Precision lifetime measurements of N II levels with the beam-foil–laser method

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Precision lifetime measurements using laser excitation of a fast ion beam preexcited in a carbon foil are reported for two levels in N II. The cascade-free decays of the fluorescence intensities give lifetimes of 0.249 ± 0.004 and 0.267 ± 0.010 ns for the N II $2p3d\ ^1F^{\circ}$ and $2p3s\ ^1P^{\circ}$ levels, respectively. The lifetime result for the $2p3d\ ^1F^{\circ}$ level—which is weakly repopulated by long-lived cascades—is in good agreement with beam-foil values and with the theoretical lifetime of McEachran and Cohen [J. Quant. Spectrosc. Radiat. Transfer **27**, 119 (1982)]. The lifetime result for the $2p3s\ ^1P^{\circ}$ level—which is strongly repopulated by cascades—differs significantly from most of the previous experimental values but is in good agreement with the theoretical lifetimes of Luken and Sinanoglu [J. Chem. Phys. **64**, 3141 (1976)], Beck and Nicolaides [Phys. Lett. **56A**, 265 (1976)], McEachran and Cohen, and Fawcett [At. Data Nucl. Data Tables **37**, 411 (1987)]. The f-value trend for the $2p^2\ ^1D-2p3s\ ^1P^{\circ}$ transition along the C I sequence is discussed.

I. INTRODUCTION

The beam-foil–laser (BFL) method recently proposed for cascade-free lifetime measurements leads, in principle, to the determination of accurate lifetime values.[1,2] This method is described in detail in Refs. 1 and 2 and will be only briefly recalled here.

The BFL method combines a first nonselective excitation of a fast monoenergetic ion beam by a carbon foil with a second selective excitation of the beam by the intracavity radiation from a cw dye laser. The laser radiation is tuned to resonance with a transition in an ion emerging from the foil and induces population changes in the two levels of this transition. The population change of one of these levels is measured by monitoring the intensity change of a line emitted from this level, in the two situations: "laser on" and "laser off." This change of intensity is measured as a function of the distance traveled by the ions downstream from the laser interaction region. The lifetime of the level is easily extracted from the analysis of the decay curve of this intensity change (BFL decay curve) which reduces to one exponential or a sum of two exponentials, according as the level studied is the upper level or lower level of the laser-induced transition.[1,2]

The BFL method has been recently applied to the measurements of the lifetimes of two levels in multiply ionized carbon.[2] However, the population changes of these levels in the laser field amounted to only a few percent and their lifetimes have been measured with a low precision (error $\approx 10\%$). Indeed, prohibitive times of measurements would have been necessary to obtain more statistically significant data and thus more precise lifetime values.

The population changes of the levels coupled by the laser field depend strongly on the ratio of the populations of these levels at the entrance of the field.[3] If the lifetimes of the levels are very different (e.g., by an order of magnitude), it is possible to change this ratio—by modifying the distance between the foil and the laser beam—and thus to increase the "laser effect": $(I^L-I^0)/I^0$ (I^L and I^0 denote the observed intensities, laser on and laser off, respectively, at the exit of the laser interaction region). In these conditions, very precise BFL lifetimes can be obtained.

In the present work, strong laser effects have been found for two levels in N II ($2p3d\,^1F^{\circ}$ and $2p3s\,^1P^{\circ}$) using this method. These strong effects allow us to report here the first precise lifetime measurements obtained with the BFL method.

II. EXPERIMENT

A. Experimental arrangement

The experimental setup is very similar to that described in detail previously (see Refs. 1 and 2). A N_2^+ beam is supplied by a 2-MV Van de Graaff accelerator equipped with an rf source into which N_2 gas was admitted. The ion beam crosses at right angles successively a carbon foil and the intracavity beam of a cw dye laser (modified Coherent CR 599) pumped by a 20-W argon laser (Coherent Innova 20). The wavelength—in the visible region—of the dye laser is tuned by a birefringent filter and analyzed by an Ebert-Fastie spectrometer acting as a wavemeter. The photons of deexcitation of the upper (or lower) level of the transition induced by the laser are analyzed in the vacuum ultraviolet region using a Seya-Namioka-type spectrometer and detected by channeltron detectors. A beam chopper and a lock-in device allow the counting of photons corresponding to the two situations: "laser on" and "laser off." The carbon foil and the interacting laser beam are movable together parallel to the ion beam axis with a precision of about ±0.01 mm.

The intracavity power of the dye laser is controlled

TABLE I. Summary of experimental conditions.

	N II $2p3d\ ^1F^\circ$	N II $2p3s\ ^1P^\circ$
N_2^+ beam energy (MeV)	1.5	1.0
N_2^+ beam current (μA/cm^2)	≈ 8	≈ 14
Ion beam cross section (mm^2)	0.6×6.0	0.6×6.0
Dye	DCM	DCM
Dye intracavity power (W)	≈ 20	≈ 15
Doppler-corrected dye wavelength (nm)	661.09	648.23
Dye-laser line FWHM (nm)	≈ 0.09	≈ 0.04
Dye-laser beam diameter (mm)	≈ 1	≈ 1
Carbon foil thickness (μg/cm^2)	≈ 5	≈ 5
Carbon foil size (mm^2)	1.5×7.0	1.5×7.0
Ion beam portion viewed (mm)	≈ 0.6	≈ 1
Distance between foil and laser beam axis (mm)	≈ 2 or ≈ 5	≈ 3 or ≈ 8
Doppler broadening due to the angular divergence of the ion beam in the foil (FWHM) (nm)	≈ 0.09	≈ 0.11

during the measurements so that photon counts can be normalized to take into account small variations of this power. Moreover, the dye-laser wavelength is repetitively measured during the whole experiment and corrected if necessary, to remain tuned to the Doppler-corrected wavelength of the induced transition.

The distance between the foil and the laser beam is longer than that used in previous works,[1,2] in order to increase laser effects. As outlined in Sec. I, this is possible because the lower and upper levels of the induced transitions considered in the present work have very different lifetimes (see Secs. II B and II C).

The details of the experimental conditions are summarized in Table I.

B. Principle of the $2p3d\ ^1F^\circ$ lifetime measurement

The dye laser was tuned to resonance with the $2p3p\ ^1D-2p3d\ ^1F^\circ$ transition in N II ($\lambda=661.06$ nm) and the laser-on and laser-off intensities of the $2p^2\ ^1D-2p3d\ ^1F^\circ$ transition ($\lambda=57.46$ nm) were recorded. These transitions are shown in Fig. 1 together with approximate values of the level lifetimes. The Doppler-corrected wavelength of the laser radiation is given in Table I. The distance between the foil and the laser beam axis was ≈ 2 or ≈ 5 mm. In these conditions, the population, at the laser field entrance, of the short-lived $2p3d\ ^1F^\circ$ level was drastically reduced whereas that of the long-lived $2p3p\ ^1D$ lower level was practically the population at the exit of the foil. Thus, during the passage of the ions in the laser field the $2p3d\ ^1F^\circ$ level was abundantly repopulated. The intensities, with and without laser excitation, for the 57.46-nm line were measured as a function of the distance traveled by the ions downstream from the foil. The difference of intensities—with and without laser interaction—after the laser interaction region gives a BFL decay curve which is a single exponential.[2] As the foil thickens during the irradiation and breaks after some irradiation time, it is necessary to record a BFL decay curve in a "sufficiently" short time. To obtain statistically significant data for the intensity changes (laser on and laser off), many BFL decay curves have been recorded. Moreover, in order to cancel small systematic errors which could be due to the thickening of the foil during the irradiation time required to record a single decay curve, half of the curves were taken where the foil-laser interaction region was moved upstream along the ion beam direction and the other half downstream.

C. Principle of the $2p3s\ ^1P^\circ$ lifetime measurement

The dye laser was tuned to resonance with the $2p3s\ ^1P^\circ-2p3p\ ^1P$ transition in N II ($\lambda=648.20$ nm) and the laser-on and laser-off intensities of the $2p^2\ ^1D-2p3s\ ^1P^\circ$ transition ($\lambda=74.70$ nm) were recorded. These transitions are shown in Fig. 2 together with approximate

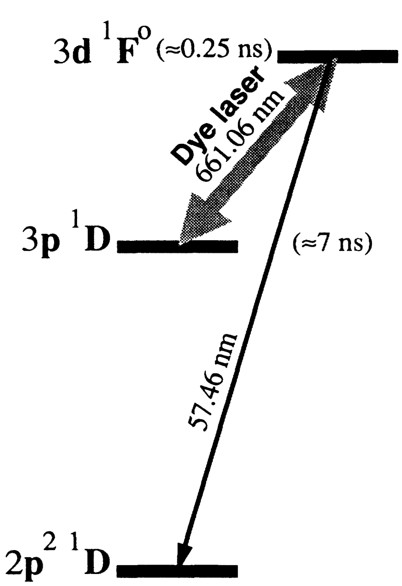

FIG. 1. Partial energy-level diagram of N II showing the levels involved in the $2p3d\ ^1F^\circ$ lifetime measurement.

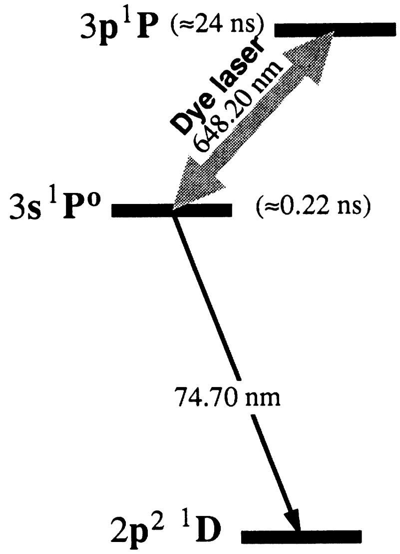

FIG. 2. Partial energy-level diagram of N II showing the levels involved in the $2p3s\ ^1P^o$ lifetime measurement.

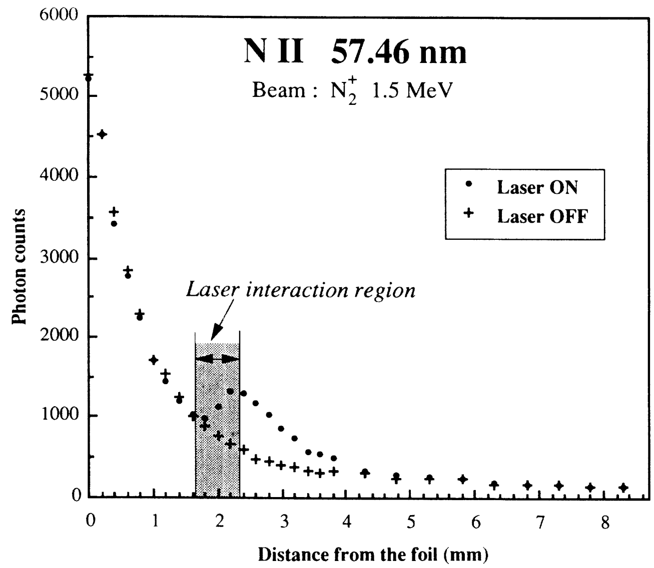

FIG. 3. Decay curves recorded, with and without laser excitation, for the N II 57.46-nm line using 1.5-MeV $N_2{}^+$ beam.

values of the level lifetimes. The Doppler-corrected wavelength of the laser radiation is given in Table I. The distance between the foil and the laser beam axis was ≈ 3 mm or ≈ 8 mm. The population, at the laser field entrance, of the short-lived $2p3s\ ^1P^o$ level was then considerably weaker than this population at the exit of the foil whereas the population of the very-long-lived $2p3p\ ^1P$ upper level was nearly unchanged. In these conditions, the laser field induced strong repopulation of the $2p3s\ ^1P^o$ level.[3]

We have recorded the intensities, with and without laser excitation, of the 74.70-nm line as a function of the distance traveled by the ions downstream from the laser interaction region. The BFL decay curve of the $2p3s\ ^1P^o$ level—which is the lower level of the laser-induced transition—is a sum of two exponentials corresponding to the lifetimes of the levels coupled by the laser field.[2] As the lifetime of the $2p3p\ ^1P$ upper level is very long, the second exponential reduces to a constant over the 1-mm distance where the intensity was measured. To obtain a precise BFL decay curve, it was necessary to record many BFL decay curves as explained in Sec. II B.

III. RESULTS AND DISCUSSION

A. $2p3d\ ^1F^o$ level in N II

The intensities, with and without laser excitation, for the 57.46-nm line are plotted in Fig. 3 as a function of the distance traveled by the ions downstream from the foil. A strong ($\approx 100\%$) laser effect (see definition in Sec. I) appears. A precise BFL decay curve was obtained by accumulating ≈ 100 BFL decay curves containing each ≈ 250 photon counts at their maximum and 15 data points along a total beam length of 2 mm. Figure 4 shows the resulting BFL decay curve. A precise lifetime value for the $2p3d\ ^1F^o$ level ($\tau = 0.249 \pm 0.004$ ns) has been deduced from a least-squares fitting of the data to one exponential function. The error represents the statistical error (one standard deviation) estimated from the dispersion of the individual lifetime results (note that in this case the same error has been obtained by the fitting program) combined with an uncertainty of 1% in the ion velocity.

We have also measured in the present work the $2p3d\ ^1F^o$ lifetime by the standard beam-foil (BF) method using a good spatial resolution (length of ion beam viewed by the spectrometer ≈ 0.4 mm). Thirteen decay curves—each containing ≈ 1000 photon counts at their

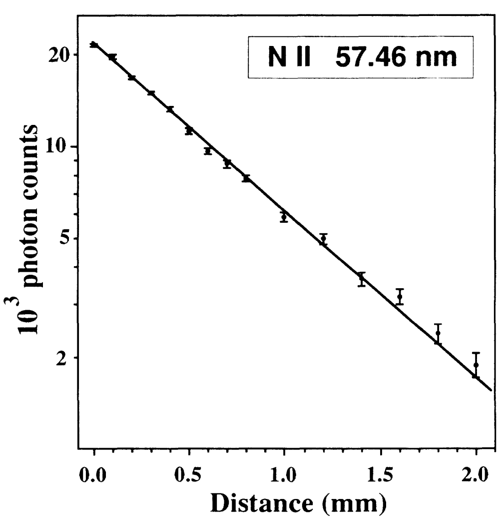

FIG. 4. BFL (laser-on–laser-off) decay curve for the N II 57.46-nm line using 1.5-MeV $N_2{}^+$ beam. The data are fitted to a single exponential function. The error bars represent the statistical errors (one standard deviation).

TABLE II. Lifetime results for the N II $2p3d\ ^1F^\circ$ level.

τ (ns)	τ_{casc} (ns)	Experiment
0.22±0.03	≈0.98,9.4	Beam-foil (Ref. 4)
0.246±0.012	≈1.5	Beam-foil (Ref. 5)
0.254±0.004[a]	≈7	Beam-foil (this work)
0.249±0.004		Beam-foil–laser (this work)
τ (ns)		Theory
0.28		Restricted Hartree-Fock (Refs. 6,7)
0.258		Polarized frozen-core Hartree-Fock (Ref. 8)
0.149		Model potential (Ref. 9)
0.220		Hartree-Fock relativistic (Ref. 10)

[a]See text for the error estimation.

maximum and 27 data points along a total beam length of 8.5 mm—were recorded and well adjusted to a sum of two exponentials. The mean values of the primary and cascade lifetimes deduced from this sample are given in Table II. The amplitude of the second exponential is weak and its lifetime is ill defined. The error indicated for the primary lifetime represents one standard deviation of the mean value estimated from the dispersion of the results combined with 1% uncertainty in the ion velocity. It is worth noting that this error is underestimated because it does not take into account the (nonestimable) uncertainty due to an incorrect analysis of the decay curve.

The lifetime value obtained with the BFL method together with the experimental[4,5] and theoretical[6–10] results available are also quoted in Table II. The lifetimes obtained in different BF experiments (Buchet *et al.*,[4] Kernahan *et al.*,[5] and this work) are in agreement, within the one standard deviation experimental error. Moreover, there is a surprisingly good agreement between the BFL result and the most recent BF values (Ref. 5 and this work). This agreement can be explained by the following comments. The $2p3d\ ^1F^\circ$ level is repopulated by cascading principally from the $2pnf\ ^1F$, ^1G, ^1D and $2pnp\ ^1D$ levels ($n \geq 3$) whose lifetimes are at least one order of magnitude longer than the primary lifetime (see Ref. 6). If, in addition, repopulation by cascading is weak, BF lifetimes are known to be reliable. These conditions are probably fulfilled for the $2p3d\ ^1F^\circ$ level.

The precise BFL lifetime value is in good agreement with the lifetime calculated by McEachran and Cohen.[8]

B. $2p3s\ ^1P^\circ$ level in N II

The laser effect recorded for the 74.70-nm observed transition was ≈20% for both foil-laser distances considered in this experiment. This steady laser effect is due to the fact that the population of the short-lived $2p3s\ ^1P^\circ$ level at a few mm downstream from the foil is principally due to repopulation of this level from long-lived cascades. The resulting BFL decay curve obtained by accumulating ≈100 BFL decay curves (each containing ≈500 photon counts at their maximum and 6 data points along a total beam length of 1 mm) is shown in Fig. 5. This curve was well fitted to one exponential indicating that the (approximately constant) second component is very weak (see Sec. II C).

A precise lifetime value for the $2p3s\ ^1P^\circ$ level (τ=0.267±0.010 ns) has been deduced from a least-squares fitting of the data to one exponential function. The uncertainty represents the statistical error (one standard deviation) combined with an uncertainty of 1% in the ion velocity.

The experimental[5,11–14] and theoretical[7,8,10,15–17] results available for the $2p3s\ ^1P^\circ$ level are presented in Table III. The experimental BF values[5,12–14] are very scattered and the phase-shift value[11] is highly uncertain. The $2p3s\ ^1P^\circ$ level is repopulated by cascading, most probably arising from $2pnp\ ^{1,3}D$, $^{1,3}P$, and $^{1,3}S$ terms ($n \geq 3$). Strong cascades are expected from $2p3p\ ^1D$, ^1P, and ^1S levels whose lifetime values are ≈6, ≈24, and

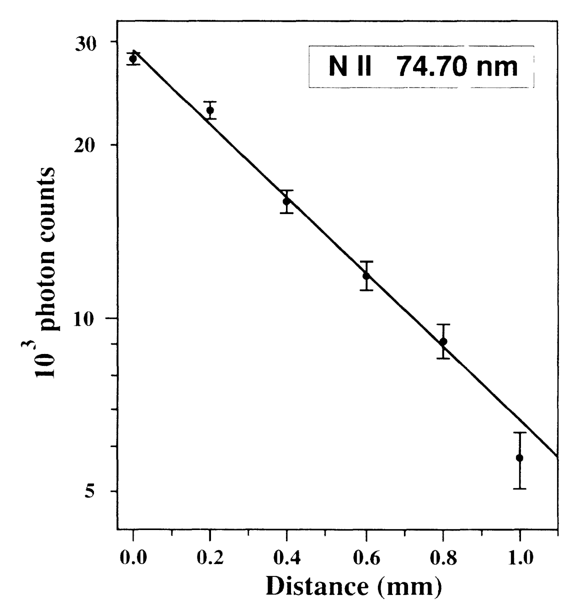

FIG. 5. BFL (laser-on–laser-off) decay curve for the N II 74.70-nm line using 1-MeV N_2^+ beam. The data are fitted to a single exponential function. The error bars represent the statistical errors (one standard deviation).

TABLE III. Lifetime results for the N II $2p3s\,^1P^o$ level.

τ (ns)	τ_{casc} (ns)	Experiment
0.8±0.4	≈24	Phase shift (Ref. 11)
0.43±0.10[a]	≈14	Beam-foil (Ref. 12)
0.213±0.010[a]	≈0.7, ≈10.4	Beam-foil (Ref. 5)
0.22±0.03	≈0.7, ≈11	Beam-foil (Ref. 13)
0.35±0.10		Beam-foil (Ref. 14)
0.267±0.010		Beam-foil–laser (this work)
τ (ns)		Theory
0.5[b]		Restricted Hartree-Fock (Ref. 7)
0.50		Scaled Thomas-Fermi (Ref. 15)
0.29(length)/0.24(velocity)		Nonclosed shell many-electron theory (Ref. 16)
0.252		First-order theory of oscillator strengths (Ref. 17)
0.286		Polarized frozen-core Hartree-Fock (Ref. 8)
0.257		Hartree-Fock relativistic (Ref. 10)

[a]The observed 74.70-nm line was not resolved from the 74.58- and 74.84-nm lines.
[b]Neglecting the weak $2p^2\,^1S-2p3s\,^1P^o$ transition probability.

≈4.5 ns, respectively (see Refs. 6, 18, and 19). The cascade of 0.7 ns observed in the BF results of Refs. 5 and 13 does not correspond to any strong cascading possibility and is probably due to an incorrect decomposition of the decay curves. This can explain why the most precise BF results[5,13] are shorter than the BFL lifetime value. The BFL lifetime is in good agreement with the most recent calculations.[8,10,16,17]

C. Oscillator strength of the $2p^2\,^1D$-$2p3s\,^1P^o$ transition along the C I sequence

Smith *et al.* have discussed the oscillator strength trend for the $2p^2\,^1D-2p3s\,^1P^o$ transition along the C I sequence.[20] They have noted that as the upper state $2p3s\,^1P^o$ can mix with the $2p^3\,^1P^o$ state, and as the energy

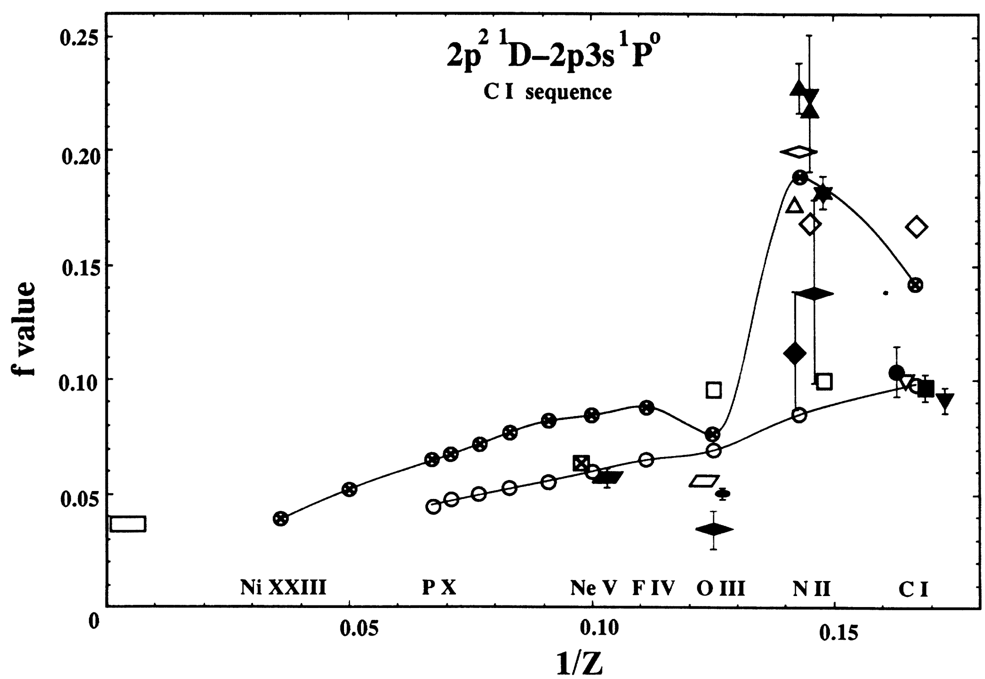

FIG. 6. Oscillator strengths as a function of $1/Z$ for the $2p^2\,^1D-2p3s\,^1P^o$ transition of the carbon isoelectronic sequence. Branching ratios from Fawcett (Ref. 10) are used for experimental values. The theoretical data sources (open symbols) are □, Ref. 7; ⊠, Ref. 26; ▽, Ref. 27; ○, Ref. 15; ▱, Ref. 28; ▭, Ref. 29; △, Ref. 16; ⬭, Ref. 17; ◇, Ref. 8; ⊗, Ref. 10. The experimental data sources (solid symbols) are ●, Ref. 21; ■, Ref. 22; ▼, Ref. 23; ◆, Ref. 12; ▲, Ref. 5; ▰, Ref. 25; ⧗, Ref. 13; ▬, Ref. 24; ⬬, Ref. 14; ★, this work. The solid curves have been drawn to guide the eye to see the trends of theoretical f values from Refs. 10 and 15.

levels do cross between N II and O III, the f-value curve for this transition might exhibit something unusual in the vicinity of the level crossing. We have plotted in Fig. 6 theoretical and experimental oscillator strengths versus $1/Z$ for the $2p^2\,{}^1D - 2p3s\,{}^1P^\circ$ transition.

The $2p3s\,{}^1P^\circ$ level has a second deexcitation branch: to the $2p^2\,{}^1S$ level. Thus, theoretical branching ratios must be used to deduce f values from experimental lifetimes. Lifetimes have been measured in C I,[21–23] N II,[5,12–14] O III,[14,24] and Ne V[25] using the beam-foil or/and the phase-shift methods. All these lifetimes are corrected for cascading and their reliability is questionable; only the cascade-free BFL result in N II of the present work is expected to be accurate. As discussed in Sec. III B, this BFL lifetime is in good agreement with the most recent calculations.

Warner and Kirkpatrick[15] using intermediate coupling calculations have obtained the f values of the $2p^2\,{}^1D - 2p3s\,{}^1P^\circ$ transition for all the elements from C I to P X and Fawcett[10] using the Hartree-Fock relativistic method has reported these values for ions between C I and Ni XXIII. A few other theoretical f values for this transition have also been reported for other elements of the C sequence.[26–29] As can be seen from Fig. 6 there are large discrepancies between theoretical f values.

We have used the very recent branching ratios from Fawcett[10] to estimate experimental f values. However, it is worth pointing out that the theoretical branching ratios for the $2p^2\,{}^1D - 2p3s\,{}^1P^\circ$ transition are large ($\approx 0.8-1$) for the elements studied experimentally so that the $2p^2\,{}^1S - 2p3s\,{}^1P^\circ$ transition does not contribute much to the $2p3s\,{}^1P^\circ$ lifetime. We must also note that the branching ratio for the $2p^2\,{}^1D - 2p3s\,{}^1P^\circ$ transition in N II has been measured by Morrison *et al.*[30] and found larger than 0.98. Fawcett's value[10] for this branching ratio—which is the same as that of McEachran and Cohen[8]—is equal to 0.967.

Examination of Fig. 6 shows that the N II f value corresponding to the accurate BFL result of this work is clearly outside the smooth trend established from the theoretical values of Warner and Kirkpatrick.[15] The N II f value deduced from the present experiment which is in agreement with the most recent calculations is about two times greater than the f value of nearby higher-Z elements of the carbon sequence.

IV. CONCLUSIONS

We have obtained the first precise (error of about a few %) lifetime values with the BFL method for two levels in N II:$2p3d\,{}^1F^\circ$ and $2p3s\,{}^1P^\circ$. The improvement of the precision relative to previous BFL lifetime measurements[2] is due to strong population changes in the laser field obtained in the present work by increasing the distance between the foil and the laser beam. This technique is applicable when the two levels coupled by the laser field have very different lifetimes.

For the $2p3d\,{}^1F^\circ$ level, the BFL lifetime is in good agreement with the BF results. This indicates that this level is weakly repopulated by cascading from levels whose lifetimes are much longer than the primary lifetime. The BFL result is in good agreement with the lifetime value calculated by McEachran and Cohen[8] using the polarized frozen-core version of the Hartree-Fock approximation (see Table II).

The BF lifetimes of the $2p3s\,{}^1P^\circ$ level—which is strongly repopulated by cascading—are very scattered and the BFL lifetime is the only reliable experimental result. Good agreement was found between the BFL lifetime and the most recent calculations of Luken and Sinanoglu,[16] Beck and Nicolaides,[17] McEachran and Cohen,[8] and Fawcett[10] (see Table III).

Analysis of the oscillator strength for the $2p^2\,{}^1D - 2p3s\,{}^1P^\circ$ transition along the C I sequence confirms the irregularity expected to occur in the vicinity of N II, O III. However, precise measurements for ions other than nitrogen are needed to establish definitively the validity of the f trend obtained by Fawcett's calculations[10] (see Fig. 6).

ACKNOWLEDGMENTS

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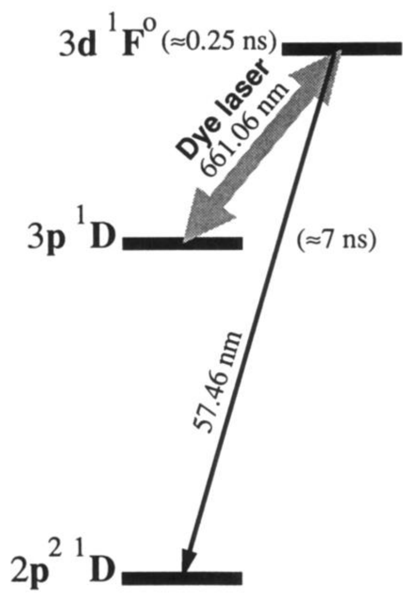

FIG. 1. Partial energy-level diagram of N II showing the levels involved in the $2p3d\ ^1F^o$ lifetime measurement.

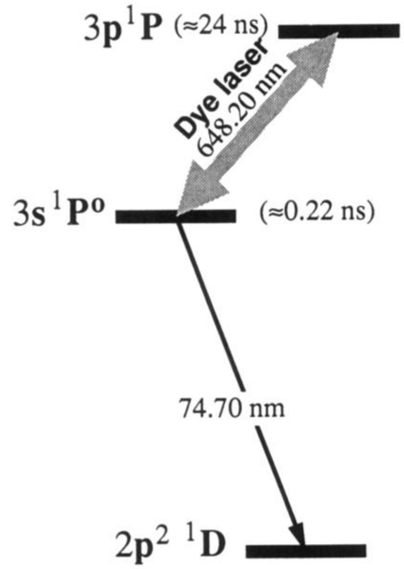

FIG. 2. Partial energy-level diagram of N II showing the levels involved in the $2p3s\ ^1P^\circ$ lifetime measurement.

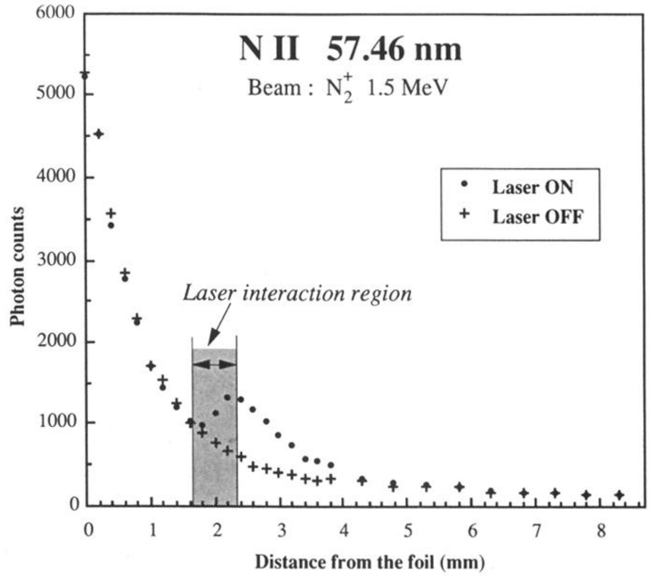

FIG. 3. Decay curves recorded, with and without laser excitation, for the N II 57.46-nm line using 1.5-MeV N_2^+ beam.