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Projectile energy dependence of *L* X-ray emission in collisions of Xe^{23+} with In target: role of Coulomb ionization and quasi-molecular effects

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Letter to the Editor

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Abstract

The X-ray emissions in the interaction of 3–6 MeV Xe^{23+} ions into thick solid In target are measured. The projectile-to-target and target L_α/L_β X-ray production intensity ratios are observed to strongly depend on the projectile energy. The dependence deviates from Coulomb ionization predictions, which implies the important roles of coupling between sub-shells and the activation of $4f\sigma$ rotational couplings for projectile energy larger than 5 MeV.

The study of X-ray emissions induced from highly charged ion-atom collisions attracts continuous attentions (Kavanagh *et al.*, 1970; Datz *et al.*, 1971; Lutz *et al.*, 1972; Anholt *et al.*, 1984; Schonfeldt *et al.*, 1985, 1986; Watson *et al.*, 1999, 2006, 2008; Horvat *et al.*, 2009; Ren *et al.*, 2015), since it is one of the most efficient ways to probe the inner-shell ionization/excitation mechanisms and helps to provide basic data for various fields such as atomic physics (Mokler and Folkmann, 1978), plasma physics (Bitter *et al.*, 1993; Hill *et al.*, 1999; Chen *et al.*, 2002), and astrophysics. It has been found that for collisions either in a high energy region ($v_p \gg v_e$, where v_p and v_e are velocities of projectile and involved target orbit electrons, respectively) or $Z_p \ll Z_t$ (where Z_p and Z_t are atomic number of projectile and target, respectively), the Coulomb ionization dominates the vacancy production process (Garcia *et al.*, 1973; McGuire and Richard, 1973). In the past decades, numerous theoretical formula has been formed to calculate the ionization cross section, such as classical binary-encounter approximation (BEA) (Gryzinski, 1965), plane-wave Born approximation (PWBA) (Johnson *et al.*, 1979), and ECPSSR (Brandt and Lapicki, 1981), which complements the influence of projectile energy loss, Coulomb deflection, target ionization within the perturbed stationary state, and relativistic corrections to electrons. In an intermediate energy region ($v_p \sim v_e$), molecular effects become important (Barat and Lichten, 1972; Eichler *et al.*, 1976). Namely, when the collision partners approach each other, they do form quasi-molecular orbits, from which the electrons are ionized/excited through direct ionization or rotational coupling. On the outgoing way, the vacancies are shared between the collision partners through radial coupling. The occurrence of molecular effects actually require appropriate incident energies that, on the one hand, are high enough to allow the collision partners approaching each other and, on the other hand, low enough to ensure the interaction timescale for the molecular orbit formation. The dominant role of molecular effects in incident energy range of 0.1–10 MeV/u has been reported in a series of papers (Meyerhof *et al.*, 1976, 1977, 1979; Anholt and Meyerhof, 1977). However, for collisions with lower incident energies, experimental data are limited.

The inner-shell processes involving *L*-shell are more complicated than *K*-shell cases and are currently far to be fully understood. Previous studies show that for highly charged ion-atom collisions in the intermediate energy region, the total *L* X-ray emission intensity of projectile present obvious peak structures *versus* target atomic number, which can be qualitatively explained by molecular effects near the level-matching region (Kavanagh *et al.*, 1970; Kubo *et al.*, 1973; Woods *et al.*, 1973; Meyerhof *et al.*, 1977; Ren *et al.*, 2013). However, the L_α/L_β intensity ratio variation is still not fully interpreted (Datz *et al.*, 1971; Meyerhof *et al.*, 1977; Genz *et al.*, 1979; Saha Amal *et al.*, 1998*a*, 1998*b*), especially its dependence on the incident energy. Meyerhof *et al.* (1977) systematically investigated the L_α/L_β intensity ratio as a function of incident energy and target atomic number in collisions of highly charged ions with various solid targets. In the framework of radial coupling between molecular orbits, the variation in the L_α/L_β intensity ratio is explained by the molecular orbital correlation shift from the BL (Barat and Lichten) to Eichler rule as the asymmetry of the collision system increases. Saha Amal

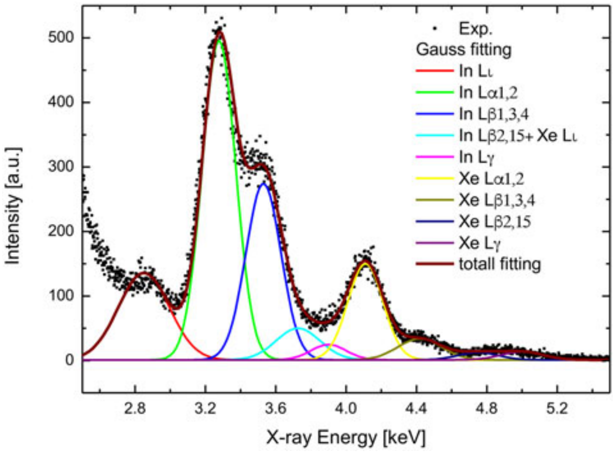

Fig. 1. Typical spectra induced by Xe^{23+} impacting on In target.

et al. (1998*a*, 1998*b*) measured the *L* X-ray emissions in collision of tens of MeV highly charged ions with thin solid target later. Combining the data from Datz *et al.* (1971), the L_α/L_β intensity ratio of iodine ions over a large incident energy range of 6–180 MeV are studied. It is found that as the asymmetry of the collision system increases, the molecular orbital correlation rule shift theories cannot even qualitatively reproduce the experimental results at low impact energies.

To extend the experimental data availability and investigate the inner-shell ionization/excitation mechanism, we measured the X-ray emissions in interaction of highly charged Xe ions with the solid target in energy range of several MeV. Here, we report about the *L* X-ray emissions induced in Xe + In collision systems. The relative intensity of target-to-projectile *L* X rays and the target L_α/L_β intensity ratio as a function of projectile energy are investigated. The results are examined in terms of direct ionization and molecular orbital couplings, whose relative roles are discussed.

The experiment was performed at the 320 kV high-voltage platform at the Institute of Modern Physics (IMP) in Lanzhou. Highly charged ions were generated from electron cyclotron resonance (ECR) ion source and directed by two quadrupole lenses to the target chamber after momentum analysis in a 90° bending magnet. The pressure in the target chamber was kept below 10^{-10} mbar. The highly charged ions impact perpendicularly onto the target. The X rays were detected at 45° with respect to the incident beam line by a silicon drifted detector, which has a resolution of 136 eV full-width at half-maximum at X-ray energy of 5.9 keV. The number of incident ions on the target was monitored temperally with a calibrated transmitting Faraday cup. More experimental details can be found in Zhou *et al.* (2019).

The typical X-ray spectra induced by 6 MeV Xe^{23+} impacting on In target is shown in Figure 1. Xe *L*ι X rays overlap with In $L\beta_{2,15}$ lines and the relative intensities are extracted according to the well-resolved lines. It was reported that in highly charged ion-atom collisions, the *L*ι/*L*α intensity ratio shows no big difference over a large range of target atomic number and changes only slightly with the incident energy. Therefore, here we use the reported data concerning 15–60 MeV iodine ions interacting with various solid target, in which *L*ι/*L*α is about 0.07, to extract the Xe *L*ι X-ray intensity. Consequently, the intensity of In $L\beta_{2,15}$ can be obtained as well.

The X-ray spectra induced by 3–6 MeV Xe^{23+} impacting on In target is shown in Figure 2. The structures of the spectra exhibit strong dependence on the projectile energy. The projectile-to-target *L* X-ray yield ratio is shown in Figure 3a as a function of incident energy. The uncertainty of about 5% mainly comes from the intensity determination from the area of Gauss fitting profiles. It can be seen that in the incident energy range of 3–5 MeV, the projectile-to-target *L* X-ray yield ratios keep nearly constant and are significantly reduced when the incident energy is increased from 5 to 6 MeV.

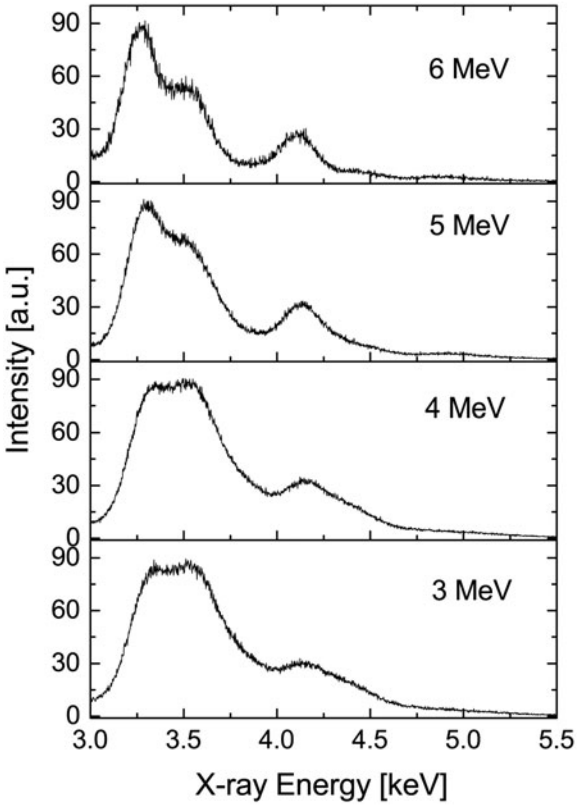

Fig. 2. X-ray spectra induced by 3, 4, 5, and 6 MeV Xe^{23+} impacting on In target.

The projectile-to-target *L*-vacancy production cross sections are calculated by the well-known BEA function developed by Gryzinski (1965) as well as the vacancy sharing model formulated by Meyerhof *et al.* (1977). In the vacancy sharing model, the In L_1 electrons are promoted through 4*f*σ molecular orbit (MO), and on the outgoing part of the collision, the vacancies are shared by the nearby levels such as In L_2, In L_3, Xe L_1, Xe L_2, and Xe L_3. If we do not differentiate the subshells, the overall projectile-to-target *L*-vacancy production cross-section ratio $\sigma_{\text{Xe-}L}/\sigma_{\text{In-}L}$ can be simplified as $\omega_L/(1-\omega_L)$:

$$\omega_L/(1-\omega_L) = \exp(-2\lambda_L), \quad 2\lambda_L = \frac{\pi|\sqrt{I_{\text{Xe}}} - \sqrt{I_{\text{In}}}|}{\sqrt{\frac{1}{2} m v_{\text{p}}^2}}, \qquad (1)$$

where ω_L and $1-\omega_L$ are the total vacancy sharing ratio of Xe *L*- and In *L*-shell from 4*f*σ orbit, respectively, I_{Xe} and I_{In} are the binding energy of Xe and In *L*-shell, respectively, *m* is the electron mass, and v_{p} is the projectile velocity.

As shown in Figure 3b, the vacancy sharing model predictions of Xe *L*/In *L*-vacancy production cross-section ratios are the orders of magnitude lower than the BEA result, and the predicted increasing trend disagrees with the measurement. Therefore, we

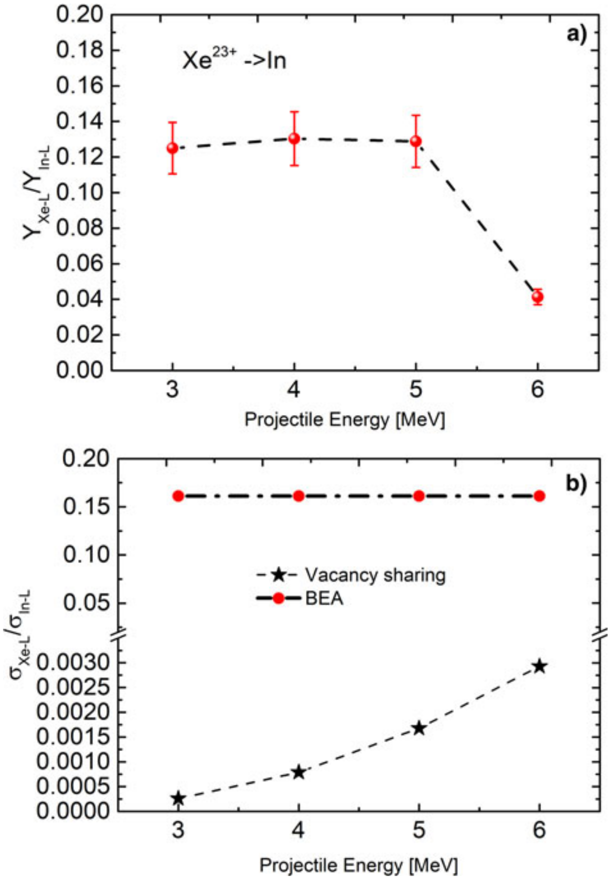

Fig. 3. (a) Projectile-to-target L X-ray yield ratio in collision of Xe^{23+} with In target. (b) The theoretical predictions of the projectile-to-target L-vacancy production cross-section ratios based on the vacancy sharing model as well as the BEA.

conclude that the vacancy sharing contribution of $4f\sigma$ MO to Xe L-shell can be neglected. The BEA formula, which describes the Coulomb ionization process, predicts a constant projectile-to-target L-vacancy production cross-section ratio. This corresponds to the constant X-ray emission intensity ratio neglecting the variation of the fluorescence yield in the incident energy region. Therefore, the observed nearly constant projectile-to-target L X-ray intensity ratio for incident energy smaller than 5 MeV can be qualitatively explained by the Coulomb ionization process.

As for the observed ratio reduction for incident energy larger than 5 MeV, we attribute it to the activation of rotational coupling of $4f\sigma$ orbit. Unlike vacancy sharing, which happens at the large and wide range of internuclear distance, rotational coupling mostly occurs at much smaller internuclear distances; therefore, sufficient incident energy is needed to trigger this process. Since $4f\sigma$ MO correlates to the In L_1 shell, the triggering of this process greatly increases the In L-shell vacancy production cross section. Consequently, the reduced Xe/In L X-ray intensity ratio is expected. The experimental data indicated that the threshold of $4f\sigma$ MO rotational coupling process in the Xe and In collision system is about 5 MeV.

The $L\alpha/L\beta_{1,3,4}$ intensity ratio of In target is observed to increase obviously with the incident energy, as shown in Figure 4. Since Coulomb ionization theory predicts a constant ratio, the results indicate the importance of the relationship between the subshells. As discussed above, the vacancies generated through $4f\sigma$ MO, which correlates to the In L_1 shell, are shared by In L_2 and In L_3 shells, when reasonably neglecting the sharing process of Xe L shells. Namely, the vacancy production cross sections of In subshells could be described as below:

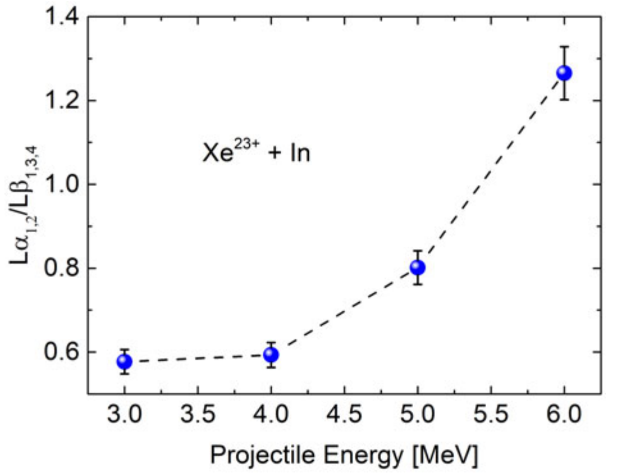

Fig. 4. In $L\alpha/L\beta_{1,3,4}$ intensity ratio as a function of projectile energy in collision of Xe^{23+} with In target.

$$\sigma_{L3} = \omega\sigma_{4f\sigma}, \quad \sigma_{L_1+L_2} = (1-\omega)\sigma_{4f\sigma}, \tag{2}$$

where σ_{L_3} is the vacancy production cross section of the In L_3 shell, $\sigma_{L_1+L_2}$ is the sum of vacancy production cross sections of In L_1 and In L_2 shells, and ω is the vacancy sharing probability from $4f\sigma$ MO to In L_3 shell. The vacancy production cross-section ratio of the In L_3 shell to In $L_1 + L_2$ shells can be simplified as $\omega/(1-\omega)$, which is formulated to increase with the incident energy. In view of the fact that the $L\alpha$ lines emit from the electron transition to L_3 vacancies, and $L\beta_{1,3,4}$ lines from the electron transition to In L_1 and In L_2 vacancies, increasing $L\alpha/L\beta_{1,3,4}$ intensity ratios are expected consequently. The experimental results can be qualitatively explained by the vacancy sharing model. On the other hand, the agreement validates Meyerhof's proposal that the molecular orbital correlation relationship tend to obey Eichler rule for asymmetric collision.

In summary, the X-ray emission induced in interaction of Xe^{23+} with solid In target is measured. It is found that the projectile-to-target L X-ray intensity ratio keeps nearly constant in the incident energy region of 2–5 MeV, which could be well explained by Coulomb ionization theory. When the incident energy is larger than 5 MeV, the projectile-to-target L X-ray intensity ratio is greatly reduced due to the activation of rotational coupling process of $4f\sigma$ MO, which correlates to the In L-shell. The incident energy-dependence $L\alpha/L\beta_{1,3,4}$ intensity ratio shows the importance of vacancy sharing between L subshells and validated Meyerhof's proposal that the $4f\sigma$ MO tends to correlates to L_1 subshell for asymmetric collisions.

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