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Radiative Lifetimes for uv Multiplets of C II and C III

L. Heroux

Air Force Cambridge Research Laboratories, Laurence G. Hanscom Field, Bedford, Massachusetts 01730

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The beam-foil method has been used to measure radiative lifetimes of 11 uv multiplets of C II and C III. The wavelengths of the measured multiplets were from 372 to 1176 Å. All of the multiplets were transitions among low-lying terms of the ions. Several were transitions to the ground term. For the majority of the multiplets, corrections for cascading were necessary to obtain their radiative lifetimes. The transition probabilities for the multiplets obtained from the lifetime measurements were found to be in good agreement with recent theoretical values.

INTRODUCTION

The use of beam-foil spectroscopic techniques for determining the radiative lifetimes of excited levels of multiply ionized atomic species is now well established.[1] With this technique, a high-energy ion beam is passed through a thin foil from which the beam emerges in various stages of ionization and in various states of excitation. The decay in the population of an excited level as a function of distance l downstream from the intersection of the foil and beam is obtained directly from the decay in intensity of any emission line that originates from the excited level. Because the velocity of the emergent beam is known, a measurement of the spatial decay of the excited level is equivalent to the measurement of its temporal decay. Thus the mean lifetime of the excited level can be determined. A major source of error in the determination of lifetime by this method is introduced by cascading transitions from higher levels which repopulate the level being measured along the length of the ion beam. Cascading processes are generally important in any lifetime experiment. However, in contrast to other methods of measuring lifetimes, both depopulation of the excited levels by collisional processes and reabsorption of the emission lines are completely negligible when the beam-foil method is used.

This paper presents experimental lifetime measurements, obtained with the beam-foil method, for several low-lying terms of C II and C III. The most intense multiplets corresponding to transitions among the lower levels of atoms and ions often fall in the extreme ultraviolet (xuv) region of the spectrum. The multiplets of C II and C III measured here have wavelengths shorter than 1200 Å. With the exception of the C III multiplet at 372 Å, the measured multiplets are observed by means of either the only transition or the dominant transition from the excited term. The transition probabilities of the multiplets therefore could be obtained directly from the measured mean lifetimes.

THE EXPERIMENTAL METHOD

The experimental method has been described previously.[2,3] To produce the emission spectra of C II and C III, a C^+ beam was passed through a thin carbon foil. The energy of the incident ion beam was (1.00 ± 0.02) MeV and the foil thickness was either 5 or 10 $\mu g/cm^2$. The direction of the ion beam was perpendicular to the plane of the foil. The cross-sectional area S of the ion beam determined by the carbon foil was 1.25 cm^2. The spectra were observed with a grazing-incidence vacuum monochromator that viewed the beam at 90° to the beam direction between the distances l and $l + \Delta l$ downstream from the intersection of the foil and beam. The incremental length Δl of the beam was 1 mm, Δl being determined by the angular aperture of the monochromator and the distance between the entrance slit of the monochromator and the ion beam. By moving the foil in small steps along the axis of the beam, the photon flux of a particular multiplet emitted within the volume element $S\Delta l$ could be measured as a function of distance l along the beam. Since the velocity of the beam is known, knowledge of l is equivalent to a measurement of time. The monochromator covered the wavelength region from about 250 to 1250 Å with an instrumental linewidth of about 2 Å. Photoelectron-counting techniques were used to measure the photon flux. The only important difference in the characteristics of the previous and present detection systems is that the background noise was reduced from its previous value of about 4 counts/sec to less than 1 count/sec. A low background is particularly important when the decay of the multiplet is influenced by cascading processes having lifetimes longer than the mean lifetime of the upper level of the multiplet being measured. When this type of cascading occurs, the cascading processes can be studied only by examining the decay of the multiplet at large distances from the foil where the signal levels are generally extremely low.

TREATMENT OF DATA AND ERRORS

When the multiplet is intense and also free from cascading, the lifetime measurement can be highly accurate. An example of the experimental data for this type of multiplet that corresponds to the transition $2s2p^2\ ^4P$-$2p^3\ ^4S^0$ in C II at 1010 Å is given in Fig. 1. The thickness of the carbon foil was 10 μg/cm^2. The plotted points give the experimental measurements corrected for a constant background (1 count/sec). Because of the low counting rates, which are characteristic of beam-foil measurements, a counting interval of 100-sec duration was used for each foil position. The mean lifetime of the $2p^3\ ^4S^0$ term was obtained from the logarithmic slope of the straight line fitted to the data by the method of least squares. The error indicated for the mean lifetime is the estimated total experimental error which includes the rms error of the least-squares fit and the error in the determination of the beam velocity, discussed below. This multiplet of C II was the only multiplet of the group studied here that appeared to be completely free from cascading. This might have been predicted because the terms in C II that could cascade into the $2p^3\ ^4S^0$ term have not been observed spectroscopically.[4] These terms, tabulated by Moore (Ref. 4) as possible terms for ions in the B I isoelectronic sequence, are the terms $2p^2ns\,^4P$ and $2p^2nd\,^4P$ for $n \geq 3$. For the C II ion, these terms probably lie above the ionization limit of the ion and, therefore, would not be expected to contribute to cascading into the $2p^3\,^4S^0$ term.

Corrections for cascading were necessary for determining the radiative lifetimes of the remaining multiplets of C II and C III. Figure 2 gives the experimental data for one of those multiplets

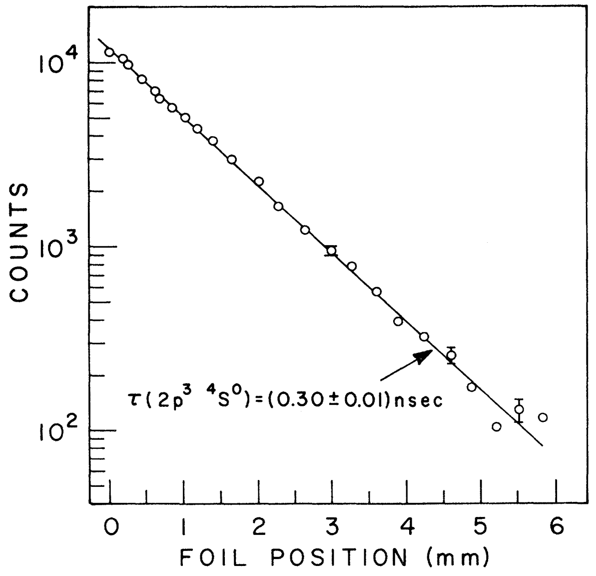

FIG. 1. Counts versus foil position l for the cascade-free multiplet of C II at 1010 Å. The data have been corrected for a constant background only. The counting interval for each point was 100 sec, the foil thickness was 10 μg/cm^2, and the incident C$^+$ beam energy was (1.00 ± 0.02) MeV.

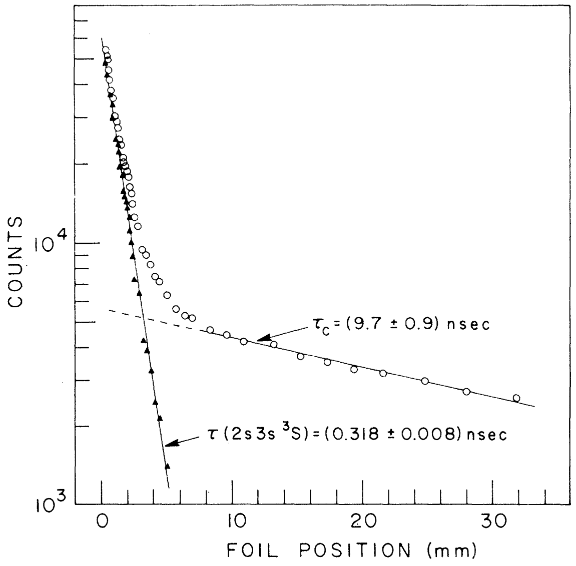

FIG. 2. Counts versus foil position l for the C III multiplet at 538 Å. The counting interval was 100 sec, the foil thickness was 10 μg/cm^2, and the incident beam energy was (1.00 ± 0.02) MeV. ○, data corrected for background; ▲, data corrected for both background and cascading. The error indicated for the lifetime is only that part of the total error associated with the cascade correction.

that is clearly influenced by cascading. The data are for the $2s2p\,^3P^0$-$2s3s\,^3S$ multiplet of C III at 538 Å. The foil thickness, beam energy, and counting interval used to obtain these data are identical to those given for the data in Fig. 1. The data in Fig. 2 were also corrected for a constant background only. The limiting slow decay which sets in at foil positions greater than 8 mm is assumed to originate from cascading transitions into the upper $2s3s\,^3S$ term of C III.

The cascading analysis for this multiplet is typical for the multiplets of C II and C III. The analysis assumes that the observed spatial decay of the multiplet can be constructed from the sum of two exponential components consisting of a decay owing to radiative transitions from the upper term of the multiplet and a decay associated with cascading transitions into the upper term of the multiplet. The cascading component is obtained by fitting a single straight line to the experimentally observed cascading as illustrated in Fig. 2. From the slope of this line, one can assign an effective lifetime τ_c to the cascading processes. Because the cascading generally will be a superposition of several cascading transitions from higher terms, each having a different lifetime, the fit of a single straight line to the complex cascading decay will be an approximation only. In this case τ_c will assume a value in the range of the mean lifetimes of the upper terms from which the cascading transitions originate (Ref. 3). The radiative decay is then obtained by subtracting the cascading component from the measured decay of the multiplet. The radiative decay of the upper term of the C III 538 Å multiplet is shown by the solid triangles plotted in Fig. 2. The indicated mean lifetime for the $2s3s\,^3S$ term of C III was obtained from the logarithmic slope of the straight line fitted to these corrected data.

The method of least squares was used to fit the two straight lines to the cascading decay and the term decay. The data for cascading were not weighted to obtain the line parameters. However, the data corrected for cascading (the term decay) were weighted according to their standard deviation to obtain the best line fit. An error analysis was also carried out to estimate the error introduced into the lifetime measurement by the cascade correction. This was done by obtaining the parameters of the two error lines that would double the rms error associated with the best line fit to the cascade decay. The slopes of the two error lines bracket the slope of the best line fitted to the cascade decay. The error limits for τ_c in Fig. 2, for example, were obtained from the slopes of the two error lines determined in this manner. The experimental data were then corrected by using these error lines for cascading to obtain a corresponding pair of error lines for the decay of the upper term of the multiplet. For the multiplets of C II and C III, the magnitude of the error in the mean lifetime introduced by the error in the cascade decay was always considerably less than the error in τ_c. This behavior is illustrated in Fig. 2, where it can be seen that an error of ± 10% in τ_c introduces an error of less than ±3% in the mean lifetime of the $2s3s\,^3S$ term. The error indicated in Fig. 2 is only that part of the

TABLE I. Results for C II and C III .

Ion (1)	λ (Å) (2)	Multiplet[a] (3)	τ_p This experiment (10^{-9} sec) (4)	A_p This experiment (10^9 sec^{-1}) (5)	A_{pk} This experiment (10^9 sec^{-1}) (6)	A_{pk} Calculated[f] (10^9 sec^{-1}) (7)
C II	1036.8	$2s^22p\,^2P^0 - 2s2p^2\,^2S$	0.45 ± 0.03	2.2	2.2 [b]	2.34
	904.1	$2s^22p\,^2P^0 - 2s2p^2\,^2P$	0.32 ± 0.03	3.1	3.1 [b]	4.2
	687.3	$2s^22p\,^2P^0 - 2s^23d\,^2D$	0.24 ± 0.03	4.2	4.1 [c]	2.7
	595.4	$2s^22p\,^2P^0 - 2s^24d\,^2D$	0.60 ± 0.04	1.7	1.6 [c]	
	1010.2	$2s2p^2\,^4P - 2p^3\,^4S^0$	0.30 ± 0.01	3.3	3.3 [b]	3.6
C III	977.0	$2s^2\,^1S - 2s2p\,^1P^0$	0.66 ± 0.03	1.5	1.5 [b]	1.9, 1.4
	1175.7	$2s2p\,^3P^0 - 2p^2\,^3P$	0.80 ± 0.04	1.25	1.25 [b]	1.3, 1.2
	538.2	$2s2p\,^3P^0 - 2s3s\,^3S$	0.32 ± 0.02	3.1	3.1 [b]	2.1, 2.8
	459.6	$2s2p\,^3P^0 - 2s3d\,^3D$	0.12 ± 0.02	8	8 [d]	7.9, 9.5
	371.7	$2s2p\,^3P^0 - 2s4d\,^3D$	0.21 ± 0.02	4.8	4.2 ± 0.5 [e]	3.4
	574.3	$2s2p\,^1P^0 - 2s3d\,^1D$	0.19 ± 0.01	5.3	5.3 [d]	6.3, 6.1

[a] The C II multiplet at 1010.2 Å is the only cascade-free multiplet.

[b] Branching fraction = 1.0.

[c] Branching fraction = 0.97.

[d] Branching fraction > 0.99.

[e] Branching fraction = 0.87.

[f] The calculations for C II are from Weiss (Ref. 8). The first entry in the column for C III is from calculations by Weiss listed in National Bureau of Standards tables (Ref. 6). The second entry for C III is from Pfennig, Steele, and Trefftz (Ref. 9).

total error in the mean lifetime resulting from the cascade correction.

The chief source of error in the measurements of mean lifetimes by the beam-foil method is introduced by cascading. Generally the magnitude of this error ranges from about 5 to 15% depending upon the intensity of the multiplet and the magnitude of the cascading processes. An uncertainty in the beam velocity downstream from the foil also contributes a small error to the measurement of the mean lifetime. Apart from a 1% uncertainty in the velocity of the ion beam incident on the foil, a small uncertainty in the downstream velocity arises from the determination of the energy loss $\Delta E/E$ of the ion beam in passage through the foil. For a 1-MeV C^+ beam, the energy loss is small, being about 5% for a 10 $\mu g/cm^2$ carbon foil and about 2% for a 5 $\mu g/cm^2$ foil.[5] The corresponding velocity loss $\Delta v/v$ is therefore about 2.5 and 1% for the 10 and 5 $\mu g/cm^2$ foils, respectively. Because the correction for the velocity of the beam is small, errors in the determination of $\Delta v/v$ will have only a small effect on the accuracy of the determination of the beam velocity. The total uncertainty in the beam velocity for the present measurements is estimated to be less than 2%.

The measurements of the radiative lifetimes are summarized in Table I. The experimental value of the mean lifetime τ_p of the upper term p of the measured multiplet is given in column 4 of the table. The error indicated for τ_p is the estimated total error of the measurement. This error includes the error introduced by cascading, the rms error of the least-squares fit of a straight line to the corrected term decay, and the error in the determination of the beam velocity.

TRANSITION PROBABILITIES

The determination of the mean lifetime of the excited term p also determines its total transition probability A_p through the reciprocal relationship $1/\tau_p = A_p$. The values of A_p in column 5 of Table I are therefore experimental values for the upper terms of the multiplets listed in column 3. The quantity A_p is defined as $A_p = \sum_i A_{pi}$, where the A_{pi} are the probabilities of all transitions from term p to the lower terms i. The measured multiplet corresponding to the transition from term p to the specific lower term k is one of these transitions. For convenience, its transition probability A_{pk} can be separated from the above sum and A_p expressed as

$$A_p = A_{pk} + \sum_{s \neq k} A_{ps} , \qquad (1)$$

where the sum $\sum A_{ps}$ includes the probabilities of all transitions from term p other than the measured multiplet. The transitions from the excited level p of a hypothetical ion are illustrated in Fig. 3.

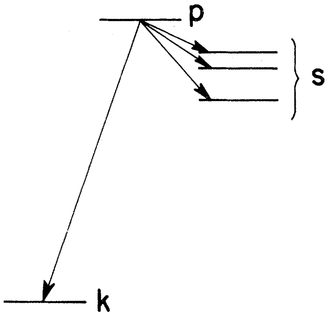

FIG. 3. Simplified term diagram that represents the general decay scheme for the uv multiplets of C II and C III. The transition probability of the measured multiplet ($p \rightarrow k$) is A_{pk}, while the transition probabilities of other multiplets from term p are the A_{ps}.

Usually only a very few transitions are possible from the lower levels of atoms and ions. When one transition alone, the measured multiplet, is possible from the excited term p, the lifetime measurement explicitly determines the transition probability of the multiplet since in this case $A_p = A_{pk}$. Several multiplets in Table I fall in this category. For these multiplets, the transition probability A_{pk} in column 6 is identical to the measured quantity A_p. The uncertainty in A_{pk} is also identical to the experimental error in the determination of A_p (or τ_p). These multiplets can also be identified in the table by a branching fraction of unity, where the branching fraction of the multiplet ($p \rightarrow k$) is defined as the ratio A_{pk}/A_p.

The remaining multiplets in Table I are not the only transitions from their respective upper terms. Therefore, according to Eq. (1), the measured quantity A_p must be corrected by subtracting the sum $\sum A_{ps}$ from A_p to determine the transition probability A_{pk} of the listed multiplet. However, with the exception of the C III multiplet at 372 Å, each of these multiplets is the dominating transition from its upper term. Thus the term $\sum A_{ps}$ in Eq. (1) is very small in comparison to A_{pk} (or A_p), and the corrected value of A_{pk} in column 6 is almost identical to the measured quantity A_p in column 5. The ratios A_{pk}/A_p for these multiplets are also included in Table I. The degree to which A_{pk} is dominant in the transition probability of the upper term is also evident by the closeness of this ratio to unity. The sum $\sum A_{ps}$ for each of the upper terms was determined by using theoretical values for the A_{ps}. The theoretical values were obtained from tables by Wiese *et al.*[6] or were calculated here by using the method of Bates and Damgaard.[7] Because the magnitude of the correction, $\sum A_{ps}$, is so small in comparison to A_p, relatively large errors in the calculated value of the sum will have little effect on the accuracy of the corrected value of A_{pk}. As an example, for the two multiplets of C II that have branching fractions of 0.97, an uncertainty of 50% in the calculated value of $\sum A_{ps}$ will introduce an uncertainty of only 1.5% in the corrected value of A_{pk}. The relative errors assigned to the corrected values of A_{pk} in column 6 are therefore essentially identical to the experimental errors given for τ_p in column 4.

The C III multiplet at 372 Å is the only multiplet in the table that has a branching fraction significantly less than unity (0.87). In addition to this multiplet having the transition probability A_{pk}, two other multiplets originate from the excited term $2s4d\,{}^3D$. The transition probabilities of these latter two multiplets were calculated to establish the correction term $\sum A_{ps}$ in Eq. (1). The transition probability of one of these multiplets, the $2s4p\,{}^3P^0$-$2s4d\,{}^3D$ transition in the infrared region, is 0.16×10^8 sec^{-1} and is therefore negligible in comparison to A_p (48×10^8 sec^{-1} from Table I). The transition probability for the other multiplet, the $2s3p\,{}^3P^0$-$2s4d\,{}^3D$ transition at 1620 Å, is 6.1×10^8 sec^{-1} which represents an appreciable contribution to the measured value of A_p. These transition probabilities calculated by the method of Bates and Damgaard should be reasonably accurate (± 25%) because the transitions are between excited terms and do not involve equivalent electrons. The value of A_{pk} for the C III multiplet at 372 Å was obtained by correcting the measured value A_p for the calculated transition probability of the 1620 Å multiplet. A 25% uncertainty in the calculated transition probability would introduce an additional 4% uncertainty in the corrected value of A_{pk}. This error is included in the total error assigned to the transition probability for the C III 372 Å multiplet in Table I.

DISCUSSION

Theoretical values of A_{pk} are included in the last column of Table I. The majority of the multiplets in Table I correspond to transitions involving shell-equivalent electrons. As pointed out by Wiese *et al.*[6] and Weiss,[8] calculations of the transition probabilities (or oscillator strengths) for transitions of this type are very sensitive to interelectronic interactions. The theoretical values of A_{pk} for C II in Table I were obtained from Weiss,[8] who used self-consistent-field calculations with a superposition of configurations to obtain the oscillator strengths of these correlation-sensitive transitions. The mean of his dipole-length and dipole-velocity values of oscillator strengths were used to obtain the values of A_{pk} in Table I. The accuracy of the calculated values, according to Weiss, should be approximately ± 25%. As can be seen from the table, the experimental and theoretical values for A_{pk} generally agree within their respective limits of error.

The theoretical values of the transition probabilities for C III in Table I are from two sources. Those of Weiss, the first entry in column 7 of Table I, are listed in the National Bureau of Standards tables by Wiese *et al.*[6] These values are from self-consistent-field calculations which did not include the effects of configuration interactions. The values are an average of the dipole-length and dipole-velocity forms of the oscillator strengths. According to Wiese *et al.*, the uncertainty in these calculations is within 50%. It can be seen from Table I that the agreement between Weiss's theoretical values of A_{pk} for the C III multiplets and the experimental values measured here is generally within 50%. The other source of theoretical transition probabilities for C III in the table is from the self-consistent-field calculations by Pfennig *et al.*[9] Again the effects of configuration interactions were not included in these calculations. The values in Table I are their "recommended" values which are close to the dipole-velocity value of the oscillator strength. The agreement between these theoretical values for C III and their experimental values is within 15%.

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