Lifetime measurements in multiply ionized atoms using beam-foil–laser excitation

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We have applied the recently proposed beam-foil–laser method to determine lifetimes in multiply ionized atoms. The lifetimes of two levels in multiply ionized carbon have been measured. The results are, for C III $2s3d\ ^1D$, 0.15±0.01 ns and for C IV $3s\ ^2S$, 0.21±0.02 ns. These values are in good agreement with the theoretical predictions. We have also determined these lifetimes using the classical (nonselective) beam-foil method. These latter lifetime results are in agreement with the beam-foil results reported previously but ≈20% longer than the beam-foil–laser values. The factors limiting the accuracy of the lifetime determinations by the beam-foil–laser method are discussed.

I. INTRODUCTION

Cascade-free lifetime measurements using fast-ion-beam–laser spectroscopy (see, e.g., Ref. 1) are restricted to levels that are optically coupled to the ground state or to a metastable state. These states are populated, prior to the laser interaction, either by collisions directly in the ion source or by beam-gas collisions. These precise lifetime determinations are restricted to neutral or singly ionized atoms.

The beam-foil–laser (BFL) method recently proposed for cascade-free lifetime measurements[2,3] is applicable, in principle, to multiply ionized atoms formed at the foil exit. This method is based on a two-step-excitation process: first, a nonselective excitation of a fast ion beam in a carbon foil and second, a selective excitation obtained by tuning the intracavity radiation of a *cw* dye laser to a transition in an ion emerging from the foil. The difference of intensities "laser on" and "laser off" of a line emitted from one level of this transition is recorded as a function of the distance travelled by the ions downstream from the foil-laser excitation region. The lifetime of this level is easily extracted from this decay curve. Indeed, let $i=1$ (2) be the lower (upper) level of a transition induced by the laser field, the differences of level populations, with and without laser excitation, $N_i^L-N_i^0$, downstream the laser interaction region are evolving according to the relations

$$\frac{d}{dt}(N_1^L-N_1^0)=-A_1(N_1^L-N_1^0)+A_{21}(N_2^L-N_2^0)\ , \tag{1}$$

$$\frac{d}{dt}(N_2^L-N_2^0)=-A_2(N_2^L-N_2^0)\ , \tag{2}$$

where $1/A_i=\tau_i$ denotes the lifetime of level i ($i=1,2$) and A_{21} the spontaneous transition probability. In these relations, all the cascade contributions from levels not affected by the laser field have disappeared. The integration of the differential system of Eqs. (1) and (2) gives

$$\frac{\Delta N_1(t)}{\Delta N_1(0)}=Ce^{-A_1t}+(1-C)e^{-A_2t}\ , \tag{3}$$

$$\frac{\Delta N_2(t)}{\Delta N_2(0)}=e^{-A_2t}\ , \tag{4}$$

where

$$\Delta N_i=N_i^L-N_i^0,\quad C=1-\frac{A_{21}}{A_1-A_2}\frac{\Delta N_2(0)}{\Delta N_1(0)}\ . \tag{5}$$

Note that $t=x/v$ where x is the distance travelled by the ions after the laser interaction region and v the ion velocity; $\Delta N_i(0)$ is the population change of the level i at $t=0$ induced by the laser field. We have reported previously the expression of $\Delta N_i(0)$ as a function of the relevant parameters (intracavity "power" of the dye laser, ratio of populations of levels 1 and 2 when the ions enter the laser field, dye laser "effective" linewidth, time spent by the ions in the laser field, etc.).[2]

Equations (3)–(5) show that the laser-on–laser-off population difference of the upper level 2 is a single exponential corresponding to lifetime τ_2 and the population change of the lower level 1 is a sum of two exponentials corresponding to lifetimes τ_1 and τ_2. The feasibility of the BFL method has been previously tested by measuring the lifetimes for the $2p$ and $3p$ terms in hydrogen.[3]

In the present work, the BFL method is applied to lifetime measurements of two short-lived levels in multiply ionized carbon atoms. The limitations of the method in application and precision are also discussed.

II. EXPERIMENT

The experimental arrangement described in detail previously[3] has been slightly modified for the present experiment. This arrangement illustrated in Fig. 1 will be briefly described here. However, the modifications of the setup and the specific conditions of this work will be given in detail.

A C^+ beam is supplied by a 2-MV Van de Graaff accelerator equipped with an *rf* source into which CO gas was admitted. This beam crosses at right angles first, a thin carbon foil and second, the intracavity beam of a modified *cw* dye laser (Coherent CR 599) pumped by an argon laser (Coherent Innova 20). The laser beam is situated immediately after the foil. The dye laser intracavity beam, folded at 90° by the mirror $M1$, crosses the ion beam before being retroreflected at the cavity ending mirror $M2$.

The crossing region between the ion and laser beams is

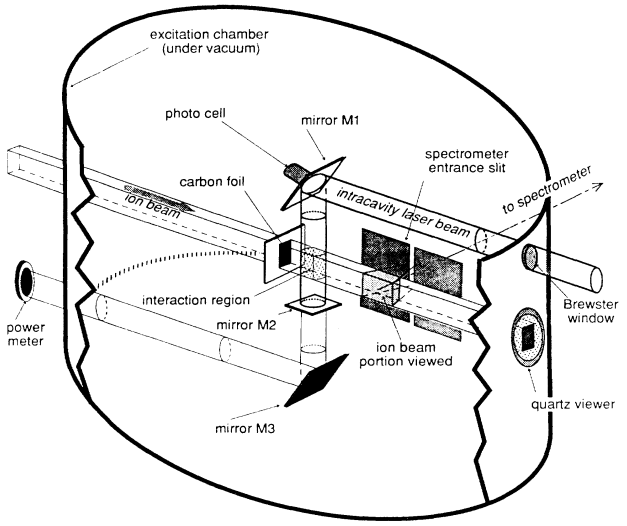

FIG. 1. Partial schematic diagram of the experimental arrangement. (Not to scale.) The mirrors $M1$ and $M2$ and the carbon foil can be moved together parallel to the ion beam.

situated inside the resonator to increase the "effective" light intensity. The carbon foil holder and the mirrors $M1$ and $M2$ can be moved together parallel to the ion beam axis by a stepper motor controlled by a microcomputer. The position of this movable system is determined with a precision of about ± 0.01 mm.

The wavelength of the dye laser radiation tuned by using the rotation (controlled by a microcomputer) of a birefringent filter is analysed by a high-resolution Ebert-Fastie spectrometer. During the whole experiment, this wavelength is repetitively controlled and corrected if necessary.

The intracavity power of the dye laser is determined by two devices. The first one is a photocell situated on the back of the mirror $M1$. The photocell signal is used to normalize the fluctuations induced in our measurements from small changes in the laser intensity. The second device is a power meter which measures the extracavity power of the dye laser. For this measurement, the laser beam transmitted through the end of cavity mirror $M2$ leaves the excitation chamber after having being folded at 90° by the total reflection mirror $M3$ (see Fig. 1). From the determinations of the mirror $M2$ transmittance and of the laser beam diameter, the intracavity photon density can thus be evaluated. This result is used to normalize the photocell readings corresponding to different measurements.

Fluorescence photons from the interaction region are analysed in the *VUV* region (where the laser stray light does not perturb the measurements) with a Seya-Namioka type spectrometer and detected by a channeltron detector (negligible background). The photon counting is made for a preset number of particles passing through the foil. The dye laser beam is chopped at ≈ 180 Hz and a lock in device allows the recording of photons corresponding to the "laser-on" and "laser-off" states.

The specific experimental conditions of the present work are summarized in Table I.

III. RESULTS AND DISCUSSION

A. $2s3d\ ^1D$ level in C III

The dye laser was tuned to resonance with the $2s3p\ ^1P^\circ - 2s3d\ ^1D$ transition in C III ($\lambda = 569.6$ nm) and the "laser-on" and "laser-off" intensities of the $2s2p\ ^1P^\circ - 2s3d\ ^1D$ transition ($\lambda = 57.43$ nm) were recorded (see Fig. 2).

TABLE I. Summary of experimental conditions.

	C III	C IV
Beam energy (MeV)	1.0	1.6
C^+ beam current (μA/cm^2)	≈ 3	≈ 3
Ion beam cross section (mm^2)	0.6×6.0	0.6×6.0
Ar-laser pumping power (all lines) (W)	18	18
Dye	Rhodamine 110	Rhodamine 6G
Dye intracavity power (W)	≈ 25	≈ 20
Doppler-corrected dye wavelength (nm)	569.64	580.19
Dye laser FWHM (nm)	≈ 0.1	≈ 0.08
Dye laser beam diameter (mm)	≈ 1	≈ 1
Carbon foil thickness (μg/cm^2)	≈ 3	≈ 3
Carbon foil size (mm^2)	1.5×7.0	1.5×7.0
Seya-Namioka spectrometer slit widths (mm)	0.8	1.5
Spectrometer slit-ion beam axis distance (mm)	13.0	13.0
Ion beam portion viewed (mm)	1.6	2.3
Foil-laser beam axis distance (mm)	0.5	1.0
Doppler broadening due to the angular divergence of the ion beam in the foil (FWHM) (nm)	≈ 0.04	≈ 0.03

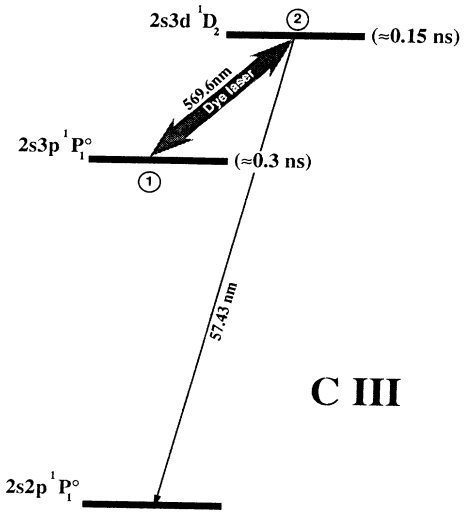

FIG. 2. Partial energy-level diagram of C III.

The difference of these intensities when the ions leave the laser interaction region, $I^L(0)-I^0(0)$, is proportional to the population change $N^L(0)-N^0(0)$ of the $2s3d\ ^1D$ level. In our experimental conditions (see Table I), the relative intensity change measured $[I^L(0)-I^0(0)]/I^0(0)$ is $\approx 4\%$. This result is consistent with the value estimated from our calculations[2] if we assume that the populations at the foil exit of the levels coupled by the laser field (which are here the populations at the entrance to the laser field) are proportional to their statistical weights $(2J+1)$. It is worth noting that the relative population change in the laser field depends strongly on the ratio of level populations at the foil exit and this is generally not known. It must also be mentioned that, in the calculations,[2] repopulations of levels 1 and 2 from cascading transitions during the passage of the ions in the laser beam have been neglected, with the exception of the spontaneous transition $2 \rightarrow 1$.

The intensities—with and without laser excitation—for the 57.43 nm line have been recorded as a function of the distance travelled by the ions downstream from the foil-laser interaction region. The laser-on–laser-off decay curve ("BFL decay curve") is a single exponential function [see Eq. (4)]. We have recorded 23 such decay curves containing each ≈ 1000 photon counts at their maximum. Measurements were made at 11 positions along a total path length of 1.3 mm. The recording of one data point (at a given position) lasted ≈ 15 min. The statistical error in each data point is large because each data is a difference of two large and nearly-equal photon numbers. This error (one standard deviation), at the position $x = vt$, is $s(x) \approx [n^L(x)+n^0(x)]^{1/2}$ where $n^L(x)$ and $n^0(x)$ are the numbers of photons counted, with and without laser excitation, respectively. In order to cancel small systematic errors which could be due to the thickening of the foil during the long irradiation time required for recording a decay curve, half of the curves were taken using one direction of travel along the ion beam direction and the other half using the opposite direction. Each BFL decay curve of the set has been well fitted to one exponential function.[4] A mean value of the $2s3d\ ^1D$ lifetime was calculated from the individual lifetime results and the standard deviation of this mean value was estimated from the sample (other contributions to the error are negligible). These results are given in Table II.

The laser effect on the decay curves is shown in Fig. 3 where typical mean decay curves corresponding to laser-off, laser-on, and laser-on–laser-off data are plotted. The data points plotted in this figure are mean values of all our decay data and the error bars on the BFL data represent one standard deviation of the mean values. The slope of the BFL curve, not perturbed by cascades, is clearly steeper than that of the standard beam-foil curve.

We have also measured the lifetime of the $2s3d\ ^1D$ level by the "classical" beam-foil method. In this case, the

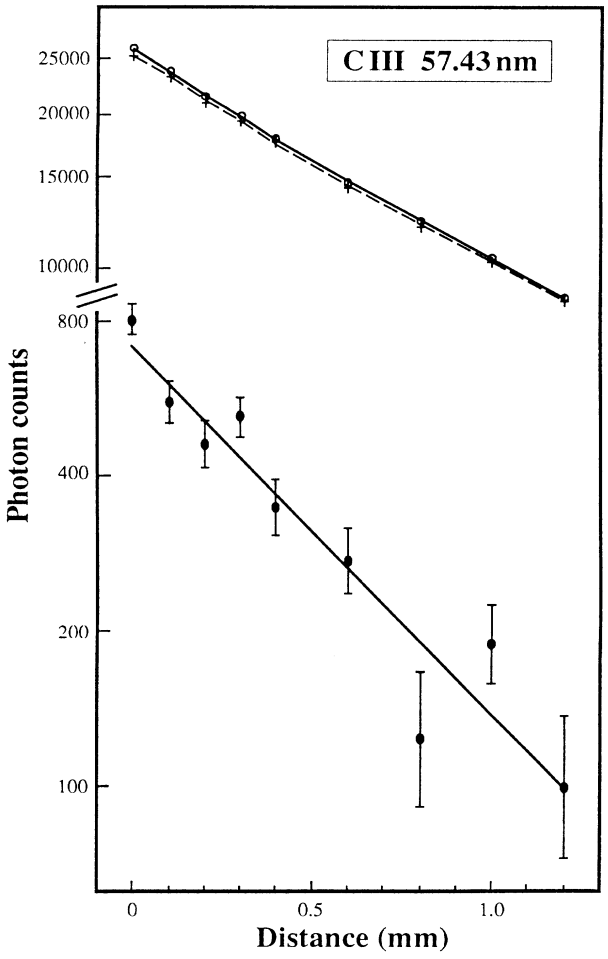

FIG. 3. Typical mean decay curves (+, laser-off; ○, laser-on; and ●, laser-on–laser-off) for the C III 57.43 nm line obtained from all our decay curves. The laser-off and laser-on data are fitted to a sum of two exponentials (dotted and solid upper curves, respectively). The laser-on–laser-off data are fitted to a single exponential (lower solid line).

TABLE II. Lifetime results for the C III $2s3d\ ^1D$ level.

τ (ns)		Method
0.19±0.01	(corrected for cascading)	Beam foil (Ref. 10)
0.14±0.02	($\tau_c \approx 0.4;8$)	Beam foil (Ref. 9)
0.169±0.014	($\tau_c \approx 0.5$)	Beam foil (Ref. 11)
0.175±0.004[a]	($\tau_c \approx 0.7$)	Beam foil (This work.)
0.15±0.01		Beam-foil laser (This work.)
0.179		Config. Interaction (Ref. 12)
0.115		Config. Interaction (Ref. 13)
0.130		Config. Interaction (Ref. 14)
0.154		Model potential (Ref. 15)
0.139		Config. Interaction (Ref. 16)

[a]See text for error estimation.

length of ion beam viewed by the spectrometer was reduced to ≈0.4 mm in order to improve the time resolution.[5] Measurements were made at 40 positions along a total beam length of 18 mm. We have recorded 16 such decay curves containing each ≈4000 photon counts in the maximum of the peak. All the decay curves were analyzed by a least-squares-fitting program.[4] They have been well adjusted by a sum of two exponentials and a small constant. The mean lifetime result is presented in Table II. The quoted uncertainty represents the standard deviation of this mean value estimated from the dispersion of the lifetime results combined with a 2% uncertainty in the ion velocity. This uncertainty does not take into account the error due to incorrect analysis of the decay curve.[6,7] The cascade lifetime result (τ_c) is also given in Table II. The level studied is repopulated from cascading levels which lifetimes are close to the primary lifetime. The principal direct cascading levels are $2s4f\ ^1F^\circ$ [$\tau \approx 0.3$ ns (Ref. 8)], $2p3d\ ^1F^\circ$ [$\tau \approx 0.1$ ns (Ref. 8)], $2s4p\ ^1P^\circ$ [$\tau \approx 0.6$ ns (Ref. 8)], $2p3s\ ^1P^\circ$ [$\tau \approx 0.3$ ns (Ref. 8)], and $2p3d\ ^1D^\circ$ [$\tau \approx 0.1$ ns (Ref. 9)]. In this situation, a correct decomposition of the decay curve is not generally possible and the fit is not unique.

Beam-foil lifetimes[9–11] and theoretical lifetimes[12–16] reported previously are also quoted in Table II for comparison. There is a perfect agreement between our beam-foil results, for the primary and cascade lifetimes, and those of Donnelly, Kernahan, and Pinnington.[11] Our BFL lifetime is ≈15% shorter than our beam-foil result and is in good agreement with the most recent calculations of Laughlin, Constantinides, and Victor[15] and Glass.[16]

B. $3s\ ^2S$ level in C IV

The dye laser was tuned to resonance with the $3s\ ^2S_{1/2}$–$3p\ ^2P^\circ_{3/2}$ transition in C IV ($\lambda = 580.1$ nm) and the $2p\ ^2P^\circ_{1/2,3/2}$–$3s\ ^2S_{1/2}$ transition ($\lambda = 41.96$ nm) was observed (see Fig. 4).

The relative laser-on–laser-off intensity change observed for the 41.96 nm line at the exit of the interaction region, $[I^L(0) - I^0(0)]/I^0(0)$, is ≈ −2% (in the experimental conditions given in Table I). The ratio of populations N_1/N_2 at the foil exit deduced from the observed relative intensity change and from our previously reported calculations[2] (using theoretical lifetimes) is found to be ≈0.7. This result seems to indicate that the population at the foil exit of the $3s\ ^2S_{1/2}$ state relative to that of the $3p\ ^2P_{3/2}$ is stronger than that corresponding to a statistical distribution. Equation (3) shows that the BFL decay curve for the $3s\ ^2S_{1/2}$ level is a sum of two exponential functions. We have simulated that curve using Eqs. (3) and (5) where the values of A_{21}, A_1, and A_2 were the theoretical ones and the value of $\Delta N_2(0)/\Delta N_1(0)$ was deduced from the calculations[2] (using our deduced value for the ratio of populations at the foil exit). The amplitude of the second exponential (corresponding to τ_2) in Eq. (3) was found to represent only ≈10% of the amplitude of the first exponential (corresponding to τ_1). A least-squares fitting of the simulated BFL decay curve with one exponential instead of two reproduces the theoretical lifetime of the $3s\ ^2S$ level with an error of only ≈1%. Note that as the lifetimes of the two levels are approximately equal [$\tau_{3s} \approx 0.23$ (Ref. 17), 0.24 ns (Ref. 18); $\tau_{3p} \approx 0.22$ ns

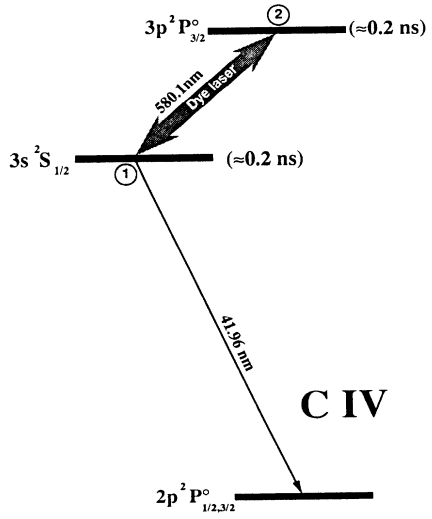

FIG. 4. Partial energy-level diagram of C IV.

TABLE III. Lifetime results for the C IV $3s\ ^2S$ level.

τ (ns)		Method
0.26±0.03	($\tau_c \approx 1.2$)	Beam foil (Ref. 9)
0.22±0.02	($\tau_c \approx 0.73$)	Beam foil (Ref. 8)
0.258±0.004[a]	($\tau_c \approx 1$)	Beam foil (This work.)
0.21±0.02		Beam-foil laser (This work.)
0.23		Z expansion (Ref. 17)
0.23		Numerical Hartree Fock (Ref. 17)
0.242		Coulomb approx. (Ref. 18)

[a]See text for error estimation.

(Refs. 17 and 18)], a two-exponential adjustment would have no meaning.

We have recorded 15 BFL decay curves containing each ≈ 1000 photon counts at their maximum. Measurements were made at six positions along a total path length of 3 mm downwards the interaction region. The recording of one data point (at a given position) lasted ≈ 10 min. Half of the curves were taken using one direction of travel along the ion beam direction and the other half using the opposite direction (see Sec. III A). All the decay curves are well fitted to one exponential function.[4] A mean value for the $3s\ ^2S$ lifetime and the standard deviation of this mean value were calculated from the sample. These results are given in Table III. As outlined in Sec. III A, only the statistical error is important for the BFL measurement.

The $3s\ ^2S$ lifetime was also determined in the present work by the classical beam-foil method using a good spatial resolution (length of ion beam viewed by the spectrometer ≈ 0.4 mm). Eight recorded decay curves containing each ≈ 4000 photon counts in the maximum of the peak (measurements made at 38 positions along a total beam length of 40 mm) have been well adjusted to a sum of two exponentials and a small constant.[4] The mean lifetime result obtained is presented in Table III. The quoted uncertainty represents the standard deviation of this mean value estimated from the dispersion of the lifetime results combined with a 2% uncertainty in the ion velocity and does not take into account the error due to incorrect analysis of the decay curve. The $3s\ ^2S$ level is repopulated by direct cascading from $np\ ^2P^\circ$ levels ($n \geq 3$). The lifetime of the repopulating $3p\ ^2P^\circ$ term is undistinguishable from the primary lifetime. The observed "cascade" lifetime ($\tau_c \approx 1$ ns) represents some mean value of the other cascading level lifetimes.

Beam-foil[8,9] and theoretical[17,18] lifetime results previously reported are also quoted in Table III. Our BFL lifetime is $\approx 20\%$ shorter than our classical beam-foil value (which reproduces the value of Poulizac and Buchet[9]) and is in agreement, within experimental errors, with the theoretical values.

IV. CONCLUSIONS

The BFL method has been applied for the first time to lifetime measurements in multiply charged ions. The C III $2s3d\ ^1D$ and C IV $3s\ ^2S$ lifetimes have been determined. The observed population changes "laser on"-"laser off" for the two levels studied amount to a few percents, at the exit of the interaction region. Long-lasting times are thus necessary for recording statistically significant BFL decay data (see Sec. III). We have obtained BFL lifetimes with an uncertainty (one standard deviation) of about 7% and 10% for the $2s3d\ ^1D$ and $3s\ ^2S$ levels, respectively. Our BFL lifetime results are in good agreement with the theoretical values. It must be pointed out that the precision of the BFL lifetimes is mainly limited by the statistical uncertainties in the photon counting.

For comparison purposes, we have also measured the lifetimes of the $2s3d\ ^1D$ and $3s\ ^2S$ levels using the classical beam-foil method. For both levels, decompositions of the multiexponential beam-foil decay curves have given lifetimes in good agreement with the previously published beam-foil values but about 20% longer than the BFL values (see Tables II and III).

The present experiment has demonstrated the feasibility of the BFL method for measuring lifetimes in multiply ionized atoms. However, in the present state of experimental conditions (dye lasser power, ion beam current, detection efficiency, etc.), its applicability is limited to levels (excited in the foil) for which the induced population change in the laser field and the emitted transition observed are "sufficiently" strong to give rise to significant differences in the intensities "laser on" and "laser off" of the observed transition. This method will be particularly useful when the level studied is repopulated by strong cascading from levels whose lifetimes are close to the lifetime studies. It is indeed well known that in this case, the decomposition of the classical multiexponential beam-foil decay curve can lead to erroneous lifetimes.

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