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K-shell vacancy production by direct Coulomb ionization in 47-MeV $Ca^{17+}+Ar$ collisions

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Calculations employing the independent-electron approximation suggest that direct Coulomb ionization is the dominant mechanism for Ar *K*-vacancy production by 47-MeV Ca^{17+}, rather than *K*-*K* vacancy sharing as proposed by Schlachter *et al.* [J. Phys. B **21**, L291 (1988)]. Using electron-capture probabilities for zero impact parameter given by the classical-trajectory Monte Carlo model, good agreement with the experimental charge-state distribution for multielectron capture in coincidence with Ar and Ca *K* x rays is achieved.

The *K*-shell vacancy production mechanism in atomic collisions has been investigated intensively over the past 20 years. Two main theories are used to describe the vacancy production mechanism in two different velocity and colliding-partner symmetry regime. At low velocities where the projectile velocity v is a lot less than the mean velocity of a *K*-shell electron v_k, the molecular-orbital (MO) model is successfully used for describing experimental results.[1] In particular, if the collision partners have similar atomic numbers ($z_1 \simeq z_2$), some mechanisms like electron promotion and vacancy sharing can be calculated using the MO model.[1] At high velocities where $v \geq v_k$ and $z_1 \ll z_2$ the direct Coulomb ionization (DI) is the dominant process and it can be modeled by several theoretical approaches such as plane-wave Born approximation, semiclassical approximation, binary-encounter approximation, and others.[2] In intermediate cases where $z_1 \simeq z_2$ and v is in the order of v_k usage of the above models is questionable and some conflicting experimental results exist.[1] It was shown by Hansteen,[3] following experimental results and calculations by Woods *et al.*,[4] that in the collision of $F^{q+}+Ne$, the MO mechanism dominates when v/v_k is smaller than 0.36, while above this ratio the DI mechanism is dominant. It is expected that for higher z collisions the DI will be dominant, even at a smaller velocity ratio. Indeed, it was shown by Maor[5] that even in the Cu+Fe system at 28 MeV Cu energy where $v/v_k \simeq 0.15$, about 25% of the *K*-shell vacancy production is due to direct ionization. If the "heavy" colliding partner in a symmetric collision has vacancies in its $2p$ orbital, the probability for electron promotion in the MO model is increased as the number of such incoming vacancies is increased and much experimental evidence for this have been shown.[1] However, this increased probability cannot be considered as a proof for the applicability of the MO model, since similar behavior was also found for very asymmetric colliding partners, indicating a similar vacancy production mechanism, i.e., direct Coulomb excitation.[6,7]

In a recent paper, Schlachter *et al.*[8] have reported experimental results for multiple electron capture by 47-MeV Ca^{17+} incident on Ar in coincidence with Ar and Ca *K* x ray emission. Additional data are given in a later publication by Schlachter *et al.*[9,10] In these reports it was shown that the inner-shell vacancy production process is independent of the outer-shell electron capture process. The Ca charge state displayed a binomial distribution, which is a signature of independent electron behavior in the capture process as well. The *K* x-ray (or *K*-vacancy) production mechanism was suggested to be a MO electron promotion followed by *K*-*K* vacancy sharing between the Ca and Ar.

The important finding of Schlachter *et al.* on the independence between the inner-shell vacancy production and the outer-shell capture process enables the usage of independent-electron-approximation (IEA) formalism for estimating the *K*-vacancy production mechanism. In the present Brief Report the IEA has been employed to predict the Ca charge-state distribution expected for *K*-vacancy production.

The cross section for the capture of n out of N independent electrons can be written:

$$\sigma_{n\ \text{capture}} = 2\pi \begin{bmatrix} N \\ n \end{bmatrix} \int_0^\infty p(b)^n [1-p(b)-p_i(b)]^{N-n} b\, db \ , \tag{1}$$

where $p(b)$ is the probability for one-electron capture as a function of the impact parameter b, $p_i(b)$ is the probability for ionization, and $\binom{N}{n}$ is the binomial coefficient. If $p_i(b)$ is a lot smaller than $p(b)$ (Ref. 11) then

$$\sigma_{n\ \text{capture}} = 2\pi \begin{bmatrix} N \\ n \end{bmatrix} \int_0^\infty p(b)^n [1-p(b)]^{N-n} b\, db \ . \tag{1a}$$

For measurements performed in coincidence with *K*-vacancy production (i.e., *K* x-ray emission), the cross section may be expressed using the approximation that $p(b) \simeq p(0)$ in the impact-parameter range where *K*-vacancy production occurs:

$$\sigma_{x+n\ \text{capture}} \simeq 2\pi \begin{bmatrix} N \\ n \end{bmatrix} p(0)^n [1-p(0)]^{N-n} \int_0^\infty P(b) b\, db \ . \tag{2}$$

In the above equation, $P(b)$ is the probability of K-vacancy production.

By fitting the experimental total capture cross sections (measured without x-ray coincidence) to Eq. (1a), with $p(b)$ represented by the empirical function $p(b) = p(0)\exp(-b/r)$ (see Heber *et al.*[11]), values of the parameter $p(0)$ and r were obtained. The best-fitted values are $p(0)=0.6$ for eight electrons in the Ar L shell (the most probable shell to capture from) and $r \simeq 0.3$ Å with a total error of about 20% between the best fit and the experimental results. Schlachter *et al.*[8] used $p(0)=0.41$ along with eight electrons or $p(0)=0.45$ with seven independent electrons (i.e., the number of L vacancies in Ca^{17+}) to represent the experimental charge-state distributions. However, it can be argued that the capture process is not necessarily limited by the number of vacancies in the Ca L-shell, since electrons can be captured to many other states, including Rydberg and metastable excited states. Moreover, in the case of the MO mechanism for Ar K x-ray production, the Ca ion captures an electron into its $2p$ subshell and therefore only six "independent holes" are left. According to the IEA formalism[12] the relevant number of electrons to be considered here is the number of Ar L-shell electrons only. The best fit to the experimental capture cross sections using $p(0)=0.41$ leads to discrepancies of about a factor of 2 for some experimental points. In Fig. 1 the fitted curves are shown. The upper solid curve is fitted by using the exponential function with $p(0)=0.6$ along with eight electrons, the dashed curve is the same with $p(0)=0.41$, while the dotted curve is for $p(0)=0.45$ and seven independent electrons (the best fit for each curve is with slightly different values of r). The lower solid curve is the Ca charge-state distribution in coincidence with the Ar x-ray production, calculated using Eq. (2). It can be seen from Fig. 1 that the Ca charge-state distribution is better presented by the Ar data; therefore the Ca K x-ray data should be shifted about one charge state to the left in order to follow Eq. (2) and the upper fitted curve. This finding suggests that an additional mechanism is involved in the K-vacancy production that can shift the Ca charge-state distribution by about one charge state compare to the MO prediction alone.

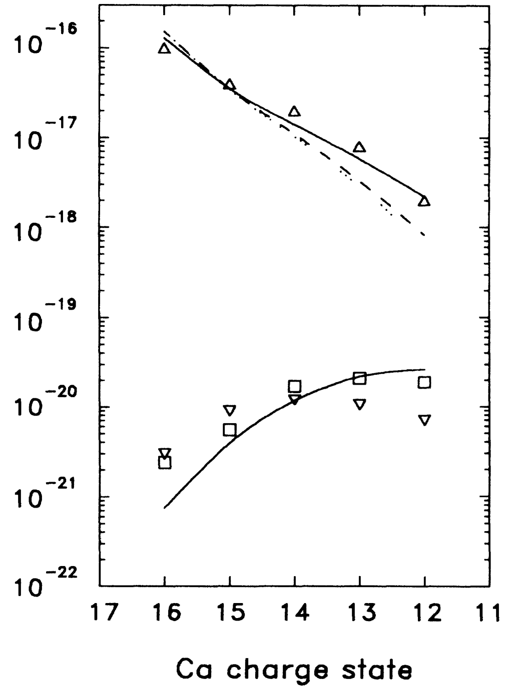

FIG. 1. Comparison between the experimental data from Ref. 1 and fitted curves of exponential probabilities and the IEA formalism (see text). The upper solid curve is the best fit to the experimental data resulting in $p(0)=0.6$. The dashed curve is the same with $p(0)=0.41$ and the dotted curve is a fit with $p(0)=0.45$ with seven independent electrons. The lower solid curve is the calculated Ca charge-state distribution using the best-fit parameters from the upper curve.

Another problem in the original work concerns the probability w for K-vacancy sharing. The measured w is about 0.29 (for the single- and double-electron capture by the Ca) when using neutral-atom fluorescence yields or 0.26 when using fluorescence yields corrected for charge state,[13] which the authors compare to the value of 0.3 calculated by Meyerhof[14] using the neutral-atom binding energy. In such a highly charged ion the change in the binding energy can affect the vacancy sharing compared to the solid target or the neutral atom (see, for example, Ref. 15); therefore a corrected value is considered here. If the corrected binding energy for the Ca^{17+} K-shell electron is taken into account (estimated from the Dirac-Fock calculation to be 5.027 keV), then $w=0.14$, which is considerably smaller than the measured value.

In view of the above difficulties, an additional mechanism is proposed here to be the dominant process for the x-ray production in this collision system; namely, the direct Coulomb ionization mechanism. The MO and DI mechanisms are distinctly different processes, since they lead to different final states when the capture of electrons from the Ar outer shell is treated as an independent process [Eq. (2)]. For example, in the case of Ca^{17+} K-vacancy production, the DI will produce Ca^{18+}, while the MO will produce Ca^{17+} in an excited state because the K vacancy results from electron promotion. In the case of Ar K-vacancy production, the DI will leave the Ca in the 17+ charge state but the MO requires the transfer of an Ar K electron to the Ca L shell producing Ca^{16+}. The total x ray production cross section for the two cases is given by

$$\sigma_x(\mathrm{Ca}) = [w\sigma_{\mathrm{MO}} + \sigma_{\mathrm{DI}}(\mathrm{Ca})]\omega(\mathrm{Ca}) , \tag{3a}$$

$$\sigma_x(\mathrm{Ar}) = [(1-w)\sigma_{\mathrm{MO}} + \sigma_{\mathrm{DI}}(\mathrm{Ar})]\omega(\mathrm{Ar}) , \tag{3b}$$

where ω is the K-shell fluorescence yield and σ_{DI} is the K-shell direct ionization cross section. Reliable estimates of the DI cross sections are not readily attainable since perturbation theory is not valid in this velocity regime, and also the K-shell binding energies change during the collision. Nevertheless, scaling laws given by the binary-encounter approximation[16] and perturbed-state-state theory with energy-loss, Coulomb deflection, and relativistic corrections[17] yield 7×10^{-19} and 4×10^{-19} cm^2, respectively, which are similar to the experimental value for the Ar K-vacancy production cross section of 8×10^{-19} cm^2.

Using the IEA formalism [Eq. (2)] only the probability $p(0)$ for capturing from the Ar L shell by the Ca is needed to reconstruct the Ca charge-state distribution after the collision. In order to approximate $p(0)$ in an additional way, the classical trajectory Monte Carlo (CTMC) method was used. The Ar L-shell effective charge, as well as the method of calculation, was taken exactly as in Ref. 18. The results of using the CTMC code with Ca^{16+}, Ca^{17+}, and Ca^{18+} as projectiles (Ca^{16+} is the projectile when Ar K-shell vacancy production is done by MO electron transfer; Ca^{17+} is the projectile when it is done by DI; and Ca^{18+}, when Ca K-shell vacancy production is done by the DI mechanism) are 0.436, 0.49, and 0.528, respectively. The probability dependence on small impact parameter was checked and it was found to be constant below $b=0.02$ a.u., within 0.5% error; and below 0.25 a.u., within 4% error.

Solving Eqs. (3a) and (3b) for the MO and DI cross sections using the experimental x-ray cross sections, a fitting to the Ca charge state, the Ar and Ca fluorescence yields (corrected for ionic charge in the case of Ca), and $w=0.14$ gives $\sigma_{\rm MO}=2.14\times10^{-19}$ cm^2, $\sigma_{\rm DI}({\rm Ca})=2.7\times10^{-19}$ cm^2, and $\sigma_{\rm DI}({\rm Ar})=6.5\times10^{-19}$ cm^2. Therefore, these estimates indicate that the K-vacancy production by the MO mechanism is only about 10% of the total for Ca and 22% for the total for Ar. Using these cross sections and the CTMC values, the Ca charge-state distribution, in coincidence with the Ar x-ray production, was calculated with Eqs. (3a) and (3b). The charge-state distributions are compared with the experimental distributions in Figs. 2 and 3. The agreement between the calculated and experimental results is found to be quite good. The addition of the DI mechanism causes a shift of the charge-state distribution from that predicted only on the basis of the MO mechanism. For example, Ca K x-ray production by DI leading to Ca^{16+} as an end product involves two-electron capture, while the production of Ca^{16+} by the MO mechanism involves one-electron capture.

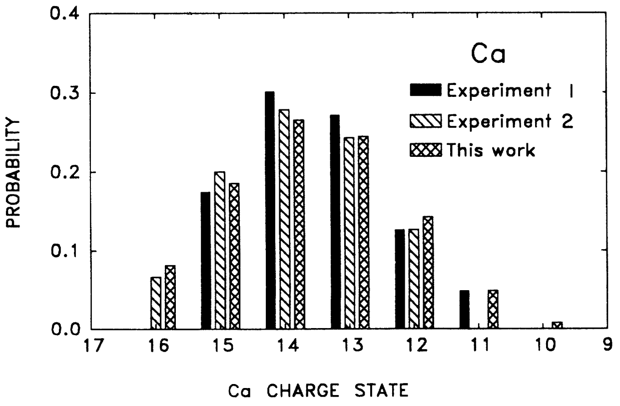

FIG. 2. Comparison of the calculated and experimental charge-state distributions for Ca in coincidence with the Ca K x ray. Experiment 1 shows data from Ref. 9, and experiment 2, from Ref. 8.

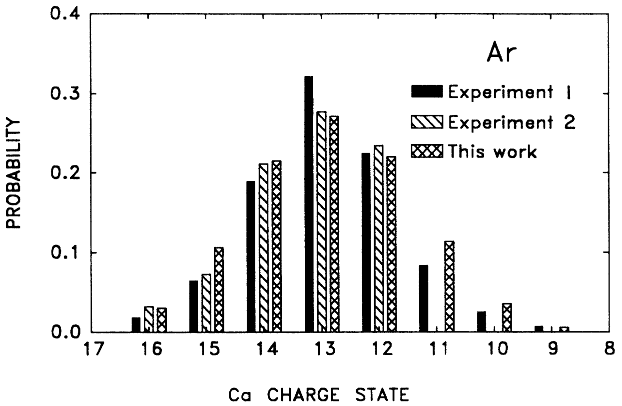

FIG. 3. Comparison of the calculated and experimental charge state distributions for Ca in coincidence with Ar K x rays. Experiments 1 and 2 as in Fig. 2.

A similar calculation using neutral-atom fluorescence yields, vacancy sharing ratio ($w=0.3$) with the CTMC values, and Eqs. (3a) and (3b) results in the same 10% contribution to the total cross section of the Ca by MO, and only 11% of the Ar K x ray is produced by the MO mechanism. This time the distribution in Fig. 3 is not as good as with the ionic corrections. This finding supports the DI dominance in the collision and the need for ionic corrections. Refined calculations of the exact fluorescence yields and vacancy sharing ratio, as well as the probability $p(0)$ estimation, should be a good subject for further investigation.

In Fig. 4 of Ref. 9, the ratio $\sigma_k({\rm Ca})/[\sigma_k({\rm Ca})+\sigma_k({\rm Ar})]$ (where σ_k is the K-vacancy production cross section) is shown as a function of the number of captured electrons. When only the MO mechanism is important, this ratio reduces to the K-K vacancy sharing probabili-

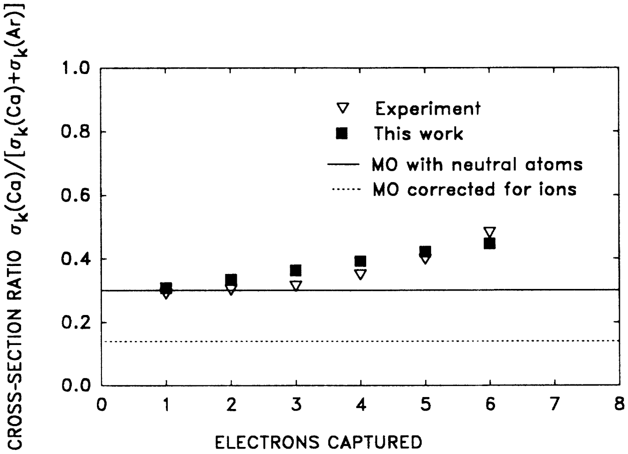

FIG. 4. The experimental (▽) and the calculated (■) ratios $\sigma_k({\rm Ca})/[\sigma_k({\rm Ca})+\sigma_k({\rm Ar})]$ as a function of the number of electrons captured by the Ca ion. The Ar data were shifted by one charge state, as in Ref. 9. The ratios predicted by the MO mechanism alone are shown by the solid line (with $w=0.3$) and the dotted line (with $w=0.14$).

ty, which should be independent of the number of captured electrons. In Fig. 4 it is shown that, instead of remaining constant, the experimental ratio slowly increases as the number of captured electron increases; any fluorescence yield change, as a function of the captured electrons, would shift the ratio to the opposite way. Moreover, if the corrected K-binding energy is taken into account, the cross-section ratio can be 0.14 rather than 0.3. By including the DI mechanism in the calculation of the cross-section ratio, good agreement with the experimental values is achieved (see Fig. 4).

In summary, the importance of direct Coulomb ionization in 47-MeV Ca^{17+} on Ar collisions has been postulated. It appears that the MO mechanism contributes only weakly to the production of K-shell vacancies in this nearly symmetric collision system. The effect of the ionic properties of the collision partners encourages further work. An experiment where the Ar recoil-ion charge-state distribution is also measured could give additional information about the vacancy production mechanisms in such collisions.

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