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Research Article

Energy Dependency of Proton-Induced Outer-Shell Multiple Ionization for $_{48}$Cd and $_{49}$In

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L subshell X-rays of $_{48}$Cd and $_{49}$In have been measured for the impact of protons with energies from 75 to 250 keV. Obviously, it is found that $L\gamma_2$ (abbreviation $L\gamma_{2,3}$ for $_{48}$Cd and $L\gamma_{2,3,4}$ for $_{49}$In) X-ray emission is enhanced in comparison with $L\gamma_1$ X-ray emission. The relative intensity ratios of $L\gamma_2$ to $L\gamma_1$ X-ray are larger than the atomic data and increase with decreasing proton energy. This is caused by the multiple ionization of outer-shell electrons. To verify this explanation, the enhancements for relative intensity ratio of $L\iota$ and $L\beta_2$ to $L\alpha$ X-ray in experiments are discussed, and the direct ionization cross sections of 4d, 5s, and 5p electrons are calculated using BEA theory.

1. Introduction

X-ray emission, an important consequential result from ion-atom collisions, involves several inner-shell processes, from primary inner-shell ionization by the incident ions up to the subsequent vacancy decay, including intrashell transitions. The detailed knowledge of X-ray emission provides important information to understand the charged ion-atom interaction mechanism and to test relevant ionization theories [1–4]. Furthermore, accurate parameters of X-ray are required for the application of trace element analysis known as particle-induced X-ray emission (PIXE), which has application in many fields, such as environmental studies, archaeology, biomedicine, and forensic science [5–8].

Multiple ionization, which means more than one orbital electron is knocked off during ion-atom collisions, can be caused by concurrent direct single ionization or subsequent Coster–Kronig (CK) or Auger transitions. Such action can reduce the screening of nuclear charge and alter the fluorescence yield because of the absence of some outer-shell electrons. Consequently, the blue shift of the X-ray energy may occur, and the relative intensity ratio of the subshell X-ray will be changed. As we all know that such multiple ionization can be induced by heavy ions [9–12], that has been also tested and verified in our previous work [13–15]. Besides, this can also be produced by relativistic electron impact and is dependent on the incident energy [16–18]. Generally, the ionization produced by high-energy protons is considered to be single ionization, and the corresponding X-ray data are chosen as the standard atomic data. However, recently, it has been found that the multiple ionization can also be produced by low-energy protons, which is not only discussed in our earlier work [19] but has also been mentioned by other researchers [20–22]. In spite of that, the dependence of multiple ionization on the proton energy is still not clear. Therefore, in this work, such a phenomenon is further investigated, and special attention is devoted to the incident energy dependency. In order to verify such a question clearly, the targets $_{48}$Cd and $_{49}$In are selected based on their atomic structure, in which the Lγ emission involves the outermost electrons.

1.1. Description of the Experiment. The measurements have been carried out at the 320 kV high-voltage experimental platform at the Institute of Modern Physics, Chinese Academy of Sciences (IMP, CAS) in Lanzhou, China. More details of the experimental system have been described in the previous work [23]. In brief, the protons are produced by the electron cyclotron resonance (ECR) ion source. In order to ensure single collision, the current intensity of the ion beam is regulated and controlled in the magnitude of only few nA. The emitted X-rays are detected by a silicon drift detector (SDD). The SDD is placed at 80 mm far away from the target surface in the chamber and at 135° to the beam direction. The number of incident projectiles, which could not be measured immediately by recording the target current due to the influence of the secondary electron emission, is detected indirectly by the combined use of a penetrable Faraday cup and a common one.

The maximum projected range of the proton in the present work is 1.45 μm and 1.65 μm for Cd and In, respectively. Those are all shorter than the target thickness. The experimental target can be taken as a thick target. The X-ray production cross section can be derived to measure the X-ray yield and the proton's stopping power [24]. The principal uncertainties for the experimental data result from the X-ray count statistics 5%, incident ions recording 3%, detector efficiency 10%, solid angle 2%, slope calculation of the experimental yield curve 2%, and stopping power calculation 10%. The maximal uncertainty of the yield is about 12%, that of the ratio of the subshell X-rays is about 14%, and that of X-ray production cross section is about 16%.

2. Results and Discussion

In Figure 1, the typical X-ray emission spectra of $_{48}$Cd and $_{49}$In produced by proton are presented as a function of the proton energy and well fitted by a nonlinear curve Gaussian fitting program. The structure of the spectra is similar for the two targets under different incident energies as a whole. It mainly consists of six distinct lines which are the L subshell X-rays arising from the relative deexcitation of target atomic L vacancies. The first five transitions are the same for both elements and identified as L_ι ($3s_{1/2}$-$2p_{3/2}$), L$\alpha_{1,2}$ ($3d_{5/2,3/2}$-$2p_{3/2}$), L$\beta_{1,3,4}$ ($3d_{3/2}$-$2p_{1/2}$, $3p_{3/2,1/2}$-$2s_{1/2}$), L$\beta_{2,15}$ ($4d_{5/2,3/2}$-$2p_{3/2}$), and Lγ_1 ($4d_{3/2}$-$2p_{1/2}$). Due to the difference in the electron configuration for the two elements, there is a small distinction for the sixth line. Electronic configuration of $_{48}$Cd atom is $1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6 4d^{10} 5s^2$, but that of $_{49}$In atom is $1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6 4d^{10} 5s^2 5p^1$. Therefore, the last peak is $L_{\gamma 2,3}$ ($4p_{3/2,1/2}$-$2s_{1/2}$) and $L_{\gamma 2,3,4}$ ($4p_{3/2,1/2}$/$5p_{1/2}$-$2s_{1/2}$) for $_{48}$Cd and $_{49}$In [25], respectively, and they are abbreviated as "Lγ_2" in order to present the following discussion clearly and briefly.

One can see in Figure 1 that although the spectrum of the two targets is similar for various proton energies, there is an obvious decrease in the Lγ_2 X-ray emission with increasing incident energy, compared to Lγ_1 X-ray emission. For further quantitative analysis, the relative intensity ratios of Lγ_2 to Lγ_1 X-ray are extracted from the original data after taking into account the detection efficiency of the detector and the self-absorption inside the target material. As shown in Figure 2, the ratios are larger than the theoretical data of single ionization which is calculated based on the ECPSSR theory [26], and it decreases rapidly and is closer and closer to the atomic data as the proton energy increases, in the present experimental energy region. For example, the enhancement factor, namely, the ratio of the experimental result to the atomic data for Lγ_2/Lγ_1, is about 6 for $_{48}$Cd and 7 for $_{49}$In at the energy of 100 keV, but those drop approximately to 2 when the energy is 250 keV.

It is proposed that the enhancement effect of Lγ_2 X-ray can be understood by the outer-shell multiple ionization. As mentioned in the Introduction, such multiple ionization can also occur for the impact of lower energy proton, except for the case of highly charged heavy ions. When it takes place, owing to the absence of outer-shell electrons, some of the nonradiative transitions, such as Auger transition, CK transition, and super CK transition, are restrained. Accordingly, the radiative transition probability of the X-ray emission is changed [9–12]. As a result, the measured relative intensity ratio of subshell X-ray is altered. Figure 3 gives the experimental results of the intensity ratios of Lι and Lβ_2 to Lα X-ray as a function of the proton energy, respectively, which are all larger than the atomic data [26], decrease with the increase of proton energy, and present a similar trend as that of I(Lγ_2)/I(Lγ_1). Based on that result, an estimation can be deduced that the M and N subshell electrons of $_{48}$Cd and $_{49}$In are multiply ionized by lower energy proton impacting, as similarly discussed in our previous work [19].

Lι and Lα X-rays originate from the main transitions for M_1 and $M_{4,5}$ electrons filling the same lower energy vacancy of L_3, respectively. The multiple ionization on M_1 subshell can be regarded as to be insignificant compared to that on $M_{4,5}$ shells because of three main reasons. The first is the large deviation of direct ionization due to the difference in binding energy and electron number between various subshells; for instance, the ionization energy of M_1 electrons is about 1.7 times larger than that of M_5 for $_{48}$Cd element [27–29], and the single ionization cross section of M_5 electrons is higher than that of M_1 by almost one to two orders of magnitude in the present experimental energy region [26]. The second is the high decay probability of M_1 vacancies via CK effects being promoted to higher M subshells. The last one is that the vacancy production in M_1 shell by CK rearrangement among the L subshells is energetically forbidden. One assumes that, besides the fluorescence yield, the X-ray emission intensity is proportional to the electronic number in the upper energy shell for filling the identical vacancy. The enhancement of ratios of Lι to Lα to the atomic data in Figure 3 indicates directly the multiple ionization in $M_{4,5}$ shells, and the diminished trend denotes that the extent of that multiple ionization decreases with the increase of proton energy. For example, the average number of multiple vacancies in M $_{4,5}$ shells is estimated using the relationship between the experimental ratios of Lι to Lα and the

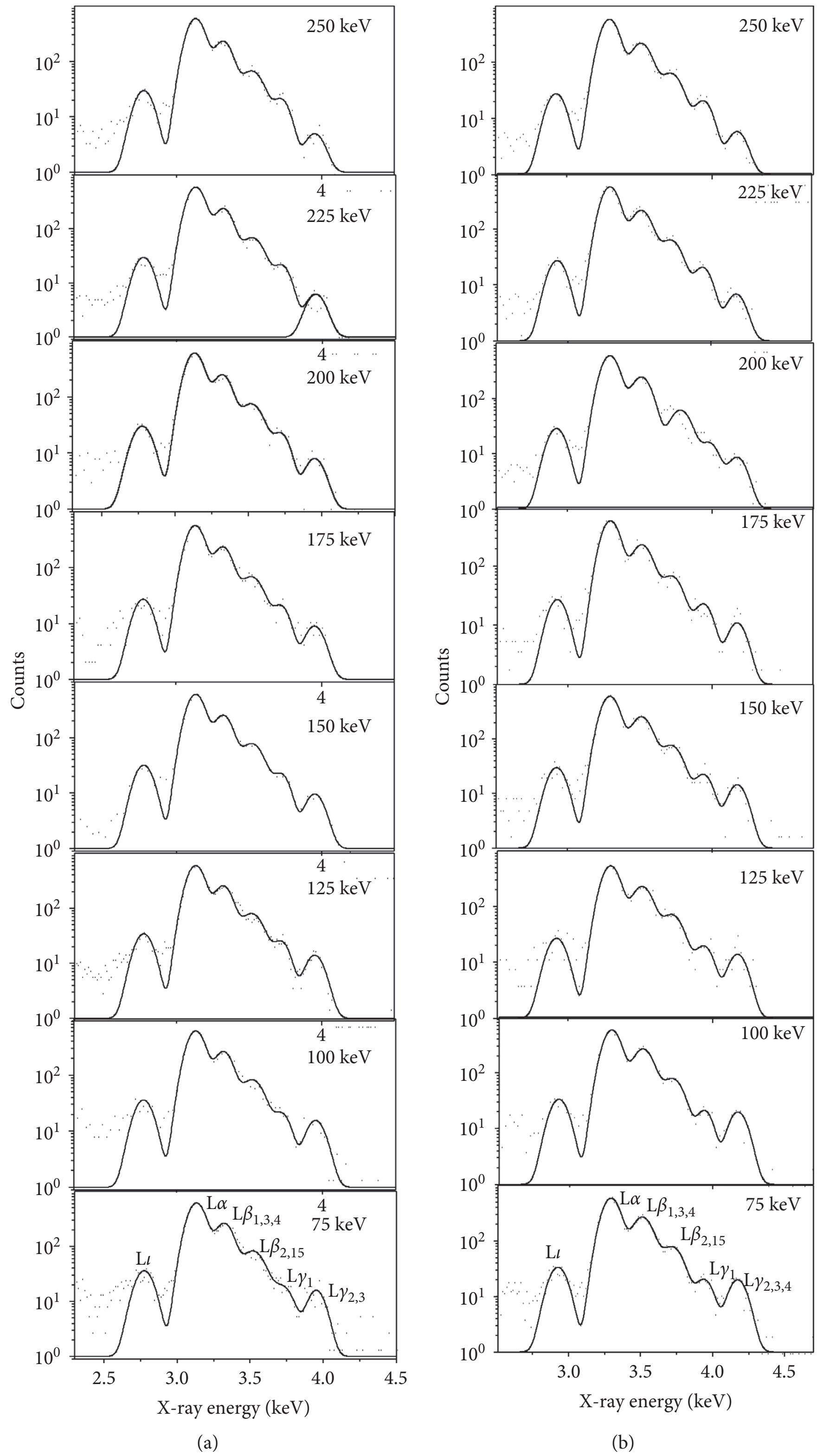

Figure 1: Characteristic L X-ray spectra of (a) $_{48}$Cd and (b) $_{49}$In for proton impacting, which are normalized by the counts of Lα X-ray.

theoretical values [20, 21, 30, 31]. That is about 5.6 ± 0.8, 5.1 ± 0.7, 4.8 ± 0.7, 4.4 ± 0.6, 3.8 ± 0.5, 4.1 ± 0.6, 4.0 ± 0.6, and 4.1 ± 0.6 for $_{49}$Cd and 6.1 ± 0.9, 4.8 ± 0.7, 4.2 ± 0.6, 4.0 ± 0.6, 3.9 ± 0.5, 3.9 ± 0.5, 3.8 ± 0.5, and 3.8 ± 0.5 for $_{49}$In, with the proton energy of 75, 100, 125, 150, 175, 200, 225, and 250 keV, respectively.

Lβ_2 and Lα X-rays mainly come from the transitions having the same lower energy level L_3 but different upper

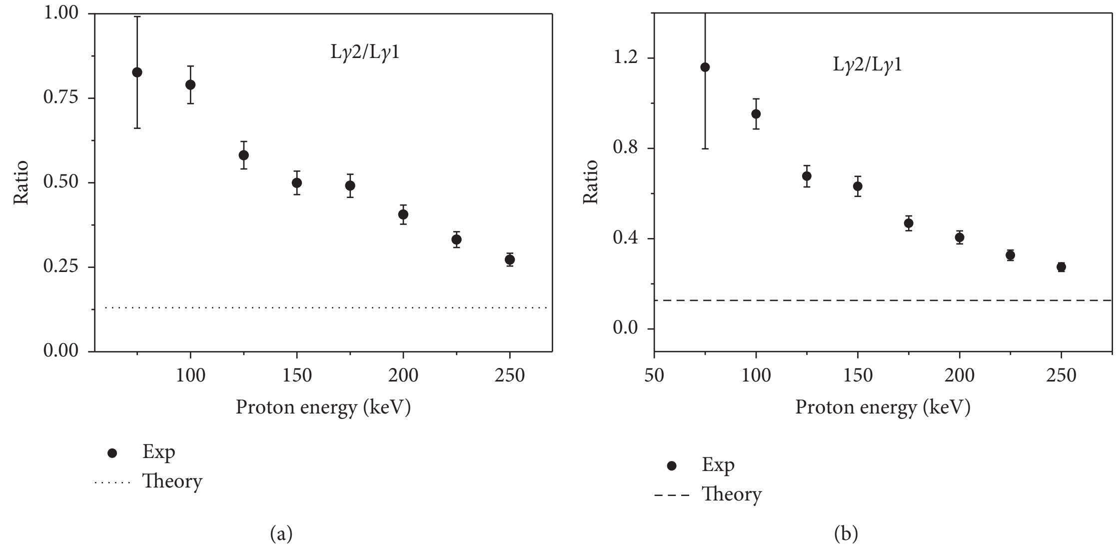

FIGURE 2: Lγ X-ray relative intensity ratios of (a) $_{48}$Cd and (b) $_{49}$In for proton impact. Circle dots are the experimental data and dotted lines are the theoretical calculation for the singly ionized atom (atomic data) [26].

levels, i.e., $N_{4,5}$ and $M_{4,5}$ shells, respectively. When the outer-shell multiple ionization occurs, with the absence of some M and N shell electrons, the Auger transition ratio for filling L_3 vacancies will decrease, while the fluorescence yield ω_3 will be enlarged [9–12, 27–29]. The Auger transition probability a_3 of L_3 vacancies for Cd is almost a factor of 20 and 170 larger than the fluorescence yield of Lα ($\omega_{L\alpha}$) and Lβ_2 ($\omega_{L\beta 2}$) X-ray, and that factor is about 18 and 149 for In, respectively [27–29]. Therefore, $\omega_{L\beta 2}$ will have a larger increase than $\omega_{L\alpha}$; as a result, the actual emission of Lβ_2 X-ray will present an enlargement compared to that of Lα, namely, the measured relative intensity ratio of Lβ_2 to Lα X-rays is enhanced than the atomic data of the singly ionized atom. Conversely, the results of Lβ_2/Lα in Figure 3 illustrate the multiple ionization of $M_{4,5}$ and $N_{4,5}$ shells, and it decreases with the increase of incident energy.

Multiple ionization can be produced by concurrent direct Coulomb ionization, charge transfer, or subsequent excitation by CK and Auger transitions. The cross section for the loss of outer-shell electron due to charge transfer can be calculated on the basis of Oppenheimer–Brinkman–Kramer (OBK) approximation [32–35]. For $_{48}$Cd impacted by proton with energy of 75–250 keV, this cross section is about 1.96×10^{-12}~3.92×10^{-10} barn for N shell and 2.57×10^{-12}~2.01×10^{-10} barn for O shell. For $_{49}$In, that is about 8.49×10^{-12}~2.80×10^{-10} barn for N shell and 3.11×10^{-12}~1.43×10^{-10} barn for O shell. That is at least 19 orders of magnitude smaller than the direct ionization cross section. So, the multiple ionization results from charge transfer can be neglected for the present result. If leaving the subsequent excitation by nonradiative transitions and the effect of electron correlation out of account, the multiple ionization probability is positive to the single ionization cross section for the direct Coulomb collision. Single ionization is inversely proportional to the binding energy of the related orbital electrons. Besides, the bounding energy of outer-shell electrons is diminished with ascending energy level. So, in consideration of the above confirmed multiple ionization in M and N shells, it is easy to deduce that the O shell electrons of $_{48}$Cd and $_{49}$In are also multiply ionized by lower energy protons in the present work. That is just the reason for the enhanced emission of Lγ_2 X-ray as shown in Figures 1 and 2.

In order to further clarify the dwindling trend of the multiple ionization reflected in Figure 2, the direct ionization cross sections of some outermost-shell electrons for $_{48}$Cd and $_{49}$In are simulated theoretically by binary encounter approximation (BEA) model [36], in which the ionization cross section is proportional to the function of the scaled velocity $G(V)$ ($V = v_p/v_i$, where v_p is the velocity of the projectile, v_i is the velocity of the ionized orbital electron) for a certain collision. Here, the v_p of proton is approximately 1.42–3.15 a.u. (a.u. is atomic units), and the v_i is about 1.10 and 0.73 a.u. for the 4d and 5s electrons of $_{48}$Cd, and that is about 1.28 and 0.82 a.u. for the 4d and 5s electrons of $_{49}$In and 0.59 a.u. for the 5p electron of $_{49}$In, respectively. V is in the region of about 1.1~5.3. The $G(V)$ is diminished as a function of V. So, those cross sections decrease with the increase of proton energy as given in Figure 4. Taking into account the positive proportion between the single and multiple ionization, one can deduce easily the decrease of the extent for the multiple ionization from the above calculated result of single ionization, which leads to the change of ratios of Lγ_2 to Lγ_1 X-ray as in Figure 2.

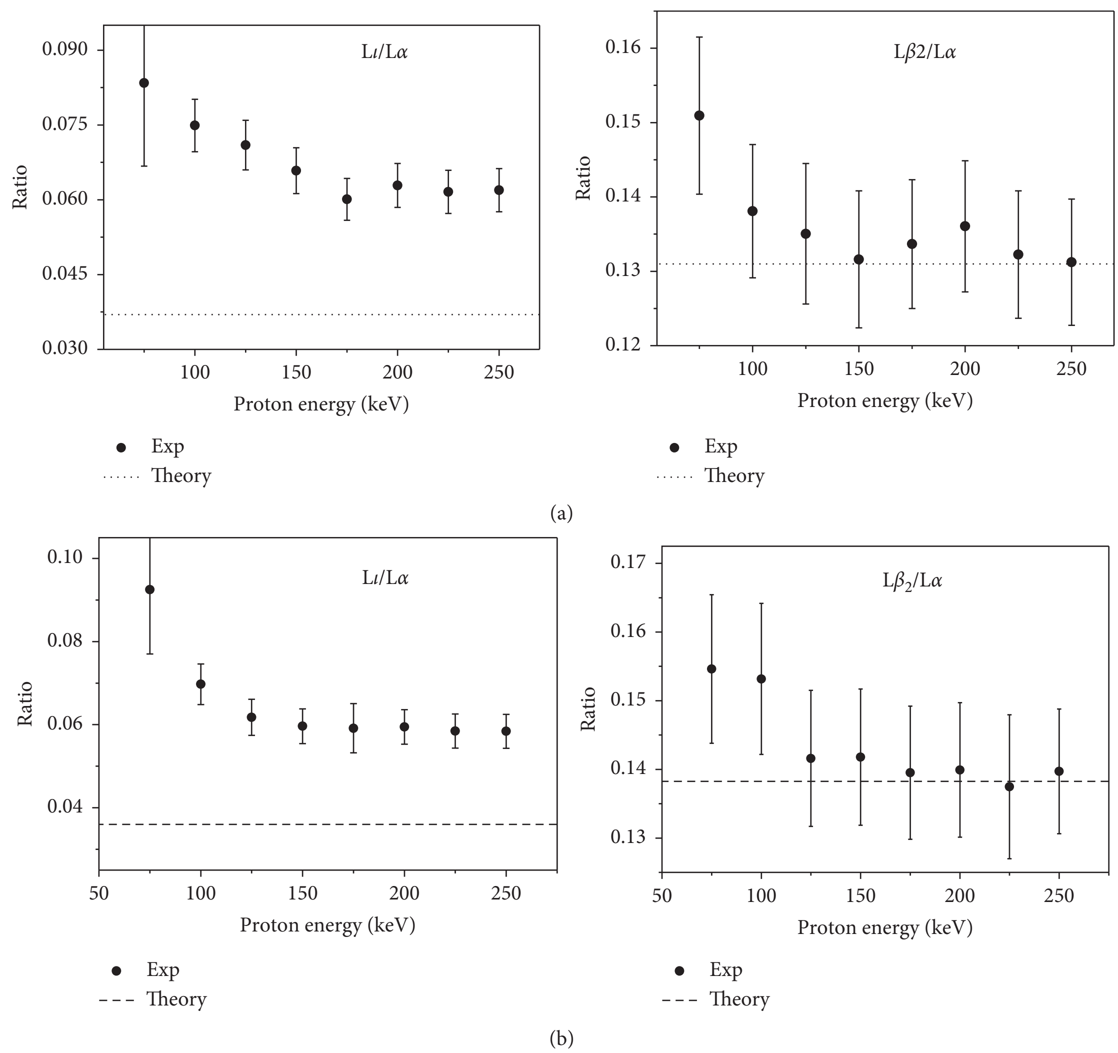

Figure 3: L-subshell X-ray relative intensity ratios of (a) $_{48}$Cd and (b) $_{49}$In for proton impact. Circle dots are the experimental data and dotted lines are the theoretical calculation for the singly ionized atom (atomic data) [26].

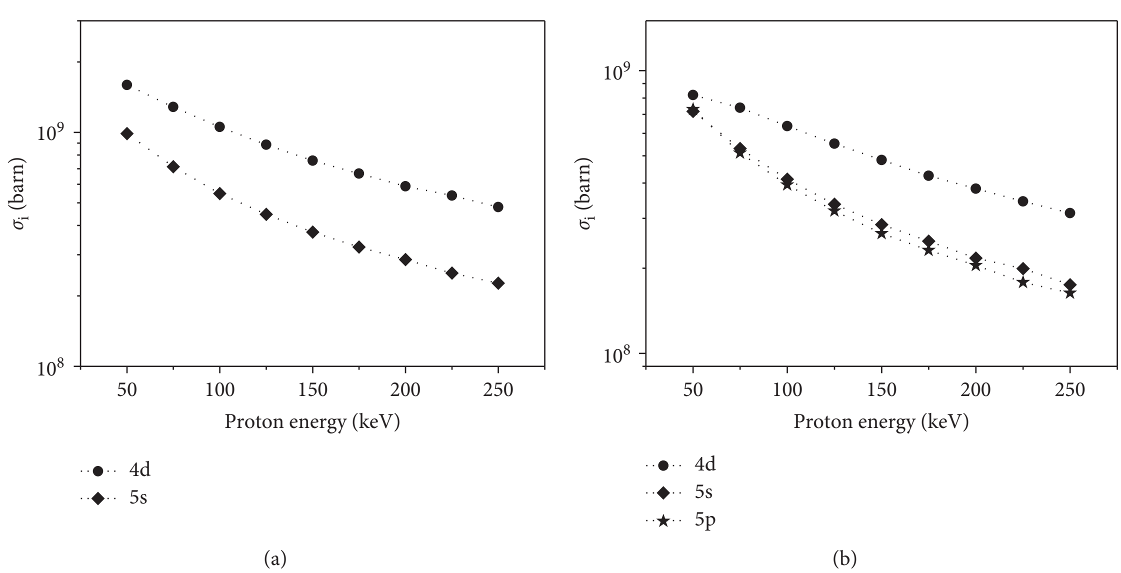

Figure 4: Theoretical (a) $_{48}$Cd and (b) $_{49}$In outer-shell electrons ionization cross sections induced by proton.

3. Conclusions

The Cd and In L subshell X-ray emission has been investigated for 75–250 keV proton impact. The relative intensity ratios of some L subshell X-rays are analyzed. The ionization cross sections of some outermost shells are simulated theoretically. The results indicate that, besides the L shell single ionization, the M, N, and O shell electrons of the target atoms are also multiply ionized. The multiple ionization is strongly dependent on the collision energy, and the extent of that is decreased with the increase of proton energy, which results in an obvious enhanced emission of $L\gamma_2$ X-ray compared to $L\gamma_1$ and the enlargement of the relative intensity ratios of $L\iota/L\alpha$ and $L\beta_2/L\alpha$; however, such enhancement is gradually diminished as a function of incident energy.

Data Availability

The basic data used to support the findings of this study can be found in the figures of the study.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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