

Article

The Laboratory Measurement of the Line Ratios in X-Ray Emission Resulting from the Charge Exchange Between Mg^{11+} and Helium

Kebao Shu ^{1,2}, Caojie Shao ^{2,3,*} , Shuo Zhang ⁴, Ruitian Zhang ^{2,3} , Cheng Qian ², Yingli Xue ^{2,3} , Mingwu Zhang ^{2,3} , Jinlei Tian ^{1,2}, Zhenqiang Wang ^{1,2}, Xiaolong Zhu ^{2,3} , Liangting Sun ^{2,3} , Junxia Ran ¹ and Deyang Yu ^{2,3} 

¹ College of Physical Science & Technology, Hebei University, Baoding 071002, China; kebaoshu@impcas.ac.cn (K.S.); 17838529707@163.com (J.T.); 20238015043@stumail.hbu.edu.cn (Z.W.); rjxhbu@hbu.cn (J.R.)

² Institute of Modern Physics, Chinese Academy of Sciences, Lanzhou 730000, China; zhangrt@impcas.ac.cn (R.Z.); qianc@impcas.ac.cn (C.Q.); xueyl@impcas.ac.cn (Y.X.); zhangmingwu@impcas.ac.cn (M.Z.); zhuxiaolong@impcas.ac.cn (X.Z.); sunlt@impcas.ac.cn (L.S.); d.yu@impcas.ac.cn (D.Y.)

³ School of Nuclear Science and Technology, University of Chinese Academy of Sciences, Beijing 100049, China

⁴ Advanced Energy Science and Technology Guangdong Provincial Laboratory, Huizhou 516007, China; shuozhang@gdlhz.ac.cn

* Correspondence: c.shao@impcas.ac.cn

Abstract: The line ratios in X-ray emission resulting from charge exchange between highly charged ions (HCIs) and neutral atoms are not only crucial for accurately modeling astrophysical X-ray emissions but also offer a unique perspective on the charge exchange processes happening during collisions. The *K* X-ray spectra following charge exchange between Mg^{11+} and He are presented for a collision velocity of 1489 km/s (11.5 keV/amu). The spectra were measured by two Silicon Drift Detectors capable of resolving the Mg^{10+} $K\alpha$, $K\beta$, $K\gamma$, and $K\delta$ lines. The line intensity ratios of $K\beta$, $K\gamma$, and $K\delta$ relative to the $K\alpha$ line, as well as the hardness ratio, were obtained. The experimental results were compared with the theoretical results from a cascade model that utilizes the state cross-sections produced by multichannel Landau–Zener (MCLZ) calculation. It was discovered that the *K* X-ray spectrum features can be reproduced well by MCLZ theory when the contributions of both single electron capture (SEC) and autoionizing double capture (ADC) processes are included. This finding implies that the ADC feeding mechanism is significant and should be taken into account for the X-ray emission during charge exchange between highly charged ions and multielectron atoms.

Keywords: X-ray spectra; line ratio; charge exchange; highly charged ion (HCI); ion atom collision; single electron capture (SEC); autoionizing double capture (ADC)



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1. Introduction

Charge exchange (CX) between highly charged ions (HCIs) and neutral atoms or molecules is a process in which one or more target electrons are transferred to an excited state of the resulting ion. The subsequent radiative de-excitation of excited HCIs results in characteristic X-ray emission. Such CX-induced X-ray emissions have been identified as the source of or larger contributors to X-ray emissions from comets [1–4], Jovian auroras [5], and diffuse soft X-ray backgrounds [6,7]. They have also been applied to many other astronomical environments ranging from supernova remnants [8,9] to extragalactic cooling

flows [10], aiming to explain the “anomalous” X-ray emission characteristics that are not likely to be ascribed to electron impact excitation (EIE) or electron radiative recombination (RR). Therefore, comprehensive and reliable CX-induced X-ray data are of crucial importance in diagnosing these astrophysical nonequilibrium plasmas where hot flows meet cold gases.

The spectra or line ratios of CX-induced X-rays have been widely studied both theoretically and experimentally over the past several decades. Theoretically, various methods, like the classical trajectory Monte Carlo [11], multichannel Landau–Zener (MCLZ) [12], atomic orbital close coupling (AOCC) [13], molecular orbital close coupling (MOCC) [14], two-center basis generator method (TC-BGM) [15], and the quantum molecular orbital close coupling (QMOCC) method [16], among others, have been developed to compute the n -, ℓ -, and even S -resolved state-selective cross-sections. Here, n and ℓ are the principal quantum number and orbital angular momentum of the captured electron, and S is the total spin angular momenta of the product ion. Subsequently, by incorporating the cascade decay of the formed excited electrons, the line intensity of the X-ray emission induced by CX can be determined. Experimentally, diverse types of photon emission spectroscopy, including the use of grating spectrometers [17,18], crystal spectrometers [19,20], semiconductor detectors [21,22], the recently rapidly evolving microcalorimeters [23–26], and the like, have also been used to measure the photon energy spectra emitted in the CX process. In addition, there are also some methods that combine theory and experiment, such as using energy-gain spectroscopy [27–30] and cold target recoil-ion momentum spectroscopy [31–42] to experimentally measure the state capture cross-section, followed by obtaining the relative intensity of the emission spectral lines through the cascade de-excitation model. However, only a few of the previously reported experimental studies are of relevance to X-ray emission for low-energy CX processes involving bare or hydrogen-like ions having an atomic number Z greater than 10.

Here, we report on the measurement of K X-ray emission resulting from the CX interaction between 11.5 keV/amu Mg^{11+} and He using Silicon Drift Detectors (SDDs). Mg^{11+} ions and He atoms are, respectively, important components of the solar wind ions and neutrals of the solar system [43]. Meanwhile, previous research by Liu et al. showed that up to 30% of the $K\alpha$ triplet of helium-like Mg ions in the XMM–Newton RGS spectra of M82 comes from the charge exchange process between hydrogen-like Mg ions and neutral species [44]. The aim of this experiment is to provide reliable line ratios for modeling astrophysical X-ray emissions and to benchmark only the available theoretical calculations made by MCLZ models [45].

2. Experiment

This experiment was performed at Low Energy high intensity highly charged ion Accelerator Facility (LEAF) at the Institute of Modern Physics (IMP) in Lanzhou. A Mg^{11+} ion beam was produced by a Hybrid superconducting Electron Cyclotron Resonance ion source [46], focused by a Glaser lens, and deflected by a 90° bending magnet towards the interaction chamber. Additionally, the beam position could be adjusted by means of two-directional correction magnets. The beam intensity after interaction with the gas target was measured using a Faraday cup with a specific suppression voltage of -500 V. During the entire experiment, the beam current intensity stabilized around 300 epA.

A schematic illustration of the experimental set-up is shown in Figure 1. An effusive He gas target was introduced into the interaction chamber via a stainless steel capillary. The features of the effusive gas target were thoroughly outlined by Lamour et al. [47], and a brief depiction will be presented here. The capillary has a length of 25 mm and an inner diameter of 0.1 mm, and its exit is located 10 mm from the ion beam center. The target thickness could be regulated by an INFICON VCC500 flow meter. In order to reduce the influence

of injecting the target gas on the beam line vacuum and eliminate the entry of ions with similar charge-to-mass ratios into the collision chamber, a $0.5 \text{ mm} \times 6.0 \text{ mm}$ rectangular slit was installed upstream of the collision zone. The background vacuums of the beam line and the reaction chamber are, respectively, 1.47×10^{-8} and 1.62×10^{-8} mbar. After injecting the target gas, the vacuum in the target chamber dropped to 2.1×10^{-5} mbar, while that of the beam line was only slightly affected and dropped to 1.2×10^{-7} mbar. Additionally, the target density that could be approximately inferred from the target chamber pressure is $3 \times 10^{12} \text{ atoms/cm}^3$. The length of the interaction region between the ion and the target is approximately 6.7 cm. The theoretical capture cross-section given by Lyons et al. is $1.52 \times 10^{-15} \text{ cm}^2$ [45]. Therefore, the corresponding capture probability P is about 0.03, which satisfies the single-collision condition that P should be much smaller than 1 [47].

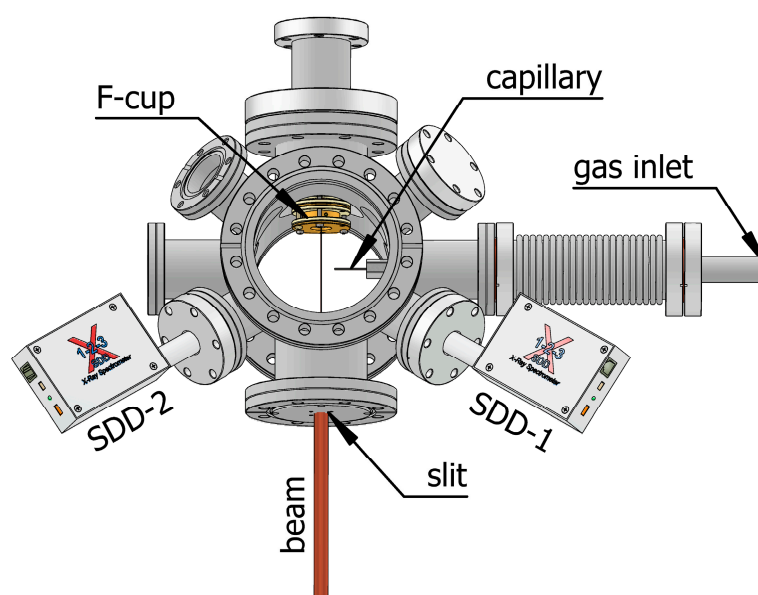


Figure 1. A schematic illustration of the set-up. The two Silicon Drift Detectors (SDDs) were both mounted at 120° observation angles with respect to the ion beam direction.

The X-rays produced in the collisions of the ions with the target were detected by two Silicon Drift Detectors (SDDs) both mounted at 120° observation angles with respect to the ion beam direction. The first SDD (No. 1), having an effective detection area of 17 mm^2 , was placed in the interaction chamber and at a distance of 147 mm from the collision center. The second SDD (No. 2), having an effective detection area of 50 mm^2 , was placed outside the interaction chamber and at a distance of 145 mm from the collision center. The detectors were calibrated by using the K diagram lines of Al, Ca, Cr, Fe, and Cu and the L diagram lines of Ag, which were induced by a mini-X-ray tube.

3. Results and Discussion

The spectra of Mg ion K X-rays after colliding with the He target recorded by the SDDs are shown in Figure 2. In addition, the positions of the Mg^{10+} $1s2p \rightarrow 1s^2$, $1s3p \rightarrow 1s^2$, $1s4p \rightarrow 1s^2$, $1s5p \rightarrow 1s^2$, and $1s6p \rightarrow 1s^2$ transitions, correspondingly labeled as the $K\alpha$, $K\beta$, $K\gamma$, $K\delta$, and $K\epsilon$ lines, are indicated by vertical lines. At the same time, the detection efficiencies of the SDDs are also denoted by a gray dashed line. It is obvious that the $K\alpha$ line is directly distinguished from the remaining lines, while $K\beta$, $K\gamma$, $K\delta$, and $K\epsilon$ have different degrees of overlap with each other. Since the energy difference between the $K\delta$ and $K\epsilon$ lines is only about 10 eV, they were analyzed as one peak (namely, $K\delta+$) in the following data processing procedure. Accordingly, the peaks in the spectra were fitted by four Gaussian

distributions, with all parameters being unrestrained except that the full widths at half maximum (FWHMs) of the $K\beta$, $K\gamma$, and $K\delta+$ peaks were set to be identical.

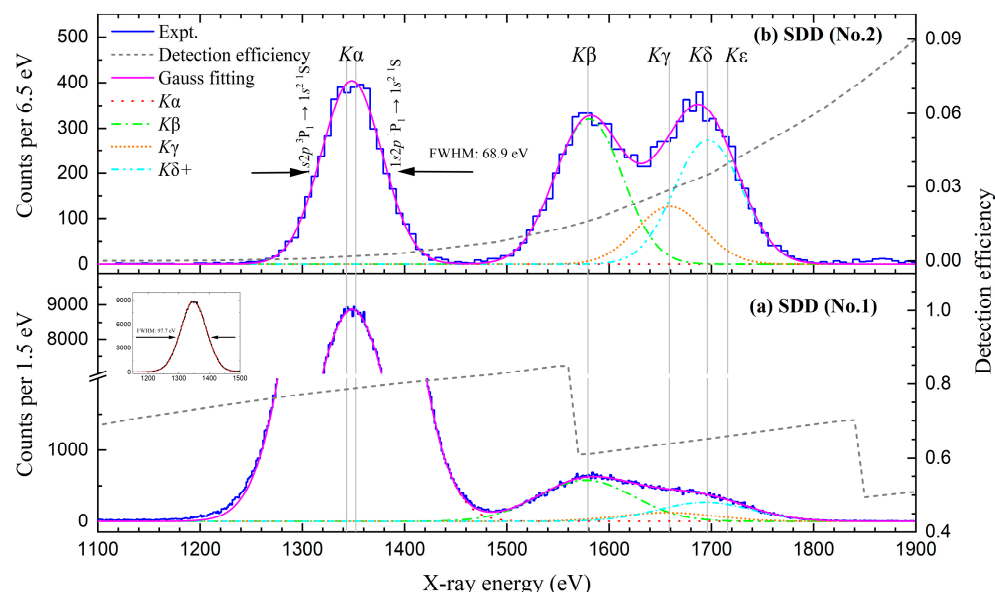


Figure 2. The spectra of the X-rays emitted from a projectile in the 11.5 keV/amu Mg^{11+} collisions with He, obtained by the (a) inner and (b) outer SDDs at a 120° observation angle. The detection efficiency of each SDD is given as a gray dashed line.

The fitting results show that the energy resolutions (FWHMs) of the inner and outer SDDs for Mg $K\alpha$ (with an energy of 1348 eV) are 97.7 eV and 68.9 eV, respectively. The spectra of metastable ions, namely lithium-like ions, generated during the collision with the residual gas in the beam line chamber are indistinguishable. Given that the vacuum degree of the beam line is two orders of magnitude better than that of the target chamber, the influence of these spectral lines should be minimal. From the areas of each fitted peak, the line emission ratios of $K\beta/K\alpha$, $K\gamma/K\alpha$, and $K\delta+/K\alpha$, as well as the hardness ratio, were obtained after efficiency correction and are listed in Table 1. The detection efficiencies of the X-ray detectors were carefully analyzed, and the uncertainty of the relative efficiencies in the energy region from 1.2 to 1.7 keV is estimated to be 4%. This result indicates that all the line intensity ratios measured by the two detectors are in mutual agreement within the error range.

Table 1. Experimentally determined line ratios as well as theoretical ratios for $\text{Mg}^{11+} + \text{He}$ collision. Experimental errors are determined by standard errors in fitting in fitted areas and uncertainties in relative detection efficiency.

Line Ratios	Experiment		Theory	
	SDD (No. 1)	SDD (No. 2)	MCLZ-II	Statistical
$I(K\beta)/I(K\alpha)$	0.091 ± 0.003	0.095 ± 0.003	0.233	0.047
$I(K\gamma)/I(K\alpha)$	0.021 ± 0.001	0.022 ± 0.001	0.250	0.019
$I(K\delta+)/I(K\alpha)$	0.038 ± 0.001	0.037 ± 0.002	0.311	0.057
Hardness ratio	0.150 ± 0.005	0.154 ± 0.006	0.794	0.122

To theoretically predict the line intensity ratio of the X-ray emissions following CX, the initial state population and all possible transitions to the lowest energy level of transferred electrons are required. Since the decay energies and rates of excited ions could be calculated well by several programs such as FAC [48], $n\ell S$ - and even J -resolved state-selective capture

cross-sections become some of the most important components for producing realistic CX X-ray emissions. Some advanced programs such as AOCC can in principle give accurate cross-sections, but the results for the current collision system are not available. Here, we adapted the MCLZ approach to generate $n\ell$ S-resolved cross-sections.

It should be noted that there are two specific implementation methods for generating $n\ell$ -resolved cross-sections in CX collisions using MCLZ theory [49]. One involves calculating the n -resolved cross-sections with MCLZ theory and then adding the different ℓ -distribution models to obtain the $n\ell$ -resolved cross-sections. The other involves directly calculating the $n\ell$ -resolved cross-sections with MCLZ theory. The first method is suitable for use in CX collisions with bare ions because the ℓ states of the product ion are degenerate within a given n state. In principle, the second method should be recommended for the CX process with non-bare projectiles. However, due to the general lack of data on ℓ -distribution functions, astrophysical modelers often use the first method for all CX collisions.

Hence, in Figure 3, we make a comparison between the results of X-ray spectra from $n\ell$ S-resolved cross-sections via MCLZ and the $n\ell$ -resolved distribution functions for Mg^{11+} collisions with He. The experimental spectrum was efficiency-corrected. This spectrum was acquired through superimposing the efficiency-corrected $K\alpha$, $K\beta$, $K\gamma$, and $K\delta$ peaks. The data of the cross-sections calculated via MCLZ are from the calculation results obtained by Lyons et al. [45]. A total of five ℓ -resolved distribution functions were adopted in the first calculation method. They are the so-called even, Landau–Zener I (LZ-I), low-energy modified Landau–Zener I (i.e., LZ-II), separable, and statistical models [12]. The ℓ -resolved distribution functions of the even, LZ-I, LZ-II, separable, and statistical models are given by the corresponding expressions $1/n$, $(2\ell + 1) \frac{[(n-1)!]^2}{(n+\ell)!(n-\ell-1)!}$, $\ell(\ell + 1)(2\ell + 1) \frac{(n-1)!(n-2)!}{(n+\ell)!(n-\ell-1)!}$, $\frac{2\ell+1}{q} \exp(\frac{-\ell(\ell+1)}{q})$, and $\frac{2\ell+1}{n^2}$, respectively. Here, q is the charge state of the projectile ion. It should be noted that the decay rate results calculated by the FAC program indicate that for $n \geq 3$, only the singlet $1sn\ p\ ^1P_1$ state decays to the ground state, while triplet states $1sn\ p\ ^3P$ nearly exclusively decay to the metastable state $1s2s\ ^3S_1$. This will give rise to a situation wherein the proportional relationship between singlet and triplet states under the same $n\ell$ -distribution exerts a significant influence on the relative intensities of the emitted spectral lines. In Figure 3a, MCLZ-I and MCLZ-II, respectively, signify the setting of the population ratio of singlet and triplet states in accordance with the calculated outcomes and the setting as 1:3 under the same $n\ell$ -population. The theoretical X-ray spectra are yielded by the line intensity ratios after convoluting the experimental detection resolution with an FWHM of 68.9 eV.

As is clearly shown in Figure 3a, all the $n\ell$ -distribution functions except the statistical model overestimate the experimental intensity of the $K\delta$ lines. Even when the population branching ratio of the singlet state was reduced by setting the ratio of the singlet and triplet states, the spectral lines predicted by the MCLZ-II model still considerably overestimate the relative intensity of the $K\delta$ lines. This can be understood by considering the comparison of the $n\ell$ -distribution functions in Figure 3b. Among the states with the principal quantum number 5, which is the dominate population state during CX, the proportion of the p state predicted by MCLZ is the largest among all the distributions. The MCLZ and LZ-I models predict almost the same ℓ -distribution, and this result is consistent with the calculation results of Mullen et al. for the CX between Fe^{25+} collisions with N_2 . However, as the population ratio of the $5\ ^1P$ singlet and $5\ ^3P$ triplet states in MCLZ-I is greater than that in MCLZ-II and LZ-I, this leads to the relative intensity of $K\delta$ predicted by the former also being considerably greater than that of the latter.

Additionally, the proportions of the p state predicted by the even, separable, and LZ-II models are similar, and the relative intensities of the $K\delta$ lines are also similar in the simulated spectra. Among all these models, the energy spectrum predicted by

the statistical model comes the closest to that measured experimentally. This result is not surprising since the statistical ℓ -distribution was initially recommended for collision energies $\gtrsim 10$ keV/amu, while models like LZ-I and LZ-II were recommended for collision energies $\lesssim 10$ –100 eV/amu. However, somewhat unexpectedly, although the ℓ -distribution function is not recommended to be used in the CX process with non-bare projectiles, the simulation spectra obtained via this method with the statistical ℓ -distribution, compared to those obtained via the MCLZ method, are more in line with the experimental spectral lines in the present CX collision. The line intensity ratios under MCLZ-II and the statistical ℓ -distribution are also listed in Table 1. One point that needs to be noted here is that since the $1s2s\ ^1S$ state is a metastable state, the $K\alpha$ line measured both in the present experiment and theory does not include the line when it decays to the $1s^2\ ^1S$ state, while in astronomical observations, this spectral line may possibly be included. Therefore, when the intensity ratios obtained in the present experiment are adopted for astronomical modeling, the contribution of $K\alpha$ by $1s2s$ - $1s^2$ should be additionally evaluated.

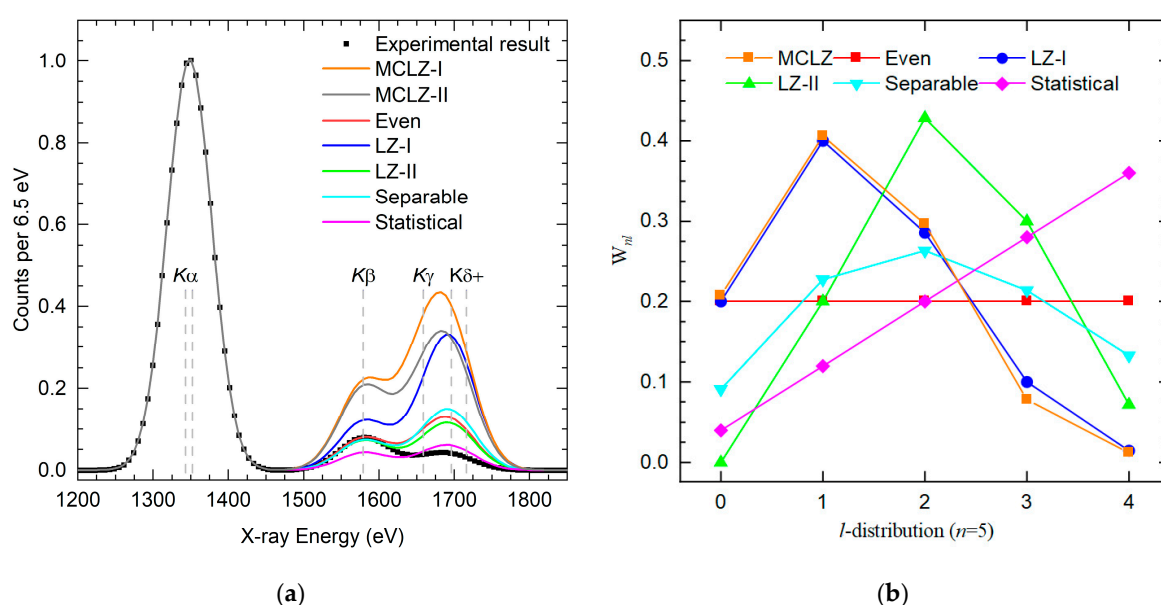


Figure 3. (a) A comparison between theoretical and experimental spectra. The intensity is normalized with respect to the $K\alpha$ line. The experimental spectrum was obtained from an SDD (No. 2) and efficiency-corrected. (b) A comparison of ℓ -resolved MCLZ cross-sections with analytical ℓ -distribution functions under $n = 5$ for 11.5 keV/amu Mg^{11+} SEC collisions with He. The MCLZ distribution was obtained by summing the singlet and triplet cross-sections multiplied by 1/4 and 3/4, respectively.

However, there are still significant differences between the theoretical results predicted by the statistical ℓ -distribution and the experimental results. Specifically, the theoretical intensity of the $K\delta+$ line relative to the $K\alpha$ line is greater than that of the experiment, while that of $K\beta$ relative to $K\alpha$ is less than the experimental one. The reason for this result should be that the current theoretical method only takes into account the single-electron capture process and does not include the double-electron capture process. Considering that the single-electron capture is directly responsible for the $1s5p$ to $1s6p$ population and thus the emission of $K\delta+$ lines [47], therefore, the double-electron capture process will not enhance the emission intensity of $K\delta+$ lines, while the double-electron capture followed by Auger de-excitation to lower energy levels will enhance the emission intensities of the $K\alpha$ and $K\beta$ lines. These two factors taken together will result in a theory that neglects the double-electron process overestimating the relative intensity of the $K\delta+$ lines with respect to the $K\alpha$ lines.

Given that these calculations involve CX between multiple electrons, they are inherently difficult and have not been widely adapted for extended use in CX modeling. Here, we only make a simple estimate. First, it is assumed that autoionizing double capture (ADC) is the dominant contribution of the double-electron capture process, and other multielectron processes such as true double capture (TDC) are less significant than ADC. Second, since double capture commonly results in doubly excited states on the projectile, with n values close to or slightly lower than those in single capture, states such as $(4\ell, 5\ell')$, $(4\ell, 6\ell')$, and $(5\ell, 6\ell')$ should be predominant among autoionizing doubly excited states. Third, the autoionization transitions of these states tend to favor the smallest energy jumps. The autoionization of these states to the 4ℓ state is energy-constrained, while that to the 3ℓ state is energy-permitted. Therefore, it can be approximately assumed that all these doubly excited states autoionize to the 3ℓ state. Finally, by assuming that the distribution of the states with different ℓ in the 3ℓ states produced by the ADC process was statistical, we can obtain the X-ray spectra or line intensity ratios resulting from the ADC process through a cascade model.

Figure 4 illustrates a comparison between the X-ray spectra measured experimentally and those simulated theoretically. The theoretical results both excluding and including the ADC process are given. In the theoretical results including the ADC process, the ratio of the ADC cross-section to the total cross-section is given as 0.267. It can be seen that the theoretical spectral line after adding the ADC process is in very good agreement with the experimental results. Although setting the ratio of the ADC cross-section to the total cross-section and assuming the autoionization of doubly excited states as mentioned above could result in the experimental results being reproduced well, the contribution of ADC could be overestimated. On the one hand, the current estimation method of the ADC process is very rough. On the other hand, both SEC and ADC will lead to the generation of a $1s2s$ metastable state that cannot contribute to the $K\alpha$ line. The proportion of the $1s2s$ metastable state to the overall $1s2\ell$ state produced during the cascading process will notably impact the estimation of the ratio of ADC to SEC via the current method. Precisely evaluating the contribution of ADC requires a combination of methods such as absolute cross-section measurement in subsequent experiments.

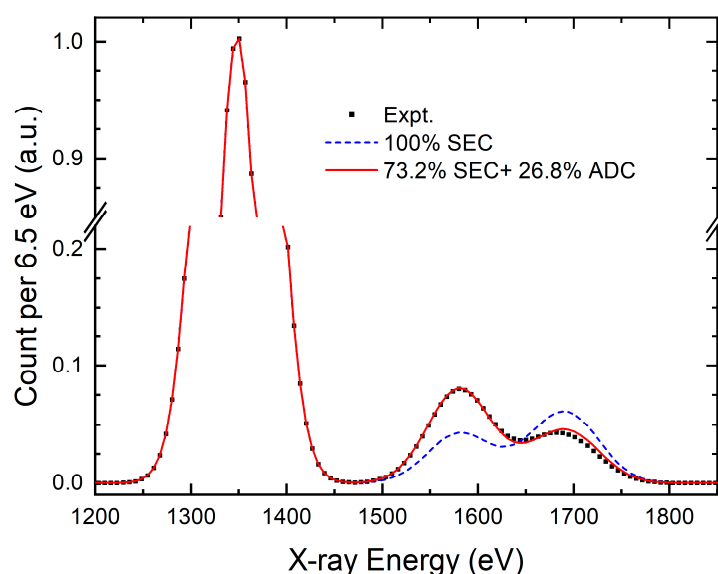


Figure 4. Comparison between experimental and theoretical X-ray spectra: black dots represent experimental results, blue dashed line represents SEC-only theoretical spectral line, and red solid line represents SEC + ADC theoretical spectral line. Experimental spectrum was obtained from SDD (No. 2) and efficiency-corrected.

4. Conclusions and Outlook

K X-ray spectra and line ratios for soft X-ray emission following charge exchange between Mg^{11+} and He were measured using a beam–gas technique and two SDDs at a collision velocity of 1489 km/s. The $K\alpha$, $K\beta$, $K\gamma$, and $K\delta$ lines of the Mg^{10+} ion were identified, and the line intensity ratios of $K\beta$, $K\gamma$, and $K\delta$ relative to $K\alpha$ lines as well as the hardness ratio were obtained. The experimental results were compared with the theoretical results resulting from a cascade model that makes use of the state cross-sections produced by MCLZ calculation. The results indicate that the spectra obtained by applying statistical ℓ -distribution models to MCLZ n -resolved cross-sections, even though this is not advisable for non-bare projectiles, are in accordance with the experimental outcome in the current CX collision. Moreover, it was found that the K X-ray spectrum features can be reproduced well by MCLZ theory when the contributions of both the SEC and ADC processes are included. This result implies that the ADC feeding mechanism is significant and should be considered for the X-ray emissions produced during charge exchange between highly charged ions and multielectron atoms.

Emission spectra from CX are dependent on the initial distribution of the angular momentum states of the captured electron, which is strongly dependent on collision velocity. More experimental measurements using different collision velocities are needed to provide reliable line ratios for modeling astrophysical X-ray emissions and to validate the predictions made by various models. The newly built LEAF experimental platform can produce low-energy, fully stripped, and hydrogen-like ions with Z up to 26, which will provide opportunities for experiments in this field.

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Data Availability Statement: All data needed to evaluate the conclusions in this work are present in the paper. Additional raw data are available from the corresponding authors upon reasonable request.

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