Mean Lives of Some Excited States in Multiply Ionized Oxygen and Neon*

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The beam-foil technique has been used to measure the mean lives of some excited levels in multiply oxygen and neon. The values for the mean lives are

Level	Spectrum	Mean Life (sec)
$2p^3\ ^3D^0$ ($2p^4\ ^4P$)	O III (O II)	$(17.1\pm0.4)\times10^{-10}$
$2p^2\ ^2D$	O IV	$(17.1\pm0.4)\times10^{-10}$
$2p^4\ ^4P$	Ne IV	$(6.6\pm0.2)\times10^{-10}$
$2p^3\ ^3D^0$	Ne V	$(8.5\pm0.1)\times10^{-10}$

Cascades that affected the decay of the $2p^4\ ^4P$ level in the Ne IV have been taken into account in the determination of the mean life of that level. The decays of other states were not affected by cascades. Possible effects from unresolved spectral lines from two different levels were considered and found to be negligible for the cases treated herein.

I. INTRODUCTION

IT has been shown that the beam-foil technique is an excellent method for the accurate determination of the mean lives of excited states of atoms and ions.[1–4] The technique employs a thin foil to excite a beam of ions that have been accelerated to velocities up to a few percent of the speed of light. Mean lives are determined by a time-of-flight measurement in which the radiation emitted by excited atoms and ions in the beam is directly related to the population of excited states that have yet to decay. In addition to lending itself to accurate measurements of mean lives, the beam-foil excitation can produce excited atoms in very high stages of ionization. This paper describes two experiments in which the beam-foil technique was used to determine some mean lives of excited levels in multiply ionized oxygen and neon.

II. EXPERIMENTAL CONSIDERATIONS

The ionic beam consists of atoms and ions that have been put into various stages of excitation upon passing through a thin self-supporting carbon foil. Light from the beam was observed at right angles with a one-meter model 225 McPherson vacuum uv scanning wavelength monochrometer equipped with a 600/mm grating. The detector was a sodium-salicylate coated EMI 9541S photomultiplier, radiation-cooled with liquid nitrogen. The current output from the photomultiplier was fed into an Elcor model A309B current integrator, where it was either integrated for certain time intervals or amplified and displayed on a strip chart recorder.

The signal $S_T(x)$ from the photomultipler can be considered to be the sum of three signals. One is the signal S_{dc} due to the dark current of the photomultiplier which can arise from within the photomultiplier itself or be caused by external radiation from the Van de Graaff, cosmic rays, etc. The second is the signal S_{bg} due to background from the beam. This is attributed to light from decaying excited states formed by collisions between beam particles and residual gas particles in the target chamber. The third signal $S(x)$ is attributed to light emitted by decaying excited states in the beam excited by the foil. Here x is the distance downstream the foil to the observation point.

The dark current S_{dc} was $\sim1.2\times10^{-9}$ A. Because this signal is a function of background radiation in the laboratory, it was measured with the entrance slit to the spectrometer closed, the Van de Graaff in operation, and with the beam of desired current and energy directed through the foil in the target chamber. The combined signal (S_{dc} and S_{bg}) was determined by deflecting the beam into the chamber, but with the exciter foil removed and entrance slit open. This signal was also $\sim1.2\times10^{-9}$ A, indicating that collisions with residual gas atoms were negligible. One might argue that with a foil in the beam, and with neutrals in the beam which originate at the foil, the collisional cross section might be different, thereby causing a different signal S_{bg} than that obtained with the foil removed. That this is not the case has been checked experimentally by deflecting both molecular and ionic beams at different energies into the chamber. The result is that the signal S_{bg} is negligible when the pressure in the target chamber is below 10^{-5} Torr.

For all scans of the spectrum of neon and oxygen, the spectrometer entrance slit width was set equal to its exit slit width. Several scans were obtained for various slit widths ranging between 2000 and 100 μ. With wide slits, the signal $S(x)$ is large and the gross features of the spectrum can be observed. As the slits become narrower, the signal decreases but the finer details of the spectrum

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[1] W. S. Bickel and A. S. Goodman, Phys. Rev. **148**, 1 (1966).
[2] W. S. Bickel and S. Bashkin, Phys. Letters **20**, 488 (1966).
[3] L. Heroux, Phys. Rev. **153**, 156 (1967).
[4] A. S. Goodman and D. J. Donahue, Phys. Rev. **141**, 1 (1966).

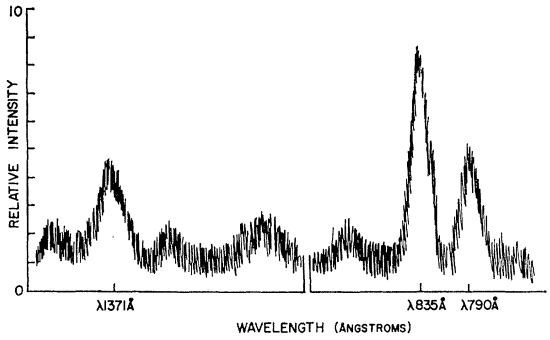

FIG. 1. A tracing from a strip chart showing intensity as a function of wavelength for two different wavelength regions between λ750 and λ1220 Å. The incident beam consisted of O_2^+ ions (mass 32) at an energy of 1.5 MeV.

become apparent as individual spectral lines become resolved. While the light emitted by the beam is proportional to the beam current, the length of time a foil lasts before being destroyed by the beam decreases with increasing beam current. A compromise is made which consists of selecting a beam current that gives a reasonable intensity and at the same time allows the foil to last long enough to make a complete wavelength scan. In general, the strongest signals obtained with the narrowest slit (100 μ) were about two orders of magnitude above the noise yielding photomultiplier currents of $\sim 10^{-7}$ A. All wavelength scans are plots of $S_T(x)$ as a function of wavelength.

The mean lives are determined from the change in $S(x)$ as a function of distance downstream from the foil. For mean-life measurements, a 0.5-cm section of the beam was observed by the spectrometer with the grating set to observe a spectral line which arises from the decay of the excited state of interest. Photomultiplier counts $S_T(x)$ were recorded for various positions x of the foil relative to the entrance aperture of the spectrometer and normalized to equal numbers of beam counts. This was done to insure that changes in the signal $S_T(x)$ were due only to changes in $S(x)$ as the foil was moved upstream, and therefore were independent of beam fluctuations. The beam current was determined by collecting beam particles in a shielded Faraday cup equipped with an electron suppressor. The current was fed into an Elcor model A309B current integrator and integrated until a final predetermined amount of charge was collected. A set of typical experimental values taken at the beginning of an oxgyen mean-life measurement in this experiment is beam current through foil ~ 0.7 μA; photomultiplier current ~ 0.005 μA; integrating time ~ 430 sec; charge collected by Faraday cup, 3.0×10^{-4} C; charge collected from photomultiplier, 2.2×10^{-6} C. The incident-particle energy has been determined in the usual way by calibrating the bending magnet used to deflect the ions into the target chamber.[5] This was done by employing the $F^{19}(p,\alpha\gamma)O^{16}$ reaction and detecting the γ resonance at 872.11 ± 0.20 keV. By plotting magnetic field as a function of energy, the incident-particle energy was determined to within 1% .This uncertainty results in a $\frac{1}{2}$% uncertainty in particle velocity and mean life.

[5] J. B. Marion, Rev. Mod. Phys. 38, 660 (1966).

III. OXYGEN WAVELENGTH STUDIES IN THE VACUUM ULTRAVIOLET

In this experiment O_2^+ ions (mass 32) were accelerated to a kinetic energy of 1.38 MeV with a Van de Graaff. The molecular ions were dissociated into atomic ions upon passing through a thin (7 ± 4 μg/cm^2) carbon foil. The emergent particles, which had lost 36 ± 20 keV in the foil[6] were in various stages of ionization and excitation. In the spectral region between λ500 and λ3000 Å, four relatively strong signals at λ702, λ790, λ834, and λ1371 Å, were observed. A typical scan over this region is displayed in Fig. 1. These signals were attributed to decays of excited states in *O* II through *O* V. Spectral lines which can give rise to these signals are listed in Table I.[6,7] With an inverse linear dispersion of 16.6 Å/mm and 0.2-mm entrance and exit slits, the narrowest possible consistent with a tolerable noise level, the individual spectral lines in each multiplet could not be resolved. The signal-to-noise ratio for some of the weaker lines is about 1, while for the stronger lines at λ834 and λ790 Å, the signal-to-noise ratio was as large as 15. (See Fig. 2.) One weak signal was attributed to the resonance line of *O* I at λ1355 Å and λ1358 Å. Special attention was given to the signals in the spectral region between λ790 and λ850 Å, including the resonance lines at λ834 Å from *O* III, λ790 Å from *O* IV, and a very weak signal at λ796 Å. The signal at λ796 Å which was scarcely discernable above the noise is attributed to λ796.66 Å in *O* II. The signal at λ790 Å is clearly due to the $2p\ ^2P^0$ multiplet in *O* IV, while λ834 Å is ascribed to a blend of the multiplets $2p^2\ ^3P - 2p^3\ ^3D^0$ in *O* III and $2p^3\ ^4S^0 - 2p^4\ ^4P$ in *O* II. All other signals observed in the spectral region between λ500 and λ3000 Å were half or less than those at λ834 and λ790 Å, with the

TABLE I. Atomic transitions in *O* II through *O* V that can yield experimentally measured wavelengths.[a]

Observed wavelength	Charge state	Transition	Multiplet wavelength range (Å) min	max	Number of lines in multiplet
λ834 A	*O* II	$2p^3\ ^4S^0 - 2p^4\ ^4P$	832.754	834.462	3
λ834 Å	*O* III	$2p^2\ ^3P - 2p^3\ ^3D^0$	832.927	935.202	4
λ702 Å	*O* III	$2p^2\ ^3P - 2p^3\ ^3P^0$	702.822	703.850	4
λ790 Å	*O* IV	$2p^2\ ^2P^0 - 2p^2\ ^2D$	787.710	790.203	3
λ1371 Å[b]	*O* V	$2p\ ^1P^0 - 2p^2\ ^1D$	1371.287		1

[a] References 6 and 7.
[b] A study of the signal at λ1371 Å is reported in Ref. 2 along with details regarding the measurement of the $2p^2\ ^1D$ level lifetime in *O* V.

[6] R. L. Kelly, University of California Radiation Laboratory Report No. UCRL 5612 (unpublished).
[7] C. E. Moore, Natl. Bur. Std. (U. S.) Circ. 488, (1950), Sec. 1.

exception of λ1371 Å from $2p\ ^1P^0$–$2p^2\ ^1D$ in *O* v. This transition has been observed before and its mean life reported in Ref. 2.

Mean-Life Measurements in Oxygen

The mean lives of the $2p^2\ ^2D$ and $2p^3\ ^3D^0$ levels were measured using techniques that have received detailed description elsewhere.[1–3]

The $2p^2\ ^2D$ Level in O IV

To measure the $2p^2\ ^2D$ level, a 0.5-cm section of the beam was observed by the spectrometer with its grating set to pass λ790 Å. Figure 3 shows the relative intensity of λ790 Å light as a function of the position downstream from the exciter foil. The eight points, which represent the decay of the $2p^2\ ^2D$ level over two mean lives, have been corrected for background following the procedure discussed earlier. The mean life of the $2p^2\ ^2D$ level, determined from a least-squares fit to the data, is 17.1 $\pm 0.4\times 10^{-10}$ sec. There is no evidence of cascades or unresolved spectral lines from other excited states.

The $2p^3\ ^3D^0$ Level in O III

The measurement of the mean life of the $2p^3\ ^3D^0$ level in *O* III was made in the same way as described above for the $2p^2\ ^2D$ level in *O* IV. In this case the signal at λ835 Å was measured as the foil was moved upstream. The results are also shown in Fig. 3, where the mean life of $(17.1\pm 0.4)\times 10^{-10}$ sec represents a least-squares fit to the data. That the data fall on a straight line, even though they most likely arise from signals from two different multiplets, might be attributed to one or both of two causes: 1. The observed decay is from a single level—either the $2p^3\ ^3D^0$ level in *O* III or the $2p^4\ ^4P$ level in *O* II. 2. The mean lives of the levels in *O* III and *O* II are the same to within 3%.

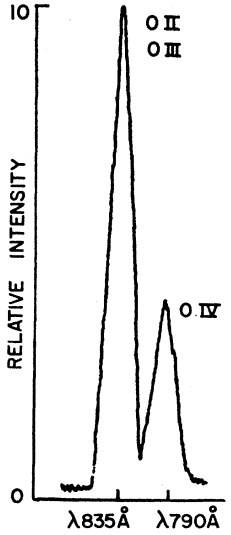

FIG. 2. A strip chart tracing showing the relative intensity of the two signals at λ835 and λ790 Å, which were involved in the determination of mean lieves of excited states in *O* III and *O* IV.

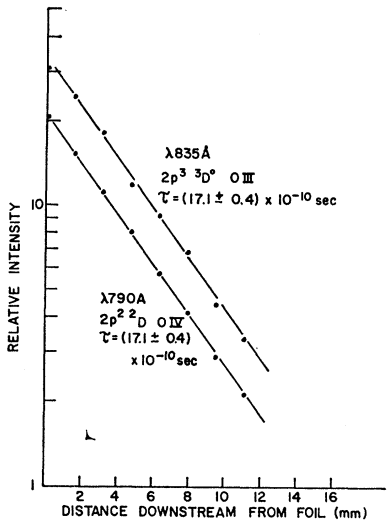

FIG. 3. Relative intensity as a function of foil positions for the signals at λ835 and λ790 Å from oxygen. These data have been corrected for background noise.

Since the resonance line from *O* I was seen, it is likely that both resonance lines from *O* II and *O* III were present. The most one can safely infer from these data is that either the $2p^4\ ^4P$ level of *O* II or the $2p^3\ ^3D^0$ level of *O* III or *both* have a mean life of $(17.1\pm 0.4)\times 10^{-10}$ sec. The possibility that the *O* II $2p^4\ ^4P$ level has the same lifetime as the $2p^3\ ^3D^0$ of *O* III has been indicated in the tables by including the data for this state in parentheses.

Both oxygen mean lives discussed above have also been measured with incident O^+ particles at 1.0 MeV and O_2^+ particles at 0.7 MeV. The results are the same. However, at the lower energy the relative intensities of the O III and O IV multiplets were different. At 1.38 MeV the ratio of intensities (O III:O IV) was (10:4), while at the lower energy the ratio was about (10:2). Since both lifetimes are the same, the change in intensity represents the actual change in relative population of the two levels. In other experiments the energy dependence of multiplet intensities has been used to assist in the identification and assignation of certain multiplets to particular charge states.

IV. NEON WAVELENGTH MEASUREMENTS IN THE VACUUM ULTRAVIOLET

For wavelength and lifetime studies with neon, mass-20 Ne^+ ions were accelerated to an energy of 1.34 MeV. The subsequent energy loss in the foil was (49±28) keV.[8]

A spectral analysis was carried out for neon in the same way as for oxygen. The beam current was about 0.4 μA through the foil. Three strong signals at λ490, λ542, and λ572 Å were seen in the wavelength region between λ400 and λ600 Å and attributed to the reso-

[8] L. C. Northcliffe, Ann. Rev. Nucl. Sci. 13, 67 (1963).

TABLE II. Atomic transitions in Ne III through Ne V that can yield experimentally observed wavelengths.[a]

Observed wavelength	Charge state	Transition	Multiplet wavelength range (Å) min	max	Number of lines in multiplet
λ490 Å	Ne III	$2p^4\ ^3P - 2p^5\ ^3P^0$	488.103	490.310	6
λ542 Å	Ne IV	$2p^3\ ^4S^0 - 2p^4\ ^4P$	541.124	543.884	3
λ572 Å	Ne V	$2p^2\ ^3P - 2p^3\ ^3D^0$	568.418	572.336	5

[a] References 6 and 7.

nance lines of Ne III, Ne IV, and Ne V, respectively. The wavelengths and transitions involved that can give rise to these signals are given in Table II. Here, as in the experiment with oxygen, the entrance and exit slit were 0.2 mm and the individual lines in each multiplet were not resolved. All other signals in this wavelength region were weaker by at least a factor of 2.

A detailed study of the spectral region embracing the signals at λ572 and λ542 Å was carried out with various slit widths to ascertain the origin of these signals. By virtue of their proximity to the resonance lines of Ne IV and Ne V and the absence of other strong lines nearby, these signals were associated with the radiation which results from the decays of the $2p^4\ ^4P$ and $2p^3\ ^3D^0$ levels in Ne IV and Ne V, respectively.[6,7] The signal at λ489 Å is most likely from the resonance multiplet from the decay of the $2p^5\ ^3P^0$ level in Ne III, but it might contain some light from the $2p^2\ ^3P - 2p^3\ ^3P^0$ transition in Ne V. Because this signal was weak and because of the uncertainty in its origin, no attempt was made to measure its decay characteristics.

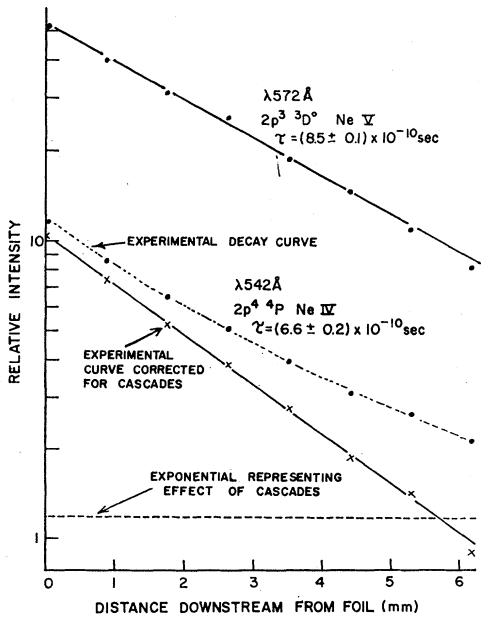

FIG. 4. Relative intensity as a function of foil position for the signals at λ572 and λ542 Å from neon. The circles represent data corrected for background noise. The crosses for λ542 Å represent the data that have been corrected for cascades by subtracting the dotted line from the experimental decay curve.

Mean-Life Measurements in Neon

Mean-life measurements of neon levels were carried out in the same way as those for oxygen.

The $2p^3\ ^3D^0$ Level in Ne V

The mean life of this level was determined by measuring the decrease of intensity of the signal seen at λ572 Å as the foil was moved upstream. The result of eight data points is given in Fig. 4. The mean life of $(8.5\pm0.1)\times10^{-10}$ sec was determined from a least-squares fit to the data. The decay curve shows no evidence of cascades or unresolved spectral lines arising from excited states with lifetimes different than the one observed. It can be shown that if the decay curve appears exponential for at least two mean lives, the effect of cascades can be considered negligible.

The $2p^4\ ^4P$ Level in Ne IV

The data acquired by measuring the decrease of radiation at λ542 Å, which are related to the change in population of the $2p^4\ ^4P$ level as a function of distance downstream from the foil, are also shown in Fig. 4. The deviation from a straight line is attributed to cascades. The curve was fitted with two exponentials, one representing the decay of the $2p^4\ ^4P$ level, the other representing the contribution due to cascades. The best fit to the curve resulted when the slope of one exponential represented a mean life of $(6.6\pm0.2)\times10^{-10}$ sec, with the other exponential representing a mean life of infinity. The two exponentials which have been extracted from the experimental curve are shown in Fig. 4. The data were not good enough for the computer to distinguish mean lives having values between infinity and about 3×10^{-8} sec.[1] The mean-square deviation in this case was as small as those obtained from exponential fits to decay curves when cascades were not apparent.

V. CONCLUSION AND DISCUSSION

The uncertainties in the lifetime measurements arise mainly from the uncertainty in the thickness of the exciter foil. The values for the incident-particle energy loss in the foil are from the experimental data of Northcliffe[8] and are uncertain by about 20%. Therefore, there is a subsequent uncertainty in the energy loss and velocity of the foil-excited ions. The thickness of the thinnest self-supporting carbon foils we have made, measured in a Rutherford scattering experiment, is less than 10 $\mu g/cm^2$. It is unlikely that the foils used in these experiments are much different, since they were prepared in the same way. Although foil thickness can vary

TABLE III. Experimental results of mean-life measurements in oxygen and neon.

Level	Spectrum	λ(Å)	τ (sec 10^{-10}) This experiment	τ (10^{-10} sec) Other methods
$2p^3\ ^3D^0(2p^4\ ^4P)$	O III (O II)	834.5(833.0)	17.1±0.4	11.9±6[a](7.15±4)[b]
$2p^2\ ^2D$	O IV	789.4	17.1±0.4	10.5±5[c]
$2p^4\ ^4P$	Ne IV	542.8	6.6±0.2	4.0±2[d,e]
$2p^3\ ^3D^0$	Ne V	571.0	8.5±0.1	7.15±3[a]

[a] Calculation of Bolotin *et al.* (Ref. 11).
[b] Calculation of Kelly (Ref. 12).
[c] Calculation of Bolotin *et al.* (Ref. 13).
[d] Calculation by Weiss (Ref. 14).
[e] Level affected by cascades.

from foil to foil, the thinnest ones have always been selected and used for mean-life measurements. It is emphasized that in these experiments all data for a single lifetime have been taken with one foil.

The largest contribution to the total uncertainty of a lifetime measurement can be caused by cascades. In general, it is difficult to identify the state or states responsible for repopulating the excited level under observation, as was the case with the decay of the $2p^4\ ^4P$ level in Ne IV. However, if the data are good, deviations of the observed decay curve from a straight line will be observed. The data can be treated analytically to extract a mean life. References 1 and 3 discuss in detail the effects of cascades in the experimental determinations of mean lives of hydrogen and some levels in N II through N V.

Problems similar to those created by cascades can be caused by unresolved multiplets. If, as in the cases for all mean lives discussed in this paper, the multiplets arise from excited states having the same quantum number n and l, then all spectral lines of the multiplet will decay at the same rate. This follows from the result that the lifetime of an excited state characterized by the quantum numbers $|nlj\rangle$ is independent of the spin of the excited state.[9] An experimental test of this theoretical prediction is discussed in Ref. 10. If the lines of the multiplet consist of spectral lines from decays of excited states with different $|nlj\rangle$, three alternatives arise: 1. Decrease the slit width until the individual lines are resolved. This has obvious experimental difficulties for weak signals. 2. Measure the simultaneous decay of all lines of the multiplet and extract the different lifetimes by a computer analysis. In this case one must also associate the proper lifetime with the proper charge state. One is assisted in this by changing the incident-particle energy and noting the changes in relative contribution of the unresolved lines to the observed decay curve. Or the multiplets can be identified in a charge-splitting experiment if they arise from different charge states.[11] 3. Conclude that a meaningful mean life cannot be determined from the data.

That a decay curve may appear exponential but yet yield a mean life that is far different from the actual mean life of the excited level under study is a danger that must be continually guarded against.

The experimentally determined mean lives are given in Table III along with the mean lives determined by other methods. For the mean lives measured here, the only values available for comparison are ones which have been theoretically determined.

The mean lives of the $2p^3\ ^3D^0$ level of O III and Ne V have been determined by Bolotin *et al.*,[12] who have used a configuration-interaction calculation as an approximation. Because of the sensitivity of these transitions on configuration interaction, large uncertainties are expected. Their theoretical value has been assigned a 50% uncertainty but lies within the experimentally determined value.

For the $2p^4\ ^4P$ level of O II, Kelly[13] has used the self-consistent-field calculations in which exchange effects have been considered. That this theoretical value for the mean life, with its estimated uncertainty of 50%, lies outside the experimentally measured value is further evidence that the lifetime measured belongs to the $2p^3\ ^3D^0$ level in O III.

Bolotin and Yutsis[14] employed analytical one-electron wave functions and approximations involving configuration interaction to determine the mean life of the $2p^2\ ^2D$ level of O IV. Large uncertainties are also expected here since transitions which involve this level are sensitive to the effects of configuration interaction.

The theoretical mean life of the $2p^4\ ^4P$ level in Ne IV is from a self-consistent-field calculation by Weiss.[15] In these calculations the average of the dipole lengths and velocity approximations is adopted, but fairly large uncertainties are to be expected since they do not include the effects of configuration interaction.

ACKNOWLEDGMENT

It is my pleasure to thank Professor S. Bashkin for frequent discussions concerning these experiments.

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