Relative level populations in beam-foil-excited C IV, N V, O VI, and F VII

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(Received 17 September 1980)

We have studied the relative level populations in beam-foil-excited C IV, N V, O VI, and F VII by optical spectrometry in the range $n = 3$–12. These Li-like ions have the configuration $1s^2\,nl$. The population distribution curves are quite different, ranging from a sharp, approximately $(n^*)^{-3}$, decrease for C IV to flat maxima at $n \sim 7$ and 8 for O VI and F VII, respectively. This gradual change may be explained as resulting from a near-resonant charge transfer from the valence band of the carbon foil to the projectile. For fixed n, states with large l are preferentially populated. The consequences of these results in cascade analyses of lifetime data from beam-foil measurements are briefly discussed.

I. INTRODUCTION

The primary purpose of this work was to measure relative level populations in multiply ionized beam-foil-excited species. For neutral or weakly ionized atoms it is now well established[1] that the relative population of different levels of the same Rydberg series varies as an inverse power law of the effective quantum number n^* of the excited term. It is remarkable that this proportionality extends downward to include valence-shell levels as well as being valid asymptotically for large values of n^*. This relation is explained qualitatively in terms of final-state wave-function amplitudes in the vicinity of the nucleus,[2] in analogy with similar findings for ion-atom collisions.[3]

It has also been found[1] that the relative level population, for fixed value of the principal quantum number of the excited term, increases more rapidly with the orbital angular momentum quantum number l than as the statistical weight $(2l+1)$, and that, superimposed on this increase, there is a preferential population of p levels. However, the increase with l is established only for s through g levels, because, for weakly ionized ions, higher l-valued levels will decay in the infrared, where quantitative beam-foil measurements at present are impossible. Therefore, by turning to projectiles in higher final charge states, one may study whether the increase in population with l occurs monotonically, because with increasing charge state such transitions gradually move into the visible part of the spectrum.

The beam-foil excitation of Rydberg states is believed to take place as an electron pickup process when the projectile leaves the back of the foil,[2,4,5] because the mean diameter of a Rydberg state is so large that the projectile cannot exist in an undisturbed eigenstate as long as it is inside the foil. The mean radius $\langle r\rangle$ of a hydrogenlike system of nuclear charge Z and in the state (n,l) is in atomic units

$$\langle r\rangle = \frac{1}{2Z}[3n^2 - l(l+1)], \tag{1}$$

and the distance between two neighbor foil atoms is approximately 0.2 nm (calculated from the density of amorphous carbon). Thus, as soon as $\langle r\rangle$ approaches half of this value, the excitation must result from electron pickup processes at the back of the foil. For ion-atom collisions involving multiply charged projectiles it has been predicted theoretically that electron pickup processes will take place predominantly into excited states and not to the ground state due to near resonance between the initial and final states.[6] The same feature is also expected to occur in beam-foil excitations. Charge transfer in ion-atom collisions is enhanced by energy degeneracy which is readily understandable from a qualitative point of view. For hydrogenlike ions with nuclear charge Ze (e being the charge of a proton, and Z the atomic number of the nucleus), the term energies T_n are given by

$$T_n = -Z^2R/n^2, \tag{2}$$

where R is the Rydberg constant, and n is the principal quantum number. Accordingly, that level of a hydrogenlike ion with $n = Z$ will have binding energy identical to that of the ground state

of hydrogen. This will cause resonance charge transfer to the level with $n=Z$ in a collision of the type

$$A^{Z+}+\mathrm{H}\rightarrow A^{(Z-1)+}+\mathrm{H}^{+}, \tag{3}$$

where A^{Z+} is a bare nucleus. There will be similar near-resonance processes in collisions where the incoming projectile is multiply ionized but not completely stripped of electrons, or where the target atom is not hydrogen. The binding energy of an electron in the valence band of a carbon foil is approximately 10 eV. Thus, by applying a similar consideration to the beam-foil process, one should expect levels with such binding energies to be preferentially excited in multiply charged species, if the beam-foil excitation predominantly results from electron pickup from the valence band of the foil. The disparity in momentum between the swift projectile ion and the foil electrons will reduce the capture probability into all states by roughly the same factor.

Charge-exchange processes involving multiply ionized atoms have been extensively studied in recent years, largely because of their relevance for fusion research.[7,8] Most theoretical work concerns the interaction between H or H_2 and totally stripped atoms. Lately Ryufuku and Watanabe[9] and Salop[10] have calculated the total as well as differential capture cross sections in such processes. The latter have been measured for a large number of ions colliding with H and H_2. See, e.g., Meyer *et al.*[11] and Crandall *et al.*[12] The cross sections for populating various n, l states in collisions of multiply ionized Ar with various gases have recently been determined by El-Sherbini *et al.*[13] It is at present not possible to obtain such cross-section data in beam-foil experiments. Here the experimental conditions are much less well defined than in ion-gas collisions. The present paper is therefore limited to studies of relative level populations. As will be discussed below, such data are quite valuable in connection with decay-curve analyses.

II. EXPERIMENTAL

The measurements were carried out at the 3-MV Pelletron tandem accelerator in Lund. The experimental setup has been described before,[14] as has the quantum efficiency calibration procedure for the detecting system[15] and the data treatment.[1,16]

Care was taken to obtain beam-foil spectra of good quality for the population analyses. Thus the linewidths were typically 1.5–2 Å in the region 2000–5000 Å. A few examples of the spectra are given in Ref. 14.

The relative population N_j of level j, measured immediately downstream from the foil, was determined from[1,16]

$$N_j=S(\lambda_{jk})/K(\lambda_{jk})A_{jk}, \tag{4}$$

where $S(\lambda_{jk})$ is the signal of the optical transition of wavelength λ_{jk} from level j to level k, $K(\lambda_{jk})$ is the quantum efficiency of the optical detecting system,[15] and A_{jk} is the transition probability for the decay from level j to level k. The transition probabilities were taken from Refs. 17 and 18 which give values calculated in the numerical Coulomb approximation.[19]

When applying Eq. (4) it is assumed that repopulation through cascade processes can be ignored. This is generally justified for higher-lying levels, and we have also estimated cascade repopulation effects to be small for the low-lying levels studied here. As an example, in O VI the $3p$ term will essentially not be repopulated from the decay of the $3d$ level, because the $3d$ level decays with a 6×10^4 higher probability[17] to the $2p$ term. Also the higher d terms decay preferentially to the $2p$ term, although not as selectively as the $3d$ level, but they have longer lifetimes, making their cascade contributions relatively small. Because of the incompleteness of the data presented here a detailed quantitative cascade correction could not be carried out, and consequently, the reported populations of the lowest-lying levels may be too high. Fortunately, such a possible error will not change the conclusions of the qualitative discussion given in Sec. IV.

Transitions between levels of high values of the principal quantum number n and the orbital-angular-momentum quantum number l could not be resolved in all cases. Fortunately, in all such cases the transition probabilities did not deviate more than 30% from an average value for fixed n. Therefore, we have, in the data treatment of such unresolved transitions, applied average values for the transition probabilities, and this has enabled us to obtain good estimates for the total populations of some upper terms with high values of n and l.

III. RESULTS

The relative level population results for C IV, N V, and O VI are shown in Fig. 1. (The carbon results are taken from Ref. 20.) The data points are labeled with the values of n and l. In the cases where transitions were unresolved (c.f. Sec. II), all possible values for l are shown.

A few additional results were obtained for O IV, O V, and F VII. Since these data sets were less complete than those presented in Fig. 1, we shall here only mention that the $3p$ level in O V was populated fairly strongly, but in F VII the $3p$

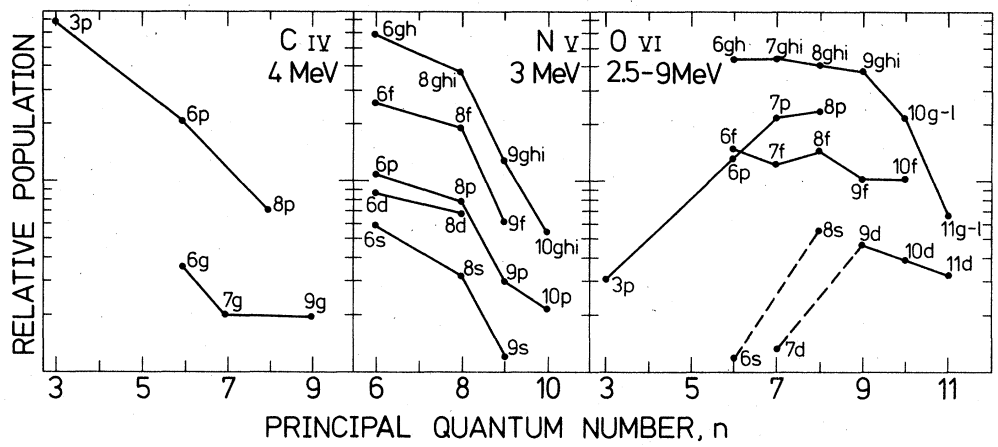

FIG. 1. Relative level populations for some terms in C IV, N V, and O VI (all of configurations $1s^2nl$) versus the principal quantum number of the excited level after beam-foil excitation. All relative level population data for one element are on the same scale, whereas the scale changes from element to element.

level population was much smaller than those of high-lying Rydberg states; levels of $n \sim 7$–8 were populated preferentially in F VII. Thus, we note that, whereas the relative level population decreases with n for fixed value of l in the less ionized species C IV and N V, there is a preferential population of medium to high-lying levels in the more strongly ionized species O VI and F VII. It is also seen that, for fixed value of n, the population increases generally with l, but in addition the p levels are overpopulated, c.f. especially O VI.

The $3p$ levels in O VI and F VII have very short lifetimes, and they may decay within less than a mm along the beam axis. This fact, together with the foil-surface roughness and shadowing effects from the foil holder, may imply that ions excited to the $3p$ level had decayed to a considerable extent before they reached the observation region, making the $3p$-level populations appear too low. However, such effects are not sufficient to explain the remarkably low populations of the $3p$ levels in O VI and F VII and thus the drastic deviation from the inverse power law that was observed for weakly ionized species.[1]

The oxygen data were recorded at different projectile energies in the interval 2.5–9 MeV and showed essentially no change with projectile energy on a relative scale.

IV. DISCUSSION

It is seen from Fig. 1 how the relative population of levels within the same Rydberg series changes with increasing core charge from being fairly steeply decreasing (C IV, N V) to showing an inverse population (O VI). It is apparent that the N V curve at $n = 6$ to 8 is leveling off. Thus there is a continuous change in population distribution from the roughly n^{-3} decrease for C IV to the strongly inverted O VI and F VII. For neutral and weakly ionized elements it is well established[1] that the relative population of levels of the same Rydberg series varies as an inverse power law with the effective quantum number of the excited term, but this is clearly not the case for the higher ionized projectiles studied here, a finding which was not unexpected, as mentioned in the introduction. The possibility stated there, namely electron pickup from the valence band of the foil, offers itself readily as an explanation.

To test this hypothesis we have in Fig. 2 plotted the $1s^2\ nl$ energy levels of all the measured species, C IV, N V, O VI, and F VII, together with those of C I as a representative of the carbon foil. All the population distribution curves in Fig. 1 can be viewed as portions of imagined extended curves

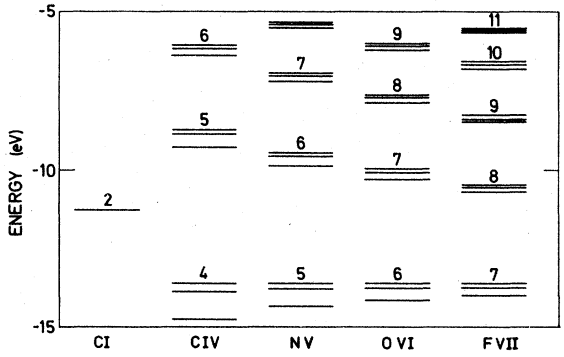

FIG. 2. Energy levels of C I, C IV, N V, O VI, and F VII (except for C I all of configuration $1s^2nl$) normalized to zero at the ionization level.

with a soft maximum. Then O VI has its maximum at $n \sim 7$ ($n = 6$–8), F VII at $n \sim 8$, N V probably at $n \sim 6$ (or 5–6), and the imagined maximum for C IV must be at $n \sim 5$ or less (the large population of the $3p$ state should not be disturbing since it may be caused by a molecular excitation effect in the collision between a carbon ion and a target atom). All these levels have binding energies around 10 eV, as does the $2p$ level of C I and therefore also the valence electrons of the graphite foil. Thus there is a strong indication that the nonmonotonic population distribution curves of Fig. 1 may be explained by preferred resonance charge transfer of a foil valence-band electron onto the projectile $1s^2$ core. The captured electron could conceivably also come from a free-electron gas or wake-riding electrons accompanying the ion.[21] The density of free electrons necessary to yield the observed capture probabilities by the former process has been found to be unrealistically high.[4] Similarly the density of wake-riding electrons is probably too low, and capture of these should not produce a preferred population of states with binding energies around 10 eV. The present experimental material thus confirms the assumption of the capture of electrons bound in the carbon foil. Additional studies of other ion-target combinations would clearly be of a great value. For example, the origin of the transferred electrons should be further traced by using foils with different binding energies of the valence-band electrons (i.e., different foil materials). Such measurements have to be performed under clean vacuum conditions in order to maintain a clean foil surface, i.e., with a rest gas pressure below 10^{-9} Torr. Until now practically all beam-foil measurements have been done at much higher pressures. A more straightforward way of further testing the interpretation given here will be to apply other projectiles with different ionization potentials and various charge states. Clearly the level structures have to be rather extensively known.

We see from Fig. 1 that the population increases with l for levels with the same value of n, in agreement with findings for neutral and weakly ionized elements.[1] Thus the excitation goes primarily to levels which are geometrically as small as possible for a given n [see Eq. (1)] and which also have remarkably large orbital periods in time. Fano[22] has pointed out that, for electron-atom collision processes, excitation of atoms to levels with high values of l may take place with substantial probability when the incoming electron energy is very close to the ionization threshold. In such a process the electron can form an intermediate negative ion complex with the target atom. The incident and the excited electrons become correlated, and this together with their low relative velocities enables them to exchange angular momentum in amounts sufficient to reach high l values. The attainment of high orbital angular momentum by one electron must result from a torque exerted on it by another electron, and it turns out[22] that high values of l can be reached only if the interaction lasts for a time comparable to the orbital period in time of the excited state. Probably a similar picture can be applied to the beam-foil excitation process. During the passage through the foil some electrons may follow the projectile. When the projectile leaves the back of the foil, an electron which has gained some correlation in velocity with the projectile, but later escapes as a free electron, may interact with the excited electron over a fairly large distance (or time). This may lead to a sufficiently large torque exerted on the excited electron, and lasting for so long a time that the excited electron ends up in a state with high value of l (close to the highest possible l if not that level itself). Thus, in such a picture, the excited electron is at the back of the foil caught from the valence band, a process taking place predominantly to levels with $n^* \gtrsim Z_{\text{core}}$, and the final distribution of excitation among the different l-valued levels is caused by long-range electron-electron interactions.

Beam-foil measurements have in recent years provided lifetimes for many levels in multiply ionized atoms. Significant systematic differences between such data and those predicted by modern theories have been noted in several cases, however.[23,24] Since some of the systems in which disagreements occur (e.g., the Na I and Cu I isoelectronic sequences) are comparatively simple to handle theoretically, it is generally belived that the disagreements are largely due to experimental shortcomings. The nonselective excitation during the beam-foil interaction may result in pronounced cascading from several higher levels into the level under study. The resulting decay curves from lifetime measurements are then the sums of many exponentials, and the extraction of the primary lifetime is a very difficult problem. Considerable systematic errors may arise even when the counting statistics are good.

A number of attempts have therefore been made to model beam-foil decay curves and compare such "data" with experimental material. In constructing the synthetic decay curves the lifetimes and initial populations for a considerable number of levels should be known. Usually theoretical lifetimes are combined with various beam-foil population and excitation models. When an experimental curve is duplicated in such a way it is

usually taken for granted that the theoretical lifetimes are essentially correct.

In connection with a study of the NaI isoelectronic sequence Crossley *et al.*[23] constructed decay curves for the $3s-3p$ resonance transition in NaI–ArVIII. A considerable number of yrast cascades ($3p$-$3d$-$4f$-$5g$...) were thus considered, and an n^{-3} dependence of the excitation was used. In a detailed study of Cu-like KrVIII Younger and Wiese[24] were able to reproduce the $4s$-$4p$ decay curve by considering cascades from about 20 yrast states and a few Rydberg states. Here a $(2l+1)/n^{*3}$ initial population model gave the best agreement with experimental data. Since the $(2l+1)/n^{*3}$ distribution diverges for large n^* values, a cutoff is needed. Younger and Wiese[24] also note that ions in very high n states cannot prevail because of disturbing microfields. They further quote some earlier work[25,26] to support the assumption that the initial populations first increase with n and then decrease.

While this has not been mentioned in Refs. 25, and 26 one may perhaps find some evidence for it in the spectra shown in the two papers. However, since the detection systems were not intensity calibrated, we do not consider previous experimental material sufficient to demonstrate the existence of a population maximum.

The model used in Ref. 24 has also been applied to the $4s^2\,{}^1S-4s4p\,{}^1P$ resonance transition in KrVII (ZnI sequence)[27] and here also the simulations essentially reproduced the experimental decay curve.

Recently Hultberg *et al.*[28] recorded many decay curves for levels in ZnII (CuI isoelectronic sequence) using very favorable counting statistics. Furthermore, the initial populations of the ZnII levels were also determined by means of the method used in the present paper. These experimental data were further compared with synthetic decay curves using theoretical level lifetimes and a variety of population models. The $(n^*)^{-5}$ population model described the decay curve for the $4s-4p$ resonance transition, whereas the $(2l+1)\,(n^*)^{-5}$ populations gave reasonable approximations of the decay curves for the $4d$ and $4f$ levels. Using new high-quality beam-foil data for the $4s-4p$ resonance transition in Cu-like KrVIII Curtis, Livingston *et al.*[29] obtained agreement between the measured and simulated decay curves by assuming a $(2l+1)\,(n^*)^{-2}$ population distribution. These examples show that it is very difficult to obtain a population model that can be applied to a large number of systems. Furthermore, the change in n^* dependence with increasing Z along an isoelectronic sequence is consistent with the findings of the present paper. Since binding energies of about 10 eV correspond to higher and higher n states when Z increases, it is clear that high-lying states tend to be strongly populated in this case. Indeed, a lifetime curve for the $3p\,{}^2P$ level in Na-like CuXIX[30] is reproduced by a $(2l+1)\,(n^*)^{-1}$ model.[29] There is additional evidence of the excitation of very-high-n states in highly ionized species.[31] We would also like to draw attention to some interesting theoretical results of Ellis[32] which predict substantial populations of high n, limited l in the beam-foil excitation process.

ACKNOWLEDGMENTS

E.V. is grateful to the Department of Physics, Lund University, for the hospitality he enjoyed during his stay, as well as for financial support from the Nordic Committee for Accelerator-Based Research. We wish to thank the Danish Natural Science Research Foundation for placing the monochromator at our disposal. This work was also supported by the Swedish Natural Science Research Council (NFR) and the National Swedish Board for Energy Source Development (NE).

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