PHYSICAL REVIEW A VOLUME 7, NUMBER 2 FEBRUARY 1973

Beam Fractions in the Lowest-Quartet Metastable Autoionizing State for O^{5+} and F^{6+} Beams after Passage through Foils*

Winthrop W. Smith
University of Connecticut, Storrs, Connecticut 06268
and
Bailey Donnally
Lake Forest College, Lake Forest, Illinois 60645
and
D. J. Pegg, Matt Brown,† and I. A. Sellin
University of Tennessee, Knoxville, Tennessee 37916
and Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830
(Received 6 April 1972)

When fast beams of oxygen and fluorine ions are incident on carbon and metal foils thick enough to produce charge-state equilibrium, a significant portion of the lithiumlike beam fraction emerges in excited metastable autoionizing states. We have studied the yield of these states with beams in the range 2–20 MeV incident on nominal 10-$\mu g/cm^2$ carbon foils and on various metallic foils. The decay in flight of the lowest of these states, the $(1s2s2p)\ ^4P^0_{5/2}$, is monitored with observation of the spectrum of ejected electrons. We find, for example, that the maximum yield of this single state per incident beam particle is approximately 0.8% and 0.6% for oxygen and fluorine, respectively, on carbon. The energy dependences and target-material dependences are measured and discussed. Comparison is made to related work of Dmitriev *et al.*

I. INTRODUCTION

Recent measurements of electron spectra[1] resulting from the production of autoionizing states of lithiumlike ions in fast beams that have passed through carbon foils reveal appreciable populations of various metastable states of high electronic spin and high excitation energy. The lowest of these states is $1s2s2p\ ^4P^0$, which is metastable against Coulomb autoionization but can decay via the spin-orbit and spin-spin interactions.[2] The spectral identification of the higher-quartet states in three-electron oxygen and fluorine and our lifetime measurements have been discussed elsewhere.[1,3] Decay times are such that several of the quartet states persist for distances of the order of centimeters (in a 1-MeV/amu beam with $v \sim 10^9$ cm/sec) after leaving the foil target. Measurements of the beam fraction in specified metastable states may be useful in the interpretation of data on beam charge-state distributions[4] and electron-shakeoff lifetimes following stripping in gas and solid targets.[5] The data we report here confirm the energy dependence of our preliminary data on carbon foils[6] and extend the measurements to several metallic foil targets. In Sec. III, we compare these measurements with the work of Dmitriev *et al.* at Moscow State University.[7] This group first observed the quartet metastable states in highly stripped ions.

II. EXPERIMENT

The apparatus used in this work is essentially identical to that shown in Fig. 1 of Ref. 3. Analyzed ^{16}O and ^{19}F ion beams in the range 2–20 MeV were obtained in various charge states from the Oak Ridge tandem accelerator as in our earlier work. Measurements of the intensity of energy-analyzed electrons emitted from the beam versus distance from the excitation point (foil target) permit the decay in flight of the metastable autoionizing states to be measured separately for each component of the electron spectrum. Figure 1 shows a typical spectrum obtained with an 11.2-MeV oxygen beam on a carbon foil target using a resolution of approximately 0.8% (full width at half-maximum). The electron energies are given in the rest frame of the emitting ions, whose laboratory velocities are comparable to the ionic-rest-frame velocities of the electrons. The $J=\frac{1}{2}$, $\frac{3}{2}$, and $\frac{5}{2}$ fine-structure levels of the $1s2s2p\ ^4P^0$ state are expected to have different decay times, but the $J=\frac{5}{2}$ component decay can be separated clearly from the other two states.[3] Quantitative yields were determined only for the $J=\frac{5}{2}$ component since these depend upon accurate lifetime data that were not available for the other J states.

A measurement of the yield of the $^4P^0_{5/2}$ state as a fraction of the number of beam particles incident

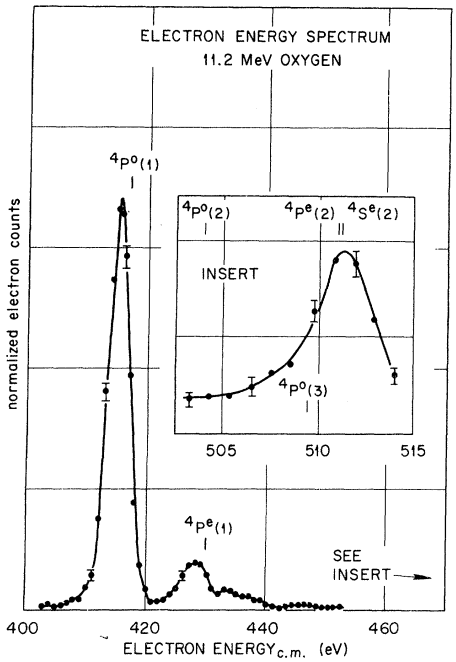

FIG. 1. Spectrum of autoionization electrons from the decay in flight of metastable 11.2-MeV oxygen ions. Examples of notation for decaying states: $^4P^0(1) = (1s2s2p)\ ^4P^0$, $^4P^e(1) = (1s2p^2)\ ^4P^e$, $^4P^e(2) = (1s2p3p)\ ^4P^e$, $^4S^e(1) = (1s2s3s)\ ^4S^e$, $^4S^e(2) = (1s2s4s)\ ^4S^e$, etc. Identifications are based on the variational calculations of Holøien and Geltman [Phys. Rev. 153, 81 (1967)]. Note that the $^4P^0(1)$ state is well resolved from the others.

on the foil depends on several factors in addition to the decay time (which enters because one must extrapolate intensities back to the foil position). The carbon foils used were 10 $\mu g/cm^2$ nominal, thick enough to produce charge-state equilibrium, as indicated by previous measurements[8] and by the fact that measured beam current did not change appreciably when a second foil was inserted between the original foil and the Faraday cup. In addition, the yield measurements reported here did not change measurably when the foil thickness was doubled. Interpretation of the charge current emerging from the foil in terms of particle current requires data on the mean beam charge after passage through the foil. In addition, one must consider the anisotropic angular distribution of the decay electrons in the laboratory frame. (We assume here that autoionization is isotropic in the ionic rest frame.) Other factors, determined from geometrical considerations and from measurements, are the length of beam viewed by the electron spectrometer (Δx), its fractional solid angle of acceptance ($\Delta\Omega/4\pi$), and the product of its transmission with the detection efficiency of the electron multiplier (η).[9] Thus the yield per particle incident on the target (Y) is given by

$$Y = C \Big/ \left[e^{-x/(v\tau)} \left(\frac{N}{\bar{q}}\right) \left(\frac{\Delta x}{v\tau}\right) \frac{d\Omega_C}{d\Omega} \left(\frac{\Delta\Omega}{4\pi}\right) \eta \right]. \tag{1}$$

Here C is the number of electrons counted, x the mean distance from foil to spectrometer, τ the mean life of the metastable state, $\bar{q}$ the mean charge of the beam emerging from the target, N the total charge collected in electron charge units, and $d\Omega_C/d\Omega$ the solid-angle transformation factor from the ionic rest frame (Ω_C) to the laboratory frame (Ω).

The factor $d\Omega_C/d\Omega$ is obtained easily by implicit differentiation of the nonrelativistic relations

$$v_L \cos\theta_L = v + v_C \cos\theta_C \tag{2a}$$

and

$$v_C^2 = v_L^2 + v^2 - 2 v_L v \cos\theta_L, \tag{2b}$$

where θ_L is the angle of emission in the laboratory frame, θ_C the corresponding angle in the ionic rest frame, v_L the laboratory-system electron velocity, v_C the emission velocity in the ionic rest frame, and v the velocity of the beam. It is not difficult to obtain a particularly simple form of the transformation that we have not found previously in the literature, namely,

$$\frac{d\Omega_C}{d\Omega} = \frac{d\cos\theta_C}{d\cos\theta_L} = \frac{(v_L/v_C)^2}{(v_L/v_C) - (v/v_C)\cos\theta_L}. \tag{3}$$

Note that Eq. (2b) gives the relation between v_L and v_C for a fixed laboratory angle as in this experiment. The condition that the denominator of Eq. (3) goes to zero corresponds to the existence of a maximum emission angle of the electrons in the laboratory frame, given by $\sin\theta_{L,\max} = v_C/v$ for $v \geq v_C$.

A number of excited metastable autoionizing states that appear in the spectrum of Fig. 1 can cascade into the lowest-quartet state by electric dipole radiation. All of the higher states that appear in the spectrum have measured lifetimes in the range 1–2 nsec or less. The lowest-quartet $J = \frac{5}{2}$ state lifetimes are much longer, approximately 15 and 25 nsec for fluorine and oxygen, respectively.[3] Yield data for the $J = \frac{5}{2}$ state were in every case taken far enough downstream to give the cascades from other states that we see in Fig. 1 negligible influence on the measurement. The $J = \frac{5}{2}$ decay time (τ) was then used to extrapolate the yields back to the foil position. Any possible quartet autoionizing states of the three-electron system with energies lying above the $1snl$ series limit of the two-electron ion of the same Z will have very short lifetimes since ordinary Coulomb autoionization is allowed for them. Therefore these states do not make any appreciable contribution to our downstream spectrum or to our yield results.

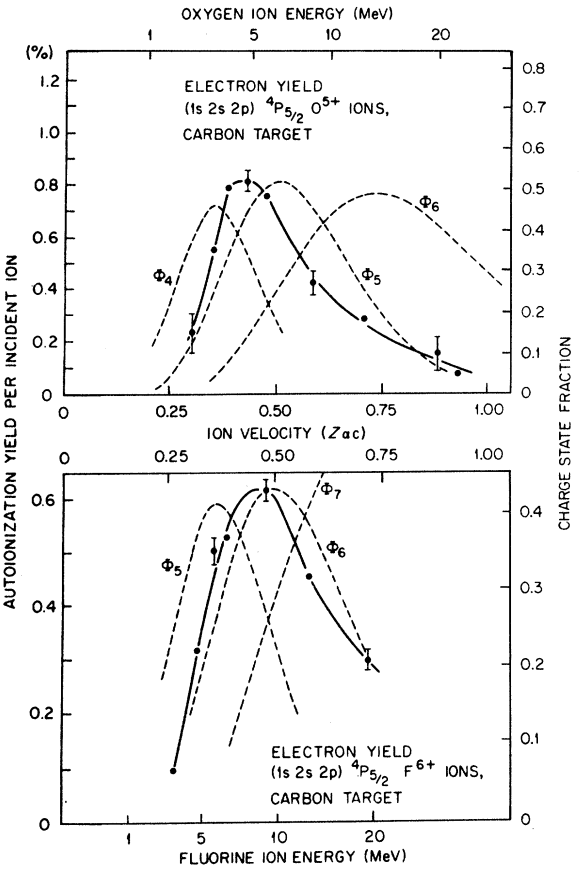

FIG. 2. Dependence of the electron yield per incident beam particle on beam velocity for oxygen and fluorine beams on carbon. Ion velocities are given in units of the K-electron Bohr velocity $Z\alpha c$, where α is the fine-structure constant and c is the velocity of light. Charge-state fractions ϕ_i, where i is the ionic charge, are also plotted (dashed curves).

There remains the possibility that very few of the ions that end up in the lowest-quartet state actually emerge from the foil in that state. The equilibrium excitation within the solid foil itself might relax very fast to the $1s2s2p\,^4P^0$ and other metastable states as the beam emerges from the foil. The states we measure are, however, quite prominent in the electron spectrum observed when looking at the foil itself.[10]

In making these measurements, the annular exit slit of the electron spectrometer was made considerably wider than the entrance slit. This resulted in spectra with approximately flat-topped peaks, eliminating the need to correct for spectral profile and simplifying the yield determination. This configuration was relatively insensitive to misalignments of the spectrometer axis with respect to the beam axis. Using the higher resolution of Fig. 1, a misalignment of $\sim\frac{1}{2}^\circ$ was easily recognized.

III. RESULTS

Our measurements for the yield of the $1s2s2p\,^4P^0_{5/2}$ state per incident beam ion on a carbon foil are given in Fig. 2 as a function of beam energy (MeV) and of ion velocity. We have also plotted, for comparison, the equilibrium charge-state fractions ϕ_i for two-, three-, and four-electron oxygen and fluorine ions, respectively, in a solid target[11]; here the subscript i refers to the actual ionic charge. Reference 4 indicates that, crudely speaking, the ϕ_i are independent of target material.[12] As the figure indicates, the maximum yield per incident ion on a $\simeq 10$-$\mu g/cm^2$ carbon foil is approximately $0.82 \pm 0.09\%$ for oxygen and $0.62 \pm 0.07\%$ for fluorine. The errors quoted are based simply on reproducibility from run to run with different foils, not limited by counting statistics. The relative yields for oxygen and fluorine and their energy dependences are believed correct to within the accuracy specified. The absolute numbers may not be this reliable because of small variations in slit-edge thickness, uncertainties in electron multiplier efficiency, etc. The combined effect on the absolute yields of all known systematic errors is conservatively believed to be less than a factor of 2.

We have also made some measurements of autoionization electron yield for oxygen and fluorine beams impinging on foils of several other materials, namely, gold, silver, aluminum, and nickel. There appears to be some systematic variation in yield with atomic number of the foil material over a range of approximately 2.5. The dependence of yield on beam energy (in the range 3–13 MeV) for these metallic foils is similar to that for carbon foils (except for a slight shift in the case of the single, rather thick, nickel foil used that can be accounted for by its greater energy loss before the beam emerges). The dependence of the *peak* yield on atomic number of the foil material is presented in Table I.[13]

Related metastable-state yield measurements by Dmitriev, Teplova, and Nicolaev at Moscow State University exist for lithiumlike ion beams up to $Z = 8$ incident on celluloid films in the MeV energy range.[7] Their measurements were made by a technique that did not permit spectroscopy of the states produced. In their work, metastables are detected by making use of the large difference in collisional ionization probability for ground-state and metastable ions. A metastable decay curve is obtained and extrapolated back to the point of excitation as in our measurements, but the autoionization electrons are not observed. The two sets of measurements can be compared only for $Z = 8$ (oxygen) and then at only two energies, 2.1 and 5.7 MeV (by interpolation of our results). Unfor-

tunately, the numbers given in Ref. 7 for oxygen do not appear to be very reliable because a substantially different decay time for the $^4P^0_{5/2}$ state is presented in their table for each beam energy. If we convert the results of Ref. 7 from $\alpha_{5/2}$ (metastable $J=\frac{5}{2}$ component of the three-electron beam fraction) to yield per incident beam particle by using the charge-state fractions from Fig. 2, we obtain the results shown in Table II. We conclude that there is quite satisfactory agreement within the combined experimental uncertainties.[14]

IV. DISCUSSION AND CONCLUSIONS

It can be seen from Fig. 2 that the energy dependence of the $^4P^0_{5/2}$ metastable state follows roughly the curve for the total equilibrium beam fraction of three-electron ions. A slight systematic shift of the peak in the curve toward lower ion velocities appears consistently in both the oxygen and fluorine cases. Too little is known about the statistical distribution of excited states of ions making multiple collisions in a solid to permit a detailed solution of the set of rate equations that determine these curves, even with the assumption of detailed balance at dynamic equilibrium. (Figure 5 of Ref. 7 suggests the complexity of the processes involved.) In spite of this complexity, some qualitative statements can be made, taking the oxygen case as an example: (i) If the $(1s2s2p)\,^4P^0_{5/2}$ state (5^+) is mainly populated from an excited configuration of O^{4+} by collisional or autoionization loss of outer-shell electrons, the 5^+ ($^4P^0_{5/2}$) threshold should lie above the excited 4^+ threshold and therefore probably above the peak for ground-state 4^+ formation. (ii) If the mechanism involves excitation from the ground state of O^{5+}, the $^4P^0_{5/2}$ population should follow the 5^+ ground-state population initially but then cut off at lower velocity because of the higher ionization (loss) cross section in the excited state. This mechanism is expected to have a relatively small cross section, especially since a spin flip is necessary to produce a quartet state. (iii) If the $^4P^0_{5/2}$ state is dominantly populated by electron capture from the $1s2s$ or $1s2p$ triplet heliumlike ion, the observed peak shift can be explained by assuming (a) a monotonic decrease in the capture cross section with ion velocity, (b) a velocity-independent geometrical loss cross section,[15] and (c) that the capture cross section in the excited state is less than the capture cross section in the ground state.[16] The ratio of (6^+) to (5^+) populations in the solid under dynamic equilibrium conditions is equal to the ratio of the effective one-electron loss cross section to the one-electron capture cross section. Assumptions (b) and (c) then imply that the equilibrium ratio between parent (6^+) and daughter (5^+) populations is *larger* in the excited state, because the capture/loss cross-section ratio is *smaller* for the excited state, at a given ion velocity. Assumption (a) then implies that the balancing between capture and loss rates that gives rise to a population peak should occur at a lower velocity for the excited-state 5^+ ions than for ground-state ions with the same charge. Mechanism (iii) is also discussed in Ref. 7 as the one most likely to dominate.

In summary, we have measured a peak absolute yield, per incident beam particle, ~1% for a single metastable state $(1s2s2p)\,^4P^0_{5/2}$, upon passage of ~1-MeV/amu oxygen and fluorine beams through several types of foils. Earlier work[3] has indicated that there is also appreciable yield of *higher*-metastable-quartet states, with a substantial fraction of these decaying by direct autoionization rather than by cascade. Thus it is highly probable that several percent of the emerging beam is in *some* metastable high-spin autoionizing state in the energy range under consideration. The energy dependences of the $(1s2s2p)\,^4P^0_{5/2}$ yields are similar to those of the total three-electron charge fractions but slightly shifted toward lower ion velocities. A variety of metallic foils give yields and energy dependences that vary only slightly from the results for carbon targets, with an apparent slight increase in the yield near $Z_{\text{target}}=13$.

TABLE I. Dependence of the apparent peak yield (Y) of autoionization electrons from the $(1s2s2p)\,^4P^0_{5/2}$ state of O^{5+} ions excited in various foils on the atomic number of the target.

Z_{target}	Sample thicknesses ($\mu g/cm^2$)	Y (peak, average) (%)
6 (carbon)	8, 9, 10, 15, 17, 18, 58	0.825 ± 0.054
13 (aluminum)	11, 12, 23	1.24 ± 0.11
28 (nickel)	369	1.025 ± 0.11
47 (silver)	73, 95	0.672 ± 0.032
79 (gold)	150, 166, 300	0.452 ± 0.065

TABLE II. Comparison of $(1s2s2p)\,^4P^0_{5/2}$ yield measurements for oxygen on carbon targets (interpolated values).

Energy (MeV)	Yield per incident oxygen ion in %	
	This work	Ref. 7
2.1	0.14 ± 0.07	0.048 ± 0.019
5.7	0.73 ± 0.04	0.57 ± 0.20

ACKNOWLEDGMENT

The authors wish to thank P. M. Griffin for assistance in the determination of the spectrometer slit geometry.

*Research supported in part by Union Carbide Corp. and by Oak Ridge Associated Universities under contract with the U. S. Atomic Energy Commission, and in part by the U. S. Office of Naval Research.

†Present address: Department of Physics, Kansas State University, Manhattan, Kan.

[1]I. A. Sellin, D. J. Pegg, M. Brown, W. W. Smith, and B. Donnally, Phys. Rev. Letters 27, 1108 (1971).

[2]This multiplet is of considerable importance in the ion He^-, where it is the ground state. The lifetime of the $J=\frac{5}{2}$ level has been measured recently [L. M. Blau, R. Novick, and D. Weinflash, Phys. Rev. Letters 24, 1268 (1970)].

[3]B. Donnally, W. W. Smith, D. J. Pegg, M. Brown, and I. A. Sellin, Phys. Rev. A 4, 122 (1971).

[4]L. C. Northcliffe, in *Annual Reviews of Nuclear Science*, edited by E. Segrè (Annual Reviews, Palo Alto, Calif., 1963), Vol. 13, p. 83.

[5]H. D. Betz, Phys. Rev. Letters 25, 211 (1970); 25, 903 (1970).

[6]W. W. Smith, Bailey Donnally, D. J. Pegg, M. Brown, and I. A. Sellin, in *Abstracts of the Seventh International Conference on the Physics of Electronic and Atomic Collisions* (North-Holland, Amsterdam, 1971), p. 513.

[7]I. S. Dmitriev, Y. A. Teplova, and V. S. Nikolaev, Zh. Eksperim. i Teor. Fiz. 61, 1359 (1971) [Sov. Phys. JETP 34, 723 (1972)].

[8]E. Hubbard and E. Lauer, Phys. Rev. 98, 1814 (1955); R. A. Brown and G. D. Simons, Nucl. Instr. Methods 58, 274 (1968). These references indicate that, in the energy range being considered, 5 $\mu g/cm^2$ of carbon is sufficient to establish charge-state equilibrium for incident oxygen ions.

[9]We have not measured this. The multiplier used was manufactured by Mullard. The very similar Bendix Channeltron cone-input unit has an efficiency of 98% for electrons of a few hundred electron volts according to the manufacturer.

[10]I. A. Sellin and D. Pegg (unpublished).

[11]J. B. Marion and F. C. Young, *Nuclear Reaction Analysis* (North-Holland, Amsterdam, 1968).

[12]The charge-state fractions ϕ_i do have a dependence on target material that may be significant in some applications. There is also considerable systematic variation in the values for the same projectile-target combinations among the results of different investigators. Results for oxygen ions on carbon, gold, nickel, and other materials are given by D. Bernard, B. Bonner, G. Phillips, and P. Stelson [Nucl. Phys. 73, 513 (1965)] and W. Booth and I. Grant [Nucl. Phys. 63, 481 (1965)]. The second paper discusses possible reasons for this dependence on target material.

[13]These data have not been corrected for possible small normalization errors due to straggling and also to the dependence of multiple beam scattering on foil thickness. It seems to us premature to speculate on the reason for this apparent dependence of Y on target material. We merely note that the yield appears to have a broad maximum near $Z_{target}=13$. The effect of foil-surface conditions in determining the yield is basically unknown. It is known, at least in the case of carbon foils, that they become thicker with time of exposure to the beam, possibly because of deposition of carbon from "cracked" pump oil in the vacuum system. This phenomenon has no appreciable effect on the yield.

[14]Using a value of 25 nsec for the $J=\frac{5}{2}$ state lifetime will raise the values of Ref. 7 relative to this work.

[15]For a background discussion of the expressions for the capture cross section obtained by N. Bohr and by H. Brinkman and H. Kramers, see the article by H. Bethe and J. Ashkin, in *Experimental Nuclear Physics*, edited by E. Segrè (Wiley, New York, 1953), Vol. 1, p. 191.

[16]N. Bohr and J. Lindhard, Dan. Mat. Fys. Medd. 28, No. 7 (1954). Experimental evidence for the decrease in the capture cross section with excitation is given (for Cl ions in low-density gases) by G. Ryding, H. D. Betz, and A. Wittkower [Phys. Rev. Letters 24, 123 (1970)]. It may be that this effect is somewhat suppressed in solids, but partial suppression will not invalidate the qualitative argument we are making.

PHYSICAL REVIEW A VOLUME 7, NUMBER 2 FEBRUARY 1973

Transition Probabilities and Multiple Ionizations of Ions by High-Energy-Electron Impact*

Yukap Hahn† and Kenneth M. Watson
Physics Department and Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720

(Received 2 August 1972)

The single-particle model for atoms and ions is used to calculate the transition probabilities to bound and continuum electronic states. The projection operators in the semiclassical approximation derived previously are applied to treat the large numbers of final states involved. Ionization cross sections of atoms and ions by high-energy-electron impact are then estimated. The ionization cross sections result both from direct transition to the continuum and from inelastic scattering followed by Auger emission.

I. INTRODUCTION

Electron impact provides a possible mechanism for production of highly ionized beams to be used for injection into heavy-ion accelerators. With most of the Periodic Table and as many as twenty to thirty steps of ionization considered of interest, it is evident that several thousand ionization cross