Time-resolved laser spectroscopy of multiply ionized atoms: natural radiative lifetimes in Ce IV.

Zhang, Zhiguo; Svanberg, Sune; Quinet, P; Palmeri, P; Biémont, E

Published in:
Physical Review Letters

DOI:
10.1103/PhysRevLett.87.273001

2001

Time-Resolved Laser Spectroscopy of Multiply Ionized Atoms: Natural Radiative Lifetimes in Ce IV

Z. G. Zhang,[1] S. Svanberg,[1] P. Quinet,[2,3] P. Palmeri,[3] and E. Biémont[2,3]

[1]*Department of Physics, Lund Institute of Technology, P.O. Box 118, S-211 00 Lund, Sweden*
[2]*IPNAS (Bâtiment 15), Sart Tilman, Université de Liège, B-4000 Liège, Belgium*
[3]*Astrophysique et Spectroscopie, Université de Mons-Hainaut, B-7000 Mons, Belgium*

(Received 7 February 2001; published 10 December 2001)

Radiative lifetimes have been measured for two excited levels of Ce IV using the time-resolved laser-induced fluorescence technique. Ce^{3+} ions were produced in a laser-induced plasma. In the measurements, a suitable magnetic field was applied to reduce the recombination between electrons and the ions and thus the background light from the recombination, and special care was exercised to avoid flight-out-of-view effects on the lifetime measurements for the high-velocity ions. The experimental lifetime results, $\tau = 30(2)$ ns for the level 49 737 cm^{-1} and $\tau = 30(3)$ ns for the level 52 226 cm^{-1}, were compared with relativistic Hartree-Fock calculations ($\tau = 30.5$ and 30.0 ns) indicating a particularly excellent agreement.

DOI: 10.1103/PhysRevLett.87.273001 PACS numbers: 32.70.Cs, 42.62.Fi

It is difficult to measure accurately radiative properties for highly ionized atoms; instead, results for radiative parameters were frequently obtained by theoretical calculations [1–3]. The difficulties stem from the fact that it is hard to achieve a stable ion source, and to avoid various detrimental effects influencing lifetime measurements. Although beam-foil techniques can be used to measure lifetimes in highly charged ions, the results are frequently quite different from those obtained from time-resolved laser spectroscopic techniques. Even if this is a rather extreme case, the data were found to differ by a factor of 2 in a recent study [4]. This is due to cascade effects from nonselective excitations in the beam-foil method. Time-resolved laser spectroscopic techniques have proven to provide accurate natural radiative lifetime measurements in a large variety of atoms and ions. In particular, a number of lifetime data in neutral, singly ionized, and doubly ionized atoms have been obtained using such techniques to meet the needs of astrophysics and test the theoretical models [5–14].

Although free ions can be obtained in a discharge, such as in a hollow-cathode lamp, it is impossible to produce highly ionized atoms due to recombination between ions and electrons. Using laser ablation, ions with different charges can be obtained, but the highly charged ions have increasingly higher speed and recombination between ions and electrons also occurs. Thus, in lifetime measurements for such ionized atoms, it is difficult to observe uniformly the decay and avoid flight-out-of-view effects, and recombination may make the observed lifetime value shorter. Background light from the recombination also constitutes a problem in lifetime measurements. Here we report, as we believe, the first lifetime measurements for triply ionized atoms using laser spectroscopic techniques in the studies of Ce IV. In the measurements, a suitable magnetic field over the plasma region was found to lead to a strong background plasma light reduction because the recombination is interrupted by the field. While the recombination light reduction in a magnetic field is not fully understood, the strongly differing mass of electrons and ions cause a magnetic-field dependent change separation. We measured the speed of the ions, and ensured that the influence of flight-out-of-view effects was eliminated by focusing the exciting beam to the center of the plasma and using a wide monochromator slit. The experimental results were compared with theoretical results obtained using the pseudorelativistic Hartree-Fock (HFR) approach of Cowan [15]. An excellent agreement was obtained. This work has opened a new perspective for radiative lifetime measurements in multiply ionized atoms using laser spectroscopic techniques.

Ce IV is the most suitable candidate to test lifetime measurements in triply ionized atoms due to its "simple" electronic structure and relatively low energy level values in the first excited configuration which has only one electron in its unfilled shell. Only a single UV laser is needed to excite the ion transitions from the ground state or metastable state to the two states in the first excited configuration, and there is a single decay channel for one level and two decay channels for the other level. Table I presents the level values measured and the excitation schemes used.

The experimental setup used in the measurements is shown in Fig. 1. Laser pulses with about 8 ns duration from a frequency-doubled and injection-seeded Nd:YAG laser (Continuum NY-82) were sent to a stimulated

TABLE I. Levels measured and excitation schemes.

Level[a]	E_{exp} (cm^{-1})	Origin (cm^{-1})	λ_{exc} $(nm)_{air}$	λ_{obs} $(nm)_{air}$
$5p^6 5d\ ^2D_{3/2}$	49 737	0.00	200.994	200.994
$5p^6 5d\ ^2D_{5/2}$	52 226	2253	200.042	200.042

[a]Level designations and energies are from NIST Atomic Spectroscopic Database [18].

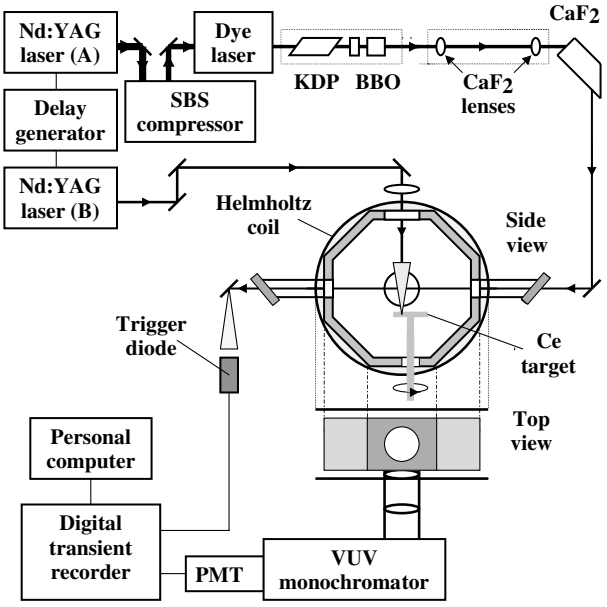

FIG. 1. Experimental setup used in the lifetime measurements for Ce^{3+} ions.

Brillouin scattering (SBS) compressor to produce laser pulses with about 1 ns duration [8]. The output was employed to pump a dye laser (Continuum ND-60), which was operated with Rhodamine 610 dye. To obtain the required excitation laser wavelength, the second harmonic of the dye laser was produced using a KDP crystal and the third harmonic of the dye laser was obtained using subsequently a retarding plate and a BBO crystal. The third harmonic was separated using a CaF_2 Pellin-Broca prism and was sent to a vacuum chamber via another CaF_2 prism. The laser beam was focused using two CaF_2 lenses, and the focal point size at the center of the vacuum chamber was less than 1 mm.

Free Ce^{3+} ions were produced utilizing a laser-induced plasma. Radiation from another Nd:YAG laser (Continuum Surelite), usually with laser pulse energy in the range of 2–10 mJ, was focused perpendicular to the surface of a Ce foil rotated in the vacuum chamber. An expanding plasma was produced above the surface of the foil. The two Nd:YAG lasers were triggered externally by a digital delay generator (Stanford Research System Model 535), which was used to adjust the delay time between the ablation and excitation pulses.

The third-harmonic beam, which passed horizontally through the plasma at a distance of 10 mm above the foil, was used to excite the ions Ce IV from the ground state or metastable state to the levels measured. Fluorescence released in the decay of the measured level was collected by two further CaF_2 lenses and imaged on the entrance slit of a vacuum monochromator (Acton Model VM502), and was detected by a Hamamatsu R3809U-58 multiplier with a rise time of about 170 ps. The signal was acquired and averaged by a digital transient recorder (Tektronix Model DSA 602) to form a fluorescence decay curve. The curve was sent to a personal computer for lifetime evaluation.

In the measurements, a strong perturbing background from the recombination between ions and electrons was found within the delay time of 500–2000 ns. In order to reduce the recombination and thus the background, a magnetic field produced by a Helmholtz coil system was added to cause the electrons and the ions in different ionization stages to run in different paths. Figure 2 shows background changes and the laser-induced fluorescence curves in different fields. From Fig. 2, it can be seen that the recombination moves to shorter delay times as the field becomes larger and there is a much lower background when a suitable field of about 100 G was applied. The magnetic field is obviously very important for eliminating the recombination and the corresponding nonconstant background. The plasma dynamics responsible for the strongly magnetic-field dependent recombination rejection remains a topic for future study and full understanding.

In fact, an important aspect in lifetime measurements in multiply ionized atoms is to avoid flight-out-of-view effects, especially when the measured lifetimes are long. In the present measurements, the entrance slit was put horizontally and parallel to the excitation beam to enhance the fluorescence collection efficiency and to prevent direct laser ablation light from the target to enter the monochromator. However, the entrance slit cannot be opened too much since the background light entering the monochromator must be controlled. In order to select optimum conditions, an estimate was made. First, the speeds of the ions in different ionization stages were determined by observing the change of the fluorescence intensity in the decay curves of ions in different ionization stages when the delay time was varied. Velocities were about 16 000 m/s for the

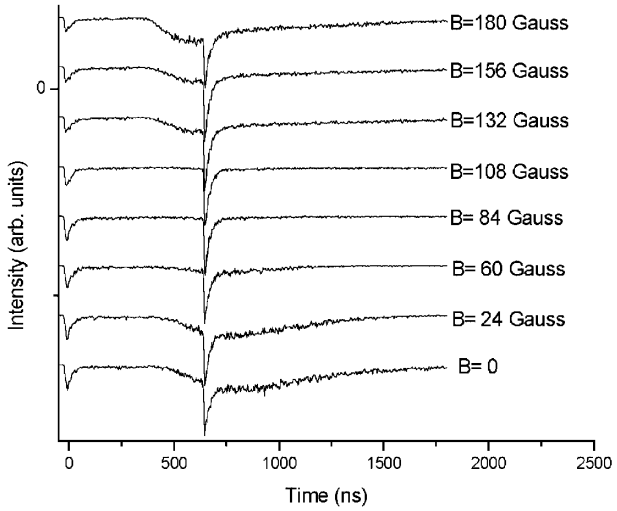

FIG. 2. Time-resolved laser-induced fluorescence signal from the level at 52 226 cm^{-1} and background light caused by the laser ablation in different external magnetic fields. $B = 0$ presents no field over the plasma. A field of about 100 G was observed to eliminate the background.

triply ionized atoms, 12 000 m/s for the doubly ionized atoms, and 5000 m/s for the singly ionized atoms. For a lifetime of 30 ns in Ce IV, it is desirable to observe the decay for 120 ns, which corresponds to 4 times the lifetime value. The ion flight distance is then 1.9 mm along the vertical direction. If the diameter of the laser beam is also considered, a slit width of more than 2.9 mm must be retained. In our experiments, the slit width was set to more than 3 mm.

In the measurements, a fluorescence decay curve needed averaging over 2000 shots to achieve a sufficient signal-to-noise ratio. The lifetime was evaluated using an exponential fit to the decay curve. A typical recording is shown in Fig. 3 including an exponential fit. About 20 fluorescence decay curves for each studied level were recorded under varying experimental conditions, using excitation and ablation laser beams with different intensities, different delay times, and high voltage supplied to the detector. Under the different conditions, intensities of the detected fluorescence signal were changed by a factor of 20, and the evaluated lifetimes of the curves scattered randomly around a certain value. No obvious systematic effects were found, showing that perturbation-free conditions prevailed, which is also our experience from several studies of doubly ionized atoms. Final lifetime values for each Ce IV level were given through averaging the evaluated lifetimes of the curves recorded. The lifetime values of the measured levels, and estimated error bars are listed in Table II. The error bars reflect a statistical scattering of the evaluated lifetimes and an estimate of possible remaining systematic errors.

The experimental data are compared in Table II with HFR calculations [15] performed in this work and with previous calculations [16]. The HFR theoretical data were obtained with the configurations $5s^25p^64f + 5s^25p^54f^2 + 5s^25p^55d^2 + 5s^25p^56p^2 + 5s^25p^54f6p + 5s^25p^44f5d^2 + 5s5p^64f5d + 5s5p^65d6p + 5p^65d^26p + 5p^64f5d^2 + 5s5p^54f^25d + 5s5p^54f5d6p$ (odd) and $5s^25p^65d + 5s^25p^54f5d + 5s^25p^55d6p + 5s^25p^44f^25d + 5s^25p^44f5d6p + 5s5p^64f^2 + 5s5p^65d^2 + 5s5p^66p^2 + 5s5p^64f6p + 5p^64f^25d + 5p^64f5d6p + 5s5p^54f5d^2$ (even). In view of the moderate complexity of the configurations considered, we have preferred to retain explicitly in the calculations all the single and double excitations in the space of the orbitals $4f$, $5s$, $5p$, $5d$, and $6p$ instead of considering implicitly the core-valence correlation effects via a core-polarization model potential with a core-penetration corrective term depending upon two parameters, i.e., the static dipole polarizability and the cutoff radius [17]. In fact, detailed comparisons between theory and experiment in Er III [10] and in Tm III [9] have shown that $4f$-$5d$ transitions deserve special attention in atomic structure calculations in lanthanides related to the fact that $4f$ electrons are deeply imbedded inside the xenon core. An attempt to solve the problem, originating from the fact that the analytical core-polarization corrections to the dipole operator [17] are no more valid, consists in applying an empirical scaling factor to the uncorrected $\langle 4f|r|5d\rangle$ radial matrix element. This semiempirical approach was not followed here, but, instead, we have preferred to consider in the model explicit interactions between the strongly interacting configurations.

No adjustment was considered for the Slater integrals [15] but, interestingly enough, it was observed that retaining the first three odd and the first two even configurations of the above list with a scaling factor of 0.85 led to numerical values (i.e., $\tau = 30.3$ and 29.6 ns) very close to those reported in Table II.

As an important conclusion, this work shows that it is possible to measure lifetimes in multiply ionized atoms following intense laser ablation. At the same time, it demonstrates ways to overcome the recombination between

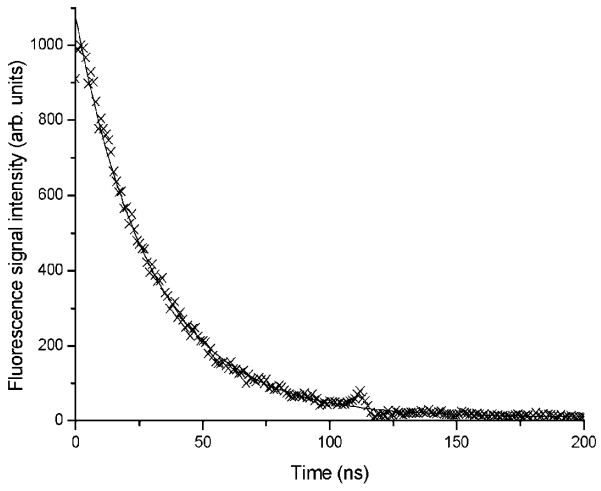

FIG. 3. Typical fluorescence signal from the level at 49 737 cm^{-1} in Ce IV showing an exponential fit. The recorded data points are marked as $\times$ in the figure. The solid curve shows the exponential fit for the LIF signal. The lifetime deduced from the fit was 30.0 ns.

TABLE II. Calculated and observed lifetime values (τ, in ns) in Ce IV. (β: Statistic error. θ: An estimate of possible remaining systematic errors.)

Level (cm^{-1})	Expt.[a]	HFR[b]	Ref. [16][c] (1)	(2)	(3)
49 737	30 (1^{β} + 1^{θ})	30.5	1.98	25.5	24.0
52 226	30 (1.4^{β} + 1.6^{θ})	30.0	1.93	23.8	22.9

[a]LIF measurements: this work.
[b]HFR calculations: this work.
[c]Reference [16]: (1) Relativistic model potential + core-polarization effects; (2) Relativistic model potential + semiclassical exchange potential + core polarization; (3) Relativistic model potential + empirically adjusted exchange + core polarization.

electrons and ions, the background from the recombination and flight-out-of-view effects. The agreement between theory and experiment observed in the specific case of Ce IV opens the door for a future better knowledge of triply and, more generally, multiply charged rare-earth ions.

This work was in part financially supported by the Swedish Natural Science Research Council and by the EU-TMR access to Large-Scale Facility programme (Contract No. HPRI-CT-1999-00041).

[1] A. Siems, F. R. T. Luna, and A. G. Trigueiros, J. Quant. Spectrosc. Radiat. Transfer **68**, 635 (2001).
[2] F. Bredice, M. Raineri, J. R. Almandos, M. Gallardo, and A. G. Trigueiros, J. Quant. Spectrosc. Radiat. Transfer **65**, 805 (2000).
[3] J. F. Wyart, W. U. Tchang-Brillet, N. Spector, P. Palmeri, P. Quinet, and E. Biémont, Phys. Scr. **63**, 113 (2001).
[4] Z. S. Li, H. Lundberg, G. M. Wahlgren, and C. M. Sikström, Phys. Rev. A **62**, 032505 (2000).
[5] K. B. Blagoev and V. A. Komarovskii, At. Data Nucl. Data Tables **56**, 1 (1994).
[6] Z. S. Li and Z. K. Jiang, Phys. Scr. **60**, 414 (1999).
[7] Z. G. Zhang, Z. S. Li, H. Lundberg, K. Y. Zhang, Z. W. Dai, Z. K. Jiang, and S. Svanberg, J. Phys. B **33**, 521 (2000).
[8] Z. S. Li, J. Norin, A. Persson, C.-G. Wahlström, S. Svanberg, P. S. Doidge, and E. Biémont, Phys. Rev. A **60**, 198 (1999).
[9] Z. S. Li, Z. G. Zhang, V. Lokhnygin, S. Svanberg, T. Bastin, E. Biémont, H. P. Garnir, P. Palmeri, and P. Quinet, J. Phys. B **34**, 1349 (2001).
[10] E. Biémont, H. P. Garnir, T. Bastin, P. Palmeri, P. Quinet, Z. S. Li, Z. G. Zhang, V. Lokhnygin, and S. Svanberg, Mon. Not. R. Astron. Soc. **321**, 481 (2001).
[11] Z. G. Zhang, A. Persson, Z. S. Li, S. Svanberg, and Z. K. Jiang, Eur. Phys. J. D **13**, 301 (2001).
[12] E. Biémont, H. P. Garnir, Z. S. Li, V. Lokhnygin, P. Palmeri, P. Quinet, S. Svanberg, J. F. Wyart, and Z. G. Zhang, J. Phys. B **33**, 521 (2001).
[13] E. Biémont, H. P. Garnir, P. Palmeri, P. Quinet, Z. S. Li, Z. G. Zhang, and S. Svanberg, Phys. Rev. A **64**, 022503 (2001).
[14] Z. G. Zhang, Z. S. Li, S. Svanberg, P. Palmeri, P. Quinet, and E. Biémont, Eur. Phys. J. D **15**, 301 (2001).
[15] R. D. Cowan, *The Theory of Atomic Structure and Spectra* (University of California Press, Berkeley, 1981).
[16] J. Migdalek and M. Wyrozumskaya, J. Quant. Spectrosc. Radiat. Transfer **37**, 581 (1987).
[17] J. Migdalek and W. E. Baylis, J. Phys. B **11**, L497 (1978).
[18] W. C. Martin, R. Zalubas, and L. Hagan, *Atomic Energy Levels—The Rare Earth Elements* (NIST, Washington, DC, 1978).