Investigation of slow collisions for (quasi) symmetric heavy systems: what can be extracted from high resolution x-ray spectra

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Abstract

We present a new experiment on (quasi) symmetric collision systems at low velocity, namely Ar\(^{17+}\) ions (\(v = 0.53\) au) on gaseous Ar and N\(_2\) targets, using low- and high-resolution x-ray spectroscopy. Thanks to an accurate efficiency calibration of the spectrometers, we extract absolute x-ray emission cross sections combining low-resolution x-ray spectroscopy and a complete determination of the ion beam–gas jet target overlap. Values with improved uncertainty are found in agreement with previous results (Tawara et al 2001 Phys. Rev. A 64 042712). Resolving the whole He-like Ar\(^{16+}\) Lyman series from \(n = 2–10\) with our crystal spectrometer enables us to determine precisely the distribution \(\{P_n\}\) of the electron capture probability and the preferential \(n_{\text{pref}}\) level of the selective single-electron capture. Evaluation of cross sections for this process as well as for the contribution of multiple-capture is carried out. Their sensitivity to the \(\ell\)-distribution of \(n\) levels populated by single-electron capture is clearly demonstrated, providing a stringent benchmark for theories. In addition, the hardness ratio is extracted and the influence of the decay of the metastable \(1s^2 3S_1\) state on this ratio is discussed.

(Some figures may appear in colour only in the online journal)

1. Introduction

The interaction between low velocity ions and atoms or molecules has been extensively investigated in the past decades. At the low velocity regime (0.4–1 atomic units (au)), the dominant process is electron capture that occurs in selective projectile excited states, the collisional system behaving like a quasi-molecule. It leads to an energy gain of the projectile ion and the populated excited states decay by emission of photons and/or electrons, both carrying information on the collision dynamics. During the 1980s, many experiments have been performed with light ions, like C, N, Ne, ... interacting with light targets as H\(_2\) and He leading to a quite complete understanding of the mechanisms involved in the collision. Besides total cross sections determination, the \(n\)- and \(\ell\)-population distributions \(\left\{P_n\right\}\) and \(\left\{P_\ell\right\}\), respectively of the single capture process have been characterized as well as the role of rotational versus radial couplings while for double-capture, the \(n\ell n'\ell'\) channels have been identified. For such studies, different detection techniques have been applied, as the kinetic energy gain of the projectile \(\left(\text{[2]}\right)\) and references therein) or the electron and the radiative emission detection \(\left(\text{[3–7]}\right)\). At the same time, extended theoretical investigations have been developed. They include simple approaches such
as the well-known classical over-barrier (COB) model or the reaction window within the Landau–Zener model [8, 9], and more sophisticated coupled state calculations using basis of either atomic or molecular orbitals [10–13]. With the advent of new ion sources, heavier projectile ions of higher charge states have become available. New techniques like the powerful cold-target recoil-ion momentum spectroscopy (COLTRIMS) were applied allowing to get unambiguous determination of partial \( n \) single-electron capture including differential cross sections for the main capture channel like in the case of \( \text{Ar}^{15−18+1} \) on He at 14 keV \( q^{-1} \) for example [14].

However, today, there is still a lack of data for (quasi) symmetric collisions, i.e. highly charged ions colliding on heavy targets, for which contribution from multi-electron exchange processes is important. The only systematic studies available are restricted to coincidence measurements between charge selected projectile and target ions [15, 16] where stabilized charge exchange mechanisms are investigated. More recently, low-resolution x-ray detection experiments have been performed [17, 1, 18, 19]. X-ray measurements provide data in well-defined conditions (in the laboratory frame) that are of interest to interpret x-ray spectra observed from different environments such as those from solar wind colliding with comets [20, 21]. However, in both cases, no information on the partial \( n \ell \) electron capture states is accessible. Furthermore, in the case of low-resolution x-ray measurements, as discussed recently in [22], the effect of single and multiple processes cannot be distinguished. Finally, all the laboratory data are used to extract a parameter called the ‘hardness ratio’ (i.e. the ratio between the x-ray intensity of \( n \rightarrow 2 \rightarrow 1 \) and \( 2 \rightarrow 1 \) transitions) that serves as a reference to determine abundance of elements when interpreting astrophysical x-ray spectra.

With the experiment described in this paper we are bringing new information on the collision dynamics for (quasi) symmetric systems, studying the collisions of low-velocity (0.53 au) \( \text{Ar}^{17+} \) ions with a gaseous Ar and N\(_2\) targets by coupling a complete characterization of the beam–target overlap with low- and also high-resolution x-ray spectroscopy measurements. On the one hand, low-resolution x-ray spectroscopy allows us for a measurement of the absolute x-ray emission cross section. In particular, the influence of the \( \ell \)-sublevels population on the recorded spectra is considered and discussed. Evaluation of single-electron capture cross section and contribution of multiple capture to the x-ray spectra are given. Finally a detailed analysis of the \( n = 2 \rightarrow 1 \) relative intensities observed with the high-resolution x-ray spectrometer is described and the significance of the measured hardness ratio is studied. General remarks and comments are given as conclusions in the last section.

2. Experimental setup and detection system

2.1. Description of the setup

The experiment has been performed at the low energy ion installation ARIBE\(^5\) [23] at GANIL\(^6\) where 255 keV \( \text{Ar}^{17+} \) ions are directed onto a gaseous jet of neutral Ar and \( \text{N}_2\). The optic equipment of the ion beamline is described in [24]. The ion charge selection is ensured by two dipoles placed after the ion beam extraction from the ECR ion source and upstream to the interaction chamber. A schematic view of the experimental setup is presented in figure 1, showing the arrangement of the x-ray spectrometers, the charge-coupled device (CCD) camera and the gaseous jet target around the interaction zone. Lyman lines (in the 3.1–4.1 keV energy range) from the ion projectile are recorded by two silicon drift detectors (SDD) placed respectively at \( +30^\circ \), \(+120^\circ \) from the ion beam axis, and by a high-resolution high-transmission Bragg crystal spectrometer located at \( -30^\circ \). To maximize the resolving power of our crystal spectrometer, the ion beam is vertically focused to 1–3 mm and limited to a beam width of 10–15 mm by reducing the beam emittance. The beam

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\(^5\) French acronym for ‘Accelerateur dedicated to research with low energy ions’.

\(^6\) Grand Accélérateur National d’Ions Lourds, Caen, France.
profile is regularly monitored detecting, with a high-sensitivity CCD camera, the fluorescence light from the ion impact on a stainless steel or alumina target (see figure 2 (left) as an example). A Faraday cup installed after the crossing zone and coupled to a current integrator measures continuously the beam current. A maximum intensity of 2 nA was obtained.

The gaseous jet crosses the ion beam at 90° and is obtained by gas diffusion through a capillary into the interaction chamber. Of effusive type, this gas source, widely used in several previous experiments, has been well characterized [25–27]. Atomic density and flow are determined by the geometry of the jet, i.e. the length L and the diameter d of the capillary, and by the thermodynamics conditions, i.e. the temperature and the backing pressure p of the gas. In our case, the characteristic dimensions are \( L = 35 \text{ mm}, \ d = 0.1 \text{ mm}, \) and working at room temperature, \( p = 0.5 – 5 \text{ mbar}. \) These conditions correspond to the so-called opaque intermediate regime for which the typical dimensions of the capillary are comparable to the gas mean free path and, more specifically, the Knudsen numbers are \( K_n \lesssim 1 \) and \( K_d < 1 \) [25, 26]. In this case, inter-atomic/molecular and wall-atom/molecule collisions inside the capillary play a role on the density profile at the exit of the capillary resulting in the so-called intermediate regime between the viscous and molecular flow. For a backing pressure \( p = 1.5 \text{ mbar}, \) it leads, at the crossing point with the ion beam (i.e. 6 mm from the capillary exit), to an atomic density of about \( 5 \times 10^{12} \text{ cm}^{-3} \) and a spatial extension of 8 mm (corresponding to an angular opening of \( 35^\circ \)).

Finally it is worth mentioning that the residual gas pressure has an influence on the detected x-ray signal through the production of metastable states in the ion beam interacting with the gaseous jet. Two contributions have to be taken into account, namely the residual vacuum in the beam line, \( 10^{-7} \text{ mbar} \) over a distance of 350 cm (the distance between the last dipole and the entrance of the interaction chamber), and within the interaction chamber itself, \( 10^{-6} \text{ mbar} \) over 20 cm (the radius of the interaction chamber) when the gaseous target is ‘on’.

### 2.2. X-ray spectrometers and experimental conditions

The two SDDs (XFlash model detector from ROENTEC and AXAS model detector from KETEK), having been extensively studied in terms of efficiency over their whole range of energy detection [28], are used to determine the absolute x-ray emission cross sections. Their efficiency (including the quantum efficiency and the window transmission) in the energy range of interest is close to 1. They are respectively \((91.70 \pm 0.35\%)\) and \((86.6 \pm 1.0\%)\) at 3 keV. They are placed at an observation angle of 30° and 120° with respect to the ion beam and are equipped with long specifically designed collimators in which a diaphragm is inserted for background reduction purpose (see figure 1). This collimation system gives rise to a solid angle of \( 8 \times 10^{-3} \text{ sr} \) and \( 1.6 \times 10^{-3} \text{ sr} \) for the detector placed at 30° and 120°, respectively. These observation angles have been chosen, on the one hand, to monitor the crystal spectrometer (the SDD at 30°, see the next paragraph) and, on the other hand, at an angle to be nearly insensitive to the possible polarization of the emitted x-rays (120° close to the magic angle). The SDDs record the complete series of Lyman transitions with an energy resolution of typically 190 eV and 210 eV at 3 keV for the SDDs placed at 30° and 120°, respectively.

A Bragg crystal spectrometer is used to reach a much higher resolution. Details about its principle of operation are given in [29]. Briefly summarized here, high transmission is achieved by using a highly oriented pyrolitic graphite (HOPG) crystal with a mosaic spread \( 2\alpha = 0.4°, \) and a large surface \((60 \times 60 \text{ mm}^2)\) position-sensitive multi-wire gas detector. The spectrometer is used in a vertical geometry and placed at an angle of 30° in order to be insensitive to the possible polarization of the emitted x-rays (12° close to the magic angle). The SDDs record the complete series of Lyman transitions with an energy resolution of typically 190 eV and 210 eV at 3 keV for the SDDs placed at 30° and 120°, respectively.

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limited to 1–3 mm (as mentioned above), while the spatial resolution of the position-sensitive detector, is better than 500 μm. Finally, depending on the Bragg angle value chosen for the crystal, a resolution power of about 1000 has been achieved at 3.1 keV (i.e., a resolution of 3.2 eV), and better than 500 on an energy range around 4 keV (i.e. a resolution of around 7.2 eV). Typically, the accessible range at a given setting is about 160 eV. Consequently all the He-like 1snp → 1s2 transitions generated during the collision are resolved as well as the fine structure of n = 2 → 1 transitions from He- and Li-like ions. The detection efficiency ratio between the Bragg angle chosen and is less than 2 counts s⁻¹.

During the data acquisition, background measurements are performed with the gas jet ‘off’, and with or without the ion beam in the chamber. In the case of the SDD, the major source of background is due to the interaction between the Ar⁺ ions and the residual vacuum in the chamber. A typical background spectrum gives rise in fact to a physical signal very similar to the x-ray Lyman lines recorded with the gas jet ‘on’ but its contribution is reduced to around 2%. In the case of the crystal spectrometer, in contrast, the background is mainly due to the hard x-ray residual radiation produced by the ion source. It leads to a smoothed shape spectrum whatever the signal very similar to the x-ray Lyman lines observed will be discussed in section 4. The data were pre-analysed and recorded with a GANIL-type acquisition⁷ consisting of a series of NIM and VME modules coupled to a computer. The dead time of the acquisition was constantly monitored by a pulse signal of well-defined rate connected to the pre-amplifier of the detectors. Additional details of the experimental set-up and the acquisition system can be found in [24, 31].

3. Absolute x-ray emission cross sections from low-resolution spectra

A typical x-ray spectrum from the SDD is presented in figure 3. Two major peaks are clearly visible in the energy region from 3 to 3.8 keV. As in previous works [1, 19], they are assigned to the 1s2p → 1s2 (high intensity line) and 1s3p → 1s2 transitions. The broad peak at higher energy is due to 1snp → 1s2 transitions with n > 3.

Quantitative peak intensities have been extracted by fitting the observed lines with a series of Gaussian profiles convoluted to an error function corresponding to the charge collection loss in the SDD detectors although its contribution is relatively small in the 3–4 keV energy region. For the extraction of the absolute value of the x-ray emission cross section, the ion beam and the gas-jet profiles have to be considered to evaluate precisely the target–projectile overlap.

3.1. Calculation of the overlap between the ion beam and gas-jet target

We based our calculation on the method developed in [32]. In our case, we consider the coordinate systems presented in

http://wiki.ganil.fr/gap.

![Figure 3](image-url)

**Figure 3.** Typical spectrum (with background subtracted) recorded by one of the solid-state detectors exhibiting transitions from Ar⁺ ions produced after collision with the argon gaseous target (p = 1.5 mbar). Experimental data are in red and the fit result is represented by the solid black line. For the sake of clarity, the different n > 3 → 1 transition contributions are shown (solid coloured lines) in the zoom inset; their relative intensities are deduced from the analysis of the high-resolution spectra (section 4).
from equation (3). The observed deviation from this curve at high targets. The solid grey line corresponds to the expected emission pressure indicates that multiple-collision starts to play a role.

Dependence and reduces to a pure Poiseuille law (p² corresponding to viscous flow) at large pressure values. As can be observed, equation (3) reproduces well the experimental data evolution for pressures up to 3 mbar (continuous grey line in the figure), where single collision condition is fulfilled. In this region, the data obtained allow us to extract with great precision the absolute value of the x-ray emission cross section, \( \sigma_{\text{x-ray}} \) (the only adjustable parameter in equation (3)), found to be \( 11.4 \times 10^{-15} \text{ cm}^2 \pm 15\% \). The experimental point at \( p = 3 \text{ mbar} \) can obviously be affected by multiple capture contribution, but its exclusion, from the fit, increases only by 1.7% the extracted cross section value. Furthermore, spectra recorded with the two SDDs lead, within the error bars, to the same value. It compares well with the unique previous experiment on this system performed by Tawara et al [1]. It can be noted that the uncertainty has been improved here by more than a factor of 2 and without referring to any external calibration.

Finally, the number of detected photons \( N_{2p} \) and \( N_{3p-np} \) of the transitions \( 1s2p \rightarrow 1s^2 \) and \( 1snp \rightarrow 1s^2 \) with \( n \geq 3 \), taking into account the efficiency \( \epsilon \) of the detector for the respective x-ray energy.

For our calculation, we take

\[
N_X = \frac{N_{2p}}{\epsilon_{2p}} + \frac{N_{3p-np}}{\epsilon_{3p-np}}. \tag{4}
\]

### 3.2. Extraction of the x-ray cross section values

Measurements are performed with two kinds of target gases: molecular nitrogen and argon. For both cases, systematic studies at different backing pressures \( p \) are performed to determine the single collision condition regime.

The evolution of the x-ray rate \( N_X(p)/(IT) \) is presented in figure 4. Theoretically [26], at low pressure, it displays a linear \( p \) dependence and reduces to a pure Poiseuille law \( (p^2 \) corresponding to viscous flow) at large pressure values. As we can observe, equation (3) reproduces well the experimental data evolution for pressures up to 3 mbar (continuous grey line in the figure), where single collision condition is fulfilled. In this region, the data obtained allow us to extract with great precision the absolute value of the x-ray emission cross section, \( \sigma_{\text{x-ray}} \) (the only adjustable parameter in equation (3)), found to be \( 11.4 \times 10^{-15} \text{ cm}^2 \pm 15\% \). The experimental point at \( p = 3 \text{ mbar} \) can obviously be affected by multiple capture contribution, but its exclusion, from the fit, increases only by 1.7% the extracted cross section value. Furthermore, spectra recorded with the two SDDs lead, within the error bars, to the same value. It compares well with the unique previous experiment on this system performed by Tawara et al [1]. It can be noted that the uncertainty has been improved here by more than a factor of 2 and without referring to any external calibration.

In addition, as seen in figure 4, no noticeable differences, within the error bars, are observed when using argon or molecular nitrogen gaseous targets. This is easily explained considering the very close ionization potentials of the less bound electrons: \(-15.8 \text{ eV for the argon } 3p \text{ electrons, and } -15.5 \text{ and } -16.8 \text{ eV for the } N_1 \text{ } 1\pi_g \text{ and } 3\sigma_g \text{ electrons, respectively.} \) In those states, six electrons are available in both cases. Moreover, the typical capture radius is around \( RC = 7 \text{ Å} \) (from classical over-barrier predictions) that is larger than the size of the bi-nitrogen (1 Å) making molecular nitrogen appear very similar to the atomic argon target for the incoming ions.

For higher pressures, multi-collisions start to take place and as expected, the x-ray rate decreases considerably, since the multiple-excited states which are populated decay mainly by autoionization. In that respect, and to fulfill the ‘single collision condition’, data acquisition was preferentially performed at a backing pressure of around \( p = 1.5 \text{ mbar} \) to extract reliable results.

### 4. Analysis of single and multi-electron capture contribution from high-resolution x-ray spectra

With the high-resolution spectrometer, an efficient zoom of the SDD spectra is achievable, as shown in figure 5. A record of the full series of Lyman lines requires us to set the spectrometer at a minimum of three different Bragg angles. To ensure sufficient statistics in each peak of the line series, typical acquisition times of a few thousand seconds are required for de-excitation of states up to \( n = 4 \) (i.e. for the 3.0–3.9 keV energy region), while at least twenty thousand s are needed for the end-series region from \( n = 5 \) to 10 (i.e. for the energy range centred around 4.05 keV). The whole spectrum, as displayed in figure 5, exhibits the He-like \( 1snp \rightarrow 1s^2 \text{ } 1S_0 \) transitions extremely well resolved from \( n = 2 \) to 10. We note that for \( n \geq 3 \) only the singlet \( 1snp \rightarrow 1p_1 \rightarrow 1s^2 \text{ } 1S_0 \) state decays to the ground state while triplet states decay almost exclusively to the metastable state \( 1s2s \text{ } 3S_1 \) [35, 36]. At low energy, for \( n = 2 \), the He-like \( 1s2p \rightarrow 1p_2 \rightarrow 1s^2 \text{ } 1S_0 \) and \( 1s2p \rightarrow 1p_1 \rightarrow 1s^2 \text{ } 1S_0 \) transitions are observed and well separated. M1 \( 1s2s \text{ } 3S_1 \rightarrow 1s^2 \text{ } 1S_0 \) and Li-like \( 1s2s2p \rightarrow 1p_1, 2/2 \rightarrow 1s^22s \text{ } 3S_{1/2} \) transitions are also observable and will be discussed in section 4.4.

#### 4.1. Analysis of the line intensities

Quantitative values of the transition intensities are obtained from a fit of the lines using a series of Voigt profiles that reproduce well the response function of the crystal spectrometer over the whole energy range studied. Under our experimental conditions, mentioned before, the resolution is primarily driven by the optical quality of the ion beam and the spatial resolution of the detector (i.e. the role of the mosaic spread is negligible). The corresponding width of the Gaussian and Lorentzian terms are kept constant for the energy range explored at a given Bragg angle but their variation, fully under control, is taken into account when varying this angle. In addition, in order to check the reproducibility of the results and to improve the uncertainty on the relative \( np \) line intensities, we have also recorded overlap spectra, e.g. transitions from \( n = 4 \) to 7 for instance.
Finally, the background contribution, in terms of intensity and shape has also been precisely determined by recording the same spectra (for each Bragg angle) without the ion beam in the chamber, since, as mentioned before, the background is mainly due to the residual radiation produced by the ion source (see section 2.2). Therefore, we have been able to fit the background contribution by a polynomial curve. Its integral over the considered energy range is normalized to the counts recorded by the SDD at 30° (see the text).

4.2. Analysis of the \( n = 7 - 10 \to 1 \) transitions: single electron capture

Keeping in mind that spectra shown in figure 5 reflect ‘only’ the \( p \) state populated either through direct capture or by cascades, it clearly comes out that the preferential capture level of the Ar\(^{17+} \) ion at low impact velocity (0.53 au) is \( n_{\text{pref}} \sim 8 - 9 \). This is in good agreement with the simple classical over-barrier model [8], which predicts \( n_{\text{pref}} = 7 - 9 \) for the single-electron capture process from the 3s or 3p Ar states. Multi-electron capture populates preferentially multiple excited states lying in lower \( n, n', n'' \ldots \) values than single capture [37]. Therefore, assuming that x-ray lines from \( n p \) levels follow almost exclusively single-electron capture, the partial \( \{ P_n \} \) of single capture can be extracted directly from the measured transition intensities. In fact, the measured intensities reflect only \( 1snp^1P_1 \) state populations decaying to the ground state. Consequently, to get the population of the \( np \) levels, a factor of 4 is introduced, assuming a statistical population between \( 3P_1^1P \) (i.e. corresponding to a ratio of 3:1). The \( \{ P_n \} \) can thus be obtained considering...
different $\ell$-distributions $\{P_{\ell}\}$ population of the sublevels and taking into account the atomic cascade between levels. For this purpose, non-relativistic H-like atom branching ratios are considered [38].

The results corresponding to the Ar target are presented in figure 6. It is well known that at an ion velocity of 0.53 au the $\ell$-sublevel distribution is most likely of statistical type ([39], or more recently [40, 41], for example). Nevertheless to probe the sensibility of the extracted $\{P_{\ell}\}$ on the choice of the $\ell$-distribution, we have assumed two extreme cases: a flat and a statistical distribution. One can observe that the different assumptions on $\{P_{\ell}\}$ do not affect significantly the results, which stay within the error bars. We can conclude that the single-electron capture occurs in a reaction window that populates preferentially $n = 8 - 9$ states with a relative narrow distribution in agreement with the Landau–Zener model [9] and as observed by Knoop et al in [14], where the COLTRIM technique for Ar$^{17+}$ on He at $v = 0.4$ au has been used.

The accurate knowledge of the relative transmission of the Bragg spectrometer, compared to the SDD detector enables us to extract the absolute value of the single-electron capture cross section $\sigma_{\text{single}}$ occurring in $n = 7 - 10$. Contrary to the $\{P_{\ell}\}$, $\sigma_{\text{single}}$ depends strongly on the choice of the $\ell$-distribution. Indeed the extracted cross section value ranges from $\sigma_{\text{single}} = 4.6 \cdot 10^{-15}$ cm$^2$ (flat) to $12.8 \cdot 10^{-15}$ cm$^2$ (statistical). The result found can be satisfactorily compared to the value of about $8 \cdot 10^{-15}$ cm$^2$ obtained by Ali et al [16] from coincidence measurements of projectile–target charge exchange for Ar$^{17+}$ on Ar at $v = 0.6$ au. Note that those values are also in good agreement with the classical over-barrier model [42] that predicts $\sigma_{\text{single}}$ of the order of $0.5\pi R_c^2 = 6.6 \cdot 10^{-15}$ cm$^2$.

4.3. Analysis of the $n = 2 - 6 \rightarrow 1$: contribution from multi-electron capture processes

As mentioned above, multi-electron capture populates preferentially lower levels than the single capture. These multi-excited states decay preferentially by Auger emission followed by a possible x-ray transition that is indistinguishable from the emission resulting through cascade from single-electron capture.

Once the single capture population probabilities in $n = 7 - 10$ are extracted, the influence of the multi-electron process can be evaluated. Considering a given $\ell$-distribution, the population of the different $n\ell$ levels by single capture for $n < 7$ is uniquely determined from the $\{P_{\ell}\}$. Indeed, the related x-ray emission can be accurately calculated. In addition to the previously mentioned assumption on the atomic cascade calculation, branching ratios are introduced independently for triplet and singlet states down to $n = 2$. For $n = 2$, 1s2p $\rightarrow$ 1s2s, 1s$^2$ decays, the complete de-excitation scheme (see figure 7) is taken into account, including coupling between triplet and singlet states (i.e. calculations from [35, 36]). The resulting calculated line intensities coming from single capture in $n = 7 - 10$ are presented in figure 8 for the two extreme $\ell$-distributions (flat and statistical). Whatever the $\{P_{\ell}\}$, an intensity deficit is observed from $n = 3 - 6$ when...
He-like Ar 1s2p 3P1 and 1s2s 3S1 transitions; \( R_{n/\ell}^{\text{corr}} \) and \( R_{n/\ell}^{\text{exp}} \) refer to the ratio between the He-like Ar 1s2s 1S1 \( \rightarrow \) 1s2p 3P1 and 1s2p 1P1 \( \rightarrow \) 1s2 1S0 transitions and with and without taking into account the correction due to the residual vacuum, respectively. \( R_{\ell/\ell}^{\text{corr}} \) refers to the ratio between the Li-like 1s2p64P \( \rightarrow \) 1s2s5S/2 and the He-like Ar 1s2s 3S1 \( \rightarrow \) 1s2 1S0 transitions. Model predictions do not include multielectron.

### Table 2. Recorded relative intensities for \( n = 2 \rightarrow 1 \) transitions for Ar+ colliding with argon gas. \( R_{\ell/\ell}^{\text{exp}} \) refers to the ratio between the He-like Ar 1s2p 4P1,2 \( \rightarrow \) 1s2 1S0 and 1s2p 4P1 \( \rightarrow \) 1s2 1S0 transitions; \( R_{n/\ell}^{\text{exp}} \) and \( R_{n/\ell}^{\text{corr}} \) refer to the ratio between the He-like Ar 1s2s 3S1 \( \rightarrow \) 1s2p 3P1 and 1s2p 4P1,2 \( \rightarrow \) 1s2 1S0 transitions with and without taking into account the correction due the residual vacuum.

| Intensity ratio | Experimental values | Model for initial \( \ell \)-distribution |
|----------------|---------------------|------------------------------------------|
| \( R_{\ell/\ell}^{\text{exp}} \)     | 1.91 ± 0.03         | 1.82 1.91                                |
| \( R_{\ell/\ell}^{\text{corr}} \)   | 0.076 ± 0.002       | 0.040 0.053                              |
| \( R_{\ell/\ell}^{\text{corr}} \)   | 0.076 ± 0.002       | 0.075 0.101                              |
| \( R_{\ell/\ell}^{\text{corr}} \)   | 0.048 ± 0.001       | 0.047 0.065                              |

Comparing the simulated spectrum to the experimental one. This difference indicates clearly that multi-electron capture contributes significantly starting from the n = 6 level. Moreover, we note that the calculated intensity of the 1s2p \( \rightarrow \) 1s2 transition, which represent 88% of the total recorded x-ray emission (see table 1), is very sensitive to the choice of \( \{P_\ell\} \). Indeed, in the case of statistical distribution, population of high \( \ell \) is favoured. Such levels decay preferentially via the Yrast cascade (namely transitions from \( n, \ell = n-1 \) to \( n', \ell' = \ell - 1 \)), which results in a consequent intensity enhancement of the 1s2p \( \rightarrow \) 1s2 transition compared to a flat \( \ell \)-distribution. The role of the multi-electron capture can be evaluated for each \( \{P_\ell\} \) and we find that within a flat \( \ell \)-distribution, the single-electron capture can explain only 22% of the total x-ray emission while it reaches 71% for a statistical \( \ell \)-distribution.

#### 4.4. Detailed analysis of the \( n = 2 \to 1 \) relative intensities observed with the x-ray spectrometer and relevance of the measured hardness ratio

The high-resolution power of the spectrometer allows for separating different Ar He-like 1s2e \( \rightarrow \) 1s2 components as well as He- from Li-like x-ray transitions. As observed in figure 5, emission lines from triplet and singlet 1s2p P states can be well distinguished. The M1 transition from the metastable state 1s2s 1S1 to the ground state and the Li-like lines are also well resolved. The relative intensities between these different x-ray lines deserve a detailed analysis. First, they allow for checking the consistency of the cascade model used and the estimated contribution of metastable 1s2s Ar16+ ion beam. Second, they can be used to refine the reliability on the hardness ratio \( H \) that can be deduced from such measurements. All the values are summarized in table 2.

(a) Experimentally, we find an intensity ratio between 1s2p 4P1,2 \( \rightarrow \) 1s2 1S0 and 1s2p 4P1 \( \rightarrow \) 1s2 1S0 transitions equal to \( R_{\ell/\ell}^{\text{exp}} = 1.91 \pm 0.02 \). Applying the atomic cascade model from the single-electron capture, as described in the previous section, leads to very close values of \( R_{\ell/\ell}^{\text{corr}} = 1.82 \) and 1.91 assuming a statistical or flat \( \ell \)-distribution in the \( n = 7 \to 10 \) levels, respectively.

(b) The Li-like argon transition line observed in figure 5 comes from the de-excitation of the 1s2s2p states to the ground state. Those levels can be produced in the gaseous jet by single electron capture either from the 1s2s 3S1 Ar16+ metastable ions in the incident beam or by multiple-electron capture from the primary 1s Ar17+ ion beam. The metastable ion beam contribution can be evaluated taking into account the collision probability on the residual gas in the beam line after the last dipole magnet and before the entrance of the collision chamber as well as within the collision chamber itself before the interaction zone (see section 2.1). It gives rise to a contribution of 10% that has been indeed confirmed through a measurement of the Lyman x-ray line ratio from Ar16+/Ar17+ when using Ar18+ as the incoming ion beam. Through the capture and cascade processes, the Li-like 1s2s11L and 1L levels are populated but x-ray emission from the doublet states are strongly suppressed by autoionization during cascade processes [43]. Hence, mostly the 1s2s2p 4P1,2 \( \rightarrow \) 1s2s 3S1/2 transition for is observed on our x-ray spectra, in agreement with previous experiments performed with lighter ions [44, 45]. From the metastable ion contribution in the incoming beam, the intensity ratio between the Li-like 4P and the He-like P transitions \( R_{4P/3P}^{\text{exp}} \) can be estimated around 0.05, which is in good agreement with the experimental value \( R_{4P/3P}^{\text{exp}} = 0.048 \pm 0.001 \). Let us note that x-ray emission from the 4P states are fully visible by the spectrometer, contrary to the M1 transition, since they have a short lifetime \( 2 - 6 \times 10^{-12} \) s [35, 46].

(c) The intensity ratio \( R_{3S/3P}^{\text{exp}} \) between the 1s2s 3S1 \( \rightarrow \) 1s2 1S0 M1 and the 1s2p (4P + P) \( \rightarrow \) 1s2 1S0 transitions can also be predicted from the atomic cascade simulation. Due to the forbidden transitions from 1snp 3P states, the 1s2s 3S1 level is efficiently populated and a value of \( R_{\ell/\ell}^{\text{exp}} \) ranging from 0.87 (statistical distribution) to 1.18 (flat distribution) can be expected. These values appear at first quite far from the experimental measured ratio \( R_{\ell/\ell}^{\text{exp}} \) = 0.076 ± 0.002. This large difference is in fact due to the long lifetime, 0.209 \( \mu \)s, of the 1s2s 3S1 metastable state [47, 35, 46]. This corresponds, at a projectile velocity of 0.53 au, to a decay length of 24.5 cm while a spatial window of only \( \sim 2 \) cm around the centre of the gaseous jet is viewed by the x-ray spectrometer. As a result, only about 4.5% of the total M1 emission is detected compared to 100% for the 1s2p \( \rightarrow \) 1s2 transitions. An intensity ratio \( R_{\ell/\ell}^{\text{exp}} \) ranging from 0.040 (statistical distribution) to 0.053 (flat distribution) is thus expected. When the capture from the residual vacuum is included, as discussed previously, such values go up to 0.075 (statistical distribution) and 0.101 (flat distribution), which is in good agreement with the observed ratio, keeping in mind that the contribution of multi-electron capture is neglected.

(d) In the case of He-like transitions, the relative long lifetime of 1s2s 3S1 M1 level also has important consequences on the relevance of the measured hardness ratio \( H \), as already pointed out by Tawara et al [1]. Indeed, transposition of
Figure 9: Measured hardness ratio values $\mathcal{H}$ (with one-sigma error bar) for Ar$^{17+}$ + Ar compared with previous experiments (Tawara in 2001 [1], Tawara in 2006 [18], Allen in 2008 [19], Beierdorfer in 2000 [17].) The values of Allen [19] at $E_{\text{ion}} = 6.4$ eV/u and Beierdorfer [17] correspond to measurements obtained in magnetic trapping experiments. The others are obtained with extracted ion beams. The empty circle correspond to $\mathcal{H}_{\text{corr}}$, the corrected value of $\mathcal{H}$ to take into account the partial detection of the 1s2s $^3S_1$ to $^1S_0$ M1 transition (see the text). The horizontal solid line corresponds to an $\mathcal{H}$ value expected assuming a statistical population $\{P_i\}$.

the $\mathcal{H}$ value measured via ‘laboratory ion–atom collisions’ towards interpretation of spectra from comets and solar wind should be made with caution. We remind that the hardness ratio $\mathcal{H}$ is the ratio between the unresolved $n > 2 \rightarrow 1$ and the $2 \rightarrow 1$ transitions. From our experiment, the measured $\mathcal{H}$ values are found to be $0.106 \pm 0.005$ and $0.109 \pm 0.005$ with the two SDD detectors at 120° and 30°, consistent with previous measurements performed in the same experimental conditions as shown in figure 9. Nevertheless, great care has to be taken when dealing with x-ray spectra observed from a comet or solar wind for which 100% of the He-like M1 transition is visible. With our experiment, we have recorded only a fraction (4.5% as mentioned above) of this transition. In addition, part of the x-ray lines recorded is coming from collisions on the residual pressure, which very much depend upon the experimental conditions. Consequently the experimental hardness ratio has to be corrected and it can be done here, thanks to the precise measurements obtained with the crystal spectrometer as follows. Starting from values reported in tables 1 and 2, the measured hardness ratio can be evaluated as $10.95/\left(89.0 \times (1 + 0.076 + 0.048)\right) = 0.109$. Keeping in mind that, as mentioned above, the $^4P$ Li-like and the M1 He-like transitions are entirely or partially due to capture on the residual pressure that produces a fraction of the ion beam in the metastable state 1s2s $^3S_1$ before the ion–atom interaction zone itself, the corrected hardness ratio is given by $10.95/\left(89.0 \times (1 + 0.047/0.045)\right) = 0.060 \pm 0.007$, where 0.047 refers to the average value of $R_{\text{S}(3P+1P)} = 0.040 - 0.053$. Let it be mentioned that although the He–Lyman lines are also affected by capture on the residual pressure (a contribution of 15%—see section 3.1), this term vanishes in the hardness ratio. Finally, it leads to a correction factor of $0.55 \pm 7.4\%$. When considering ions of different Z value than argon, e.g. C, N, O, Ne, Mg and Si, which have been identified at the origin of the x-ray emissions from the interstellar medium [21], attention has to be payed since the correction factor depends on the metastable state lifetime in addition of the dependence on the collision velocity (through the decay length and the $\{P_i\}$). Finally, in this respect also, comparing measurements performed with extracted ions of typically a few keV/u and trapped ions as in EBIT experiments is not trivial. As observed in figure 9, for the Ar$^{17+}$ incident beam, the corrected $\mathcal{H}_{\text{corr}}$ value we found approaches to the statistical limit as expected at the relative high velocity of the slow collision energy regime.

5. Conclusions

Using low- and high-resolution spectroscopy, projectile x-ray emission is studied for (quasi) symmetric heavy collision systems at low energy, where Ar$^{17+}$ ions collide with Ar and N$_2$ targets at $v = 0.53$ au. The determination of the ion beams–gaseous jet overlap, coupled to a perfect knowledge of our SDD detectors efficiencies, allows us to extract the absolute x-ray emission cross section, found to be $11.4 \cdot 10^{-15}$ cm$^2$ ±15% in agreement with the only previous experiment on this system performed by Tawara et al [1] but with an improvement in the experimental uncertainty by more than a factor of 2. Moreover, with our direct method of measurement, contrary to [1], no reference cross section is needed.

Differently from experiments where low-resolution x-ray spectroscopy were employed [17, 19], the high resolving power of the crystal spectrometer enables us to have access to additional information on single- and multi-electron processes without applying coincidence with target charge state detection. In our case, we are able to resolve the whole He-like Ar$^{16+}$ Lyman series from $n = 2$ to 10 as well as the fine structure of $n = 2 \rightarrow 1$ transitions. For each $n$ value, the relative $1snp \rightarrow 1s^2$ intensity is obtained showing that the $1s2p \rightarrow 1s^2$ transition represents almost 90% of the total x-ray emission whatever the gaseous target (Ar or N$_2$). From the analysis of the $n = 7-10 \rightarrow 1$ line intensities, the preferential $\sigma_{\text{single}} = 4.6 \cdot 10^{-15}$ cm$^2$ (flat distribution) and $12.8 \cdot 10^{-15}$ cm$^2$ (statistical distribution). With the knowledge of $\{P_i\}$, the multi-electron capture can also be evaluated through the comparison between the experimental data and the expected intensities of the $n = 2-6 \rightarrow 1$ lines from single-electron capture using a cascade code. Quantitatively,
we find that the single-electron capture can explain only 22% of the total x-ray emission with a flat \{P_\ell\} while it reaches 71% for a statistical \{P_\ell\}. Those findings provide a stringent test on the consistency of a given theoretical description able to account for single- and multiple-capture processes in such symmetric systems when it will be available.

Finally, the analysis of the intensities of the different He-like 1s2\ell \rightarrow 1s^2 transitions (involving the 1s2p^1P and 3P states and the metastable 1s2 3S_1 state) as well as the Li-like 1s2s2p^4P \rightarrow 1s^22s transitions allows us to control the contamination from metastable Ar^{16+} ions in the incoming ion beam. The consistency of our cascade calculations is checked since all the evaluated relative peak intensities are found in rather good agreement with the experimental ratios. This fine analysis argues discussing on the hardness ratio \overline{\eta}_n, commonly used to analyse low-resolution x-ray spectra. In particular, we point out that special care has to be taken when experiments on the same collision system but with different setups are compared. Indeed, the hardness ratio is reduced by a factor of 2 in the case presented in this paper, mainly due to the partial detection of the 1s2 3S_1 metastable state decay.

In conclusion, it is worth mentioning that accurate determination of \eta_{sec} and \{P_\ell\} through x-ray measurements are achievable only with a high-resolution spectrum, where the different transitions can be well separated. Due to the dependence of the \{P_\ell\} on the single capture process, only rough values can be obtained for single-capture cross section and multiple-capture contribution in x-ray spectra. However, high-resolution spectroscopy can be advantageously used to test the consistency of a full calculation of the different processes involved in such symmetric collisions. The achievement of a more quantitative estimation requires to apply coincidence techniques between high-resolution x-ray spectroscopy and ion charge state detection, or at least to perform simultaneously COLTRIM-type experiments, which could be done in the future.

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