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Anomalous Target Z Dependence of Double to Single K-Shell Vacancy Production in Cl-Beam X Rays*

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We have measured X rays from 30-MeV Cl beams on various solid targets between carbon and copper. The ratio of double K-shell to single K-shell vacancy production in Cl exhibits a broad peak near $Z = 20$ (Ca). This behavior is not predicted by Coulomb ionization. This peaking is examined using a molecular-orbital mechansim. To explain the results both K-shell electrons must be promoted to the vacant orbitals of the Cl beam. This is the first evidence of double K-shell promotion.

Various groups have seen peaked Z dependence in the production of L-shell vacancies. In 1965 Specht[1] observed target Z dependence in the study of target x rays produced when fission fragments impinged on solid targets. In 1970 Kavanagh *et al.*[2] observed a similar dependence in the Cu L x rays produced when Cu targets were bombarded with various ions as well as when various solid targets were bombarded with Cu ions. In 1971 Saris[3] observed a similar Z dependence when gaseous Ar was bombarded with various ions. All these data exhibit maxima and minima as a function of either target or projectile Z. The maxima occur when the binding energy of the L electron removed is slightly smaller than that of the K, L, or M shell electrons in the conjugate atom. This has been referred to as a level-matching effect, and has been qualitatively understood in terms of the Pauli interaction causing electron promotion to higher molecular orbitals.[4-6]

A similar effect is observed here for Cl K x rays. A 30-MeV Cl beam was used to bombard C, Mg, Al, Ca, Sc, Ti, Fe, Ni, and Cu; the ratio of the Cl double K-shell vacancy production to the Cl single K-shell vacancy production was found to maximize near $Z = 20$.

The experiment consisted of bombarding thick targets with a Cl beam from the Kansas State University tandem Van de Graaff accelerator and observing the resulting Cl $K\alpha$ x rays with a curved-crystal vacuum spectrometer using an ammonium dihydrogen phosphate crystal. With this choice of crystal it was possible to separate the Cl $K\alpha$ x rays associated with double K-shell vacancies from those associated with single K-shell vacancies and retain enough intensity to make the experiment possible on several targets in a reasonable time period. Because only the ratio of the two intensities is used, difficulties due to beam integration in the different targets are avoided.

Figure 1 is a plot of the ratio of the number of Cl $K\alpha$ x rays arising from double K-shell vacancy production to the number of Cl $K\alpha$ x rays arising from single K-shell vacancy production as a function of the target Z. The data presented include thick-target corrections.[7] With the assumption that a single process does not fill both K-shell vacancies, an initial configuration with two holes in the K shell will result in two events. The ratio of the intensities, R, can be related to sin-

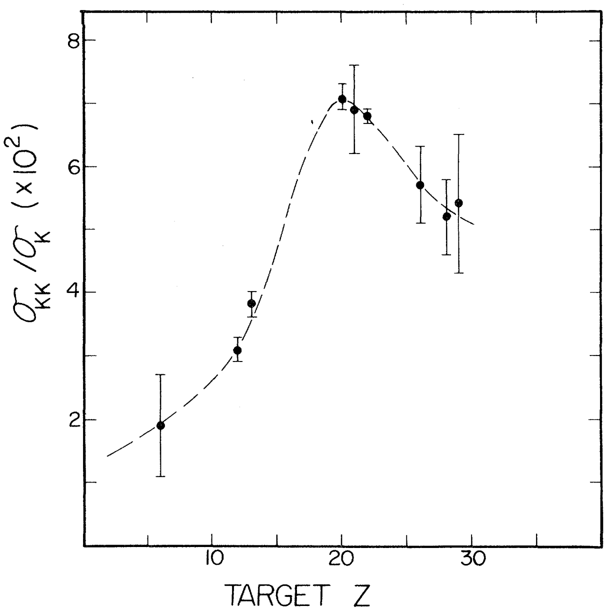

FIG. 1. Ratio of Cl double K-shell vacancy production to single K-shell vacancy production for 30-MeV ^{35}Cl beams as a function of target Z.

gle and double K-shell vacancy production cross sections, σ_K and σ_{KK}, respectively, by

$$R = \omega_2\sigma_{KK}/(\omega_1\sigma_K + \omega_1'\sigma_{KK}),$$

where ω_1 and ω_2 are the respective $K\alpha$ fluorescence yields for the single and double K-shell vacancy configurations. The quantity ω_1' is the fluorescence yield for atomic configurations with one K-shell vacancy resulting from de-excitation of configurations with two K-shell vacancies. The fluorescence yields are not well known; however, calculations by Bhalla and Hein[8] indicate, for Ne, that the ratio of fluorescence yields, ω_2/ω_1, is near unity if the number of L-shell vacancies is the same. We assume this is the case for Cl. In addition, it is reasonable to neglect the term $\omega_1'\sigma_{KK}$ in the denominator as the measured values of R are at most about 7%. The value of R is thus a close approximation to the ratio of double K-shell to single K-shell vacancy production cross sections at 30-MeV bombarding energy. In the binary-encounter approximation this ratio increases approximately as Z^2, where Z is the bare nuclear charge of the target.[9] In sharp contrast the observed ratio decreases as the target Z is increased from 20 to 29. The observed maximum in the ratio of double to single ionization probabilities occurs near a target Z value of 20.

Two groups have recently observed similar oscillatory behavior for the single K-shell vacancy production as a function of target Z. Kubo, Jundt, and Purser[10] studied total K-shell ionization of Br and Ni beams by solid targets, whereas Winters *et al.*[11] studied total K-shell ionization of Cl beams in gaseous targets. The double K-shell ionization transitions were not energetically resolved from the single K-shell ionization transitions; however, it is reasonable to assume that their measurements reflect the behavior of single K-shell vacancy production. Combining the results of Winters *et al.*[11] with the present results, it can be concluded that both single and double K-shell vacancy production are peaked at a Z slightly greater than the projectile Z.

The maxima in the L x-ray cross sections at the low bombarding energies as reported by Kavanagh *et al.* and Saris are explained using an electron-promotion mechanism. Similar arguments can be applied to the K-shell vacancy production observed with the Cl beam. At 30 MeV, Cl^{+10} and Cl^{+11} are the most probable charge states,[12] which leaves Cl with at least two vacancies in the L shell. This increased binding of the Cl orbitals due to ionization reverses the order of beam and target orbitals as discussed by Der *et al.*[13] The net result is that for certain target Z values, it is possible to promote two Cl K-shell electrons to the L-shell vacancies, which total three for a Cl^{+10} ion. This is illustrated in Fig. 2 using the binding energies of Clementi.[14] For the case of Cl+C the $1s$ orbital of Cl is correlated with the $1s$ orbital of the (V) combined atom, leading to no K-electron promotion. For atoms with $17 < Z < 42$ the neutral Cl $1s$ orbital becomes correlated with the $2p$ orbital of the target atom via the $2p$ orbital of the combined atom. For these cases no promotion can occur since the $2p$ orbital of the target is filled. However, for Cl^{+9} ions the Cl $1s$ orbital becomes correlated with the $2p$ orbital of the Cl ion and promotion can thus occur.[15] The correlation exists in this case due to the increased binding of the $2p$ orbital in the Cl ion, bringing it below the $2p$ orbital of Ca. This situation exists up to $Z = 22$, above which the target $2p$ orbital binding becomes larger than the $2p$ orbital of the Cl^{+9} ion.

This electron-promotion mechanism predicts an additional maximum in the probability of producing K-shell vacancies when the binding energy of the L and M shells of the target are such that the Cl $1s$ and $2p$ orbitals are correlated via the $3d$ orbital in the combined atom. This sec-

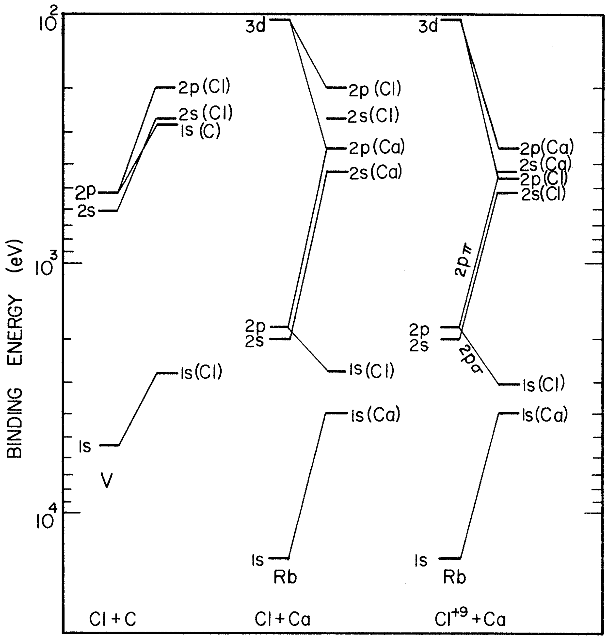

FIG. 2. Correlation diagrams based on conservation of $n-l-1$ and the Pauli exclusion principle. In Cl + C the Cl 1s orbital is correlated to itself, and in Cl + Ca it is correlated to the Ca 2p orbital. Both cases should result in no vacancies in the Cl 1s orbital. For Cl^{+9} + Ca the Cl 1s orbital is correlated to the Cl 2p orbital, and thus can lead to vacancies in the Cl 1s orbital.

ond maximum should occur near target $Z = 49$ (In).

In conclusion, the present measurements have demonstrated a peaked behavior in projectile double K-shell vacancy production similar to that for total K-shell vacancy production as observed in poor-resolution experiments. These results imply that not only the Coulomb ionization mechanism is giving rise to double K-shell vacancies in the Cl projectile. Energy-level matching between orbitals in the target atom and the incident ion is important. Electron promotion via molecular orbitals is one candidate for the level matching mechanism and is suggested as the possible explanation for the observed peaking of the ratio of double to single K-shell vacancy production.

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