Measurement of L X-ray production cross sections by impact of protons with energies between 2.5 MeV and 5.0 MeV in selected lanthanoids

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Accurate quantitative analysis with particle induced X-ray emission (PIXE) requires an accurate knowledge of the X-ray production cross sections, in particular the L lines. While there are a lot of experimental results, recent reviews have found a disagreement between several previously published papers and also by the use of different atomic parameters databases (fluorescence yields and Coster-Kronig transition probabilities). Therefore, it is very convenient to redo some of these cross sections measurements and to extend to other proton energy ranges. Thus, this paper presents results with lanthanoid elements (Ce, Nd, Sm, Eu, Gd, Dy, Ho) irradiated with protons in the energy range 2.5 MeV to 5.0 MeV. The results are compared also with predictions of the ECPSSR theory with the United Atom modification and experimental data of other authors.

Keywords: X-ray production cross sections; protons; ECPSSR; ECPSSR-UA.

La realización de análisis cuantitativos exactos con la emisión de rayos X inducida por partículas (PIXE) requiere del conocimiento de las secciones eficaces de producción de rayos X, en particular de las líneas L. A pesar de que existe una gran cantidad de resultados experimentales, en revisiones recientes se ha encontrado un desacuerdo entre diversos trabajos y la utilización de diferentes bases de datos de parámetros atómicos (producciones de fluorescencia y probabilidades Coster-Kronig), por lo cual se considera conveniente rehacer algunas de las mediciones de estas secciones y extender los intervalos de energías de los protones. Así pues, en este trabajo se presentan resultados obtenidos al irradiar elementos lantanoides (Ce, Nd, Sm, Eu, Gd, Dy, Ho) con protones en el intervalo de energías 2.5 MeV a 5.0 MeV. Se comparan, además, con predicciones de la teoría ECPSSR con modificación de átomo unido y los datos experimentales de otros autores.

Descriptores: Producción de rayos X; protones; ECPSSR; ECPSSR-UA.

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1. Introduction

The accurate knowledge of X-ray production cross sections by ion impact is necessary to adequately apply the analytical technique known as Particle Induced X-ray Emission (PIXE) [1]. As characteristic X-ray emission involves several atomic inner-shell processes, from primary ionization by the incoming radiation, up to the subsequent vacancy filling from outer shell electrons, including intrashell transitions, it is necessary to describe all of them appropriately. The description of K-shell X-ray emission is already satisfactory, so even reference cross sections have been published [2]. However, due to their increased complexity, L-shell and M-shell X-ray emissions have not been properly studied. Therefore, much effort has been dedicated to this task [3]. However, the models proposed to explain, for example, the L-shell ionization, show an increasing sophistication, and are difficult to use in any analytical applications. Moreover, the experimental data often present a very wide spread among themselves, making it difficult to decide which experimental results have a greater accuracy [3]. Additionally, for several elements the proton incident energy ranges are limited, either at low or high energies, so more experiments are required to increase and improve the existing database.

It has been frequently shown that L-shell X-ray production cross sections are acceptably described by the ECPSSR theory of Brandt and Lapicki [4]. This model improves the Plane Wave Born Approximation (PWBA) by taking into account 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 further improvement of the theory was obtained by the United Atom (UA) correction developed by Sarkadi and Mukoyama [5].

Therefore, the goal of this paper is to enlarge the existing experimental database, measuring the L X-ray production cross sections for selected elements in the atomic number range $58 \leq Z \leq 67$, for proton energies between 2.5 MeV and 5.0 MeV. An evaluation of the results is carried out through the application of the ECPSSR-UA model, by semiempirical predictions from tabulated values, as well as comparing with previously published experimental results.

2. Experimental

Samples were prepared in the form of thin films of fluorides of lanthanoids (Ce, Nd, Sm, Eu, Gd, Dy, and Ho) deposited onto carbon substrates. The advantage of using these substrates is that they may be thick, as there are low ion backscattering yields. Experiments were carried out with the 6 MV Tandem Van de Graaff accelerator at the Instituto Nacional de Investigaciones Nucleares. Proton beams were produced in energies from 2.5 MeV to 5.0 MeV, in 0.5 MeV steps. An Ortec Si(Li) X-ray detector (resolution 180 eV at 5.9 keV, located at an angle of 135° with respect to the incoming beam direction) collected the X-rays. The detector efficiency was measured using thin film standards (MicroMatter Co., Deer Harbor, WA, USA). In this case, 2.5 MeV protons were used as incident particles to induce the K X-rays, which are accurately known [2]. Backscattered ions were recorded with an Ortec surface-barrier detector (1000 μm thickness, located at an angle of 150°), as an absolute integrated charge measurement. The thickness of the lanthanoid film targets were measured previously [6] by Rutherford backscattering using ^{4}He$^+$ ions, with seven energies ranging from 2 MeV to 3 MeV. Results varied from 30 μg/cm^2 up to 150 μg/cm^2. Both the target thickness measurement and proton charge integration (with backscattered ions) is obtained through the analysis of spectra with the computer code SIMNRA 6.05 [7].

The uncertainties on the X-ray production cross sections were in the range 8% to 10% approximately. The main sources of uncertainty were the number of backscattered particles, the solid angle of the particle detector, the number of K$_\alpha$ X-rays produced in the standard films, the K-shell ionization cross sections, and the number of L$_i$ X-ray photons emitted by the lanthanoid targets.

L$_\alpha$, L$_\beta$, and L$_\gamma$ X-ray intensities were extracted from each X-ray spectrum using the non-linear least-square fitting routine QXAS [8] and then using the equation [9]:

$$\sigma_{L_i} = \frac{L_i \Omega_R \sigma_R(E_0)}{N_R \epsilon(E_{L_i})}\left[\frac{e^{-\mu t}}{1-\mu t}\right] \qquad (1)$$

Here, L_i is the number of X-ray photons in the respective peak, with $i = \alpha, \beta, \gamma$; N_R is the number of backscattered particles from the target; σ_R is the Rutherford scattering cross section at an angle of θ = 150° and the proton energy E_0, Ω_R= 3.62 (0.36) msr is the solid angle subtended by the particle detector from the target; $\varepsilon(E)$ is the X-ray detector efficiency, and the last parenthesis is a correction factor associated with X-ray absorption in the film, with μ the mass attenuation coefficient and t is the film thickness. The mass attenuation coefficient was obtained through the database XCOM [10].

3. Results and discussion

X-ray production cross sections for the target elements are available in tabular form upon request to the authors. Figs. 1 to 3 present the L$_\alpha$, L$_\beta$ and L$_\gamma$ X-ray production cross sections for Ce, respectively, as a function of the proton energy, to show an example. This plot also displays the theoretical predictions based on the ECPSSR theory and the United Atom approximation (UA), calculated with the ISICS computer code [11]. Also, the semiempirical tables published

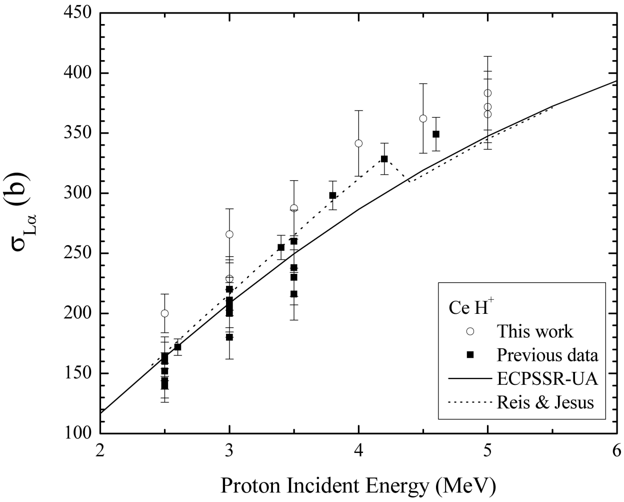

FIGURE 1. Ce L$_\alpha$ X-ray production cross sections induced by protons, as a function of proton incident energy.

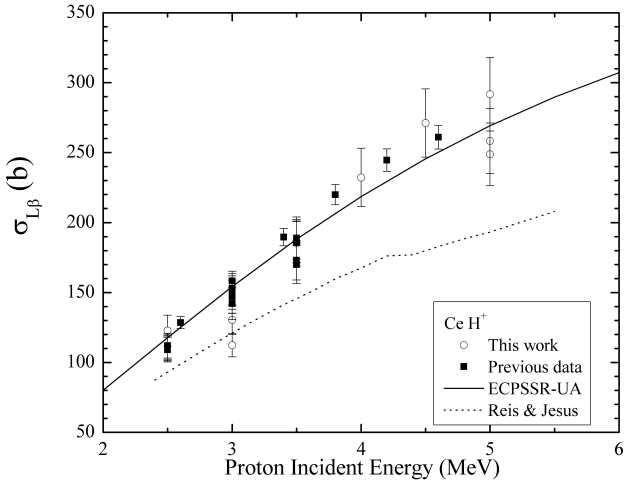

FIGURE 2. Ce L$_\beta$ X-ray production cross sections induced by protons, as a function of proton incident energy.

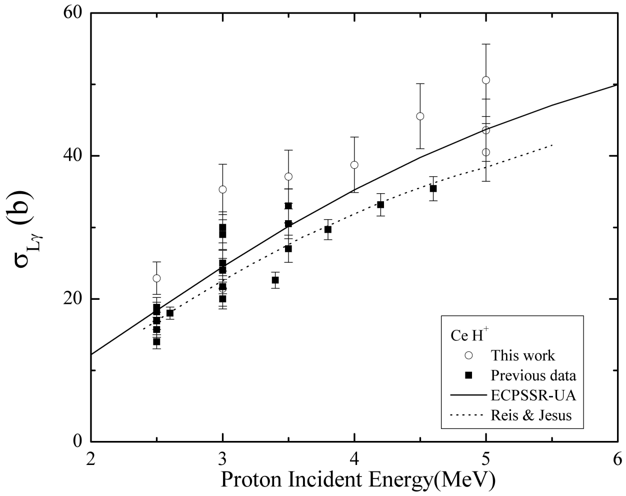

FIGURE 3. Ce L$_\gamma$ X-ray production cross sections induced by protons, as a function of proton incident energy.

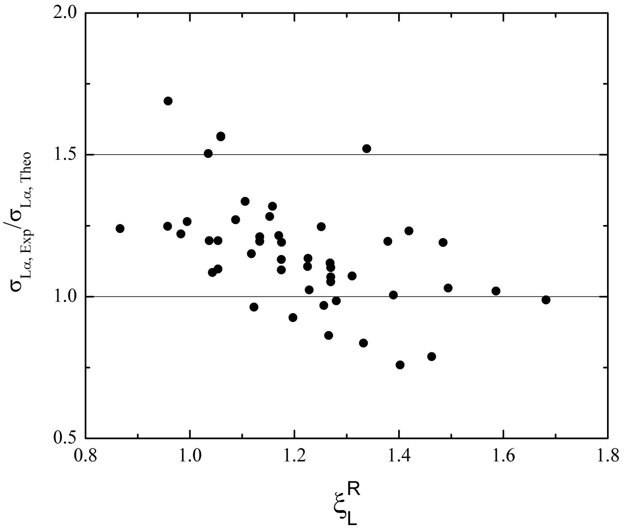

FIGURE 4. Ratios of experimental to theoretical X-ray production cross sections using the ECPSSR-UA theory, as a function of the reduced velocity parameter ξ_L^R.

by Reis and Jesus [12] were employed to make a comparison, although for the L_β lines they include only $L_{\beta1}$ and $L_{\beta2}$, and for L_γ only $L_{\gamma1}$ and $L_{\gamma2}$. The atomic parameters databases were the tables published Campbell [13] for fluorescence yields and Coster-Kronig transition probabilities, while emission rates were taken from Campbell and Wang [14]. Additionally, other experimental results are plotted in the same figures, taken form the compilation made by Lapicki and Miranda [3]. In this comparison, it is observed that the experimental results are in good agreement with previous experiments and the predictions of the ECPSSR-UA model. Furthermore, the curves obtained from the tables by Reis and Jesus [12] show a discontinuity at higher energies, which has no physical explanation.

A simple way to look at the whole set of experimental results, including those published previously, is to calculate the reduced velocity parameter ξ_L^R, defined by Rodriguez-Fernández et al. [15] as:

$$\xi_L^R = \frac{1}{4}\left(\xi_{L1}^R + \xi_{L2}^R + 2\xi_{L3}^R\right) \qquad (2)$$

where ξ_{Li}^R is the relativistic reduced velocity parameter of the L_i subshell (i =1, 2 or 3). The ratios of experimental to theoretical cross sections are then plotted as function of the parameter defined by eq. (2). Fig. 4 gives the results for this procedure using the ECPSSR-UA theory; there is an apparent trend in the experiment-to-theory ratio, decreasing as the proton incident energy is increased. The largest deviation occurs for low energies, where underestimations of the cross sections by the ECPSSR-UA may be as low as 50%, while for larger energies the values are larger than the experiment.

4. Conclusions

It is possible to conclude the following, from the results obtained above:

Results obtained in this work are in close agreement with those from other publications;

The scaling using the reduced velocity parameter ξ_L^R seems to be appropriate for the particular ion-target combinations;

The ECPSSR-UA theory, together with Campbell [13] tabulation of fluorescence yields and Coster-Kronig probabilities and Campbell-Wang emission rates [14], offers reasonably accurate predictions of the X-ray production cross sections;

A larger data set X-ray production cross sections using wider energy ranges and targets must be obtained, especially if further applications for ion beam analysis are sought;

The tables by Reis and Jesus [12] are inadequate for making predictions of the X-ray production cross sections.

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