Understanding the $L$-shell ionization mechanism through osmium atoms bombarded by 4-6 MeV/u fluorine ions

Soumya Chatterjee, Sunil Kumar, Sarvesh Kumar, M Oswal, Biraja Mohanty, D. Mehta, D. Mitra, A. M. P. Mende, D. M. Mitnik, C. C. Montanari, L. Sarkadi, and T. Nandi

1Department of Physics, University of Kalyani, Kalyani, West Bengal-741235, India.
2Goet. Degree College, Banjar, Kullu (HP) 175123, India.
3Atomic and Molecular Collisions Laboratory, CEFITEC, Department of Physics.
4Department of Physics, Dev Samaj College for Women, Sector 45 B, Chandigarh-160014, India.
5Department of Physics, Panjab University, Chandigarh-160014, India.
6Instituto de Astronomía y Física del Espacio, CONICET and Universidad de Buenos Aires, Buenos Aires, Argentina
7Institute for Nuclear Research of the Hungarian Academy of Sciences (MTA Atomki), H-4001 Debrecen, Pf. 51, Hungary.
8* Inter-University Accelerator Centre, Aruna Asaf Ali Marg, Near Vasant Kunj, New Delhi-110067, India.

The $L$-subshell ionization mechanism is studied in an ultra-thin osmium target bombarded by 4-6 MeV/u fluorine ions. Multiple ionization effects in the collisions are considered through the change of fluorescence and Coster-Kronig yields while determining $L$-subshell ionization cross sections from $L$-line x-ray production cross sections. The $L$-subshell ionization, as well as $L$-shell x-ray production cross sections so obtained, are compared with various theoretical approximations. The Coulomb direct ionization contributions is studied by (i) the relativistic semi-classical approximations (RSCA), (ii) the shellwise local plasma approximation (SLPA), and (iii) the ECUSAR theory, along with the inclusion of the vacancy sharing among the subshells by the coupled-states model (CSM) and the electron capture (EC) by a standard formalism. We find that the ECUSAR-CSM-EC describes the measured excitation function curves the best. However, the theoretical calculations are still about a factor of two smaller than the measured values. Such differences are resolved by re-evaluating the fluorescence and the Coster-Kronig yields. This work demonstrates that, in the present energy range, the heavy-ion induced inner-shell ionization of heavy atoms can be understood by combining the basic mechanisms of the direct Coulomb ionization, the electron capture, the multiple ionization, and the vacancy sharing among subshells, together with optimized atomic parameters.

I. INTRODUCTION

The measurement of emitted x-rays from targets has resulted in major advances in radiation physics, plasma physics, atomic and nuclear physics, and the particle-induced x-ray emission (PIXE) technique. Thus far, the PIXE method has used light ions such as protons or alphas; however, there is an increasing interest to employ heavy ions since their ions such as protons or alphas; however, there is an increasing interest to employ heavy ions since their cross sections are larger and have, thereby, better sensitivity. Nevertheless, this potentiality is discouraged by discrepancies observed between the theories and experiments. Although these inconsistencies are often attributed to multiple ionization phenomena, they do not account for all the discrepancies observed, for example, in experiments with an 8-36 MeV Si-ion beam on targets of Au, Bi, Th, and U with thicknesses between 12 and 40 $\mu g/cm^2$. On such occasions, theoretical approaches have been modified to include the $L$-subshell coupling effect as well as the saturation of the binding effect at the united atom limit in addition to the multiple ionization. Even though closeness between the experiments and theory is achieved, differences remain, suggesting that other physical processes are involved.

It is well-known that for asymmetric collisions, $Z_p/Z_T < 1$, the direct ionization (DI) is dominant, whereas for symmetric collisions, $Z_p/Z_T \approx 1$, the electron capture (EC) process becomes increasingly important. Although the present collisional system –F ions ($Z_p = 9$) impinging on Os ($Z_T = 76$) is asymmetric ($Z_p/Z_T = 0.1184$), the measurements cannot be accounted for without considering the EC contribution to the $L$-shell vacancy production. This phenomenon can be understood by considering the ratio of the projectile velocity $V_p$ to the orbital velocity of the $L$-shell electrons $V_L$. In our full-relativistic calculations, the mean velocities (in a.u.) of the osmium $L_i$ sub-shells are $v_i = 26.6, 37.7$, and $32.8$ for $i = 1, 2, 3$, respectively. Therefore, $0.33 \leq V_p/V_L \leq 0.58$, and the collision is asymmetric but in the slow velocity regime. Hence, we included the EC contribution along with the DI, the multiple ionization, and the vacancy sharing. However, contrary to our expectations, the EC does not account for all the discrepancies found between our measurements and the theoretical descriptions. In the final stage of this work, we found that the presently available atomic parameters are not properly described. We modified these values iteratively until a good agreement between the theory and experiment was achieved.

To reveal the $L$-shell ionization mechanism of the collisions of 4-6 MeV/u fluorion ions on a ultra thin osmium target, we provide experimental details in Section. The theoretical methods employed to describe the ionization are discussed in Section. Section addresses the ef-
fects of the single- and multiple-hole atomic parameters required for the derivation of the subshell-ionization cross sections from the measured x-ray production cross sections. Section IV describes the inclusion of ionization by the LK capture processes, and Section VI summarizes the major findings.

II. EXPERIMENTAL DETAILS AND DATA ANALYSIS

The L-shell x-ray production cross sections in the Os elements fusing the $^{19}$F ions (charge states $q = 6+$, 7+, 8+) in the 76–114 MeV energy range have been measured in the atomic physics beamline at the Inter-University Accelerator Centre, New Delhi. The heavy ions of fluorine –$^{19}$F (76 and 84 MeV), $^{19}$F$^+$ (90 MeV) and $^{19}$F$^{8+}$ (98 and 114 MeV) were obtained from the 15 UD Pelletron accelerator. The chamber has provision for two silicon surface barrier (SSB) detectors at ± 7.5° and two x-ray detectors at 55° and 125° to the beam direction, respectively. The target was mounted on a steel ladder forming a 90° angle to the beam direction. The vacuum inside the chamber was ∼ 10⁻⁶ Torr. The spot of the ion beam at the target had a diameter of approximately 2 mm. The spectra were taken at different positions of each target. Details of the experimental setup and detection system are given by Kumar et al. [21]. The ultra-thin target of 76 Os was prepared on the polypropylene backing using an ultra-high vacuum deposition setup at IUAC, New Delhi. The thickness of the target was measured using the Rutherford Back-scattering (RBS) method and its spectrum is given in Fig. 1. The target turned out to be very thin, only 1.09 µg/cm². The beam current was kept below 1 nA to avoid pile up effects and damage to the target. The spectra were collected for a sufficiently long time to get good statistical accuracy. The L x-ray spectra of natural Os bombarded by $^{19}$F$^+$ at different projectile energies (76-114 MeV) is shown in Figs. 2 and 3. The spectra were analyzed with a fitting method considering a Gaussian line shape for the x-ray peaks and a suitable background function. From the figures, it is clear that all major L x-ray components are well resolved by the Si(Li) detector. The details about the data acquisitions and the terms related to the projectile velocity are given by Oswal et al. [22].

The measured L x-ray production cross sections for the major peaks namely $L_1$, $L_α$, $L_β$, and $L_γ$ were obtained using the following relation

$$\sigma \alpha = \frac{Y_\alpha A \sin \theta}{N_A n_p t \beta},$$  

(1)

where $Y_\alpha$ is the intensity of the $\alpha$th x-ray peak, $A$ is the atomic weight of the target, $\theta$ is the angle between the incident ion beam and the target foil surface, $N_A$ is the Avogadro number, $n_p$ is the number of incident projectiles, $\epsilon$ is the effective efficiency of the x-ray detector, $t$ is the target thickness 1.09 µg/cm² and $\beta$ is a correction factor for the absorption of the emitted x-rays inside the target.

The absorption correction factor for the absorption of the emitted L x-rays in the target is written as

$$\beta = \frac{1 - \exp^{-\mu t}}{\mu t},$$

(2)

where $\mu$ is the attenuation coefficient inside the target and its unit is cm²/g [23]. The value of $\beta$ is $\geq 0.99$ for the target thickness used in the present measurements. The energy loss calculation using the SRIM code [24] for the incident beam within the target suggests negligibly small energy loss for the target thickness and the beam energies used in the present work. The ion beam changes its charge state during its passage through the target.

The role of projectile charge state in this collision regime for 4-6 MeV/u is found to be negligible [22]. Integrated charge in a Faraday cup measured by a current integrator has been used to count $N_p$ (see discussion in Section VI). The energy loss calculation using the SRIM code shows that 76 and 114 MeV fluorine ions lose 2.30 and 1.91 keV in the Os target, respectively. The peak areas $Y_\alpha$ are evaluated using the computer program CANDELE [25]. This software is an improved version of the Levenburg-Marquardt [26] non-linear minimization algorithms for the peak fitting. The energy calibration of the detector is performed before and after the in-beam measurements. A semi-empirical fitted relative efficiency curve for the present measurement is available in Oswal et al. [22].

The percentage error in the measured x-ray production cross sections is about 10–15%. This error is attributed to the uncertainties in different parameters used in the analysis, namely the photo peak area evaluation (∼ 5%)
FIG. 2: The L x-ray spectra of the Os target bombarded by $^{19}$F ions of energies 76, 84, 98, and 114 MeV. The pink lines indicate the baseline correction for the background due to Compton scattering.

FIG. 3: Same as Fig. 2 but for an impact energy of 90 MeV. Deconvoluted x-ray lines due to different transitions are shown.

for the $L_\alpha$ x-ray peak and 3% for the other peaks), ion beam current ($\sim 7\%$), target thickness ($\sim 3\%$). In the energy region of interest, the error of the absolute efficiency values $\epsilon$ ranges between 5 and 8%.

III. IONIZATION THEORIES

To calculate the direct Coulomb ionization cross section, we have employed (i) the coupled-states relativistic semi-classical approximation (RSCA-CSM), (ii) the shellwise local plasma approximation (SLPA) with fully relativistic electronic structure calculations for Os, and (iii) the ECUSAR theory, which accounts for the energy (E) loss and the Coulomb (C) deflection of the projectile, the perturbed-stationary state effect through a united and separated atom (USA) treatment of the ionization process, and by considering the relativistic (R) nature of the L-subshell electronic states. All these models are briefly described below.

A. The coupled-states relativistic SCA model (RSCA-CSM)

In the semi-classical approximation (SCA), the general form of the cross section is expressed as follows

$$\sigma_i = 2\pi \int_0^\infty b b \sum_f |a_f(t = +\infty)|^2 ,$$

where $b$ is the impact parameter, and $a_f(t)$ is the excitation amplitude of the $L_i$ sub-state to a final $f$ state. For the continuum states, the sum means integration over the electron energy. Applying the independent-particle model approximation, the many-electron excitation amplitudes are replaced by single-electron transition amplitudes.

The subshell coupling mechanism is described in a way that a “mixed” L state is considered as the initial state, instead of a “pure” atomic state [27]. The mixed state evolves in time from the initial $L_i$-substate as a result of dynamical couplings with the other $L$-substates. The time evolution is governed by the following eight coupled equations (in a.u.):

$$\frac{a_{n_L}}{t} = -i \sum_{n_L'} \gamma_{n_L n_L'} a_{n_L'} ,$$

where $\gamma_{n_L n_L'}$ is the transition matrix element.
with the initial condition
\[
\alpha_{n_L}(t = -\infty) = \delta_{n_L,i}.
\] (5)

The excitation from the mixed \(L\)-substate to the \(f\) final state is described by
\[
\frac{\partial}{\partial t} \psi_{n_L}(t) = -i \sum_{n_L} V_{fn_L} \alpha_{n_L}.
\] (6)

In Eqs. \((4)\) to \((6)\), \(n_L\) represents the quantum numbers \((l, j, m_j)\) of the \(L\) substates. The \(V_{mn}(t)\) matrix elements for the projectile-target-electron interaction are defined as
\[
V_{mn}(t) = V_{mn}(t) \exp(i\omega_{mk}t),
\] (7)
\[
V_{mn}(t) = \int \mathbf{r} \psi_m^*(\mathbf{r}) \frac{-Z_1}{|\mathbf{r} - \mathbf{R}(t, b)|} \psi_n(\mathbf{r}),
\] (8)
\[
\omega_{mk} = E_m - E_k,
\] (9)
where \(Z_1\) is the atomic number of the projectile, \(\mathbf{R}\) is the internuclear vector, and \(\psi_j(\mathbf{r})\) and \(E_j\) are the one-electron energy eigenstates and eigenvalues of the unperturbed target atom. For \(\mathbf{R}(t, b)\), a Kepler projectile orbit is applied.

In the present SCA model, the \(V_{mk}\) matrix elements are calculated using screened relativistic wave functions for both the bound and continuum states \([28]\), i.e., it is an RSCA model. Furthermore, since it includes the subshell coupling effects, the full name of the model is coupled-states RSCA, briefly: RSCA-CSM.

We stress that unlike previous works (see, e.g., Refs. \([29, 30]\)), the integration over the final states in Eq. \((3)\) is complete, and the present RSCA-CSM calculation extends to the entire range of the energy transfer. Additionally, no restrictions are imposed on the angular momentum of the ionized electron.

### B. ECPSSR/ECUSAR-CSM model

The ECPSSR model by Brandt and Lapicki \([31]\) and its evolution into the ECUSAR model \([32]\) are the most employed theories to describe inner-shell ionization cross sections. They cover an extended energy range, and are the usual input in PIXE codes \([32]\). The ECPSSR theory goes beyond the plane-wave Born approximation (PWBA) by accounting for the energy loss (E), the Coulomb deflection from a straight-line trajectory and retardation of the projectile (C) and its influence on the unperturbed and non-relativistic atomic orbitals in a perturbed stationary state (PSS) treatment that also accounts for the relativistic (R) nature of the inner shells of heavy target atom. In the ECUSAR theory of Lapicki \([32]\), the PSS treatment of ECPSSR is replaced by the united (U) and separated (S) atom (A) formula (see Eqs. \((2)\) and \((3)\) in \([32]\)).

Comparing RSCA-CSM to ECPSSR, one has to stress that the former automatically incorporates most of the effects that are included in ECPSSR as corrections. The application of Kepler orbit accounts for the Coulomb-deflection effect. The diagonal matrix elements \(V_{mk}\) determine in a first-order approximation the change of the binding energies of the \(L\) substates in the presence of the projectile, i.e., they account for the increased binding effect, which is one of the most important consequences of the PSS approach. And, of course, the electronic relativistic effects are exactly taken into account. At the same time, there are differences between the two theories (in addition to the subshell couplings). The ECPSSR model is based on the plane-wave Born approximation (PWBA). Although the PWBA and the straight-line SCA are equivalent, this feature holds only for the hydrogen atom, or hydrogenic ions. For a many-electron atom, it is known that the screening procedure is different for the two theories. It can be shown that the outer screening applied in PWBA leads to an effective potential that approaches the many-electron potential better than the corresponding effective potential in SCA. Furthermore, the energy loss effect is not included in the RSCA-CSM, i.e., one does not expect a good performance of the model at very low collision velocities.

In light of the above arguments, the best description of the ion-induced \(L\)-shell ionization is expected to be given by a modified ECPSSR that includes the subshell coupling effects. Such a model, named ECPSSR-CSM, is obtained by combining ECPSSR and RSCA-CSM in the following way. The original idea behind the consideration of the couplings between the \(L\) substates was the vacancy sharing process among the \(L\) subshells. The vacancy sharing does not change the total \(L\)-shell ionization cross section, which is supposed to be described well by ECPSSR. At the same time, the relative subshell ionization cross sections change according to RSCA-CSM. This concept can be expressed by renormalizing the RSCA-CSM subshell cross sections in a way that its sum equals the total ECPSSR cross section (see also \([33]\)).

\[
\sigma_{Li}(\text{ECPSSR-CSM}) = \sigma_{Li}(\text{RSCA-CSM}) \times \frac{\sigma_{\text{Tot}}(\text{ECPSSR-CSM})}{\sigma_{\text{Tot}}(\text{RSCA-CSM})}.
\] (10)

In the present work, we applied the improved version of ECPSSR, the ECUSAR model. According to Eq. \((10)\), the subshell coupling effects are also included in the latter model.

### C. Shellwise local plasma approximation and coupled state model (SLPA-CSM)

The shellwise local plasma approximation (SLPA) \([34]\) is an ab-initio approach for the calculation of ionization probabilities within the dielectric formalism. Based
on the quantum dielectric response theory, which is generally employed to deal with the conduction band of solids, it has been extended to account for the inner-shells by considering the density of target electrons and the binding energies. The SLPA calculates the $j$ subshell ionization cross section of certain target atoms due to the interaction with a projectile (velocity $v_1$ and nuclear charge $Z_1$) as

$$
\sigma_{j,\text{SLPA}} = \frac{2}{\pi v_1^2} \int_0^\infty \frac{Z_2^2}{p} dp \int_0^{\omega_{p1}} d\omega \left[ \frac{1}{\epsilon(p, \omega, E_j, \delta_j(r))} \right] \hat{d}r,
$$

with $\epsilon(p, \omega, E_j, \delta_j)$ being the Levine-Louie dielectric function \[39\], $E_j$ the binding energy, $\delta_j(r)$ the density of the $j$-subshell electrons around the nucleus, and $p(w)$ the momentum (energy) transferred. The $L_i$ subshells of Os were described by performing full relativistic atomic structure calculations. We solved the Dirac equation by implementing the parametric potential method \[37\] \[39\] and an optimized configuration interaction mixing. The electronic structure computed, which is then used in Eq. (11), agrees with available experimental values within 1.5\% \[40\]. It is worth noting that the SLPA has been successfully employed previously by the authors to obtain $L$-shell ionization cross sections of relativistic targets such as Pt, Ta, W, Pb, Au, Bi, Th and U \[22\] \[41\].

### D. Method for obtaining theoretical x-ray production cross section from theoretical $L$-shell ionization cross section

The theoretical $L$ x-ray production cross sections for the most commonly resolved $L_I$, $L_\alpha$, $L_\beta$, $L_\gamma$, $x$-rays are related to the $L_i$ subshell ionization cross sections, $\sigma_{L_i}$, as given below

$$
\sigma_{L_i}^x = [\sigma_{L_1}(f_{13} + (f_{12}f_{23}) + \sigma_{L_2}f_{23} + \sigma_{L_3} \omega_3 F_{31}],
$$

$$
\sigma_{L_\alpha}^x = [\sigma_{L_1}(f_{13} + (f_{12}f_{23}) + \sigma_{L_2}f_{23} + \sigma_{L_3} \omega_3 F_{3\alpha}],
$$

$$
\sigma_{L_\beta}^x = \sigma_{L_1}[\omega_1 F_{1\beta} + \omega_2 F_{2\beta} + (f_{13} + (f_{12}f_{23}) \omega_3 F_{3\beta}) + \sigma_{L_2}(\omega_2 F_{2\beta} + f_{23} \omega_3 F_{3\beta}) + \sigma_{L_3} \omega_3 F_{3\beta}],
$$

$$
\sigma_{L_\gamma}^x = \sigma_{L_1} \omega_1 F_{1\gamma} + (\sigma_{L_1} f_{12} + \sigma_{L_2}) \omega_2 F_{2\gamma},
$$

where $\sigma_{L_i}^x$ is the $x$-ray production cross sections of the different $L$ x-ray components, $\sigma_{L_i}$ is the ionization cross sections for the $L_i$ subshell, $\omega_i$ is the fluorescence yields of the $L_i$ subshells, $f_{ij}(i < j)$ is the CK yields for the CK transition between the $L_i$ and $L_j$ subshells, and $F_{ip}$ is the fractional radiative emission rates, with $i = 1, 2, 3$ and $p = l, \alpha, \beta, \gamma$. The theoretical $L$ x-ray production cross sections were calculated by combining the $L_i$ ionization cross sections obtained by the different $L$-shell ionization models in Eqs. \[12\] to \[15\].

### IV. EFFECT OF THE MULTIPLE VACANCIES ON THE ATOMIC PARAMETERS USED FOR THE CONVERSION OF THE IONIZATION CROSS SECTIONS TO X-RAY PRODUCTION CROSS SECTIONS

Multiple vacancies in the target atom change the atomic parameters: the fluorescence yields and the CK yields, which in turn alter the x-ray production cross sections. In the present work, single-hole fluorescence $\omega_i^0$ and CK yields $f_{ij}^0$ \[42\], were corrected for multiple ionization using a model prescribed by Lapicki et al. \[43\]. Each electron in a manifold of the outer subshells is ionized with a probability $P$, which is calculated using Eq. (A3) from \[43\], and replacing the projectile atomic number $Z_P$ by its charge state $q$ \[44\].

$$
P = \frac{q^2}{2\beta V_p^2} (1 - \frac{\beta}{4V_p^2}),
$$

with $\beta = 0.9$. For the charge state $q$, we take the incident charge state of the projectile. The $\omega_i^0$ values corrected for the simultaneous multiple ionization (SMI) in the outer subshells are given by

$$
\omega_i = \omega_i^0 [1 - P(1 - \omega_i^0)]^{-1},
$$

while the $f_{ij}$ values for multiple ionization are given by

$$
f_{ij} = f_{ij}^0 [1 - P]^2.
$$

Note that the fractional rates $F_{ip}$ remain unchanged because both the partial and the total non-radiative widths are narrowed by identical factors. According to Eqs. \[17\] and \[18\], the single-hole fluorescence and CK yields depend on the energy and charge state of the projectile ion. The fluorescence and the CK yields for singly- and multiply-ionized Os is given in Table \[4\]. It is clear from this table that in the extreme the $L_i$ subshell fluorescence yields are enhanced by $\sim 30\%$, and the CK yields are reduced up to $\sim 50\%$ from the single-hole to the multiple-hole atom in Os. Note that the use of different sets of atomic parameters can change the x-ray production cross section by $\sim 30\%$ or more. Recent values of $\omega_i^0$, and $f_{ij}^0$, compiled by Campbell \[42\] for the elements with $25 \leq Z \leq 96$, have been used in the present work for singly-ionized atoms.

### V. EFFECT OF THE $L$-$K$ ELECTRON CAPTURE ON THE $L$-SHELL IONIZATION CROSS SECTION

To calculate the $L$-$K$ electron capture cross sections, we used the theory of Lapicki and Losonsky \[15\], which is based on the Oppenheimer-Brinkman-Kramers (OBK) approximation \[46\] with binding and Coulomb deflection corrections at low velocities. Neglecting the change in the binding energy of the $K$ shell electron of the projectile with one versus two $K$ shell vacancies, a statistical scaling is used to calculate the electron transfer cross section
for the case of one projectile K-shell vacancy, $\sigma_{L\rightarrow K}$, resulting in $\sigma_{L\rightarrow 2K}/2$, where $\sigma_{L\rightarrow 2K}/2$ is the production cross section for two projectile K-shell vacancies. In the present experimental condition, $v_1$ ranges between 12.39 and 15.17, while $v_{2L} = Z_{2L}/n_2 = 35.925$ (in a.u.), $n_2$ and $n_1$ are the principal quantum numbers of $L$ and $K$ shell electrons of the target and the projectile atom, respectively. Following Lapicki and Losonsky [15], $\sigma_{L\rightarrow 2K}$ can be obtained as

$$\sigma_{L\rightarrow 2k} = \frac{1}{3} \sigma_{L\rightarrow 2k}^{OBK}(\theta_L), \theta_k = \frac{E_L}{v_{2L}^2 \times 13.6},$$

(19)

with $Z_{2L} = Z_T - 4.15$ and

$$\sigma_{L\rightarrow 2k}^{OBK}(\theta_k) = \frac{2\pi}{5v_1^2} \frac{\pi a_0^2 n_2^2}{(v_{1k} v_{2L})^5 Z_F 10^{24}} \left[ v_{1k}^2 + (v_1^2 + v_{2L}^2 - v_{1k}^2)^2 / 4v_1^2 \right]^5,$$

(20)

where $E_L$ is the binding energy of L-shell electron of the target (in eV), and the parameters $a_0$, $v_{1k}$, $Z_F$, and $Z_T$ are the Bohr radius, the K-shell orbital velocity of the projectile ion, atomic number of the projectile and the target atom, respectively.

Due to certain charge state distribution inside the target, the effective capture contribution will be $F(q) \times \sigma_{L\rightarrow 2k}^{OBK}(\theta_L)$, for $q = 8^+$ and $9^+$. Here, $F(q)$ is the charge state fraction for a specific $q$. To obtain $F(q)$, we employed a two-fold procedure. First, we used a Fermi-gas-model based empirical formula for the determination of the mean charge state, $q_m$, inside the target [47]

$$q_m = Z_F \left( 1 - \frac{v_F}{v_1} \right),$$

(21)

where $z_1$ and $v_F$ are the projectile atomic number and the Fermi velocity of target electrons, respectively. The value of $v_F$ for Os is $8.19 \times 10^6$ cm/sec. To showcase the difference between the ionization of the projectile ion inside and outside the target, we displayed the $q_m$ as predicted by the Fermi-gas-model [47] and by the Schwitz model [48] in Fig. 3A. This contrasting picture is governed by the solid surface [49, 50]. The charge state of the heavy projectiles inside the solid target is higher than for those outside. This feature has been described in detail by Chatterjee et al. [20]. In the second part of the procedure, the $q_m$ values inside the target are substituted by a Lorentzian charge state distribution [49] to obtain the $F(q)$ as follows

$$F(q) = \frac{1}{2\pi} \frac{1}{(q - q_m)^2 + (\Gamma/2)^2} \quad \text{and} \quad \sum_q F(q) = 1,$$

(22)

where the distribution width $\Gamma$ is taken from Novikov and Teplova [51] as follows

$$\Gamma(x) = C \left[ 1 - \exp(-x^\alpha) \right] \left[ 1 - \exp \left[ -(1-x)^\beta \right] \right],$$

(23)

being $x = q_m/z$, $\alpha = 0.23$, $\beta = 0.32$ and $C = 2.669 - 0.0098 Z_2 + 0.058 Z_1 + 0.00048 Z_1 Z_2$. The $F(q)$ for $q = 8^+$ and $9^+$ are displayed with a bar chart in Fig. 4B.
VI. DISCUSSIONS

In a recent article [52], we analyzed the major sources of errors in the measurements of $L$-subshell cross sections. We concluded that these uncertainties come from inaccurate (i) determination of target thickness, (ii) counting the number of projectile particles, (iii) background subtraction during spectrum analysis, and (iv) atomic parameters used for the conversion of (a) the measured x-ray production cross sections into $L_i$-subshell ionization cross sections and (b) the theoretical $L_i$-subshell ionization cross sections into $L$-shell x-ray production cross sections. In this work, all these aspects were considered carefully. The first source of error was managed by measuring the foil thickness by the RBS method, as shown in Fig. 1. The mass thickness (target thickness in $\mu\text{gcm}^{-2}$) is normally measured by three techniques viz., RBS, PIXE and XRF (x-ray fluorescence). Out of these, RBS is the most accurate (see Fig. 4 of Ager et al. [53]). The second source of uncertainties was controlled by measuring the integrated charge count of the projectile ions in a Faraday cup placed behind the target for a fixed duration (100 s) and under two different conditions: (a) solely the blank target frame in place and (b) the target foil in place. The ratio between the integrated charge counts for the two different conditions is $R = \frac{n q}{n q'} = \frac{q}{q'}$, where $n$ is the number of projectile ions of incident charge state $q$ in case of blank target, and of charge state $q'$ when the target is in place. If the spectra is recorded for a long duration (say, 30 minutes), then, the total measured charge divided by $q'$ will give the number of projectile ions passing through the target foil as required in Eq. (1). The third source of error was also taken into consideration, as can be evidenced from Figs. 2 and 3, the data points are well on the fitting profile, and the reduced $\chi^2$-squared values are close to 1. The fourth aspect is also addressed through a step by step evaluation, and is discussed below.

Besides the $L$-shell ionization, heavy ion collisions give rise to simultaneous ionization of the higher-shell electrons. This change of electronic environment in an atom alters the properties of the $L$ x-ray emission. As a result, the atomic parameters vary with the projectile energy as shown in Fig. 5. Although this effect was taken into account, the $L_i$-subshell ionization cross sections derived from the measured $L$ x-ray production cross sections are underestimated by the ECUSAR-CSM, ECUSAR-CSM-EC, RSCA-CSM, RSCA-CSM-EC, SLPA, and SLPA-EC models, as can be seen in Fig. 6. The vacancy sharing among the $L$ subshells described by the coupled-states model (CSM) seems to have a minor role in the present collisional system. Despite the $LK$ electron capture (EC) effects are included to the various theories, the cross sections remain lower than the measured values. The EC cross sections for the $L_i$ subshells as a function of the impact energy are plotted in Fig. 7. According to the figure, the EC contribution is the largest for the $L_3$, and is almost identical for $L_1$ and $L_2$. To estimate these contributions, we used the charge-state distributions of the projectile ions inside the solid target as presented in Fig. 4.

The atomic parameters are used to convert the $L$ x-ray production cross section to $L_i$ subshell ionization cross section as discussed above. In general, such parameters are taken from various sources, where the authors have calculated them using different theoretical methods. In some cases, the theoretical predictions considerably deviate from each other. In particular, for $w_1$, $f_{12}$ and $f_{13}$ a spread of up to a factor of two can be observed, as shown in Table IV. In the present work, we used the most
TABLE II: The $L_1, L_2, L_3$, and $L_{\text{Tot}}$ ionization cross sections (kilo-barn) for $^{19}$F on Os as a function of energy (MeV). TH1, TH2, and TH3 denote the ECUSAR-CSM, the RSCA-CSM, and the SLPA model, respectively, while EC denotes the electron capture.

| E  | Expt. $(\sigma_{L_i})$ | TH1 + EC | TH2 + EC | TH3 + EC |
|----|------------------------|----------|----------|----------|
| r = 1 |
| 76  | 4.7                    | 4.0      | 4.3      | 1.9      | 2.2      | 2.7      | 3.0      |
| 84  | 6.6                    | 5.5      | 5.9      | 2.7      | 3.1      | 3.7      | 4.1      |
| 90  | 8.4                    | 6.7      | 7.2      | 3.4      | 3.9      | 4.5      | 5.0      |
| 98  | 9.1                    | 8.4      | 9.1      | 4.3      | 5.0      | 5.6      | 6.3      |
| 114 | 13.9                   | 11.8     | 12.8     | 6.3      | 7.2      | 8.3      | 9.3      |
| r = 2 |
| 76  | 6.1                    | 5.0      | 5.3      | 2.4      | 2.7      | 3.9      | 4.2      |
| 84  | 7.1                    | 6.2      | 6.6      | 3.1      | 3.5      | 4.9      | 5.3      |
| 90  | 7.9                    | 7.2      | 7.7      | 3.6      | 4.1      | 5.7      | 6.2      |
| 98  | 9.5                    | 8.5      | 9.1      | 4.4      | 5.0      | 6.9      | 7.5      |
| 114 | 14.4                   | 11.2     | 12.2     | 6.0      | 7.0      | 9.4      | 10.4     |

TABLE III: The $L_\alpha, L_\beta, L_\gamma$, and $L_{\text{Tot}}$ x-ray production cross sections for $^{19}$F on Os as a function of energy. Same notation and units as Table. II.

| E  | Expt. $(\sigma_i^L)$ | TH1 + EC | TH2 + EC | TH3 + EC |
|----|----------------------|----------|----------|----------|
| $L_\alpha$ |
| 76  | 5.53                 | 5.52     | 5.70     | 2.65     | 2.82     | 4.64     | 4.82     |
| 84  | 7.67                 | 6.63     | 6.87     | 3.25     | 3.49     | 5.52     | 5.76     |
| 90  | 9.32                 | 7.47     | 7.75     | 3.77     | 4.02     | 6.23     | 6.50     |
| 98  | 9.38                 | 8.64     | 9.05     | 4.43     | 4.84     | 7.2      | 7.60     |
| 114 | 13.00                | 10.90    | 11.5     | 5.82     | 6.38     | 9.24     | 9.81     |
| $L_\beta$ |
| 76  | 3.95                 | 3.51     | 3.70     | 1.69     | 1.87     | 2.78     | 2.96     |
| 84  | 5.19                 | 4.30     | 4.54     | 2.12     | 2.38     | 3.41     | 3.65     |
| 90  | 6.16                 | 4.94     | 5.23     | 2.48     | 2.77     | 3.91     | 4.20     |
| 98  | 6.56                 | 5.78     | 6.16     | 2.96     | 3.35     | 4.63     | 5.00     |
| 114 | 9.21                 | 7.49     | 8.06     | 3.98     | 4.57     | 6.16     | 6.74     |
| $L_\gamma$ |
| 76  | 0.38                 | 0.32     | 0.34     | 0.15     | 0.18     | 0.22     | 0.24     |
| 84  | 0.63                 | 0.43     | 0.46     | 0.21     | 0.24     | 0.29     | 0.32     |
| 90  | 0.75                 | 0.51     | 0.55     | 0.26     | 0.30     | 0.35     | 0.39     |
| 98  | 0.57                 | 0.62     | 0.67     | 0.32     | 0.37     | 0.43     | 0.48     |
| 114 | 0.90                 | 0.85     | 0.92     | 0.45     | 0.52     | 0.61     | 0.68     |

TABLE IV: Fluorescence and CK yields for singly ionized element from different theories.

| $\gamma_{\text{Os}}$ | Fluorescence Yield | CK Yield |
|----------------------|---------------------|----------|
| $\omega_1^f$         | $\omega_2^f$        | $\omega_3^f$ | $\omega_4^f$ | $J_{12}^f$ | $J_{13}^f$ | $J_{14}^f$ |
| Krause [53]           | 0.130               | 0.295     | 0.281     | 0.16     | 0.39     | 0.128     |
| Chen et al. [55]      | 0.088               | 0.318     | 0.282     | 0.088   | 0.636    | 0.130     |
| Orlić et al. [56]     | 0.13                | 0.295     | 0.281     | 0.16    | 0.39     | 0.128     |
| Campbell [42]         | 0.15                | 0.318     | 0.282     | 0.07    | 0.33     | 0.13      |

The difference between the above mentioned methods is as follows. In the first one, an individual $L_i$ subshell ionization cross section is compared, while in the second one, a combination of the contributions of the various subshells is considered. Hence, the mathematical expression for the former is given by a differential operation while the later constitutes an integral. From the values in Tables II and III, we notice that the agreement between the theory and experiment is better for the $L_i$ subshell ionization cross sections than for the $L$ x-ray production cross sections. Thus, the differential comparison provides recent theoretical parameters [12]. The estimated uncertainties for $\omega_1, \omega_2, \omega_3, f_{12}, f_{13}$, and $f_{23}$ are $15\%, 5\%, 5\%, 10\%, 5\%, 5\%$, respectively. To resolve the above-mentioned discrepancy of the $L_i$ subshell ionization cross section between the measurements and the theoretical models, we varied these parameters iteratively until a good agreement was achieved; see Fig. 2. We present the optimized parameters in Table II and show their variation with the beam energy in Fig. 3. The difference between the original [12] values and the optimized atomic parameters is not significant, except for $\omega_1$.

There are two ways to study the $L_i$ subshell ionization by ion impact: by comparing the theoretical predictions (i) with the measured $L_i$ subshell ionization cross sections, and (ii) with the measured $L$ x-ray production cross sections. The former has been considered above, and the latter is examined below.

The measured x-ray production cross sections are compared with calculations in Fig. 5. The theoretical values in Fig. 5(A) are obtained by employing the set of atomic parameters from [12], while in Fig. 5(B) the optimized set of atomic parameters are used. The various theories underestimate the measured cross sections when the original parameters are used; however, the new set of atomic parameters allows one to obtain a better agreement.
FIG. 6: Experimental and theoretical \( L_i \)-subshell \((i = 1 - 3)\) and total \( L \)-shell ionization cross sections for \(^{19}\text{F} \) on Os collisions as a function of the impact energy. The measured data denoted by orange symbols were obtained using atomic parameters published in \([42]\), those denoted by black symbols were obtained using atomic parameters which were optimized in order to have a good agreement between the theory and experiment (see text).

VII. CONCLUSIONS

In the present work, the \( L \) x-ray production cross sections of Os were measured by 4-6 MeV/u \(^{19}\text{F} \) ions of charge states \( q = 6^+, 7^+ \) and \( 8^+ \), and compared with theoretical \( L \) x-ray production cross sections. Different ionization theories such as RSCA, ECUSAR and SLPA were used for the Coulomb direct ionization. Additionally, the contribution of the \( LK \) electron capture was added to each theory. The effect of multiple ionization was also considered through the modified atomic parameters. Furthermore, \( L_i \) \((i = 1, 2, 3)\) subshell ionization cross sections were derived from the measured \( L \) x-ray production cross sections, and compared with the corresponding theoretical counterparts. Both comparisons show the best agreement with the experiment for the ECUSAR-CSM-EC model. However, certain differences are still clearly noticed. To resolve such discrepancies, the atomic parameters were optimized to obtain a good agreement between the measurements and ECUSAR-CSM-EC model. Thus, this work gives us a convincing understanding of the \( L \)-subshell ionization mechanism by heavy ion bombardments, while states doubts on the atomic parameters used in the conversion of the x-ray production cross sections to the ionization cross sections. Further, our work suggests the urgent need for accurate measurements and theoretical calculations of the atomic parameters used.

VIII. ACKNOWLEDGEMENTS

S.C. acknowledges the University of Kalyani for providing generous funding towards his fellowship. Financial support from the Science and Engineering Research Board (SERB), New Delhi (SB/FTP/PS-023/2014), to Sunil Kumar in terms of Young Scientist scheme, and the grant from IUAC, New Delhi (UF-UP-43302 project), are highly acknowledged. L. Sarkadi acknowledges the support from the Hungarian Scientific Research Fund (Grant No. K128621). A.M.P. Mendez, D.M. Mitnik and C.C. Montanari acknowledge the financial support from the following institutions of Argentina: Consejo Nacional de Investigaciones Científicas y Técnicas (CONICET), Agencia Nacional de Promoción Científica y Tecnológica (ANPCyT), and Universidad de Buenos Aires (UBA).

The authors are grateful to the Pelletron staff for the smooth conduct of the experiments and the RBS lab for the target thickness measurements.
FIG. 8: Comparison of the measured $L_\alpha$, $L_\beta$, $L_\gamma$, and $L_{\text{Tot}}$ x-ray production cross sections with the predictions of the different theoretical models. (A) The theoretical values were obtained using atomic parameters published in [42]. (B) The calculations were obtained using optimized atomic parameters, which allow to have a good agreement between the theoretical and experimental $L$-subshell ionization cross sections (see text).

[1] T. Satoh, International Journal of PIXE 25, 147 (2015).
[2] P. Sharma and T. Nandi, Physics of Plasmas 23, 083102 (2016).
[3] N. A. Dyson and N. A. Dyson, X-rays in Atomic and Nuclear Physics (Cambridge University Press, 2005).
[4] M. Antoszewska-Moneta, R. Brzozowski, and M. Moneta, The European Physical Journal D 69, 1 (2015).
[5] A. W. Gillespie, C. L. Phillips, J. J. Dynes, D. Chevrier, T. Z. Regier, and D. Peak, Advances in agronomy 133, 1 (2015).
[6] T. B. Johansson, R. Akselsson, and S. A. Johansson, Nuclear Instruments and Methods 84, 141 (1970).
[7] A. Bertol, R. Hinrichs, and M. Vasconcellos, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 363, 28 (2015).
[8] J. Garcia, Physical Review A 1, 280 (1970).
[9] D. Joseph, S. N. Rao, and S. Kailas, Mapana Journal of Sciences 12, 1 (2013).
[10] X. Zhou, Y. Zhao, R. Cheng, Y. Wang, Y. Lei, X. Wang, and Y. Sun, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 299, 61 (2013).
[11] J. Miranda, G. Murillo, B. Méndez, J. López-Monroy, J. Aspiazu, P. Villaseñor, J. Pineda, and J. Reyes-Herrera, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 316, 113 (2013).
[12] J. Miranda and G. Lapicki, Atomic data and nuclear data tables 100, 651 (2014).
[13] H. Mohan, A. K. Jain, M. Kaur, P. S. Singh, and S. Sharma, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 332, 103 (2014).
[14] R. Siegele, D. D. Cohen, and N. Dytlewski, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 158, 31 (1999).
[15] G. N. Raju, G. R. Murty, B. S. Reddy, T. S. Reddy, S. Lakshminarayana, and S. B. Reddy, The European Physical Journal D-Atomic, Molecular, Optical and Plasma Physics 30, 171 (2004).
[16] W. Uchai, G. Lapicki, W. Milner, S. Raman, P. V. Rao, and C. Vane, Journal of Physics B: Atomic and Molecular Physics 18, L389 (1985).
[17] G. Murillo, B. Méndez, J. López-Monroy, J. Miranda, and P. Villaseñor, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 383, 89 (2016).
[18] I. Fijał-Kirejczyk, M. Jaskóła, W. Czarnacki, A. Korman, D. Banaś, J. Braziewicz, U. Majewska, J. Semañiak, M. Pajek, W. Kretschmer, et al., Physical Review A 77, 032706 (2008).
[19] M. Pajek, D. Banaś, J. Braziewicz, U. Majewska, J. Semañiak, I. Fijał-Kirejczyk, M. Jaskóła, W. Czarnacki, A. Korman, W. Kretschmer, et al., Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 383, 89 (2016).
teractions with Materials and Atoms 363, 19 (2015).
[20] S. Chatterjee, P. Sharma, D. Mitra, and T. Nandi, arXiv preprint arXiv:2103.08299v4 (2021).
[21] S. Kumar, U. Singh, M. Oswal, G. Singh, N. Singh, D. Mehta, T. Nandi, and G. Lapicki, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 395, 39 (2017).
[22] M. Oswal, S. Kumar, U. Singh, S. Singh, G. Singh, K. Singh, D. Mehta, A. Mendez, D. Mitnik, C. Montanari, et al., Radiation Physics and Chemistry 176, 108809 (2020).
[23] M. Berger, http://physics.nist.gov/PhysRefData/Xcom/Text/Xcom.html (1998).
[24] J. F. Ziegler, M. D. Ziegler, and J. P. Biersack, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 268, 1818 (2010).
[25] S. E and K. B, Nucl. phys 69, 117 (2010).
[26] D. W. Marquardt, Journal of the society for Industrial and Applied Mathematics 11, 431 (1963).
[27] L. Sarkadi and T. Mukoyama, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 4, 296 (1984).
[28] L. Lugosi and L. Sarkadi, Computer physics communications 141, 73 (2001).
[29] L. Sarkadi and T. Mukoyama, Physical Review A 37, 4540 (1988).
[30] L. Sarkadi and T. Mukoyama, Journal of Physics B: Atomic, Molecular and Optical Physics 23, 3849 (1990).
[31] W. Brandt and G. Lapicki, Physical Review A 23, 1717 (1981).
[32] G. Lapicki, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 189, 8 (2002).
[33] G. Lapicki, G. R. Murty, G. N. Raju, B. S. Reddy, S. B. Reddy, and V. Vijayan, Physical Review A 70, 062718 (2004).
[34] C. Montanari, D. Mitnik, and J. Miraglia, Radiation Effects & Defects in Solids 166, 338 (2011).
[35] C. Montanari and J. Miraglia, Advances in Quantum Chemistry 65, 165 (2013).
[36] Z. H. Levine and S. G. Louie, Physical Review B 25, 6310 (1982).
[37] M. Klapisch, Comput. Rend. Acad. Sci. 265, 914 (1967).
[38] M. Klapisch, Computer Physics Communications 2, 239 (1971).
[39] A. Bar-Shalom, M. Klapisch, and J. Oreg, Journal of Quantitative Spectroscopy and Radiative Transfer 71, 169 (2000).
[40] A. M. P. Mendez, C. C. Montanari, and D. M. Mitnik, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 460, 114 (2019).
[41] M. Oswal, S. Kumar, U. Singh, G. Singh, K. Singh, D. Mehta, D. Mitnik, C. C. Montanari, and T. Nandi, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 416, 110 (2018).
[42] J. Campbell, Atomic Data and Nuclear Data Tables 85, 291 (2003).
[43] G. Lapicki, R. Mehta, J. L. Duggan, P. Kocur, J. Price, and F. D. McDaniel, Physical Review A 34, 3813 (1986).
[44] R. Mehta, J. Duggan, F. McDaniel, M. McNeir, Y. Yu, D. Marble, and G. Lapicki, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 79, 175 (1993).
[45] G. Lapicki and W. Losonsky, Physical Review A 15, 896 (1977).
[46] R. May, PhL11 33 (1964).
[47] W. Brandt, R. Laubert, M. Mourino, and A. Schwarzschild, Physical Review Letters 30, 358 (1973).
[48] G. Schiwietz and P. Grande, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 175, 125 (2001).
[49] P. Sharma and T. Nandi, Physical Review Accelerators and Beams 22, 034501 (2019).
[50] T. Nandi, The Astrophysical Journal Letters 673, L103 (2008).
[51] N. Novikov and Y. A. Teplova, Physics Letters A 378, 1286 (2014).
[52] S. Singh, S. Chatterjee, D. Mitra, and T. Nandi, arXiv preprint arXiv:2103.07247 (2021).
[53] F. Ager, M. Ferretti, M. Grilli, D. Juanes, I. Ortega-Feliu, M. Respaldiza, C. Roldán, and S. Scrivano, Spectrochimica Acta Part B: Atomic Spectroscopy 135, 42 (2017).
[54] M. O. Krause, Journal of physical and chemical reference data 8, 307 (1979).
[55] M. H. Chen, B. Crasemann, and H. Mark, Physical Review A 24, 177 (1981).
[56] I. Orlic, C. Sow, and S. Tang, International Journal of PIXE 4, 217 (1994).