

Unexpected Charge-State Dependence of K X Rays Produced in Gaseous Targets by S^{+q} and Ti^{+q} Ions

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K x rays produced in the bombardment of gas jet targets of helium, oxygen, and argon by sulfur and titanium beams have been measured for projectile charge states of +6 to +16 and +8 to +19, respectively. Unexpectedly, the charge-state dependence is similar for nearly symmetric systems and for very asymmetric systems. The results suggest that direct Coulomb excitation into empty bound states could be a common K -shell-vacancy production mechanism.

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It is well known that K -vacancy production cross sections in heavy-ion-atom collisions exhibit a strong dependence on the projectile charge state.¹⁻⁹ For nearly symmetric collisions the common feature is a monotonic increase in the K -vacancy production of both projectile and target atoms with the number of L -shell vacancies present in the incident beam. The cross sections are charge-state independent as long as the L shell of the projectile is filled. These experimental results have been interpreted in terms of the molecular-orbital (MO) model.¹⁰ The sum of projectile and target K -shell ionization is assumed to be a measure of the total $2p\pi$ - $2p\sigma$ rotational coupling mechanisms. Direct vacancy formation in the $1s\sigma$ and the $2p\sigma$ MO is neglected. The similar behavior of projectile and target K x-ray production is explained by $2p\sigma$ - $1s\sigma$ radial coupling, more commonly known as K -shell vacancy sharing.¹¹

In the present work, we have extended, for the first time, measurements of the charge-state dependence of K x rays to both nearly symmetric and very asymmetric collisions. The interest is to search for differences in the charge-state dependence of the x-ray production to see if different reaction mechanisms operate in the two cases. In very asymmetric collisions, the K -vacancy sharing is negligible, and therefore vacancies in the K shell of the higher- Z collision partner can only be produced by direct Coulomb excitation. We have observed, however, that the results are very similar for both symmetric and very asymmetric collisions; hence this type of experiment alone cannot be considered as confirmation of the $2p\pi$ - $2p\sigma$ rotational-coupling model.

Measurements were made with beams from the Brookhaven National Laboratory Tandem Van de Graaff Facility of 95-MeV Ti^{+q} ($q=8-19$) on Ar and O_2 , and 70-MeV S^{+q} ($q=6-16$) on Ar, O_2 , and He. The high charge states were obtained using a post-acceleration stripping foil before the switching magnet. The target was a gas jet emitted by a 0.3-mm hypodermic needle with the beam passing 1 mm from the tip. The pressure was below 3×10^{-4} Torr at a distance of 5 cm from the needle and in the range of 10^{-5} - 10^{-7} Torr in the beam line. Under these conditions, single-collision production conditions for the K shell were ensured (this was checked by measurements as a function of pressure). We estimate that less than 1% of the beam underwent charge-exchange collisions between the switching magnet and the gas

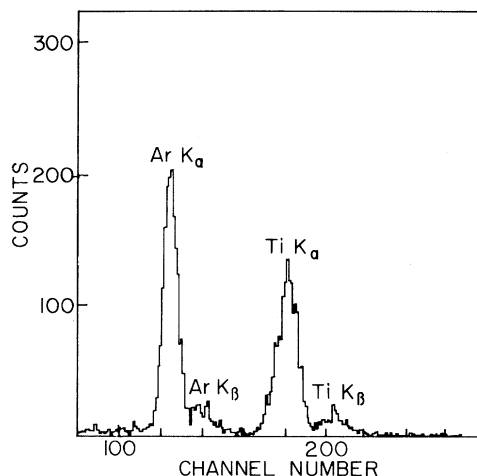


FIG. 1. Typical x-ray spectrum observed for the collision of 95-MeV Ti^{+8} projectiles with Ar.

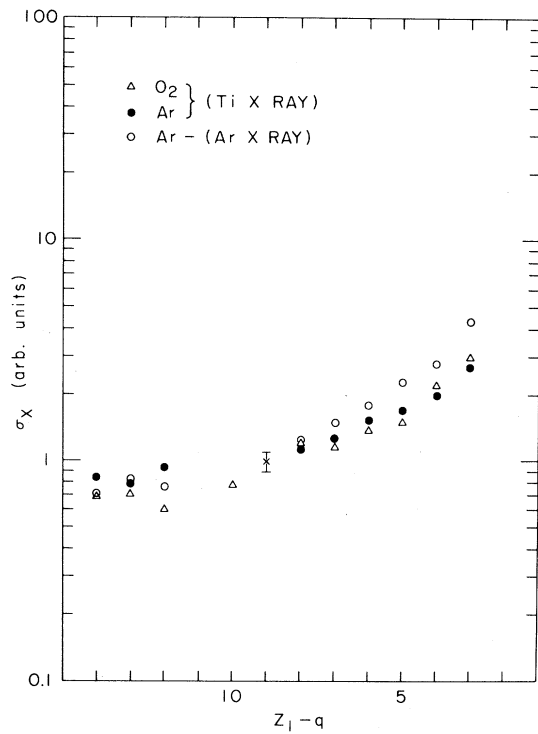


FIG. 2. Cross sections for K x rays produced by 95-MeV Ti^{+q} incident on gas targets of O_2 and Ar. The data have been arbitrarily normalized at a projectile charge of $Z_1 - q = 9$ where Z_1 is the atomic number of the projectile. The x ray observed is indicated in the figure.

target chamber. The elastically scattered projectiles were monitored with an annular silicon surface-barrier detector which subtended a laboratory scattering angle of 7° . A Si(Li) x-ray detector placed at 90° to the beam detected x rays produced in the collisions. A typical pulse-height spectrum for the 95-MeV Ti^{+8} on Ar system is shown in Fig. 1.

Relative x-ray yields are presented in Fig. 2 for the titanium projectiles and in Fig. 3 for the sulphur projectiles as a function of the number of electrons, $Z_1 - q$, for the incident projectile. The data have been arbitrarily normalized at $Z_1 - q = 9$ for the various target gases.

The charge-state dependence of the x-ray cross sections shown in Figs. 2 and 3 results from the combination of two effects. The first is a dependence of the reaction mechanism on the projectile charge state, that is, the number of electrons brought *into* the collision, and the other is the possible variation of the fluorescence yields of the post-collision projectile and target atoms. It is important to realize that the relevant charge

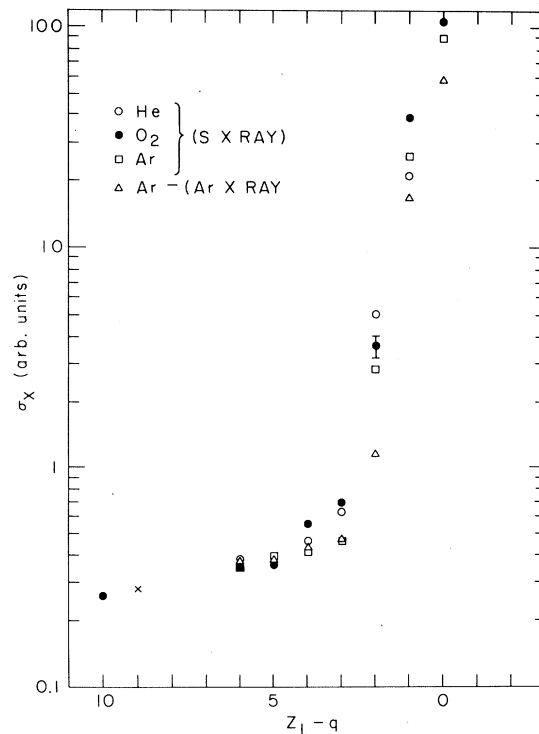


FIG. 3. Same as Fig. 2 for 70-MeV S^{+q} incident on gas targets of He, O_2 , and Ar.

states for the target and projectile x-ray production are those *following* the collision. A fluorescence yield, $\omega_k^{(q)}$, for an incident ion with one K -shell and q outer-shell vacancies is *not* appropriate for converting from measured x-ray production to vacancy-production cross sections. The relevant fluorescence yield for both projectile and target atoms is the final-state mean fluorescence yield, $\bar{\omega}_k$, which is averaged over the post-collision charge-state distributions.

In the present experiment, the final charge-state distributions were not measured, and we cannot, therefore, directly determine $\bar{\omega}_k$. Previous work, however, indicates that $\bar{\omega}_k$ is *not* a strong function of the incident projectile charge state. For example, calculations^{12,13} show that the maximum relative change in $\omega_k^{(q)}$ for argon due to vacancies in the $2p$, $3s$, and $3p$ subshells is $\sim 40\%$, and no dependence of target atom $\bar{\omega}_k$ was found¹⁴ for variation of projectile charge state from $q = 5-9$ for 30-MeV F^{+q} on Ar. Similar measurements¹⁵ for O^{+q} and Cl^{+q} on Ne show that there is only a small variation in $\bar{\omega}_k$ for the neon target, whereas large changes in $\omega_k^{(q)}$ have been measured.¹⁶ Large changes in the projectile $\omega_k^{(q)}$ were reported by Doyle *et al.*¹⁷ for Si^{+q}

on He and by Tanis and Shafroth¹⁸ for Cl^{+q} on Cu. Rather than ask whether the projectile $\bar{\omega}_k$ changes as a function of incident charge state, we can equivalently ask whether there is a change in the post-collision charge-state distributions. This question has been addressed by Rosner and Gur¹⁹ who found that under single-collision conditions, the final charge-state distributions for 10–30-MeV O^{+q} ($q=4-7$) on Ne, Ar, Kr, and Xe were insensitive to the incident q .

If the electronic configurations of the emitting atoms were sensitive to the incident q of the projectile, there should be detectable x-ray energy shifts. In the present experiment, as observed in Ref. 6 for S on Ar, we found that for S on Ar, as well as for S on O and S on He, the centroid peaks of the x-ray lines for both target and projectile *do not shift* with projectile charge state (except for $q=16$). This implies that the average electronic configurations after the collision and hence the post-collision effective fluorescence yields, $\bar{\omega}_k$, are nearly independent of charge state. Since we observed no difference in the x-ray energy shifts for asymmetric and nearly symmetric systems, we conclude that changes in $\bar{\omega}_k$ with the projectile charge state are small and are therefore unlikely to explain the general behavior of the results presented in Figs. 2 and 3. These cross sections should then be mainly determined by the reaction mechanism responsible for x-ray production and *not* by variations in the mean fluorescence yield, $\bar{\omega}_k$.

The data shown in Fig. 2 for the Ti-Ar system are typical of previous results for nearly symmetric systems. In particular, the results for the heavier system Cu-Kr,⁸ where the fluorescence yield plays a much smaller role, are very similar to those presented here. The K x-ray production cross sections for both Ti and Ar are constant as long as $Z_1 - q \geq 10$, that is, the Ti L shell is full. When $Z_1 - q < 10$, vacancies in the Ti L shell (i.e., holes in the $2p\pi$ MO) are brought into the collision, and an increase in the cross section proportional to the number of these vacancies is observed. We find that the sharing of vacancies between Ti and Ar K shells is nearly independent of charge state and in agreement (within 25%, with use of static fluorescence yields for Ti and Ar) with the Meyerhof charge-sharing formalism.¹¹ The dependence on the number of $2p\pi$ vacancies is just that expected from the rotational-coupling model and has been previously claimed as a confirmation of the applicability of the model.⁷⁻⁹ However, the results for the very

asymmetric system, Ti-O₂, shown in Fig. 2 are essentially identical to the results for Ti-Ar. The large difference in the binding energies of the K shells of Ti and O atoms implies that there will be negligible vacancy sharing, thus the production of Ti K x rays must result from a direct process. In this case a strong dependence on the projectile charge state is not expected, and the experimental results are therefore very surprising.

Similar results are observed for the S^{+q} projectiles incident on targets of Ar, O₂, and He (see Fig. 3). In all three systems the x-ray production cross sections increase with the number of L vacancies in the incident beam. Earlier work⁷ at lower energies for S-Ar collisions showed the same behavior. In the MO picture, $2p$ vacancies in the neutral S ions correlate to the $3d\pi$ orbital and can be transferred to the $2p\pi$ orbital by radial coupling.²⁰ However, for the range of charge states investigated here, the binding energy of the S $2p$ electrons shifts²¹ by over 200 eV from the neutral atom value and hence are more tightly bound than the Ar $2p$ electrons. Vacancies in the S $2p$ shell therefore correspond to vacancies in the $2p\pi$ MO even though S is lighter than Ar. The sharp increase in the x-ray production cross section as the K shell begins to empty (K vacancies can be present in the incident beam for S projectiles with $Z_1 - q = 2$ because of metastable states²²) has been observed in several works^{23,24} and is attributed to electron capture processes.

It should be noted that the relevant adiabatic condition to justify the applicability of the molecular-orbital model (namely $v_1/v_{2p} \ll 1$, where v_1 and v_{2p} are the velocities of the projectile and $2p$ electrons, respectively) is not met for the collision systems studied here, as well as those studied in Refs. 7–9. These experiments, therefore, can neither confirm nor refute the predictions of the rotational-coupling model.

The great similarity of the asymmetric and symmetric data presented here (including also the data of Refs. 7–9) suggests a *common mechanism* for the K -vacancy production in both cases. A plausible explanation would be the direct excitation of K electrons into empty bound states, essentially into $2p$ states as suggested by the results shown in Figs. 2 and 3. Recent experimental²⁵ and theoretical²⁶ works have pointed out the importance of such excitation processes. Experiments on the energy and charge-state dependence of the absolute cross sections should be of particular importance to confirm this explanation and

to understand the K -vacancy production mechanism in energetic heavy-ion-atom collisions. Direct measurements with electron and x-ray spectrometer of the q dependence of fluorescence yields of the final ions are also needed. On the theoretical side, formulation of an approach which incorporates energy dependence of the violent and complex many-body nature of the collision seems to be a *sine qua non* for further progress.

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