PHYSICAL REVIEW A VOLUME 50, NUMBER 3 SEPTEMBER 1994

Precision lifetime measurements of the $6p\ ^2P_{1/2,3/2}$ states in atomic cesium

R. J. Rafac, C. E. Tanner, A. E. Livingston, and K. W. Kukla
Department of Physics, University of Notre Dame, Notre Dame, Indiana 46556

H. G. Berry and C. A. Kurtz
Physics Division, Argonne National Laboratory, Argonne, Illinois 60439

(Received 13 June 1994)

We report measurements of the $6p\ ^2P_{1/2}$ and $6p\ ^2P_{3/2}$ state lifetimes in the ^{133}Cs atom with precisions of 0.23% and 0.27%, respectively. The results are important to the interpretation of the next generation of parity nonconservation experiments in atomic cesium. A diode laser was used to selectively excite a fast beam of neutral cesium atoms. The decay in flight of the fluorescence was observed with photon-counting detectors. The lifetime results are 34.934 ± 0.094 ns for the $6p\ ^2P_{1/2}$ state and 30.499 ± 0.070 ns for the $6p\ ^2P_{3/2}$ state. We present comparisons with previous measurements and with relativistic many-body calculations of alkali-metal-atom transition matrix elements.

PACS number(s): 32.70.Fw, 42.55.Px, 42.62.Fi, 35.10.Wb

Precision measurements of atomic structures and transition probabilities can provide stringent tests of fundamental theories and of calculational capabilities for relativistic atomic systems. In heavy neutral atoms, even the most elaborate relativistic *ab initio* theoretical models exclude known correlation effects [1]. The low-lying electronic states of neutral cesium are being studied extensively because of their importance to the interpretation of parity nonconservation (PNC) in the $6\ ^2S_{1/2} \rightarrow 7\ ^2S_{1/2}$ transition. PNC measurements [2] are approaching a sensitivity that can test the standard model at the level of radiative corrections, making it possible to look for new physical effects predicted by high-mass-scale supersymmetric and technicolor models [3]. In order to obtain precise weak-interaction information from the new generation of cesium PNC experiments, the atomic radial matrix elements between low-lying states must be known to an accuracy approaching the 0.1% level [4]. In addition, precision measurements of the same transition probabilities test directly the accuracy of atomic calculations that are used to interpret cesium PNC experiments [1].

We have reported previously [5] the first beam-laser measurement of the $6p\ ^2P_{3/2}$ state lifetime in atomic cesium using resonant diode laser excitation of the $6\ ^2S_{1/2} \rightarrow 6\ ^2P_{3/2}(D_2)$ transition at 852 nm in a fast atomic beam. This result is already of sufficient precision to verify the accuracy of relativisitc many-body calculations of the dipole transition matrix element to 0.5%, which is adequate to confirm the interpretation of past PNC experimental results in cesium. Following a series of improvements incorporated into our experimental apparatus and techniques, we have reduced the systematic uncertainties in the measurements to a level that is comparable with the statistical uncertainties. We have also extended the measurements to include the lifetime of the $6p\ ^2P_{1/2}$ state in cesium, using resonant diode-laser excitation of the $6\ ^2S_{1/2} \rightarrow 6\ ^2P_{1/2}(D_1)$ fine-structure transition at 895 nm. In this Rapid Communication, we report our measurements of the $6p\ ^2P_{1/2}$ and $6p\ ^2P_{3/2}$ state lifetimes in atomic cesium to be 34.934 ± 0.094 and 30.499 ± 0.070 ns, respectively. These results represent the most precise determinations of the cesium $6p$ state lifetimes, and test the many-body calculations [1,6] of the $6\ ^2S_{1/2} \rightarrow 6\ ^2P_{1/2}$ and $6\ ^2S_{1/2} \rightarrow 6\ ^2P_{3/2}$ transition matrix elements to better than 15 parts in 10^4.

Our technique for lifetime measurements employs a fast atomic beam crossed with an excitation laser beam and subsequent detection of the decay in flight of the atomic beam fluorescence. The advantages of the technique and the basic experiment have been described previously [5]. Briefly, Cs^+ ions are accelerated through a potential of 50 kV, velocity selected, and neutralized in a rubidium charge exchange cell. Light from a stabilized diode laser is directed perpendicular to the collimated fast beam of cesium atoms and is tuned to excite one of the fine-structure levels in the $6p\ ^2P$ state. The decay of fluorescence along the atomic beam is monitored by both a stationary detector for normalization and a translatable detector observing the decay. The collection optics for each detector consist of fiber-optic bundles shielded from scattered light and coupled to cooled photomultiplier tubes (Fig. 1). The beam velocity is determined by measuring the Doppler shift of the $6s\ ^2S_{1/2} \rightarrow 6p\ ^2P_{3/2}$ transition when excited collinearly with a second diode laser.

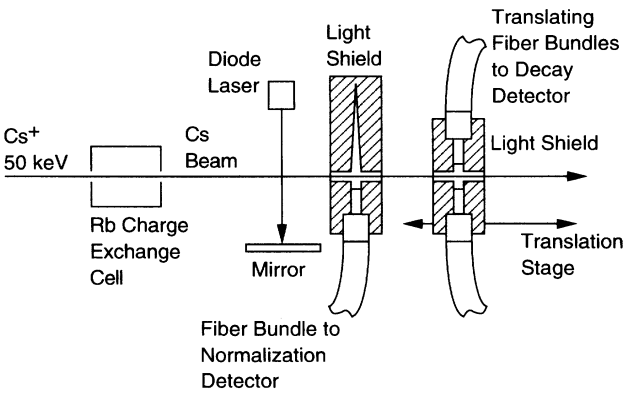

FIG. 1. Schematic diagram of the experiment. The fiber-optic bundles couple the collected light through the vacuum chamber wall and into the photomultiplier tubes.

TABLE I. Lifetimes of the $6p\ ^2P_{1/2,3/2}$ states of cesium.

Lifetime (ns)			
$6p\ ^2P_{1/2}$ state	$6p\ ^2P_{3/2}$ state	Method	Reference
34.934±0.094	30.499±0.070	fast-beam laser	This work
	30.55±0.27	fast-beam laser	Tanner [5]
	31.8±0.7	time-resolved laser	Campani [12]
	29.9±0.2	level crossing	Rydberg [9]
	32.7±1.5	level crossing	Schmieder [14]
35.2±1.5	30.8±1.5	phase shift	Dodd [8]
	30.5±0.7	phase shift	Link [13]
35.53	30.99	*ab initio*	Dzuba [6]
34.92	30.49	energy corrected	Dzuba [6]
35.22	30.81	*ab initio*	Blundell [1]
34.51	30.13	energy corrected	Blundell [1]
34.961	30.563	CAHS	Theodosiou [11]

Major improvements to the experimental geometry include more stringent control of collimation, shielding, and monitoring of the fast atomic beam. Light collection efficiency has been improved in two ways. First, another fiber-optic bundle was added to the translation assembly radially opposite the original bundle. This reduced systematic uncertainties due to possible misalignment between the translation of the detector inputs and the atomic beam direction. Second, improvements were made in the fiber-optic couplings through the vacuum wall and to the detector photocathodes. Nonresonant scattered light background was suppressed by adding narrow-bandwidth interference filters to the detectors, and the detector dark counts were reduced by lowering the temperature of the photomultiplier tubes. Sensitivity to variations in atomic beam-induced scattered light was reduced by incorporating a laser beam chopper into the apparatus. The uncertainty in the velocity determination was reduced by improving the frequency stability of the collinear laser. Complete details of the improved apparatus, techniques, calibrations, and tests of systematic errors will be reported elsewhere [7].

The lifetime data consist of about 30 decay measurements for each fine-structure component monitored at 31 separate observation points over 5–6 decay lengths. The data-acquisition sequence consisted of laser-on and laser-blocked pairs of photon-count signals in both the translating and the normalization detectors, accumulated at each point of the decay curve. Decay data were acquired for both directions of translation along the atomic beam. Following each six decay measurements, a pair of control decay runs were acquired with the atomic beam blocked in order to verify the stability of the scattered laser light and detector dark counts. Detector dark count rates and scattered laser light contributions were typically <100 Hz. Scattered light produced by the atomic beam was <2 kHz. Counts due to all of these backgrounds are recorded during data acquisition for each detector so that their contributions to the signals can be removed. The fluorescent signal rate for the D_2 transition at 852 nm was ~ 70 kHz. For the D_1 transition at 895 nm the available diode laser power was only about 15% of that for the 852-nm laser, and the detector quantum efficiency was about $\frac{1}{3}$ of that at the lower wavelength, resulting in fluorescence rates of about 4 kHz. The background-corrected and normalized decay data were fit to the function of a single exponential plus constant background. Nearly all the decay fits resulted in a background consistent with zero, as expected from the data-acquisition and treatment scheme. The reproducibility (standard deviation of the mean) of the fitted lifetimes was 0.14% for the $6\ ^2P_{3/2}$ state and 0.22% for the $6\ ^2P_{1/2}$ state. The estimated systematic errors that are common to both of the fine-structure lifetime measurements are 0.05% due to solid angle uncertainties, 0.14% uncertainty in atomic beam velocity, and $<0.05\%$ due to mechanical nonlinearities, temperature, and pressure effects. Residual quantum beat effects in the decays contribute negligible uncertainty for the $6\ ^2P_{1/2}$ state and $<0.1\%$ for $6\ ^2P_{3/2}$. Detailed discussions of the error contributions will appear elsewhere [7]. The sum in quadrature of the uncertainties yields 0.23% for the $6\ ^2P_{3/2}$ lifetime and 0.27% for the $6\ ^2P_{1/2}$ lifetime.

Previous measurements of the $6p\ ^2P_{1/2,3/2}$ lifetimes in neutral cesium are compared with our results in Table I. We have excluded measurements with uncertainties of $>5\%$ as well as measurements performed before 1960. These historical references can be found in Ref. [5]. The only previous lifetime measurement of the $6p\ ^2P_{1/2}$ state in atomic cesium is the phase shift value of Dodd and Gallagher [8], with an uncertainty of 4%. Our measurement improves upon this experimental determination of the $6p\ ^2P_{1/2}$ lifetime by a factor of 16. The $6p\ ^2P_{3/2}$ lifetime has been measured previously by several techniques. Only our first fast-beam measurement [5] and the most recent level crossing measurement [9] quote uncertainties of $<1\%$. Our present measurement agrees with our previous result and improves upon the uncertainty by a factor of 4, yielding a lifetime value that is more precise but 3σ (2%) larger than that of Ref. [9].

Two accurate *ab initio* calculations exist for the $6p\ ^2P_{1/2,3/2}$ state lifetimes in cesium [1,6]. Both apply relativistic many-body perturbation (MBPT) theory to the calculation of the electric dipole matrix elements for the two $6s \rightarrow 6p$ fine-structure transitions. Although both calculations include most of the same effects due to infinite sums of many-body diagrams, certain contributions are not included in one or both formulations, as discussed in Ref. [1]. The

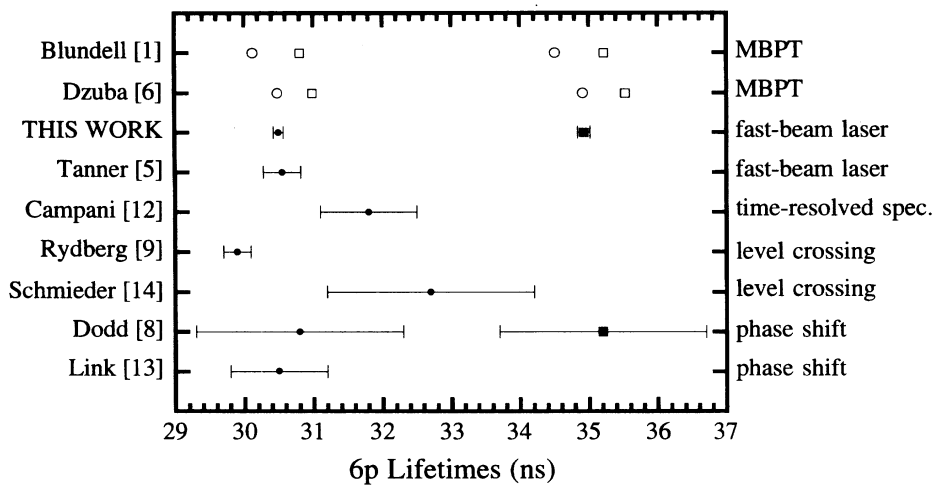

FIG. 2. Graphical comparison between our reported lifetime measurements, previous experimental work, and many-body theoretical values: ○, energy corrected theory; □, *ab initio* theory; ●, $6p\ ^2P_{3/2}$ experimental values; ■, $6p\ ^2P_{1/2}$ experimental values.

resulting *ab initio* values for the transition matrix elements from Refs. [1,6] differ from each other by 0.4% for $6s \rightarrow 6p\ ^2P_{1/2}$ and by 0.3% for $6s \rightarrow 6p\ ^2P_{3/2}$. Therefore, the corresponding *ab initio* transition probabilities, proportional to the matrix elements squared, differ by 0.8% and 0.6%, respectively (see Table I). Comparison of the calculated matrix elements with experimental lifetimes requires a conversion factor of the transition energy cubed. The relativistic many-body $6s \rightarrow 6p$ energy intervals in Refs. [1] and [6] differ from the accurately known experimental transition energies [10] by 0.5–0.8 %. Therefore, we use experimental transition energies with the *ab initio* matrix elements to provide the energy-corrected theoretical lifetime values in Table I. This energy correction is approximately 2% for the $6s \rightarrow 6p$ transitions in cesium. The comparison of our results with previous measurements and with the many-body calculations are shown in Fig. 2.

Our measured lifetimes for both fine-structure states happen to show excellent agreement with the energy-corrected values derived from Ref. [6], but are more than 1% longer than the values derived from Ref. [1], a 5σ difference from our experimental results. However, agreement between experiment and these energy-corrected theoretical lifetimes at the precision of our experiment is not necessarily to be expected. One source of inaccuracy in both theoretical calculations of transition matrix elements arises from the omission of high-order terms due to inexact electron binding energies. Estimates of the omitted contributions are given in Refs. [1] and [6] using experimental energy scaling considerations. These estimated corrections increase the calculated lifetime values of Ref. [6] by about 0.2% and those of Ref. [1] by about 0.7%, resulting in a 2σ agreement between our measured values and both many-body calculations. A more conservative interpretation would be to view the energy-corrected lifetime values in Table I as representing well-defined theoretical results, and use the above-mentioned energy-scaled contributions to the matrix elements as estimates of at least part of the theoretical uncertainty.

A semiempirical approach [11] for predicting lifetimes uses the Coulomb approximation with a Hartree-Slater (CAHS) model core potential. The predicted lifetimes are based entirely on experimentally measured data for energy levels, ionization potentials, and core polarizabilities. The lifetimes of the $6\ ^2P_{1/2,3/2}$ states in cesium predicted by this technique are in good agreement with our present experimental results (see Table I).

The comparisons between experiment and *ab initio* theory may also be presented through matrix elements directly. Since the excited $6p_{1/2,3/2}$ states each have a single decay mode, the lifetime of each is related to a single reduced radial matrix element between the excited and ground states by the following formula:

$$\frac{1}{\tau_{J' \rightarrow J}} = \frac{4}{3} \frac{\omega^3}{c^2} \alpha \frac{|\langle J \| r \| J' \rangle|^2}{2J'+1} .$$

In this expression, ω is the transition energy divided by $\hbar$, c is the speed of light, α is the fine-structure constant, J' and J are, respectively, the excited- and ground-state angular momenta, τ is the excited-state lifetime, and $\langle J \| r \| J' \rangle$ is the reduced matrix element. Using experimentally derived transition energies, our lifetimes yield values for the reduced radial matrix elements of $-4.4978(61)a_\infty$ for the $6s\ ^2S_{1/2} \rightarrow 6p\ ^2P_{1/2}$ transition and $-6.3311(72)a_\infty$ for the $6s\ ^2S_{1/2} \rightarrow 6p\ ^2P_{3/2}$ transition, where a_∞ is the Bohr radius and the signs are determined theoretically. These experimental results may be compared with the matrix elements calculated using relativistic all orders methods [1,6]. The theoretical reduced matrix elements calculated by Blundell *et al.* [1] are -4.525 and -6.370 for the same transitions, respectively, in units of the Bohr radius a_∞, and those calculated by Dzuba *et al.* [6] are -4.499 and -6.332. Scaling factors of $\sqrt{2/3}$ and $\sqrt{4/3}$ were used respectively to normalize the latter calculation in same manner as the former.

In conclusion, we have measured the lifetimes of the $6p\ ^2P_{1/2}$ and $6p\ ^2P_{3/2}$ fine-structure states of atomic cesium with sufficient precision to determine the $6s\ ^2S_{1/2} \rightarrow 6p\ ^2P_{1/2,3/2}$ transition matrix elements to better than $\pm 0.14\%$. These matrix elements are now sufficiently well established to be used in the interpretation of the next generation of cesium PNC experiments. Our lifetime results suggest that the accuracies of existing relativistic many-body calculations of these matrix elements in cesium are of order

0.3–0.6 %. We expect that the present level of experimental precision will motivate improved theoretical calculations of transition probabilities for atomic cesium as well as for lighter alkali-metal atoms.

We would like to acknowledge the assistance of L. Young, F. G. Serpa, and P. J. Koppers in the early stages of the experimental development. We thank W. R. Johnson, J. Sapirstein, and C. E. Wieman for helpful comments during the course of this project. Partial support of this work is acknowledged for R.J.R. from the Arthur J. Schmitt Foundation, for C.E.T. from National Institute of Standards and Technology, and the Luce Foundation, for A.E.L. from Research Corporation, and from the U.S. Department of Energy, Office of Basic Energy Sciences, Fundamental Interactions Branch.

[1] S. A. Blundell, W. R. Johnson, and J. Sapirstein, Phys. Rev. A **43**, 3407 (1991); Phys. Rev. Lett. **65**, 1411 (1990).

[2] M. C. Noecker, B. P. Masterson, and C. E. Wieman, Phys. Rev. Lett. **61**, 310 (1988).

[3] W. J. Marciano and J. L. Rosner, Phys. Rev. Lett. **65**, 2963 (1990).

[4] D. Cho, Bull. Am. Phys. Soc. **39**, 1082 (1994). Report of an experiment in progress with C. E. Wieman.

[5] C. E. Tanner, A. E. Livingston, R. J. Rafac, F. G. Serpa, K. W. Kukla, H. G. Berry, L. Young, and C. A. Kurtz, Phys. Rev. Lett. **69**, 2765 (1992).

[6] V. A. Dzuba, V. V. Flambaum, and O. P. Sushkov, Phys. Lett. A **142**, 337 (1989).

[7] C. E. Tanner (unpublished); R. J. Rafac, Ph.D. thesis, University of Notre Dame (unpublished).

[8] J. N. Dodd and A. Gallagher, J. Chem. Phys. **50**, 4838 (1969).

[9] S. Rydberg and S. Svanberg, Phys. Scr. **5**, 209 (1972).

[10] C. E. Moore, *Atomic Energy Levels*, Natl. Bur. Stand. (U.S.) Circ. No. 467 (U.S. GPO, Washington, D.C., 1958), Vol. III; G. Avila, P. Gain, E. de Clercq, and P. Cerez, Metrologia **22**, 111 (1986).

[11] C. Theodosiou (private communication); L. J. Curtis, Phys. Scr. **48**, 559 (1993).

[12] E. Campani, R. DeSalvo, and G. Gorini, Nouvo Cimento Lett. **29**, 485 (1980).

[13] J. Link, J. Opt. Soc. Am. **56**, 1195 (1966).

[14] R. W. Schmieder, A. Lurio, W. Happer, and A. Khadjavi, Phys. Rev. A **2**, 1216 (1970).