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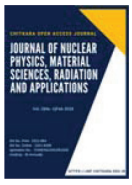

How do Uncertainties in Atomic Parameters Influence Theoretical Predictions of X-Ray Production Cross Sections By Proton Impact?

J Miranda*

Institute of Physics, National Autonomous University of Mexico, Scientific Research Circuit S/N, Coyoacan-04510, Mexico

*Email: miranda@fisica.unam.mx

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ABSTRACT

The emission of characteristic X-rays induced by proton impact is a phenomenon known since the first half of the 20th century. Its more widely known application is the analytical technique Particle Induced X-ray Emission (PIXE). Several models have been developed to calculate, first, ionization cross sections and then the subsequent X-ray production cross sections. However, to carry out the comparisons of these predictions with experimental data it is necessary to use atomic parameters databases (fluorescence yields, Coster-Kronig transition probabilities, emission rates) that also have experimental uncertainties. In this work it is demonstrated how these values do not allow to decide which model describes more accurately the cross sections, due to a final "theoretical uncertainty" obtained through the propagation of the original uncertainties.

1. Introduction

Particle Induced X-ray Emission (PIXE) is a powerful analytical technique [1], which requires a reliable knowledge of ionization cross sections by charged particle impact and the subsequent process of characteristic X-ray emission, including the atomic parameters involved, such as fluorescence yields, Coster-Kronig transition probabilities, and relative emission rates [2]. Since the first half of the 20th century, hundreds of papers regarding the measurement of X-ray production cross sections (XRPCS) and derived ionization cross sections (ICS) have been published. This includes K-, L- and M-X-rays [3-7]. Furthermore, many efforts have been dedicated to develop theories that describe appropriately those phenomena [8-12]. Some of these comprise also corrections to the models, considering second-order effects, such as the united atom approximation (UA) [13], electron capture by the incoming ion (EC) [14] or multiple ionization of atomic outer shells (MI) [15]. The results of all these theoretical predictions are usually compared among them with the purpose of deciding whether one model or correction is more adequate to explain the experimental data. This requires using the aforementioned atomic parameters, which must be, in turn, either determined experimentally or calculated by some theoretical model [16]. Therefore, those estimates need the consideration of experimental uncertainties, which are not usually taken into account to check the validity of the theories.

With this in mind, it is worth asking if the experimental uncertainties of the atomic parameters would actually allow determining if any particular model is the best to predict the experimental data. In this work, thus a description of how those uncertainties may prevent any definite conclusion about the validity of theoretical models or atomic parameters databases employed in the predictions, specifically for K and L shells.

2. X-ray production cross sections and atomic parameters databases

The process of X-ray emission induced by ion impact initiates at the atomic level by the ionization of inner shells of the target atom. Following this, a vacancy transfer to higher shells occurs, concluding afterwards with the emission either of an X-ray photon or an Auger electron. This way, the total cross section for the emission of K X-rays, $\sigma_{X,K}$, is given by [17]:

$$\sigma_{X,K} = \omega_K \sigma_{I,K} \quad (1)$$

where ω_K is the fluorescence yield and $\sigma_{I,K}$ is the ionization cross section. The theoretical models actually refer to

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ionization cross sections, while the experiments are based on the measurement of the X-ray production cross sections. Therefore, to predict the latter, it is necessary to know fluorescence yields, either empirically or theoretically.

In a similar fashion, the total XRPCS for the L-shell is expressed as:

$$\sigma_{X,LT} = \omega_1\sigma_{L1} + \omega_2\left(\sigma_{L1}f_{12} + \sigma_{L2}\right) + \omega_3\left[\sigma_{L1}\left(f_{13} + f_{12}f_{23}\right) + \sigma_{L2}f_{23} + \sigma_{L3}\right] \quad (2)$$

where ω_i are the fluorescence yields and f_{jk} are the Coster-Kronig transition probabilities; σ_{Li} is the ionization cross section of the L_i subshell.

The description of the ICS by the ECPSSR correction to the Plane Wave Born Approximation (PWBA) is the most widely accepted one [10]. The former adds rectifications due to projectile energy loss (E), Coulomb deflection of the incident ion (C), polarization and change in electron binding energies through a Perturbed Stationary States method (PSS), and relativistic values of target electron mass (R). A simple-to-use computer code to calculate the theoretical ECPSSR XRPCS was developed by Cipolla [18], known as ISICS. This tool also allows the inclusion of certain modifications to the model, like relativistic consideration of the ion velocity (RECPSSR) for K-shell ionization [19] or the UA approach. The values provided by this program are a very good basis to make comparisons among the various corrections and atomic parameters databases, as well as with experimental data.

Regarding the atomic parameters, several tables have been published, presenting empirical, semiempirical or theoretical values. For the K-shell, possibly the most widely used data come from the tables by Krause [20], which are rather old. A more recent compilation was presented by Kahoul et al. [21], using more recently published experimental data. Both databases include uncertainties in their tabulated parameters. As for the L-shell, nowadays the most relevant compilations were published by Campbell [16, 22]. His tables include recommended, Krause's [20] and theoretical Dirac-Hartree-Slater values, both for fluorescence yields and Coster-Kronig transition probabilities. A more recent contribution is the fluorescence yield database presented by Aylikci et al. [23]. Krause, Campbell and Aylikci et al. present the estimated uncertainties of the parameters.

When measuring XRPCS for individual lines it is necessary to introduce still another parameter, namely, the relative emission rate of the particular line to be used. For instance, the $L_{\alpha 1,2}$ XRPCS $\sigma_{L\alpha}$ are determined with the equation [17]:

$$\sigma_{X,L\alpha} = \left[\sigma_{L1}\left(f_{13} + f_{12}f_{23}\right) + \sigma_{L2}f_{23} + \sigma_{L3}\right]\omega_3 F_{3\alpha} \quad (3)$$

where $F_{3\alpha}$ represents the relative emission rate of the $L_{\alpha 1,2}$ line. Values for this magnitude have been published by Campbell and Wang (theoretical) [24], Puri (theoretical) [25] and Salem et al. (experimental) [26]. Only the latter contains an estimation of the uncertainties.

3. Effect of experimental uncertainties on X-ray production cross section evaluations

All the possible combinations for the calculation of the theoretical cross sections to determine whether one approach or database is better than any other usually are presented graphically in many publications. Nevertheless, the experimental uncertainties of the atomic parameters have seldom been taken into account in these comparisons.

Using the law for the propagation of uncertainty, defined in ref. [27], the "uncertainties" in the theoretical values, $u(x_i)$, where x_i is the corresponding physical quantity, are obtained from:

$$u\left(\sigma_{X,K}\right) = \sigma_{I,K}\,u\left(\omega_K\right) \quad (4)$$

$$u\left(\sigma_{X,L1}\right) = \sigma_{I,L1}\,u\left(\omega_1\right) \quad (5)$$

$$u\left(\sigma_{X,L2}\right) = \left[\left(\sigma_{L1} + f_{12}\sigma_{L1}\right)^2 u^2\left(\omega_2\right) + \left(\omega_2\sigma_{L1}u(f_{12})\right)^2\right]^{1/2} \quad (6)$$

$$u\left(\sigma_{X,L3}\right) = \left\{\begin{array}{l}\left[\sigma_{L3} + f_{23}\sigma_{L2} + \sigma_{L1}\left(f_{13} + f_{12}f_{23}\right)\right]^2 u^2\left(\omega_3\right) \\ \quad + \left(\omega_3\sigma_{L2} + \omega_3 f_{12}\sigma_{L1}\right)^2 u^2\left(f_{23}\right) \\ \quad + \left(\omega_3\sigma_{L1}u\left(f_{13}\right)\right)^2 + \left(\omega_3 f_{23}\sigma_{L1}u\left(f_{12}\right)\right)^2\end{array}\right\}^{1/2} \quad (7)$$

$$u\left(\sigma_{X,LT}\right) = \left[u^2\left(\sigma_{X,L1}\right) + u^2\left(\sigma_{X,L2}\right) + u^2\left(\sigma_{X,L3}\right)\right]^{1/2} \quad (8)$$

$$u\left(\sigma_{X,L\alpha}\right) = \left[\left(F_{3\alpha}\right)^2 u^2\left(\sigma_{X,L3}\right) + \left(\sigma_{X,L3}\right)^2 u^2\left(F_{3\alpha}\right)\right]^{1/2} \quad (9)$$

As a first step, the comparison of predictions for K X-rays is exemplified in Fig. 1. Here, PWBA, ECPSSR and UA, together with relativistic corrections to ECPSSR to compute total K XRPCS in Cu, are displayed, according to eq. (1). The fluorescence yields from Krause [20] and Kahoul et al. [21] are used with ECPSSR only. In particular, the uncertainty bars obtained from Krause and the ECPSSR are

given. Also, all the available published experimental data [3, 28-33] are plotted.

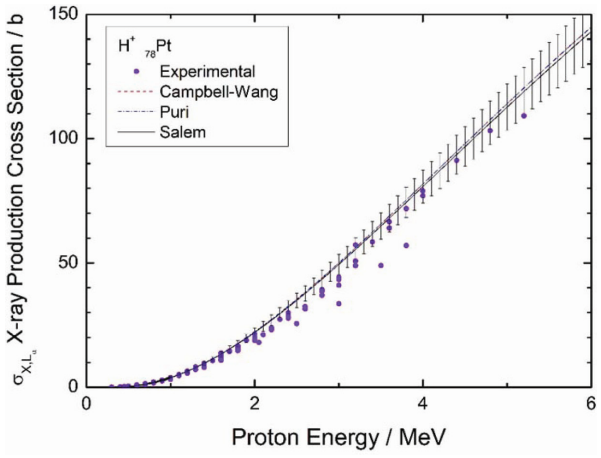

Figure 1: Total K X-ray production cross sections of Cu induced by proton impact. Experimental data were taken from the compilations by Paul and Sacher [3] and Lapicki [27]; new data from Refs. [28-33]. Curves correspond to PWBA and ECPSSR theories, ECPSSR-UA with united atom correction, RECPSSR to ECPSSR with relativistic ion velocity, and RECPSSR-UA the same with united atom correction. The last curve uses Kahoul et al. [21] data.

In this case, it is evident that the uncertainties in the ECPSSR predictions, based on Krause's tables [20], avoid any definition about which is the "best" prediction. Even at high energies, although the RECPSSR forecasts larger XRPCS, the agreement with the experimental data is not satisfactory.

Fig. 2 presents a similar comparison for total L XRPCS. Again, the ECPSSR in Pt is used as a reference, but based on Campbell's tabulations [16, 22]. The other curves employ Krause [20] and Aylkci et al. [23] atomic parameters. This time, it is apparent that it is impossible to decide which database provides the best results, as all the curves lie inside the uncertainty intervals, including the experimental points, which were taken from the compilation by Miranda and Lapicki [5, 6]. It is necessary to mention that Campbell's values present the lowest uncertainties, so the intervals shown in Fig. 2 represent the most precise theoretical prediction.

A final comparison reflects the effect of the uncertainty in the emission rates. Fig. 3 displays the experimental data for the L_α line of Pt [5, 6], and the predictions by the ECPSSR using Campbell's parameters [16, 22], as well as Campbell and Wang [24], Puri [25] and Salem et al. [26] relative intensities. Like in the other cases, the uncertainties (based on Salem's tables), limit any decision about which is the best database.

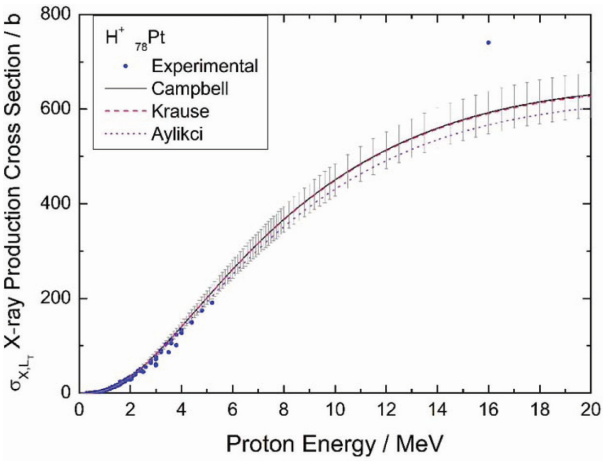

Figure 2: Total L X-ray production cross section of Pt induced by proton impact. The curves use the ECPSSR model [10], and databases by Campbell [16, 22], Krause [20], and Aylikci et al. [23].

Conclusions

The inclusion of experimental uncertainties of atomic parameters required for the calculation of theoretical values of XRPCS has not been carried out frequently in publications, at least in a rigorous quantitative manner. The present work describes how the uncertainties can be considered and presents how they influence any possible conclusion about the validity of the models or the databases themselves.

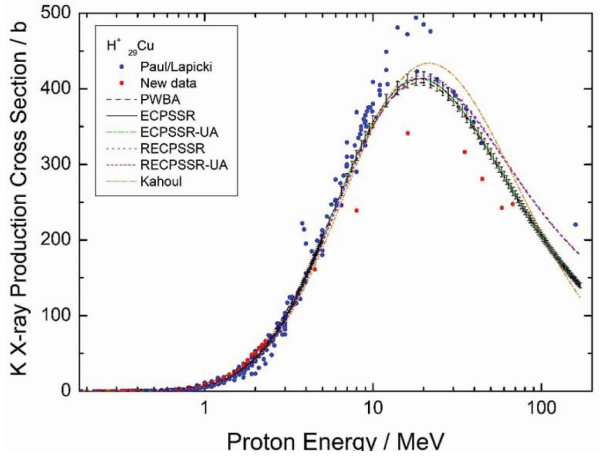

Figure 3: L_α X-ray production cross section of Pt induced by proton impact. The curves use the ECPSSR model [10], and relative intensities by Campbell and Wang [24], Puri [25], and Salem et al. [26]. Experimental data taken from the compilation by Miranda and Lapicki [5, 6].

Although many efforts have been devoted to improve the theories, it is necessary to make major progress in the

quality of the measurements of the atomic parameters. The current level of experimental uncertainties does not allow to identify adequately the differences among the models or their adaptations to add other secondary effects. The need of low uncertainty measurements of atomic parameters (fluorescence yields, Coster-Kronig transition probabilities and relative intensities) must be emphasized.

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Chitkara University, Saraswati Kendra, SCO 160-161, Sector 9-C, Chandigarh, 160009, India

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