Investigation of the distribution of (n,l) states populated by beam-foil excitation of 32-MeV oxygen ions

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The decay curves for $2p$-$1s$ and $3p$-$1s$ transitions in 32-MeV H-like oxygen projectiles excited by a thin carbon foil have been measured and compared with the results of a cascade computer program which calculates the intensities of these transitions for an assumed initial distribution of excited states as a function of decay time. The measured decay curves agreed best with those predicted by the cascade program using an l-independent population probability proportional to n^{-4} for each state.

I. INTRODUCTION

It is well documented that high Rydberg states are readily populated as an energetic ion emerges from the back surface of a thin foil. Evidence for this has been provided by numerous observations of long-lived components in the $2p$-$1s$ x-ray decay of highly stripped ions following foil excitation.[1-6] The time dependence of the emission intensity is given by a power law of the form $I(t)=at^{-b}$, as predicted for the decay of a short-lived state populated by cascading from long-lived states of high principal quantum number.[7,8]

Betz *et al.*[9] have performed cascade calculations for several different (n,l) population distributions with the result that each distribution predicted a significantly different decay curve. This finding points to the possibility of deducing the general dependence of the Rydberg-state population distribution on the principal and angular-momentum quantum numbers by carefully comparing measured decay curves with calculated decay curves. The value of such an exercise would be strengthened by studying several transitions that are populated by different routes in the cascade process.

The present work was carried out for the purpose of testing the above proposition by examining the decay curves for both the $2p$-$1s$ and $3p$-$1s$ transition in H-like oxygen. In the course of this study, a new cascade computer program was developed and used to deduce the (n,l) dependence that gave the best representation of the experimental decay curves.

II. EXPERIMENT

A beam of 32-MeV O^{2+} ions from the Texas A&M cyclotron was directed onto a 20-μg/cm^2 carbon foil which could be accurately translated along the beam axis without interrupting the vacuum. The oxygen K x-rays were analyzed at 90° with respect to the beam axis by a 12.7-cm Johansson-type curved-crystal spectrometer employing a thallium-acid-phthalate (TAP) crystal. The viewing region of the spectrometer was restricted to a 2-mm length of the beam path by an adjustable slit assembly, and the spectrometer response function was measured to be nearly Gaussian with a 2-mm full width at half maximum.

Presented in Fig. 1 is an oxygen x-ray spectrum taken with the spectrometer directly viewing the foil. Spectral features of delayed emission of oxygen x rays were similar to those obtained for Ne and Mg projectiles.[6] The decay curves for the H-like oxygen $2p$-$1s$ and $3p$-$1s$ transitions were constructed by carrying out spectral scans over their wavelength regions at a number of foil positions.

III. RESULTS AND DISCUSSION

The decay curves for the $2p$-$1s$ and $3p$-$1s$ transitions are shown in Fig. 2. They are well represented by power laws with $b=1.42\pm0.22$ ($2p$-$1s$) and $b=1.93\pm0.16$ ($3p$-$1s$). The present value of the slope of the $2p$-$1s$ decay curve is in agreement with previous high-resolution[3]

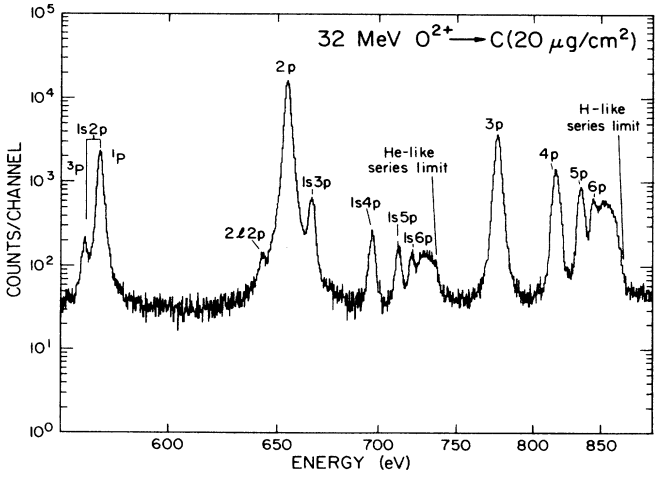

FIG. 1. K x-ray spectrum of 32-MeV oxygen ions excited by a thin carbon foil, taken with the spectrometer directly viewing the foil.

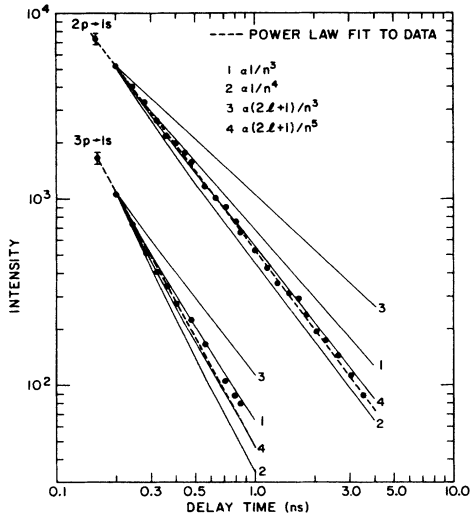

FIG. 2. Measured and calculated decay curves for $2p$-$1s$ and $3p$-$1s$ transitions in 32-MeV H-like oxygen ions. The calculated decay curves (solid lines) were normalized to the measured curves at $t=0.2$ ns.

($b=1.5$) and low-resolution[4] ($b=1.40\pm0.08$) measurements. The decay curve measured by Rothermel *et al.*[5] exhibits two different slopes: 1.1 ± 0.1 for $t\leq2.5$ ns and 1.7 ± 0.1 for $t>2.5$ ns. The only previous high-resolution measurement of the $3p$-$1s$ decay curve[3] gave $b=1.5$ while the present data indicate a steeper decay. The slopes of the present decay curves for one-electron oxygen are quite similar to those for H-like neon and magnesium measured recently by Palinkas *et al.*[6].

In order to fulfill the objective of comparing the experimental decay curves with the decay curves predicted by different assumptions regarding the initial (n,l) population distribution, a cascade computer program was written. Given an initial population distribution of excited states, the time-dependent population of a particular excited state can be calculated from the rate equation

$$dN(n,l,t)/dt=-N(n,l,t)\sum_{n',l'}A(n,l;n',l')+\sum_{n',l'}N(n',l',t)A(n',l';n,l)\ ,$$

where A is the transition rate between states labeled by the sets of quantum numbers (n,l) and (n',l'). The first and second terms on the right-hand side represent the rates of depletion and repopulation (through cascades) of state (n,l), respectively. Similar equations for each of the excited states must be written for a complete description of the cascade process. Although the set of coupled first-order differential equations obtained in this way can be solved numerically, it is more convenient to use a diagrammatic technique developed by Curtis.[10] Employing this method, the population of an excited state can be written as a sum of individual cascade contributions, grouped according to the number of steps in the cascade. As an example, consider the case of the $2p$ level which is fed by states with $n>2$ and which is depleted by transitions to the $1s$ ground state. According to the diagrammatic method, the time-dependent population of the $2p$ level is given by

$$N(2,1,t)=N(2,1,0)\exp(-\alpha_{21}t)+\sum_n N(n,0,0)A(n,0;2,1)\{[\exp(-\alpha_{n0}t)-\exp(-\alpha_{21}t)]/(\alpha_{21}-\alpha_{n0})\}$$
$$+\sum_n N(n,2,0)A(n,2;2,1)\{[\exp(-\alpha_{n2}t)-\exp(-\alpha_{21}t)]/(\alpha_{21}-\alpha_{n2})\}+\cdots\ ,$$

where the ellipsis represents higher-order terms and α_{nl} is the total transition rate of a state. In order to perform such calculations, one needs to know the initial population distribution of the excited states and the transition rate between each state.

The electric dipole transition rates used in the present work (approximately 11 500 in number) were calculated from well-known formulas[11,12] for the hydrogen atom and scaled to the H-like oxygen ion by multiplying by Z^4. In order to determine the number of excited states contributing to the repopulation of the $2p$ and $3p$ levels, cascade terms were added sequentially and in order of increasing complexity until no significant numerical changes were observed in the resulting decay curves over the range of delay times investigated experimentally (0.15 to 3.5 ns). It was found to be necessary to include states up to $n=40$ and $l=9$ in the calculations.

The cascade calculations were performed for a number of assumed initial population distribution functions. The results of some of these calculations are shown in Fig. 2 together with the experimental decay curves. The hypothetical distribution functions used in these calculations were of the form $N(n,l,t=0)=kf(n)g(l)$. According to theoretical predictions for electron capture by high-velocity projectiles within the framework of the Born approximation,[13] the dependence of the population distribution on principal quantum number should be given by $f(n)=n^{-3}$. Therefore, this dependence was examined in considerable detail. Various possibilities were considered for $g(l)$: (a) only s states are populated; (b) all l states for a given n are populated equally, $g(l)=$const; and (c) all l states are populated statistically, $g(l)=2l+1$. The results of the calculations showed that when only s states are populated, the calculated decay curves (not shown in Fig. 2), normalized to the experimental decay curves at $t=0.2$ ns, fall off much faster than the measured decay curves. The inadequacy of a pure s-state population has also been discussed by Dehmelt *et al.*[14] and by Hopkins *et al.*[15] On the other hand, calculations based on the assumptions of an equal and a statistical population of l states resulted in

slopes that were smaller than those displayed by the measured decay curves. Therefore, other hypothetical distribution functions were investigated using $f(n)=n^{-4}$ and $f(n)=n^{-5}$, together with the three assumptions considered for $g(l)$. The best agreement was obtained for the case of an equal population of l states with $f(n)=n^{-4}$, and for the case of a statistical population with $f(n)=n^{-5}$. It should be noted that both of these distribution functions [namely, $N(n,l,t=0)=k_1 n^{-4}$ and $N(n,l,t=0)=k_2(2l+1)n^{-5}$] predict that the *total* population associated with a given n (i.e., the population summed over all l states) is proportional to n^{-3}, since

$$N(n,t=0)=\sum_l N(n,l,t=0)=\begin{cases}k_1 n n^{-4}=k_1 n^{-3}\\ k_2 n^2 n^{-5}=k_2 n^{-3}\end{cases}.$$

It was found that good agreement could also be attained between measured and calculated decay curves using $f(n)=n^{-3}$ in conjunction with an l-state population that decreases as l increases. For example, the distribution function $N(n,l,t=0)=kn^{-3}l/(l+2)$ gave a reasonable representation of the experimental decay curves.

For the purpose of comparing the relative shapes of the experimental and theoretical decay curves for the two transitions investigated, the intensity ratios $R=I(2p\text{-}1s)/I(3p\text{-}1s)$ as a function of delay time are shown in Fig. 3. These ratios have been corrected for absorption in the proportional counter window and for crystal reflectivity. Since the experimental decay curve for the $3p$-$1s$ transition falls off faster than that for the $2p$-$1s$ transition, the value of R increases as delay time increases. The results of this comparison show that the predicted variation of the intensity ratio with time depends more strongly on $g(l)$ than on $f(n)$.

In all of the comparisons discussed above, including the ratio comparison shown in Fig. 3, the calculated curves were normalized to the experimental curves individually. In comparing the predictions of a particular distribution function for the two transitions of interest, however, the same normalization factor should apply to both decay curves. This requirement was fulfilled best for the assumption of an equal population of l states which gave calculated values of R about 1.2 times higher than the measured values. The assumption of a statistical population, on the other hand, gave calculated R values which were too large by a factor of 3.3. This reflects the fact that when high l states are populated, a much larger fraction of the cascade proceeds through yrast transitions, thereby populating the $2p$ level much more strongly than the $3p$ level. In a similar comparison for foil-excited sulfur ions, Betz[16] found that the experimental R values were intermediate between those predicted for low l-state and high l-state populations.

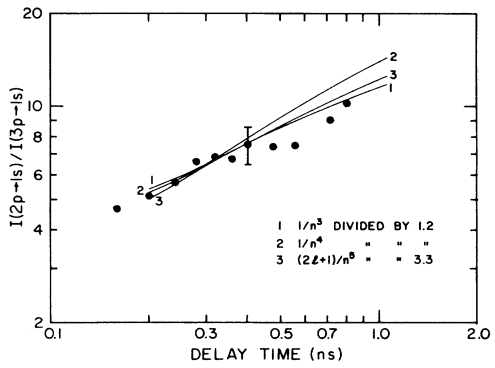

FIG. 3. Measured and calculated intensity ratios as a function of delay time. The calculated curves were normalized arbitrarily for easy comparison with the measured intensity ratios.

In conclusion, the distribution function that gave the best representation of both the slopes of the decay curves and the intensity ratios for the $2p$-$1s$ and $3p$-$1s$ transitions was $N(n,l,t=0)=n^{-4}$. An n^{-4} dependence was also observed for optical transitions in foil-excited fluorine and silicon ions by Dybdal *et al.*[17] The present results support the contention of Betz *et al.*,[18] that another mechanism besides the direct capture of electrons from the last layer of the foil plays a major role in the population of Rydberg states.

ACKNOWLEDGMENTS

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[1]P. Richard, Phys. Lett. **45A**, 13 (1973).
[2]Patrick Richard, R. L. Kauffman, F. F. Hopkins, C. W. Woods, and K. A. Jamison, Phys. Rev. Lett. **30**, 888 (1973).
[3]W. J. Braithwaite, D. L. Mathews, and C. F. Moore, Phys. Rev. A **2**, 465 (1975).
[4]A. P. Georgiadis, L. von Gerdtell, G. Dehmelt, D. Strater, R. N. Ahmad-Bitar, and P. von Brentano, Z. Phys. A **300**, 277 (1981).
[5]J. Rothermel, H.-D. Betz, F. Bell, and V. Zacek, Nucl. Instrum. Methods **194**, 341 (1982).
[6]J. Palinkas, R. J. Maurer, and R. L. Watson, Phys. Rev. A **32**, 2674 (1985).
[7]R. M. Schectman, Phys. Rev. A **12**, 1717 (1975).
[8]F. Hopkins and P. von Brentano, J. Phys. B **9**, 775 (1976).
[9]H.-D. Betz, J. Rothermel, and F. Bell, Nucl. Instrum. Methods **170**, 243 (1980).
[10]L. J. Curtis, Am. J. Phys. **36**, 1123 (1968).
[11]W. Gordon, Ann. Phys. (Leipzig) **2**, 1031 (1929).
[12]H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Plenum, New York, 1977).
[13]J. R. Oppenheimer, Phys. Rev. **31**, 349 (1928).
[14]G. Dehmelt, A. Georgiadis, L. von Gerdtell, D. Strater, P. von

Brentano, and R. N. Ahmad-Bitar, Phys. Lett. **89A**, 193 (1982).

[15]F. Hopkins, J. Sokolov, and P. von Brentano, in *Beam Foil Spectroscopy,* edited by I. A. Sellin and D. J. Pegg (Plenum, New York, 1976), Vol. 2, p. 553.

[16]H.-D. Betz, *Forward Electron Ejection in Ion Collisions,* Vol. 213 of *Lecture Notes in Physics,* edited by K. O. Groeneveld, W. Meckbach, and I. A. Sellin (Springer-Verlag, New York, 1984), p. 115.

[17]K. Dybdal, J. Sorensen, P. Hvelplund, and H. Knudsen, Nucl. Instrum. Methods B **13**, 581 (1986).

[18]H.-D. Betz, D. Roschenthaler, and J. Rothermel, Phys. Rev. Lett. **50**, 34 (1983).