

Radiative Lifetimes from Ultraviolet Lines of Ne I, Ne II, and Ne III †

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The electron-excitation, phase-shift apparatus of Lawrence has been applied to the determination of radiative lifetimes of Ne I, Ne II, and Ne III states via transitions in the ultraviolet. With the exception of the Ne II 462 Å resonance transition, the measured transitions lie in the wavelength range 2000 to 3800 Å. Stages of ionization were separated by varying the energy of the electron beam between 60 and 215 V. Eight Ne I radiative lifetimes in the series $np \rightarrow 3s$, where $n=4-7$, have been measured using ten lines in the 2775 to 3634 Å region; the lifetimes range from 51 to 335 nsec. In Ne II, ten lifetimes between 2.1 and 10.1 nsec have been measured for quartet and doublet states radiating in the 2095 to 3777 Å region, as well as for the $2p^6 2S^0$ state (462 Å transition). Finally, eight radiative lifetimes of Ne III states ranging from 1.2 to 6 nsec have been studied via transitions in the 2065 to 2680 Å interval. Assignment of three lines, previously unclassified with respect to ionization species, has been unambiguously made using the electron-beam source.

I. INTRODUCTION

This paper initiates a series of reports in which lifetimes of noble-gas states which radiate in the ultraviolet and blue regions of the spectrum are examined experimentally. Results for a number of Ne I, Ne II, and Ne III lifetimes are reported here and work completed on Ar I and Ar II is in preparation.

A great deal of research on absolute intensities of the red lines of Ne I has been carried out since the pioneering investigations of Ladenburg and his colleagues.¹ Except for the more recent work of Koopman² on relative line strengths of Ne II and of Klose³ on the radiative lifetime of one line of Ne I, very little effort has been applied to the ultraviolet and blue transitions of Ne I or Ne II; two indirect experimental investigations^{4,5} on oscillator strengths for far ultraviolet Ne II and Ne III lines have been carried out, but no experimental data on the longer-wavelength transitions among excited states have been reported. The current investigation is motivated by the numerous facets of modern spectroscopic research in which these lines may play an important role. For instance, lines of Ne II observed in hot, early-type stars, such as 10 Lacertae,⁶ are used for stellar-abundance determinations; inert-gas lines and their radiative properties are of considerable interest in laser research,^{7,8} in the investigation of laboratory plasmas,^{4,5} and in the determination of radiative-recombination-rate coefficients.⁹ Furthermore, as can readily be seen from *Atomic Transition Probabilities*,¹⁰ practically no experimental work has been done on these excited states. And finally, it is hoped that these experimental data will provide absolute calibration points for experimental and theoretical determinations of line strengths.

In this paper the phase-shift technique of Lawrence¹¹⁻¹³ is briefly reviewed in Sec. II, and in Sec. III the detailed results of the Ne investigations are presented.

II. EXPERIMENTAL METHOD

An exponentially decaying excited state has a decay constant or radiative lifetime, τ , which may

be determined by measuring the phase shift between a sinusoidally modulated excitation process and the subsequent radiation from the excited state:

$$\tau = \omega^{-1} \tan \varphi, \quad (1)$$

where φ is the phase shift and $\omega = 2\pi f$ is the circular modulation frequency. The phase-shift experiment of Lawrence¹¹ used for the measurements reported herein has been thoroughly described elsewhere.¹¹⁻¹⁵ The ultraviolet emission modulated at one of ten frequencies between 0.54 and 54 Mc/sec is produced by exciting a low-pressure gas with a current-modulated beam of low-energy electrons ($E \leq 215$ V). Subsequently, the emission spectra are analyzed in a vacuum monochromator with a band pass of ≤ 8 Å, and the phase of the modulated emission relative to that of the excitation is determined as described by Lawrence.¹¹

Lawrence^{11,12} has also shown how data which include additive phase shifts due to population of the emitting state by radiative cascading may be analyzed in favorable cases, namely, when (1) extensive phase-vs-frequency information is available, and 2) only one cascade is present from a state whose lifetime T is distinctly different from the lifetime τ being measured. Where necessary, T and the cascading strength parameter β were obtained in this work by the method described by Lawrence and Savage.¹² Typical phase-vs-frequency diagrams for a cascade-free and cascade-complicated transition are presented in Sec. III.D below.

Detection of the emitted photon signal was accomplished with an EMI 6256A phototube. Two cascade-free atomic emission multiplets were used as zero-phase references for the present measurements, the B II (1625 Å) and Ne II (1908-1935 Å) transitions.^{12,13} As expected, no variations in final results were found using either of these two standards. Typically 10 to 12 phase-difference measurements were made at each modulation frequency, and the radiative lifetimes for cascade-free transitions were determined from phase measurements at the three points nearest the "linear" portion of the $\varphi(f)$ curve. Experimental deviations from these typical procedures will be discussed where applicable below.

the emission spectra. On the basis of the spectra obtained in this work it is clear that the 2065 Å line of Fig. 2, whose stage of ionization was previously unidentified,¹⁸ arises in Ne III and not in Ne II or Ne IV; similarly, it has been possible to confirm that the 2365 Å line is due to Ne III, as previously found by Humphreys,²⁰ and not to Ne II or IV.¹⁸ Assignments to stages of ionization for the purpose of ascertaining spectral purity for the ensuing phase measurements, as well as for the removal of ambiguities from the older work on Ne,^{17,18} could be made in more quantitative fashion (than inspection of spectra made at widely separated energies) by obtaining plots of intensity in a spectral line as a function of the accelerating voltage. Typical results are shown in Fig. 4, where data obtained for two Ne I lines (2933 and 3057 Å), two Ne II lines (3230 and 3568 Å) and the 2678 Å Ne III line are presented. The behavior of these $I(V)$ curves clearly demonstrates the correctness of the present assignments to Ne III of the weak emissions excited by low-energy electron collision in the wavelength region covered by Fig. 2; this is the first unambiguous observation in our laboratory of the excitation of a doubly-ionized species under such conditions. It is not surprising, however, that in the noble-gas atoms simple electron excitation is so effective, while in excitation of other atomic emission spectra from molecules (as has been done extensively in this laboratory¹¹⁻¹⁵) only the first stage of ionization of the constituent atoms is seen, since with the inert gases one begins with atoms *per se*, with numerous equivalent electrons, and none of the available excitation energy must be utilized to destroy strong molecular bonds or to selectively excite a particular optical electron.

It is clear from inspection of Figs. 1 and 2 that, with an instrument (of only moderately greater resolution than the present one) which has been calibrated for relative intensity measurements, the simple electron source used here will be able to provide a number of useful branching ratios for comparison with theory and for direct application to the determination of individual line transition probabilities. Since this source has good stability characteristics, long life, and yields good intensi-

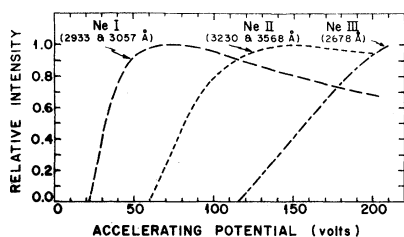


FIG. 4. Intensity (arbitrary units) versus accelerating potential for two Ne I and two Ne II lines and a Ne III line; each curve has been normalized to its maximum intensity. These curves have not been corrected for small changes in percentage modulation or for possible errors of a few percent in absolute energy scale, but they show that the first three stages of ionization are easily distinguished.

ties for photoelectric measurements, it seems that the branching-ratio problem should be a straightforward one once the calibration of the monochromator-detector is performed.

B. Ne I Radiative Lifetimes

Eight Ne I excited-state radiative lifetimes in the series $np - 3s$ ($n = 4-7$, see Fig. 3) have been measured using ten lines in the 2775 to 3634 Å region, after inspection of the emission spectra and studies of $I(V)$ curves (see Sec. A, above) revealed which lines were of sufficient purity for phase measurements. Measurements were made either (a) with 60-V electrons and the B II (1625 Å) multiplet as a phase reference for those Ne I lines which would have suffered from spectral overlap with Ne II lines if higher accelerating potentials were used; or with (b) the Ne II (1908-1935 Å) multiplet and accelerating potentials of 90-115 V for those lines with no Ne II overlap (e.g., for the 2933, 2992, and 3126 Å transitions). The same phase shifts were obtained on a given line regardless of the phase standard or potential employed.

The results of these measurements are presented in Table I, where it is seen that, with only two exceptions, the measured mean lives are quite long and tend, as expected, towards larger values as n increases. Several of the lifetimes encountered exceed the optimum modulation-frequency range of this experiment in that the amplitude of the modulated emission, even at 0.54 Mc/sec, for these lines is reduced compared to that expected at modulation frequencies lower than that corresponding to $\omega_c \tau = 1$ in Eq. 1.²¹ When the $\varphi(f)$ behavior at frequencies for phase shifts less than 45° cannot be examined, it is more difficult to ascertain by varying the modulation frequency whether or not the measured phase shifts show effects from longer-lived radiative cascading. Despite the fact that the excitation energy used in these measurements is not a threshold value, it is felt that the lines are cascade-free or very nearly so since (1) each of the lines could be observed with sufficient modulated intensity for phase measurements at three frequencies or more and each $\varphi(f)$ point yielded the same lifetime within the range of experimental error; and (2) the likely cascade transitions are infrared lines from states with relatively small total transition probabilities (approximately equal to, if not less than, those observed here) and the probabilities for specific cascades will be even smaller.

The measured mean lives found for Ne I are between 51.4 and 336 nsec and were found to be independent of Ne pressure in the excitation region over an observable range of about 0.4 to 12.0 μ Hg. Since none of the excited states measured connect with the ground state, no entrapment of radiation is expected and the pressures used for the phase measurements were sufficiently low (typically, 2 to 3 μ Hg) that no quenching effects were expected or observed. The error estimates assigned in Table I include estimates for possible systematic errors as well as for the observed standard deviations of the phase-difference measurements at a specific frequency; accounting for any undetected

TABLE I. Ne I Radiative Lifetimes.

Wavelength (Å)	Transition ^a		Radiative lifetime (nsec)	Comments ^b	
	Upper	Lower		T	β
2775	7p' [½]	-3s' [½] ₀	335 ± 50	Cascade	Free
2873	6p' [½]	-3s' [½] ₀	336 ± 50	Cascade	Free
2933	6p [½]	-3s' [½] ₀	270 ± 30	Cascade	Free
2992	5p [½]	-3s [1½] ₀	164 ± 25	Cascade	Free
3057	5p' [½]	-3s [½] ₀	157 ± 23	Cascade	Free
3126	(see 2992)				
3418	4p' [1½]	-3s [1½] ₀	150 ± 22	Cascade	Free
3520	4p' [½]	-3s [½] ₀	51.4 ± 5 ^c	Cascade	Free
3593	(see 3418)				
3634	4p [½]	-3s [½] ₀	80.4 ± 8	Cascade	Free

^aThe identifications were made using the wavelength lists of Ref. 16, while the spectroscopic designations are those of Ref. 19.

^bSee Sec. III.B of the text for a discussion of possible effects of radiative cascading on the lifetime values of this table.

^cAnother experimental value is Klose's lifetime of 63.5 ± 3.8 nsec (see Ref. 3).

phase shifts due to radiative cascading would decrease the lifetime values of Table I, but the experimental data support the conclusion that changes of lifetime in excess of the error estimates are unlikely. There is only one other lifetime measurement available for comparison with any of the results of this work, namely Klose's³ 63.5 ± 3.8 nsec lifetime for the 4p' [½] level, for which a value of 51.4 ± 5 nsec is reported here. In this single overlapping case the two experiments are seen to be in substantial agreement, although it is somewhat disappointing that the agreement is not even better, since both sets of measurements were made with equipment designed to perform well under the conditions imposed by this specific case. However, for applications in various laboratory,

astrophysical, and theoretical problems, differences of this order in the transition probabilities are usually not the limiting factor in the accuracy of the final result.

C. Ne II Radiative Lifetimes

From transitions lying in the 2095 to 3777 Å region it has been possible to determine radiative lifetimes for 10 excited states of Ne II; in addition, an attempt has been made to measure the 2p⁶ 2S-state lifetime. The results of these measurements, which range from about zero to 10.1 nsec, are given in Table II. Observations of the longer-wavelength transitions were made using the Ne II (1908-

TABLE II. Ne II Radiative Lifetimes.

Wavelength (Å)	Transition ^a		Radiative lifetime (nsec)	Comments		Other values ^b
	Upper	Lower		T	β	
462	2p ⁶ 2S	-2p ⁵ 2P ⁰	0.0 ± 0.2 ₀	Cascade	Free	0.2 ₇ ^c , 0.1 ₄ ^d
2096	unclassified		8.2 ± 0.8	Cascade	Free	
2792	4s	4P-3p 4P ⁰	2.1 ± 1.0	30	1.83	
3230	3p' 2D ⁰	-3s' 2D	5.5 ± 0.6	Cascade	Free	5.3
3345	3p' 2P ⁰	-3s' 2D	3.8 ± 0.4	Cascade	Free	5.9
3393	3p 2P ⁰	-3s 2P	5.0 ± 0.5	Cascade	Free	5.0
3482	3p 2S ⁰	-3s 2P	4.8 ± 0.5	Cascade	Free	6.7
3568	3p' 2F ⁰	-3s' 2D	8.8 ± 0.9	Cascade	Free	7.7
3664	3p 4P ⁰ _½	-3s 4P ⁰ _½	10.1 ± 1.0	45	0.29	7.7
3694	3p 4P ⁰ _½	-3s 4P ⁰ _½	10.0 ± 1.0	45	0.34	7.7
3727	3p 2D ⁰	-3s 2P	8.4 ± 0.8	50	0.19	7.7
3777	(see 3664)					

^aIdentified using the wavelengths of Ref. 17.

^bExcept where otherwise noted, the values are based upon the Coulomb approximation calculations of Ref. 10, where the uncertainties are estimated to be less than 50%.

^cRef. 4.

^dRef. 5.

1935 Å) multiplet as a phase reference. The bulk of the measurements were made with about 3μ Hg of Ne gas in the excitation region and with 120-V electrons, after it was found that the phase shifts were insensitive both to pressure variations over a range of about 0.4 to 12.0μ Hg and to variations of electron energies in the range 90 to 200 V. Because of the relative strengths of the Ne II and Ne III lines and their nearly complete natural exclusion from the same wavelength region, particular care was not required to keep the accelerating voltage lower than about 120 V to prevent overlap of stages of ionization. The principal exception to the previous statement is the 2096 Å transition; this line initially appears in spectra made with accelerating potentials >60 V and must therefore be assigned to Ne II, whereas in de Bruin's¹⁸ work on Ne III the stage of ionization of a number of lines in this region was indeterminate.

Most of the measured phase-vs-frequency curves for the transitions among excited states appeared to be cascade-free. For those $\varphi(f)$ curves which definitely deviated from exponential behavior, possible cascade transitions could be identified in the original spectra, even though they were usually quite weak. For the lines which appeared to be cascade-free, no plausible cascade transitions could be unambiguously resolved spectrally, and the conclusion that the lines are cascade-free results from the $\varphi(f)$ behavior, although in certain cases the conclusions are enhanced by the definite lack of an expected cascade. It should also be recalled that entrapment of radiation is unimportant in the electron-beam source for ionized species and that at the pressures used no quenching effects are expected or observed.

Except in the case of the 462 Å transition (see below), there are no other experimental lifetime or transition-probability determinations available for comparison to the results presented in Table II. Several of the multiplets may, however, be compared to Coulomb-approximation²² transition probabilities, as has been done in the fifth column of Table II; the multiplet spontaneous transition rates used are those tabulated by Wiese, Smith, and Glennon¹⁰ and are expected to have uncertainties $\leq 50\%$. In all cases the measured lifetimes are within 50% of the calculated values, indicating the expected result that for many applications in which high accuracy is not required the Coulomb approximation applied to Ne II is adequate.

The 462-Å resonance transition of Ne II was measured by Lawrence and Hesser²³ and that measurement is described here for the first time. Phase measurements were made with a Bendix magnetic photomultiplier in the fourth order of the grating using modulation frequencies of 30 and 54 Mc/sec and the N II (1086 Å) line as a phase reference.^{11,12} Results from phase-difference measurements at the two frequencies were the same and indicated a lifetime of about zero nsec, which, if estimates of possible errors are included as suggested by Lawrence and Savage,¹² implies that $\tau(2p^6 \ ^2S)$ is certainly ≤ 0.2 nsec. Consequently, the oscillator strength of the transition is ≥ 0.05 , which may be compared to the values of 0.035 and 0.07 deduced by Hinnov⁴ and Mickey,⁵ respectively, from studies of excita-

tion-rate coefficients in an ohmically-heated discharge, and to the value of 0.33 calculated by Varsavsky.²⁴ Although it is encouraging that Mickey's⁵ revision of Hinnov's⁴ work yields a value near the limit placed by this experiment, it is clearly desirable to re-investigate this transition with an experiment with higher frequency or time response than that used in this work. If indeed the f -value is as small as indicated by Mickey's work, it would be interesting to know why an allowed resonance transition should be described by an oscillator strength that is very much less than unity. Even with our present limited experimental knowledge of this transition, it seems that the intensity of the 462-Å multiplet combined with its near-zero mean life and its freedom from strong cascading effects²⁵ would make it an excellent zero-phase reference for phase measurements of longer-lived species radiating in this region.

Before leaving the topic of Ne II, detailed comments should be made on a few of the results presented in Table II. The large value of β necessary to account for the observed phase-vs-frequency curve of the $4s \ ^4P - 3p \ ^4P^0$ transition at 2792 Å considerably lessens the confidence that may be placed in the lifetime, since in at least one case²⁶ errors of about 30% have been found in measurements made under conditions where a substantial fraction of the upper state population is due to radiative cascading. At least two factors may contribute to the unusual phase behavior observed. The weak 2792 Å transition suffers slightly from spectral overlap with a much longer-lived and much weaker Ne I transition which could slightly distort the lowest frequency points; in addition, the data indicate that a second, much longer-lived, cascading transition than the single 30-nsec cascade already assumed in Table II also contributes to the measured phase shifts. No further attempt has been made to unfold these secondary terms in the $\varphi(f)$ diagram, and consequently larger error estimates have been assigned to the lifetime value; it should be remembered that removal of such effects as discussed above will tend to reduce the lifetime value of Table II. It is also interesting to note that the lifetimes found in this work for the $3p \ ^4P_{1/2}^0$ and $3p \ ^4P_{3/2}^0$ levels are the same, as measured using the 3664 and 3694 Å transitions.

D. Ne III Radiative Lifetimes

As shown in Sec. III.A, above, and illustrated in Figs. 2 and 4, electron excitation of low-pressure Ne gas with electrons of energy ≥ 120 V produces weak Ne III emissions in the near ultraviolet between 2000 and 3000 Å. Since these transitions are quite weak, it was necessary to use slightly higher pressures (about 5μ Hg) in the excitation region than were used for the phase measurements previously described for Ne I and Ne II. However, studies of phase differences between the Ne II reference line and the strongest Ne III transition, $3p \ ^3P - 3s \ ^3S^0$ at 2678 Å, showed no pressure dependence over a range of about 2 to 12μ Hg, indicating that quenching was not taking place. On the basis of the results of Fig. 4, electron energies of 215 V were used to obtain the maximum intensities for the phase measurements.

In Figs. 5 and 6, the phase-vs-frequency curves for two transitions in Ne III are plotted, as examples of the type of data acquired in this investigation. Figure 5 is an example of a cascade-free transition, the $3p' \ ^3F-3s' \ ^3D^0$ multiplet at 2613 Å, while Fig. 6 illustrates the method used to correct for cascading from a single, higher state which populates the measured radiating state, in this case for the $3p \ ^5P-3s \ ^5S^0$ multiplet at 2593 Å. Eight excited-state lifetimes were determined in this manner for Ne III and are given in Table III. Two of the transitions are unclassified in de Bruin's work¹⁸ (see also the discussion in Sec. III.A, above) and a third, the 2473 Å multiplet, was given the classification, $a \ ^3D^0-3p' \ ^3P$. However, the $a \ ^3D^0$ level has not been included in the *Atomic Energy Levels*,¹² and its classification status is in doubt at the present time.^{8b} As found in Ne II, cascading effects were, in general, absent or quite small for the Ne III transitions studied (the corrections made to the τ values in Table III were on the order of 5% or less). In those multiplets which were found experimentally to be cascade-free, no expected cascade transitions were observed in the original spectra; even for the lines with cascading it was not possible to isolate unambiguously any specific cascades in the spectra, which was probably due to the weakness of the possible cascade transitions. It should be noted, however, that there are in principle many possible cascade transitions which, because of the rather incomplete state of the present knowledge of the Ne III spectrum,¹⁹ have not yet been observed.

As was generally the case in Ne I and Ne II, there are no other experimental data with which to compare the present results. Lifetime estimates based on the Coulomb approximation^{10,22} are listed in Table III for three transitions and are found to

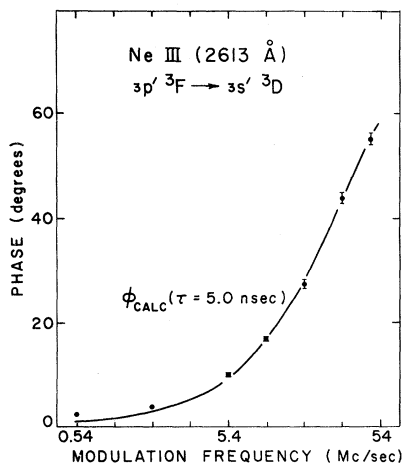


FIG. 5. An example of a cascade-free phase-shift-versus-frequency curve for a Ne III transition. The solid line was calculated from Eq. (1) for a $3p' \ ^3F$ state lifetime of 5.0 nsec. The error bars on the individual phase measurements indicate probable errors, while those points without flags are based upon five or fewer phase measurements.

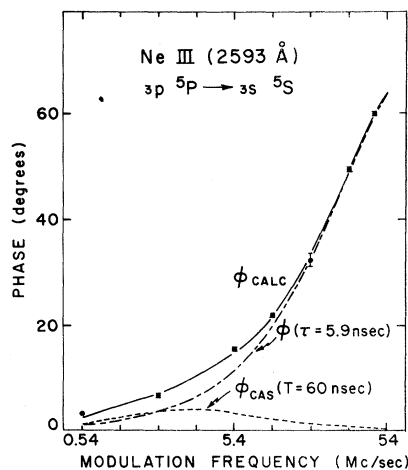


FIG. 6. An example of a transition in Ne III which shows a decay complicated by a small amount of radiative cascading. The $\phi(f)$ curve for an exponential decay of 5.9 nsec for the $3p \ ^5P$ state is shown by the short-long dashed curve, while the additional phase shift due to cascading ϕ_{CAS} from a state with a nominal lifetime of approximately 60 nsec is shown by the dashed curve. The sum of the first two curves yields the solid curve which fits the observed data points well.

be within 35% or better agreement with the experimental values.

IV. SUMMARY

In this work 27 excited-state radiative lifetimes in Ne I, Ne II, and Ne III have been measured for the first time. With the exception of the $2p^6 \ ^2S^0$ state in Ne II, which radiates in the far ultraviolet, the transitions measured are among excited states which do not connect directly with the ground states of the atoms. An interesting result of this work is that contributions to the measured phase shifts from radiative cascading in general do not appear to be large, and consequently, it seems that the applicability of nonselective excitation mechanisms to the study of transitions among excited states is in general not seriously limited by cascading. It should be noted, however, that some authors^{7,27} have called particular attention to the errors that may arise from radiative cascading when one is forced by intensity considerations to work near the peak of the excitation cross section (as is the case of this experiment¹¹) rather than at threshold; but several recent experiments²⁸ have indicated that in most cases encountered cascading is not a strong function of the energy of the exciting electrons. Nevertheless, it would be desirable to repeat at least a portion of the present experiments by another method using more selective excitation techniques.

In addition to the number of new transition probabilities reported here, more evidence has been accumulated concerning the utility of the simple electron-beam source as a spectroscopic light source,¹⁴ and in particular, it has been possible to assign three transitions, observed in earlier work¹⁸

TABLE III. Ne III Radiative Lifetimes.

Wavelength (Å)	Transition ^a		Radiative lifetimes (nsec)	Comments		Other values ^b
	Upper	Lower		T	β	
2065	unclassified		1.6 ± 0.2	Cascade	Free	
2181	3p' ³ P-3s' ³ D ⁰		1.2 ± 0.2	Cascade	Free	
2216	3d' ³ G ⁰ -3p' ³ F		2.1 ± 0.2	90	0.24	
2365	unclassified		2.3 ± 0.2	100	0.10	
2473	a ³ D ⁰ -3p' ³ P		1.9 ± 0.2	Cascade	Free	
2593	3p ⁵ P-3s ⁵ S ⁰		5.9 ± 0.6	60	0.14	4.0
2613	3p ³ F-3s ³ D ⁰		5.0 ± 0.5	Cascade	Free	4.2
2678	3p ³ P-3s ³ S ⁰		3.7 ± 0.4	Cascade	Free	4.2

^aThe notation of Ref. 18 is used to designate the transition.

^bBased upon the Coulomb-approximation calculations of Ref. 10, where the uncertainties are estimated to be less than 50%.

on Ne, to appropriate stages of ionization by use of this source.

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¹For a comprehensive bibliography on past work see: B. M. Glennon and W. L. Wiese, Bibliography on Atomic Transition Probabilities, Natl. Bur. Std. Misc. Publ. No. 278 (1966).

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²⁰C. J. Humphreys, private communication to Dr. C. M. Sitterly; I am especially indebted to Mrs. Sitterly for providing me with this information.

²¹As pointed out in Ref. 11, the amplitude of modulated emission varies as $(1 + \omega^2 \tau^2)^{-1/2}$ and begins to fall off as $1/\omega$ beyond the corner frequency, $\omega_c \equiv 1/\tau$, corresponding to a 45° phase shift for an exponential decay.

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²⁸In addition to the present results, J. Z. Klose (private communication) has not observed the strong effects reported in Ref. 27 as being due to cascading, although his experiment is in principle quite similar to that used in Ref. 27 and has been used on many of the same lines. Similar conclusions have been suggested by recent work of G. M. Lawrence (private communication) on Ar I lifetimes, where he has also found that the strength of cascading is not a strong function of electron energy.