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Experimental study of relative level populations in beam-foil-excited Ar VIII and Kr VIII

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The relative level populations in beam-foil-excited sodiumlike argon (Ar VIII) and copperlike krypton (Kr VIII) have been studied at 2-MeV projectile energy. In Ar VIII, the $3p$ and the $3d$ terms are strongly excited. The level population decreases rapidly with increasing value of the principal quantum number n, reaches a minimum for levels with $n = 5$ or 6, but increases then and reaches a strong maximum for levels with $n \simeq 11$. As an example, the $11d$ level is populated a factor of 2 more than the $3d$ level. Also the Kr VIII data show a population maximum for levels with $n = 11$. The strong $3p$ and $3d$ level excitations in Ar VIII are explained as selective inner-shell processes which can be understood in the molecular-orbital electron-promotion picture. The preferential population of high-lying Rydberg states is explained as resulting from a near-resonance electron transfer from the valence band of the carbon foil to the projectile. This process takes place when the projectile leaves the back of the foil, and the distribution of population upon different n levels will vary with charge state. The level population for levels with fixed n increases rapidly with increasing value of the orbital-angular-momentum quantum number for levels in Kr VIII, Kr X, Kr XI, and Kr XIII. The results are discussed, and their influence on decay curves as well as their relations to multiply charged ion-atom collisions are pointed out.

I. INTRODUCTION

It has recently been found in an experimental study of relative beam-foil level populations in Li-like oxygen (O VI) and fluorine (F VII) that there is a preferential excitation of levels with binding energies of approximately 10 eV or, in other words, of levels with principal quantum number $n \simeq 6-8$ in these multiply ionized projectiles.[1] As an example, in O VI the $7p$ level was found to be populated approximately 10 times more than the $3p$ level.[1]

Such a preferential population of rather highly excited levels is very different from what is observed in neutral or weakly ionized projectiles, where it is well established[2] that the relative population of levels within the same term series decreases monotonically as $(n^*)^{-p}$, where n^* is the effective quantum number of the excited level, and the power p is slightly larger than 3 ($3 < p < 5$ generally[2]). The results of Ref. 1 clearly called for further experiments. Although the data indicated that the power law observed for neutral and weakly ionized species breaks down for the multiply charged ions O VI and F VII, the data presented in Ref. 1 are incomplete. That experiment indicated the need for a study in which the relative level population is determined for a large number of excited terms, including several values of the principal quantum number n as well as the orbital-angular-momentum quantum number l. In addition, the results given in Ref. 1 are rather uncertain because the low-lying levels in O VI and F VII have very short lifetimes. Therefore, in that study,[1] non-negligible parts of the primary decays took place in the region where foil-holder shadowing effects occurred, so that part of the initial level populations had died out before the projectiles reached the observation region, and a certain amount of repopulation through cascade processes had taken place too. The total errors caused by these two counteracting processes are difficult to estimate. Also from this viewpoint, a more elaborate work is justified.

We shall present here a fairly complete set of relative level populations for Ar VIII (sodiumlike argon), together with a less complete set for Kr VIII (copperlike krypton), neither of which is contaminated noticeably from the above-mentioned foil-holder shadowing effects.

The preferential population of levels with binding energy of approximately 10 eV in O VI and F VII was explained as a result of near-resonant electron pickup in the following way.[1] The beam-foil-excitation process for Rydberg states is believed to take place as electron pickup into the excited state, when the projectile leaves the back of the foil.[3,4] The valence-band electrons in the carbon foil have binding energies of approximately 10 eV. Electron pickup processes are expected to occur preferentially for near-resonance processes.[5] Since pickup of an electron from the valence band in the foil into an excited projectile state will be near resonant for projectile states bound approximately 10 eV, the population of such levels will be favored. This explanation has been adapted from the theory of ion-atom collisions involving multiply charged incoming projectiles, where electron transfer is believed to take place predominantly through near-resonance processes.[5] We mention here that although attempts have been made to study such ion-atom collisions, the experimental results are meager.[6] As we shall see, the beam-foil excitation of multiply charged species offers a similar, but experimentally much simpler situation than that of ion-atom collisions for studying quasiresonant electron transfer to excited levels.

A study of level populations in medium-to-highly ionized, beam-foil-excited species has its own inherent interest, because no experimental data are yet available, except for the sparse data on C IV, N V, O VI, and F VII presented in Ref. 1, and a similarly incomplete set using 1.5-MeV Fe projectiles.[7] Further experiments similar to the ones presented here will yield valuable information concerning penetration of heavy particles through matter, a large field which, although having been studied frequently by different experimental techniques and theoretical approaches, still has many unexplained features. However, besides its large academic interest, there is a practical need for experimental relative beam-foil level-population data for performing cascade corrections when deducing lifetime values from experimental beam-foil decay curves.

There have been some attempts made recently[8] to synthesize beam-foil decay curves. Two data sets are necessary for such synthesization processes, namely, a complete set of transition probabilities, and a complete set of relative, initial beam-foil level populations. If such reliable sets of data are available, one may hope through synthesizing of decay curves to reveal how the different cascade processes influence the experimental decay curve in question. Of all such works performed, only one, namely, on Zn II (Ref. 8) has been based on an experimentally determined set of initial level populations. Another work, on Kr VIII,[9,10] clearly indicated the need for experimental data for medium-to-highly ionized species, causing a special motivation for us to study Kr VIII.

Beam-foil measurements are normally carried out in vacuum chambers with a residual pressure so high that the foil surface is contaminated with impurities, and also no efforts are normally made to prevent the foils from being coated by a film of pump oil. This work is no exception from such "dirty excitation conditions." The excitation probabilities may be sensitive to the presence of impurities inside as well as on the foil surface, and therefore, one warning is appropriate concerning the relative-level population data presented here. The results need not be reproducible by other laboratories working under other experimental conditions.

In this article, the expressions "relative initial level population" or just "relative level population" refer to the situation of the projectiles at the initial part of a beam-foil decay curve.

II. EXPERIMENTAL

The measurements were carried out at the 2-MV van de Graaff accelerator facility at the University of Arizona in Tucson. Singly charged ions of Ar or Kr were accelerated to an energy of 2 MeV and sent through thin carbon foils (5–10 $\mu g/cm^2$). Spectral scans were performed with the observation region immediately downstream from the foil, and photon intensities of the different optical transitions were read from the charts. The analytical device was a 1-m normal-incidence scanning monochromator.

The relative population N_j of level j was determined from the signal $S(\lambda_{jk})$ of the optical transition of wavelength λ_{jk} from level j to level k through[2]

$$N_j = S(\lambda_{jk})/[K(\lambda_{jk})A_{jk}], \tag{1}$$

where $K(\lambda_{jk})$ is the overall detection efficiency[11] at wavelength λ_{jk}, and A_{jk} is the transition probability for the transition under study. The transition probabilities used for Ar VIII were taken from Ref. 12, which lists theoretical values calculated in the numerical Coulomb approximation, whereas those used for Kr VIII-Kr XIII were scaled hydrogenic values, also taken from Ref. 12, because, as we shall see, only transitions from levels with high value of l were analyzed for krypton.

The efficiency calibration of the optical detection system is easy to carry out at wavelengths above about 200 nm (Ref. 11) by use of a filament lamp, but there is no similar, easy method of calibration in the vacuum ultraviolet region of the spectrum.[11] This is especially unfortunate when dealing with such highly ionized species as Ar VIII and Kr VIII, because many of the transitions of high interest, e.g., the resonance transitions, are at wavelengths below 200 nm. Fortunately, we were able to include some vacuum ultraviolet (VUV) transitions in the data analysis by using the branching-ratio method.[11] As an example in Ar VIII, the $3s$-$3p$ transition at 71 nm is so close to the $4f$-$5d$ transition at 67.5 nm that the quantum efficiency can be assumed to be constant over this wavelength range. Thus, by applying Eq. (1) to these two transitions, the population of the $3p$ term can be determined relative to that of the $5d$ term. However, the $5d$

term has another decay branch at a wavelength of 309 nm ($5p$-$5d$), which is in the middle of the wavelength region easy to calibrate.[11] Thus, from the latter transition, the $5d$ level population is determined relative to all other Ar VIII levels which have an observable transition in the wavelength region 200–600 nm, and consequently, the $3p$ level population can be determined relative to all these levels.

Another important transition in Ar VIII is the $3p$-$3d$ decay, which is close to the $5p$-$8s$ transition (at 53 and 50 nm, respectively), but the $7p$-$8s$ transition is at 299 nm, so from analyzing these three optical transitions by use of Eq. (1), the $3d$ level population can be linked to the other relative-level-population determinations. The overall uncertainty in the relative-level-population determination is naturally larger when such a branching-ratio method is applied, but additional valuable information is, on the other hand, obtained in such a way.

The total uncertainties related to our experimental-level-population data are difficult to evaluate precisely because of the transition probabilities involved in the data reduction [cf. Eq. (1)]. We believe that for most of the levels, the relative uncertainty in level population is not larger than 20%. For low-lying excited levels, which deacy by emitting radiation in the VUV so that the branching-ratio method has to be applied as described above, the overall uncertainty may well be up to 50%. These uncertainties do not mask the main features of the population distributions.

Application of Eq. (1) is justified when the following conditions are fulfilled. The length along the beam axis of the observation region must be so short, compared with the distance corresponding to a mean life, that the level population under study can be regarded as being constant over the observation region. In addition, the observation region must be located so close to the exit surface of the foil that the fraction of decays taking place between the foil and the observation region is small compared to the initial level population. Also, repopulation through cascade processes must be small. These conditions were fulfilled in this work by masking the spectrometer grating, so that the length of the observation region as well as its location from the foil was approximately 0.1 mm. Therefore, no corrections due to dying out in front of the observation region or to cascade repopulations had to be made. This was checked by recording the initial parts of the decay curves

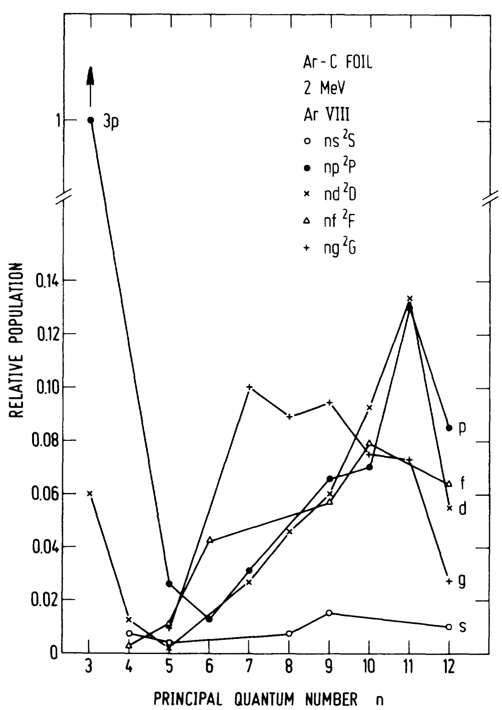

FIG. 1. Relative level populations for some terms in Ar VIII versus the principal quantum number of the excited term, after beam-foil excitation at a projectile energy of 2 MeV. All the relative-level-population data are on the same scale. They have been normalized so that the $3p$ level population is unity. Note the broken scale for the $3p$ level.

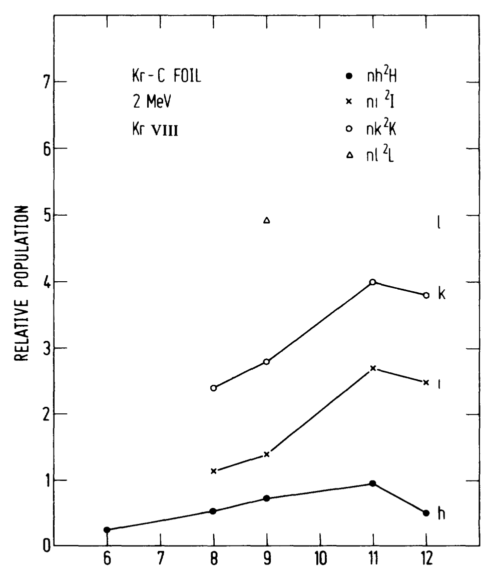

FIG. 2. Relative level populations for some terms in Kr VIII versus the principal quantum number of the excited term, after beam-foil excitation at a projectile energy of 2 MeV. All the relative-level population data are on the same scale.

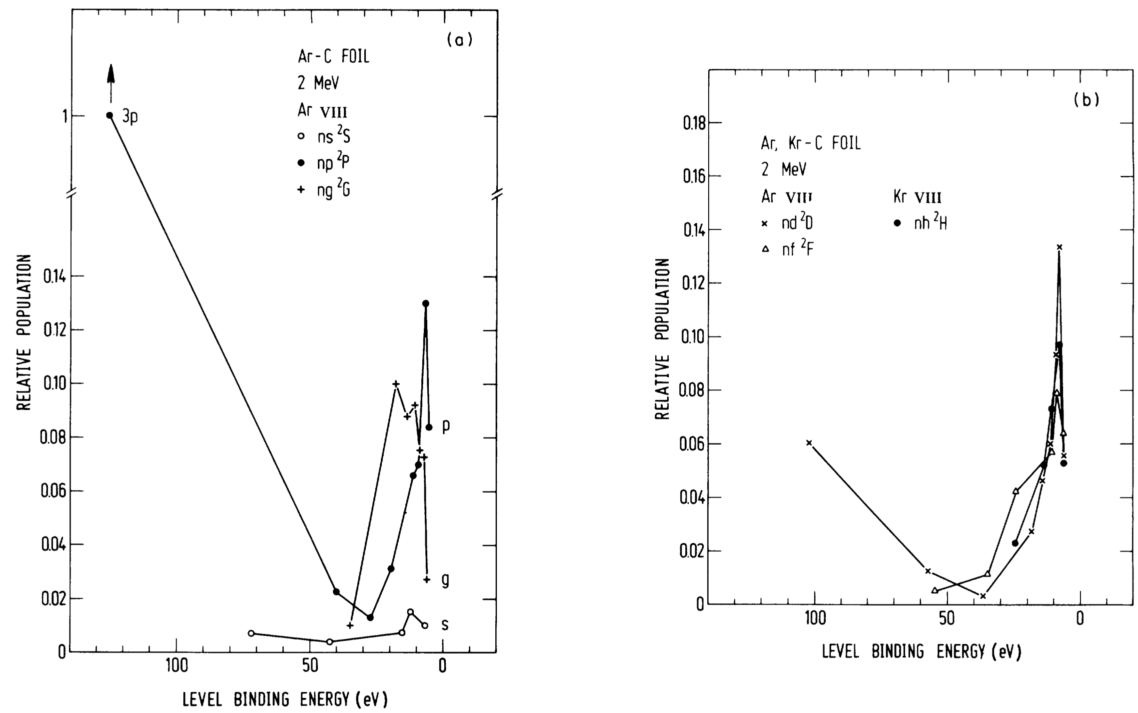

FIG. 3. (a) Relative level populations for s, p, and g levels in Ar VIII versus the binding energy of the excited level. Note the broken scale for the $3p$ level. (b) Relative level populations for the d and f levels in Ar VIII and the h levels in Kr VIII versus the binding energy of the excited level.

for some of the fastest transitions, in which way the foil position was also determined.

Many of the spectral lines belonging to transitions between the high-lying Rydberg states in Ar VIII and Kr VIII have not been reported in the literature. The extrapolated wavelengths given in Ref. 12 for Ar VIII were used for line identifications, together with the wavelength lists for krypton given by Cardon.[13]

III. RESULTS

The relative-level-population results for Ar VIII and Kr VIII are plotted versus the principal quantum number of the excited term in Figs. 1 and 2. The same data in Fig. 3 are plotted as functions of the level binding energy. The ionization level and the excitation energies for Ar VIII are taken from Ref. 12, whereas the corresponding data for Kr VIII can be found in Ref. 14. We have used scaled hydrogen excitation energies for the high-l levels of Kr VIII.

The results presented here are of a higher quality than those given in Ref. 1 because the lifetimes in the present investigation were in a better range (c.f. the introduction). In addition, the Ar VIII data set is more complete than that presented in Ref. 1, and our study of Kr VIII yields further information for levels with high values of l. We have determined the populations of 35 levels out of the 46 levels existing in Ar VIII with $3 \leq n \leq 12$ and $0 \leq l \leq 4$. In Ref. 12 transition probabilities only for levels with $n \leq 12$ are listed. Therefore, we have been unable to treat possible decays from levels with $n > 12$, albeit transitions from such levels may be present in our spectra.

IV. DISCUSSION

A. Relative level populations as functions of the level binding energy

For the p and d terms in Ar VIII we see from Fig. 1 that the lowest-lying levels, namely, the $3p$ and the $3d$ terms, are strongly excited. Then, the population has a minimum for n equal to 5 or 6, and thereafter increases dramatically to a maximum for levels around $n = 11$. Note that the populations of the $11p$ and $11d$ levels are slightly above that for the $3d$ term and only surpassed by that of the $3p$ term.

The f- and g-term populations increase with n from very small values for the $4f$ and $5g$ levels to a maximum around $n = 10$ for the f series and a broad plateau for the g series. The population maxima for these two term series are 10 times or more above the populations of the $4f$ and the

5*g* terms. The *s*-level-population data, which are much more sparse, follow similar trends, but whereas it can be said that there is no strong population of the lowest-lying *s* level (4*s*), the height and the location with regard to *n* of the maximum cannot be determined precisely. We only note that the maximum occurs for *n* equal to 9, 10, or 11, and that it is at least a factor of 2 above the 4*s*-level population.

It is clear from Fig. 1 that two excitation processes are active for Ar VIII, one causing the strong 3*p*- and 3*d*-level populations, but apparently not active for the 4*s* and 4*f* levels, and the other excitation mechanism causes the maxima in level populations for high values of *n*. To proceed further with the discussion of these features, we first estimate the values of the mean radii of the different orbitals and compare them with an estimate for the average distance between two neighbor foil atoms.

The mean radius $\langle r\rangle$ of a hydrogenlike ion of nuclear charge Z is in atomic units (a.u.) given by

$$\langle r\rangle = [3n^2 - l(l+1)]/2Z\,. \qquad (2)$$

From this relation one can estimate the expectation value of the radius of the 3*p* and 3*d* levels in Ar VIII to be approximately 1.6 and 1.3 a.u., respectively. These values are so small compared to the mean distance between two neighbor foil atoms (which is around 4 a.u., calculated from the density of amorphous carbon) that the 3*p* and 3*d* orbitals can exist fairly undisturbed when the projectile penetrates the foil. Therefore, excitation of these states must be discussed in the well-developed inner-shell excitation picture.[15] One arrives at the same conclusion concerning the starting point of an excitation-mechanism discussion from inspecting the level binding energies, which are 126 and 102 eV for the 3*p* and 3*d* terms, respectively.[12] The levels are tightly bound, leading to an excitation-mechanism discussion in terms of the inner-shell excitation picture,[15] although the 3*p* and 3*d* levels in Ar VIII in fact are outer-shell excitations from a spectroscopist's viewpoint.

The projectile velocity is so low (1.4 a.u.) compared to the classical orbital velocities of the inner-shell electrons that inner-shell excitation will be well described by considering the transient quasimolecule formed momentarily by the collision partners carbon and argon.[15] (Coulomb excitation can be ignored.) The dominating processes in such encounters can be found from inspection of the molecular orbital (MO) description.[16] Figure 4 is a correlation diagram for the C-Ar collision, constructed from the rules given in Ref. 16. The correlation diagram shows

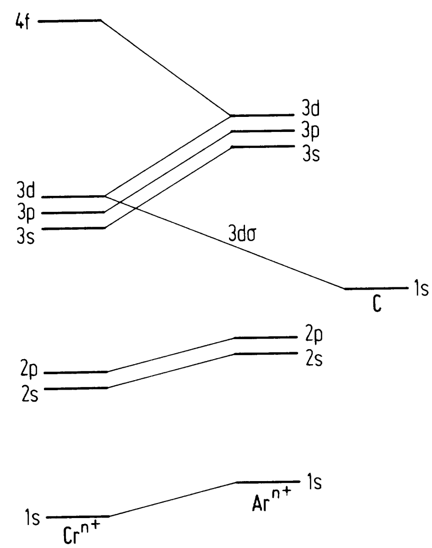

FIG. 4. Diabatic molecular-orbital diagram for the Ar^{n+}-C collision shown schematically. To the right of the figure the inner-shell terms of the separated atoms (not to scale) are shown, and the united-atom inner-shell terms are given to the left (not to scale). The terms of the separated and the united atoms have been connected by straight lines to indicate how the levels correlate during a collision.

schematically how the different levels of the separated collision partners (shown to the right of the figure) correlate at different internuclear distances (the abscissa). By constructing the correlation diagram in Fig. 4, one ambiguity arises, because in neutral or weakly ionized argon, the binding energy of a 2*p* electron is smaller than the binding energy in neutral carbon, whereas in highly ionized argon the 2*p* electrons have higher binding energies than the *K*-shell electrons in carbon. Thus, with increasing ionization stage of argon, a "swapping" in the relative position of the argon 2*p* and the carbon 1*s* levels occurs.[16] This problem has been discussed by Fortner and Garcia.[17] However, when dealing with excitations leading to Ar VIII, the 2*p* binding energy of argon to be used when constructing the correlation diagram in the incoming channel will be that of Ar^{8+}. By doing so, the 2*p* level in Ar is clearly below the 1*s* level in carbon, resulting in the diagram shown in Fig. 4, which agrees with that shown in the bottom of Fig. 4 of Ref. 17.

It is readily seen from Fig. 4 that during an Ar^{8+}-C collision, the carbon 1*s* electrons will be promoted to the 3*dσ* orbital, and one (or both) of the 1*s* electrons in carbon can, through curve crossings or interactions in the united-atoms limit, be transferred to the 3*s*, 3*p*, or 3*d* levels in argon, the latter two processes being exactly what is needed to explain the relatively strong $3p$- and $3d$-level populations in Ar VIII. We also learn from Fig. 4 that levels above the $3d$ term in Ar VIII are not populated through MO electron promotion mechanisms, and this is in agreement with our observations of low populations of the $4s$, $4f$, and the $n = 5$ levels.

The decrease in level populations from the $3p$ to the $5p$ level, or from the $3d$ to the $5d$ level, is indeed noteworthy, but this is slightly difficult to appreciate from Fig. 1. If for these levels we fit the level populations with power laws of the form

$$N_j = \text{const} \times (n^*)^{-p}, \qquad (3)$$

we find $p = 6.4$ and 5.8 for the p and the d terms, respectively. Such a power-law fitting is qualitatively in analogy with the finding for neutral or weakly ionized projectiles[2] that the population of levels within one term series can be well approximated with an expression of the form presented in Eq. (3). However, generally a much slower decrease in population with increasing n^* has been found ($2 < p < 4$) for neutral or weakly ionized projectiles.[2]

After having described the strong $3p$- and $3d$-level excitations in Ar VIII as caused by inner-shell MO electron promotion processes, let us now focus our attention on the strong population of high-lying Rydberg levels in Ar VIII (Fig. 1) and Kr VIII (Fig. 2). This is clearly the same phenomenon as that observed earlier[1] for O VI and F VII, and shall therefore find the same explanation.[1] These high-lying levels are geometrically large in size. The expectation value of the orbital radius for a level with $n = 10$ and $0 \leq l \leq 4$ is approximately 18 a.u., estimated from Eq. (2); and this is so large that a projectile cannot exist undisturbed in such an eigenstate when it traverses the foil, but the excitation must either take place when it leaves the back of the foil, or downstream from the foil. A variety of such processes can be imagined, namely, capture of an electron from the valence band of the foil,[1,3,4] capture of a wake-riding electron,[18] and radiative recombination with a free electron.[19] However, as already discussed in Ref. 1, the latter two processes are improbable explanations because the appropriate electron densities are too low, and also because such processes will not produce a preferential population of terms with binding energy of approximately 10 eV, as that seen in Fig. 3. On the other hand, electron pickup from the valence band of the carbon foil into excited states of the projectile can well account for the preferential population of levels with binding energies around 10 eV. This is because electron pickup processes are favored by energy resonance.[5] The valence-band electrons have binding energies of approximately 10 eV. Therefore, electron transfer will preferentially take place in projectile states of this binding energy, and this is in close agreement with our experimental finding, as shown in Fig. 3. The level populations for O VI (Ref. 1) are also found to reach maximum values for levels bound approximately 10 eV, as seen for Ar VIII and Kr VIII, indicating that the same excitation mechanism is acting in these ions of different elements and of different charge states.

We have mentioned above that for neutral or weakly ionized projectiles, the population of levels within the same term series has been found empirically to decrease as a power law[2] [cf. Eq. (3)]. But for all such cases,[2] the levels studied had binding energies less than 10 eV or, in order words, are in a region where also the level populations in O VI, Ar VIII, and Kr VIII are decreasing functions of the level excitation energy, or effective quantum number. Thus, there is no conflict between the preferential excitation of high-lying levels in multiply ionized projectiles observed here and the power-law population observed previously.[2]

Although this work substantiates the ideas proposed in Ref. 1, showing a much more complete picture, there remains the study of how the level population behaves in highly charged ions for the level binding energy tending asymptotically to zero. The asymptotic behavior of the level population is important to study, because various experimental findings indicate that the projectiles are subject to a rather strong local electric field as long as they are in the vicinity of the foil, the field being produced by the beam-foil interaction itself.[20] Such a field will reduce the ionization level of the projectile. Therefore, the highest-lying projectile energy levels will become unbound and this will lead to a net reduction in population for levels close to the ionization level for the free projectile, causing a motivation to study the asymptotic behavior of the level population.

Our relative-level-population data for Ar VIII indicate the presence of two beam-foil-excitation mechanisms. The lowest-lying, geometrically small levels, are very selectively populated through inner-shell excitations inside the foil. Contrary to this, levels of geometrically large size are populated through transfer of electrons from the carbon-foil valence band into the projectile excited states taking place when the projectile leaves the back of the foil, and populating preferentially levels with binding energies close to that of the valence band through near-resonance pickup. These findings are in agreement with the ideas proposed in Refs. 3 and 4, and are also in agreement with the weak $3p$ excitation in O VI, reported in Ref. 1, because of the following two reasons. Inner-shell processes will not lead to $3p$ excitations in O VI because, in the initial channel, the projectile will only carry with it two $1s$ electrons. This makes the appropriate diabatic MO correlation diagram extremely simple, and the result is that molecular orbitals which might lead to promotion of an electron into the $3p$ level of oxygen are initially empty, so that no $3p$ excitation will occur through inner-shell excitations. Also, the electron pickup process acting at the back of the foil will be relatively weak, because the $3p$ binding energy is 56 eV, so electron pickup will be far from resonance. In fact, from Fig. 3 we learn that pickup so far from resonance is reduced roughly a factor of 10 from the maximum populations occurring at resonance, and this reduction of a factor of 10 is very close to what was found for the $3p$ level in O VI in Ref. 1.

B. Relative level population as a function of l

It has been found[2,4,21] that the relative level population, for a fixed value of the principal quantum number of the excited level, increases with the orbital-angular-momentum quantum number l, although not in proportion to the statistical weight factor $(2l+1)$. In addition, there is a preferential population of p levels, superimposed on the general increase with l. This is substantiated with our findings for krypton, but, as is evident from Fig. 1, not for Ar VIII.

For medium- to high-lying levels in Ar VIII, we observe that s states are weakly excited, and the p, d, f, and g level populations come fairly close to each other for a fixed value of n. However, for $5 \leq n \leq 9$, the f and g levels are somewhat more populated than the p and d terms. A similar weak excitation of s states has been noted in neon by Bashkin *et al.*[22]

The Kr VIII data are in much better accord with the systematical trends observed previously.[2,4,21] The populations of the observed levels with $n=9$ in Kr VIII are plotted in Fig. 5 semilogarithmically versus the orbital-angular-momentum quantum number of the excited term, and the points have been connected with a straight line. In Fig. 5 some data are also presented for Kr X, Kr XI, and Kr XIII, calculated from the line intensities given by Cardon.[13] The values of n are indicated in the figure. We see that the general increase in level population with increasing value of l is generally much stronger than the statistical weight factor $(2l+1)$, which is indicated at the bottom of Fig. 5. Rather, the level population increases exponentially with l:

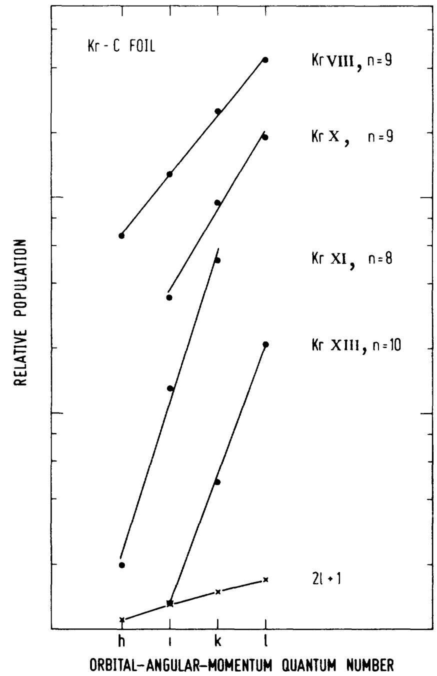

FIG. 5. Relative-level-population data for some terms in Kr VIII, Kr X, Kr XI, and Kr XIII plotted versus the orbital-angular-momentum quantum number of the excited term. Data for levels of same value of the principal quantum number are connected by straight lines, and the values of the principal quantum number are indicated on the figure. At the bottom of the figure the statistical weight factor $(2l+1)$ is also shown.

$$N_j = \text{const} \times P^{\,l}, \tag{4}$$

and we find $P \simeq 2$ for Kr VIII, but P reaches a value as high as 5 for Kr XI. Our data for krypton clearly extend systematical trends to higher values of l than previously reported.

The distribution of population among the different l-valued levels with the same value of n has been discussed in Ref. 1 and suggested an explanation in terms of long-range electron-electron interactions. We shall offer here an additional explanation, which is based on a classical visualization of the electrons moving in their orbits. In such a picture, the electron moves in an orbit which, for fixed value of n and increasing l, will come closer and closer to a circular orbit. Also, the velocity will be more and more independent of the position in the orbit. Now, electron pickup will be favored not only by energy matching, as discussed in subsection A, but also by velocity matching. In the classical picture, an electron which is picked up from the foil will clearly find it easier to adapt its velocity to an orbit with high value of l, in accord with our experimental findings.

V. RELATIVE LEVEL POPULATIONS AND DECAY CURVES

There has been a great concern whether cascade processes can account for the long tails observed in a number of beam-foil decay curves, and also whether the large discrepancies observed in some cases between theoretical and experimental lifetimes can be explained from insufficient cascade corrections.[8,9,23]

There have been some attempts to estimate amounts of repopulations caused from cascade processes.[8,9,14,24,25] In all cases except one,[8] various assumptions concerning the initial level populations were made, because of lack of experimental data, and population models in fair agreement with experimental findings for neutral or weakly ionized projectiles have been applied to highly ionized projectiles like Kr VII (Ref. 24) and Kr VIII (Ref. 9). Of the articles published, we shall concentrate here on those[9,14] which simulated the decay of the $4s\,^2S$-$4p\,^2P$ transition in Kr VIII.

Younger and Wiese[9] observed a good agreement between the experimental and the synthesized beam-foil decay curves for the above-mentioned transition when using a relative level population proportional to $(2l+1)(n^*)^{-3}$, where n^* is the effective quantum number of the excited term. However, since the total level population summed over all excited terms diverges when such a population model is applied, they had to truncate at a chosen maximum value of n. Later, Livingston *et al.*[14] remeasured as well as remodeled the decay of the $4s\,^2S$-$4p\,^2P$ transition in Kr VIII. They found a good agreement by using a relative level population proportional to $(2l+1)(n^*)^{-2}$. They concluded that the decay curve measured by them contained a tail which could not be explained by cascades from levels populated according to a distribution of type $(2l+1)(n^*)^{-p}$, with p greater than the limiting value for normalizable populations.[14] The reason that they reached such a negative conclusion is obvious from Figs. 1 and 2. It is clearly impossible to cast our experimentally determined relative level populations into simple, analytical expressions. Also, we must repeat here from Sec. IV that for Ar VIII we find two excitation mechanisms active. Therefore, the magnitudes of the cascade repopulations of the $3p$ level in Ar VIII will be projectile energy dependent. Our experimental results indicate that one cannot expect to obtain reliable, synthesized beam-foil decay curves on the basis of simple, analytical level population models as those given by $(2l+1)(n^*)^{-p}$. Such models give neither a good l dependence nor a reasonable n^* dependence. Whereas Livingston *et al.*[14] mention that the experimental situation regarding the appropriate population model to be used is not clear at present, we conclude that experimentally determined level populations must at present be used when modeling beam-foil decay curves.

The case of Kr VIII is not the only one in which the existence of long tails is observed, which can only be explained in terms of bound-state cascades through inclusion of a very heavy population of high-lying states.[26,27] We shall not give here a detailed calculation of cascade repopulations for the $3p$ level in Ar VIII, but only mention that the $nd\,^2D$ levels in Ar VIII with $n \simeq 9$–12 have theoretical lifetimes[12] which come very close to the lifetime of the $3p$ level,[12] and the $3p$-nd branching ratios are around 60% for these d levels.[12] They will clearly repopulate the $3p$ level noticeably through cataract processes. Such cataract decays have to be included in a detailed ANDC decay-curve analysis.[28] The $ns\,^2S$ levels with $n \leqslant 11$ have theoretical lifetimes shorter than those of the $3p$ level,[12] and also, their branching ratios to the $3p$ level are quite large[12] (30–60%), but, as seen from Fig. 1, their populations are of the order of 1% of that of the $3p$ term, so their cascade contributions to the $3p$ level will be small.

VI. COMPARISON WITH ION-ATOM COLLISIONS

Several theoretical articles on multiply charged ion-atom collisions have predicted that electron pickup processes will take place predominantly in excited states and not in the ground state, due to near resonance between the initial and final states.[5] There have been some attempts to observe such an effect,[6] but such measurements are rather difficult, and the results are still sparse and uncertain.[6] This is, among other things, because accelerators available cannot produce intense beams of highly charged projectiles at the desired projectile energies. The situation is much more tractable with beam-foil excitation. The projectile flux can be high enough to ensure favorable signal-to-noise ratios, and, as evident from Fig. 1, fairly complete data sets can be obtained (we note parenthetically that already the poor beam-foil data set presented in Ref. 1 surpasses the best data set from ion-atom collisions[6] in completeness). Also, by a suitable choice of projectile like argon, cascade processes can either be ignored or accounted for in a beam-foil excitation mechanism study. On the contrary, in ion-atom collisions a proper cascade correction is very difficult or virtually impossible to carry out, because contributions from a great number of the many cascades occurring cannot be determined.

We want to emphasize that what we have observed and are presenting here is essentially what a great number of theoreticians have been working on, namely, near-resonance electron pickup processes.[5] Admittedly, we have not performed ion-atom collisions with highly charged ions impinging on an atomic hydrogen target, as most of the theoretical works are concerned with, but we do in our beam-foil study observe the essence of the theoretical predictions, that electron pickup by multiply charged ions takes place preferentially to those excited levels which are close in binding energy to the initial state of the electron.

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