

Influence of Multiple Ionization on L X-ray Intensity Ratio in Pt Induced by Proton

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Abstract—Ion-atom interaction provides a valuable insight into the study of atomic properties of matter. But it involves complex mechanisms which can also be understood by the inner-shell ionization. For charged particle as a projectile not only inner-shell but also the outer-shell ionization takes place which is referred as multiple ionization (MI). The effect serves as a basis for a systematic understanding of X-ray emission process. The atomic parameters viz. Fluorescence yields and Coster-Kronig probabilities, which are required for their investigation, have been affected by the multiple ionization and plays the significant role for their precise calculations. In this paper an attempt is made to understand the importance of MI effect and its influence on the L X-ray intensity ratios in Pt (Platinum).

Index Terms—Inner-shell ionization, Line ratios, Multiple ionization, X-ray yields.

I. INTRODUCTION

IONIZATION by the charged particle has attained recent attention during the present decades [1, 2]. The rapid intensification in applications of the most potent technique PIXE (particle induced X-ray emission) has made researchers to work enthusiastically for the accurate knowledge of X-ray yields over wide energy region [3, 4]. The growing amount of experimental [5-7] and theoretical work [8, 9] on ion-induced inner-shell ionization has solved many analytical problems [10-12] and revealed many interesting features of the interaction processes. For light ion impact the fundamental process of interaction is dominated by direct Coulomb ionization resulting in creation of single-vacancy configuration, which decays radiatively via emission of X-rays or nonradiatively by the Auger or Coster-Kronig processes. In case of heavy targets, the radiative transitions dominate over

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the non radiative. As the charged particle moves through the matter it induces multiple vacancies in the inner shell through the outer shell. As a matter of fact, additional vacancies in the outer shell substantially influenced the atomic parameters viz. Fluorescence and Coster-Kronig (CK) yields.

II. THEORETICAL METHODOLOGY

To describe the mechanism for production of inner-shell vacancies, several theoretical models have been developed. Brandt and Lapicki [13, 14] modified the Plane Wave Born Approximation (PWBA) by incorporating polarization and binding effects in the PSS (perturbed stationary state approximation) and including relativistic (R), energy loss (E), and coulomb deflection (C) effects in it. This is generally referred as the ECPSSR theory [15]. It reduced the disagreement between experiment and theory but only for the K shell and for light projectiles. In the case of L-shell ionization, the deviations from the theory have been observed in the low ion velocity range. When the collision energy decreases the discrepancy between these simple theories and experiments becomes significant and modifications should be made. Further, at lower energies, a refinement has been made in ECPSSR theory which is entitled as “united atom” approximation (UA). This corrects the unphysical behaviour of atomic level binding energy in ECPSSR theory using the procedure suggested by Cipolla [16] and termed it as ECPSSRUA. Accordingly, the UA effect increases rapidly as proton energy decreases. Based on this theory, experimental results are compared with the theoretical predictions. It is observed that even this theory gives the erratic results. This is because the factors (Fluorescence and Coster-Kronig yields) required for the extraction of cross sections are strongly affected by the predominance of multiple ionization by incoming ions which leads to a necessity of some refinements. Keeping in view of this and to improve the results, multiple ionization effect has been included in ECPSSR theory. By computing with these modifications, the present calculated results are compared with the experimentally measured values.

A. Multiple Ionization

In collisions of ions with atoms the strong Coulomb field of the projectile can cause simultaneous ejection of several electrons. This effect is known as the multiple ionization [17]. The multiple vacancies created in the outer shells during the ion-atom collision act as spectators and may not all be filled prior to the radiative filling of inner-shell vacancy. As a result,

the screening of nuclear charge get reduces and results in the increase of binding energy of all energy levels. Consequently, the X-rays emitted from the multiple ionized atoms are considerably shifted in both position and width [18-19]. This effect also leads to change in (a) Fluorescence yields (b) Coster-Kronig probabilities. Miranda *et al.* [20] showed that, at least in the case of protons with energies below 1 MeV, the correct database of atomic parameters (fluorescence yields and Coster-Kronig probabilities) improves agreement with theoretical predictions. Lapicki *et al.* [21] suggested a method to revise these yields so as to account this effect. This amendment considers a basic assumption that the Fluorescence yields are modified by creation of holes in outer shells by the incoming projectiles, with an equal probability for each shell which is calculated through the binary encounter approximation [22]. The equation for the modified Fluorescence and Coster-Kronig yields is given by

$$\omega_s = \frac{\omega_s^0}{1 - (Z_1^2/2\beta v^2)[1 - (\beta/4v^2)](1 - \omega_s^0)}$$

$$f_s = f_s^0 [1 - Z_1^2(1 - \beta/4v^2)/2\beta v^2]^2$$

where,

ω_s^0 is the single hole Fluorescence yield.

Z_1 is the atomic number of the projectile.

f_s^0 is the single hole Coster-Kronig yield.

β is a parameter fixed from electron binding energies, it takes the value 0.9 for the L-shell.

v is the projectile ion velocity.

The multiple ionization effect results in the increase of Fluorescence yields and it decrease with the decreasing velocity of projectile. The modified values of Fluorescence yields and Coster-Kronig probabilities computed from above equations are used for the calculation of X-ray intensity ratios on the basis of ECPSSR ionization theory [23, 24].

III. EXPERIMENTAL PROCEDURE

The experimental measurements have been performed using AN - 400 Van de Graaff accelerator which yields a proton beam current up to 50 μ A. The beam energy is calibrated using the $^{15}\text{F}(p, \alpha\gamma)^{16}\text{O}$ resonance at 340.5 keV. The H^+ beam current of continuously variable energy up to 400 keV on the target is measured by current integrator Elcor-A309F (having high sensitivity, accuracy, low drift and an internal calibrating source). The experimental set up includes the target chamber and X-ray detection system. The accelerated particles are passed through a 15° bending magnet to ensure that the beam which strikes the target is isotopically pure. Target is mounted at 45° to the beam direction. The high purity Germanium

(HPGe) detector placed at right angle to the beam is used to detect L X-rays. The L X-ray spectra from the targets are recorded with the help of an ORTEC multichannel analyzer. The beam current employed is 10 nA to 1 μ A so as to obtain X-ray yields compatible with the input characteristics of detecting electronics and thus avoiding the problem of dead time. The details of experimental procedures and data analysis are given in our earlier work [25-27]. The error in the measured cross sections is estimated at 10-12% and is attributed to uncertainties in the peak area evaluation, charge collection, target thickness and the X-ray attenuation coefficients.

IV. RESULTS AND DISCUSSION

In the light of multiple ionization, the calculations for intensity ratios L_β/L_α , L_γ/L_α and L_ℓ/L_α of Pt are performed at selected proton energies ranging from 260 to 400 keV and are plotted as shown in the Figs 1-3 respectively. This energy range is chosen because it is the range where the theories have shown larger discrepancies with experiments. These calculated results are compared with the experimental values. For calculation, data basis of Campbell [28, 29], Krause [30], and Puri's Dirac Hartree Slater (DHS) [31] have been used in ECPSSR model with different effects i.e. ECPSSR+UA, ECPSSR+MI. These are abbreviated as UAWOMI-C, UAWOMI-K, UAWOMI-DHS and EWMI-C, EWMI-K, EWMI-DHS respectively.

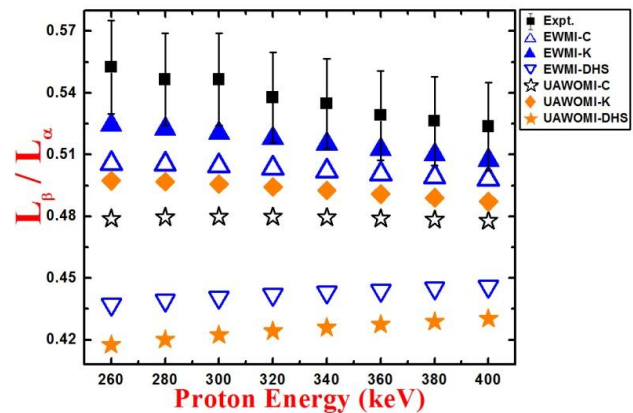


Fig. 1. L_β/L_α for Pt as a function of proton energy.

In Fig. 1, L_β/L_α results show that the Krause's values with multiple ionization effect are in better agreement with the experimental values than Campbell's values. This is due to inclusion of multiple ionization effect. However Dirac Hartree Slater values are far away from the experimental measurements. The deviation is comparatively more at the lower energy side and it decreases as the projectile energy increases.

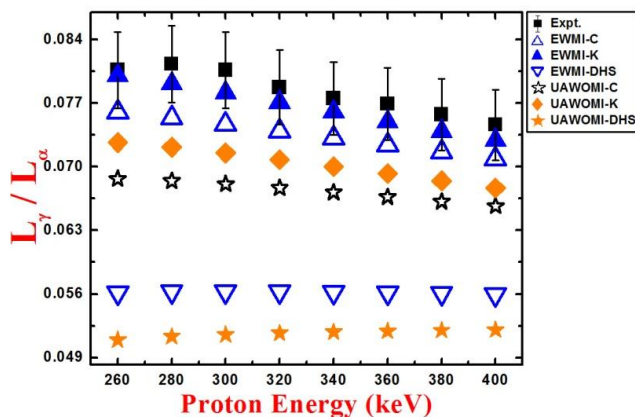


Fig. 2. L_γ/L_α for Pt as a function of proton energy.

The trend is similar for L_γ/L_α (Fig. 2). It is found that Krause's as well as Campbell values with same effect i.e., MI approach the experimental measurements whereas Puri *et al.*

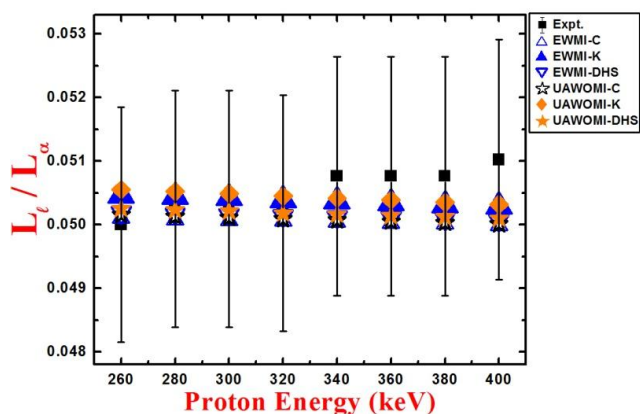


Fig. 3. L_t/L_α for Pt as a function of proton energy.

(DHS values) lag behind. On the other hand in case of L_t/L_α (Fig. 3), almost all the results are within the range of the experimental measurements.

The present results clearly demonstrate the importance of the modification of Fluorescence and Coster-Kronig yields in multiple ionized atoms for the accurate estimation of the line ratios. It clearly indicates that the ECPSSR+MI calculations give a well prediction to the experimental data than ECPSSR+UA. For calculations with approach ECPSSR+UA, almost all the results with all data points under estimate the experimental data and show a remarkable discrepancy at low proton energy regime. However the effect i.e. MI comparatively improves the results especially at lower energy region.

V. CONCLUSION

The multiple ionization effect is very important for proper interpretation of X-ray emission and intensity ratios in ion-

atom collision. The ECPSSR+MI calculations agree well with the experimental values, generally close over the entire energy region except at lower energy edge. It improves our results considerably and offer relatively good opportunity to understand the atomic properties in a reliable way. It can be consider as a positive step to bring theoretical predictions closer to the experiment.

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