2P levels in alkali atoms are ideal for this method since a single mode-locked laser can be used to both excite atoms, and with a portion of the beam split off and frequency-doubled, ionize atoms in the excited ^2P states but not the ground state. A Coherent Mira 900 titanium:sapphire (Ti:S) ultrafast oscillator produces excitation pulses centered at 852 nm of $\sim$ 5 nJ energy and 150 fs duration. These pulses will excite about 0.5% of the Cs atoms present, taking into account most photons are not in resonance with the atomic transition due to the broad bandwidth (8 nm FWHM) of the laser pulses. The probability for ionization is much smaller than for excitation, requiring the use of higher energy pulses. This is accomplished using a Coherent RegA 9000 Ti:S regerative amplifier. These pulses are subsequently frequency-doubled by single-passing through a beta barium borate (BBO) nonlinear crystal. Pulses centered at 426 nm of $\sim$1.2 μJ are achieved, which will ionize approximately 0.2% of the atoms in the $6P_{3/2}$ state.

Pulse selection is accomplished by passing the excitation laser beam through a series of three electro-optic modulators (EOMs). Individually the EOMs achieve a pulse selection contrast ratio of typically 200:1, while combined it is $\geq 10^5$:1. Before pulse selection the excitation laser repetition rate is 75.5 MHz. Ionization pulses from the regenerative amplifier come at a much slower pulse repetition rate of 250 kHz and each of these pulses is used. The EOMs are triggered by the ionization pulses, using one SRS DG535 pulse/delay generator to select a particular excitation pulse with respect to the ionization pulses, and multiple DG535 pulse/delay generators to fine-tune the delay of each EOM onto the center of the selected pulse. Initially we select the excitation pulse closest to the ionization pulse in time while counting the ions produced. We then select the next mode-locked excitation pulse further away from the ionization pulse in time and again count ions, repeating this process out to about 500 ns (with the next data set starting at 500 ns and going to short delays). A typical data set is illustrated in Fig. 2, with a fitting function of the form,

$$N_{ion}(t) = Ae^{-t/\tau} + B \tag{1}$$

where the initial height of the curve, A, the background, B, and the lifetime, τ, are determined by a nonlinear least squares fit to the data. The data set in Fig. 2 resulted in a lifetime of 30.5 ns with a statistical error of 1%. Background ion counts come primarily from residual excitation by laser pulses which are not completely extinguished by the EOMs. In order to achieve statistical errors below 0.1% we take many such data sets (see inset of Fig.

TABLE I: Summary of contributions to the uncertainty in the cesium $6P_{3/2}$ lifetime.

Source	Error (%)
Mode-locked laser pulse timing stability	≤ 0.0015
EOM pulse selection	≤ 0.005
Overlap of excitation/ionization laser beams	$\pm$ 0.05
Hyperfine quantum beats:	
(a) angle θ between laser beam polarizations	$\pm$ 0.06
(b) linear/circular polarization components	$\pm$ 0.04
Zeeman quantum beats	≤ 0.01
Pulse pileup	$\pm$ 0.003
Statistics and extrapolation to zero Cs density	$\pm$ 0.088
Total	$\pm$ 0.12

2) and use the weighted mean of the results to determine the overall lifetime and uncertainty.

Cesium atoms originate from ovens at well controlled temperatures and pass through two collimation slits (height = 675 μm, width = 17 mm, and a slit separation of 55 cm) before entering the laser interaction region. The beamline and slits are cooled with liquid nitrogen in order to reduce background Cs and maintain a well defined atomic beam. With the laser beams oriented perpendicular to the atomic beams, atomic motion is possible between the excitation and ionization laser pulses which can systematically affect lifetime measurements. For this reason, the ionization beam has a greater diameter (2 mm FWHM) than the excitation beam (1 mm). The overlap of the laser beams along the atomic beam axis is also very important since a difference in overlap position corresponds to a difference in time. By incorporating counter-propagating atomic beams this effect is greatly reduced for Cs beams of approximately equal densities. We also use a computer-controlled translatable mirror (see Fig. 1c) to reoptimize the laser beam overlaps after each $\sim$ 40 minute data run.

The primary systematic errors considered are summarized in Table 1. We measure the mode-locked laser pulse repetition rate before and after every data run with a typical variation of less than 1 part in 10^6. Using such broad laser pulses we coherently excite all four of the $6P_{3/2}$ hyperfine states which causes quantum beating in the photoion signal. This results in an oscillatory modulation in the exponential decay of the $6P_{3/2}$ state due to the

interference of the wavefunctions of the coherently excited levels, with the frequency of modulation related to the energy separation between the excited hyperfine states. However, if the angle between the excitation and ionization laser beam polarizations are aligned at the "magic angle" the modulation will be suppressed [14] ($\theta_{magic} = 54.74°$). Since the fitting function does not account for quantum beats, the measured lifetime will vary depending on the amount and phase of modulation present. Figure 3 illustrates the observed change in lifetime as the angle (θ) between excitation and ionization laser beam polarizations is varied about 54.7°. The modulation clearly appears in the residuals of the data shown in Fig. 3 for $\theta = 0°$, while at $\theta = 54.7°$ the residuals are small and random. We precisely control the laser beam polarizations through the use of rotatable linear polarizers, with a possible deviation in $\theta = 54.7°$ of $\pm$ 1.2°, which corresponds to an uncertainty in the lifetime of 0.06%. This comes from both an uncertainty in alignment of the linear polarizers along with birefrigence from the vacuum chamber viewports, which can also add a small amount of circular polarization to the laser beams. Quantum beating due to Zeeman splitting of atomic levels can also occur. This effect is minimized by nulling the magnetic field in the interaction region to $\leq$ 20 mG.

We test for Cs density dependent effects by taking lifetime measurements at Cs oven temperatures from 25.5°C to 50°C, shown in Fig. 4. Evidence of radiation trapping is observed which causes in an increase in the measured lifetime as the Cs density increases, since a photon from the decay of the $6P_{3/2}$ state does not freely exit the interaction region but excites other Cs atoms to the $6P_{3/2}$ state. We note that 25.5°C is barely above room temperature, however, since we cool the Cs beamlines with liquid nitrogen this results in a much lower Cs background vapor pressure, and allows us to maintain well-defined atomic beams even with "ovens" at approximately room temperature. A final lifetime value of 30.460(38) ns is determined from an extrapolation of the lifetime to zero Cs density, although at Cs oven temperatures of 25.5°C the increase in the excited state lifetime from the extrapolated value is only 0.09%. Further details of the experimental apparatus and associated systematic errors will be given in a future publication.

The result of this work is compared to previous high precision measurements in Table 2. Our value falls between the two other direct measurements using position-correlated [6] and time-correlated [5] photon counting. High precision measurements of the Cs $6P_{3/2}$ lifetime have also been obtained indirectly from the value of the van der Waals coefficienct C_6 [15],

TABLE II: Comparison of precision lifetime measurements of the cesium $6P_{3/2}$ state.

Method	Lifetime (ns)
Ultrafast excitation and ionization	30.460(38)[a]
Fast-beam position-correlated photon counting	30.57(7)[b]
Time-correlated single-photon counting	30.41(10)[c]
Van der Waals coefficient C_6	30.39(6)[d]
$6S_{1/2}$ static dipole polarizability	30.32(5)[e]
Theoretical calculation of $\left\langle 6P_{1/2} \right\| D \left\| 6S_{1/2} \right\rangle$	30.35[f]

[a]This work; [b]Ref. [6]; [c]Ref. [5]; [d]Ref. [15]; [e]Ref. [16]; [f]Ref. [17] and Ref. [18]

deduced from high-resolution Feshbach spectroscopy, and also from the Cs $6S_{1/2}$ static dipole polarizability measured in an atomic fountain experiment [16].

From the measured lifetime we can determine the important dipole matrix element, $\left\langle 6P_{1/2} \right\| D \left\| 6S_{1/2} \right\rangle$, which enters into the calculation of atomic parity violation in Cs. The reduced dipole matrix elements can be calculated from

$$\frac{1}{\tau_J} = \frac{4}{3}\frac{\omega^3}{c^2}\alpha\frac{\left|\left\langle 6P_J \right\| D \left\| 6S_{1/2} \right\rangle\right|^2}{2J+1} \tag{2}$$

where ω is the transition frequency, c is the speed of light, α is the fine-structure constant, and J is the excited state angular momentum. Using the above equation and our measured Cs $6P_{3/2}$ lifetime we determine $\left\langle 6P_{3/2} \right\| D \left\| 6S_{1/2} \right\rangle = 6.3351(40)$ a.u.(atomic units). We can obtain a corresponding matrix element for the $6P_{1/2}$ state by combining the $6P_{3/2}$ result with a high precision measurement of the relative line strength ratios between these states [18], giving $\left\langle 6P_{1/2} \right\| D \left\| 6S_{1/2} \right\rangle = 4.5012(30)$ a.u. Of particular interest is a recent theoretical result by Porsev *et al.* [3] of 4.5093 a.u., which deviates from our result by 0.18%, and is also given in Table 2 as a lifetime using the above procedure in reverse.

It has also been demonstrated that atomic radiative lifetimes can be determined from high-resolution spectroscopy of photoassociated cold atoms [19]. An extremely accurate value of 30.462(3) ns was initially given for the Cs $6P_{3/2}$ lifetime [20], while a more recent study carefully investigating the estimation of the error bars in the measurement obtained a lifetime of 30.41(30) ns [11]. As this method is further developed, interesting comparisons with direct lifetime measurements such as this work can be made which may illuminate

small radiative QED effects which differ between atomic and molecular systems [21].

Using ultrafast pulses from a mode-locked laser we have measured the excited state lifetime of the Cs $6P_{3/2}$ state. The extremely uniform rhythm of the mode-locked pulse train provides a high precision time base for the measurements. An overall uncertainty of 0.12% is achieved, which may be improved using lower density atomic beams, with more careful control of the laser beam polarizations, along with reducing the offset between the excitation and ionization laser beams due to drift during data collection. This technique may also be applied to radiative lifetimes requiring very short wavelengths to reach, using pulses from high harmonic generation or soft x-ray (free electron) lasers. The method described here can be adapted to measure a wide variety of atomic lifetimes as long as suitable synchronization of the excitation and ionization laser pulses can be achieved.

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FIG. 1: (color online) The experimental setup. (a) Relevant Cs energy levels. (b) Particular excitation pulses are selected with respect to the ionization pulses in time. The interval between the excitation and ionization pulses is varied by an integer multiple ($N\Delta t$) of the mode-locked pulse repetition rate. (c) Mode-locked laser pulses at 852 nm are used to excite Cs atoms to the $6P_{3/2}$ state. These pulses are also sent to a regenerative amplifier and frequency-doubled to 426 nm which will ionize atoms which remain in the $6P_{3/2}$ state. The laser beams cross perpendicularly to Cs atomic beams with a Channeltron detector counting the ions produced.

FIG. 2: (color online) A typical Cs $6P_{3/2}$ lifetime decay curve. The fitting function is shown as a solid line, representing an exponential decay with a constant background. The inset is a histogram of 140 such data sets at a Cs oven temperature of 25.5°C, resulting in a lifetime of 30.487 ns with a statistical error of 0.085% (the solid line is a Gaussian curve illustrating the mean and standard deviation of the measurements).

FIG. 3: (color online) The variation in the measured lifetime from adjusting the angle between excitation and ionization laser beam polarizations. The residuals of the data fit are shown for θ = 0° and 54.7°, which illustrate the modulations which cause the observed lifetime variation at angles deviating from θ_{magic}.

FIG. 4: (color online) The measured Cs $6P_{3/2}$ lifetime at a variety of Cs oven temperatures. The solid line is a weighted linear fit through the data points.

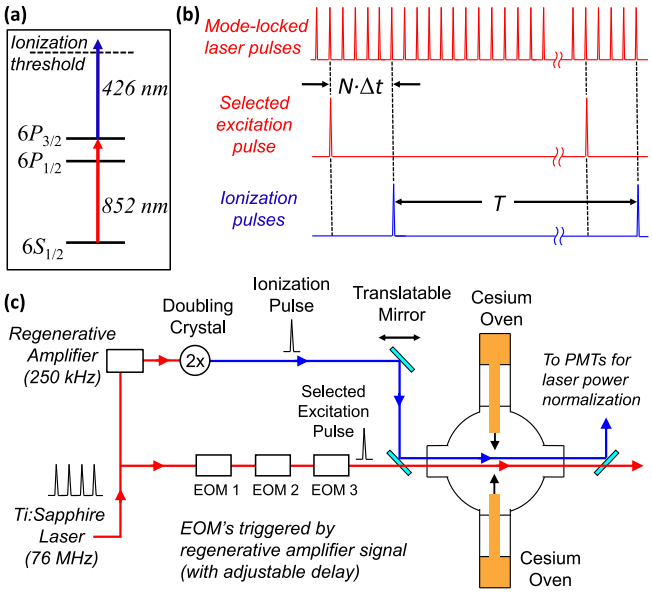

Figure 1 17May2011

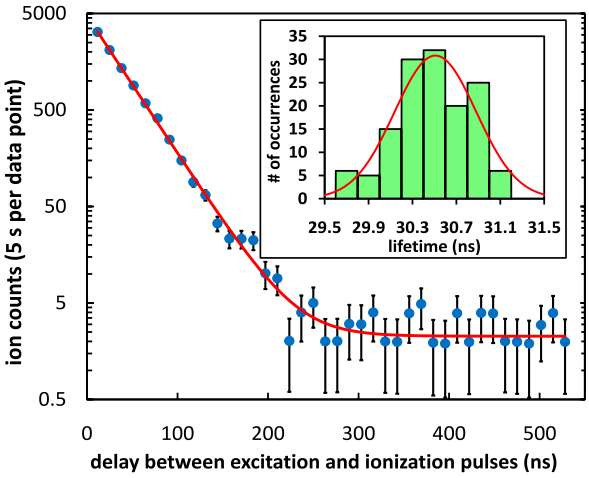

Figure 2 17May2011

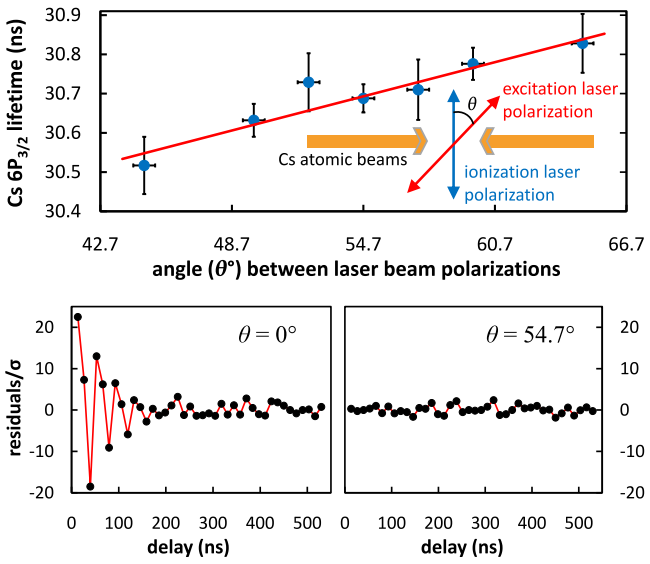

Figure 3 17May2011

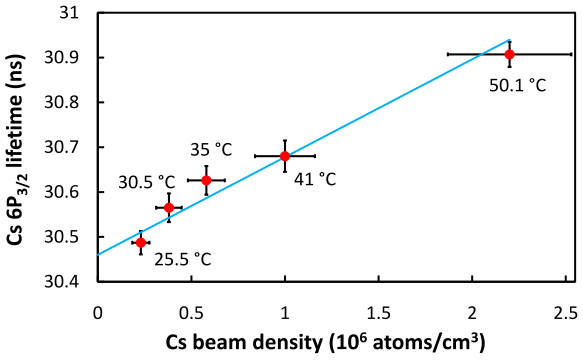

Figure 4 17May2011