Radiation Physics and Chemistry 95 (2014) 22–25

Contents lists available at ScienceDirect

Radiation Physics and Chemistry

journal homepage: www.elsevier.com/locate/radphyschem

Detailed calculation of inner-shell impact ionization to use in photon transport codes

Jorge E. Fernandez [a,*], Viviana Scot [a], Luca Verardi [b], Francesc Salvat [c]

[a] Laboratory of Montecuccolino-DIN, Alma Mater Studiorum University of Bologna, via dei Colli 16, I-40136 Bologna, Italy
[b] Department of Electrical Engineering (DIE), Alma Mater Studiorum University of Bologna, Viale del Risorgimento 2, I-40136 Bologna, Italy
[c] Facultat de Física (ECM), University of Barcelona, Diagonal 647, 08028 Barcelona, Spain

HIGHLIGHTS

- Some photons interactions produce also electrons as secondary particles.
- Inner-shell impact ionization (ISII) enhances XRF characteristic lines.
- PENELOPE is used to quantify the ISII in terms of angle, space and energy.
- A new photon kernel comprising the correction due to ISII is introduced.
- The impact of the ISII correction is discussed for the most intense K and L lines.

ARTICLE INFO

Article history:
Received 28 September 2012
Accepted 22 December 2012
Available online 31 December 2012

Keywords:
Inner-shell impact ionization
Determinist transport
Monte Carlo simulation
Photon-electron transport
XRF
Characteristic lines

ABSTRACT

Secondary electrons can modify the intensity of the XRF characteristic lines by means of a mechanism known as inner-shell impact ionization (ISII). The ad-hoc code KERNEL (which calls the PENELOPE package) has been used to characterize the electron correction in terms of angular, spatial and energy distributions. It is demonstrated that the angular distribution of the characteristic photons due to ISII can be safely considered as isotropic, and that the source of photons from electron interactions is well represented as a point source. The energy dependence of the correction is described using an analytical model in the energy range 1–150 keV, for all the emission lines (K, L and M) of the elements with atomic numbers $Z=11$–92. It is introduced a new photon kernel comprising the correction due to ISII, suitable to be adopted in photon transport codes (deterministic or Monte Carlo) with a minimal effort. The impact of the correction is discussed for the most intense K ($K_{\alpha1}$,$K_{\alpha2}$,$K_{\beta1}$) and L ($L_{\alpha1}$,$L_{\alpha2}$) lines.

1. Introduction

Compton scattering and photoelectric effect produce both photons and electrons as secondary particles. Secondary electrons may contribute to the photon field through emission mechanisms like bremsstrahlung (which produces a continuous photon spectrum) and inner-shell impact ionization (ISII) (which modifies the intensity of the characteristic lines). In particular, ISII is a characteristic emission produced as a consequence of electron induced ionization followed by spontaneous relaxation. Therefore, the most accurate description of the radiation field in X-ray spectrometry requires the modelling of coupled photon–electron transport. The whole problem is very complex because all of these mechanisms (plus other not mentioned here involving electron–electron interactions) have to be considered together. Moreover, since electrons interact continuously and locally (compared to the photons), it is necessary to consider also the multiple scattering of the electrons. A practical approach to describe the mentioned mechanisms in presence of multiple scattering is to use a coupled photon–electron Monte Carlo (MC) code with a detailed description of the electron and photon physics. In this work, the MC code PENELOPE (Salvat et al., 2008) has been used to calculate the effect of the secondary electrons into the photon transport in terms of angle, space and energy. In particular, PENELOPE has been used to compute a corrective term to the photon kernel which fully describes the effect of ISII. Since ISII modifies the intensity of the characteristic lines, it should be considered as a correction in X-ray fluorescence analysis.

It is shown that the characteristic photons contributed by ISII can be safely considered as an isotropic and point wise local source centred at the point of the primary interaction. The energy dependence of the correction is then computed in the range 1–150 keV, for all the emission lines (K, L and M) of the elements with atomic numbers $Z=11$–92. For each characteristic line, the energy dependence is described by a simple parametric model which uses an analytical expression for each one of the 5 energy

* Corresponding author. Tel.: +39 0516441718; fax: +39 0516441747.
E-mail address: jorge.fernandez@unibo.it (J.E. Fernandez).

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http://dx.doi.org/10.1016/j.radphyschem.2012.12.030

intervals delimited by the K, L1, L2 and L3 absorption edges. The formal representation of the photon kernel comprising the ISII correction is given. The new photon kernel is suitable to be adopted in photon transport codes (deterministic or Monte Carlo) with a minimal increase in complexity.

2. The new kernel

PENELOPE is a subroutine package, and cannot operate by itself. The user needs to build a main program for his/her particular problem, to read the parameters introduced through the input file, control the evolution of the generated tracks and keep score of relevant quantities during simulation. For our study, the ad-hoc code KERNEL (Verardi, 2011) was used to simulate a forced first collision producing photons and electrons at the origin of coordinates. A point source of monochromatic photons was considered. The physics of the interactions was described using the PENELOPE subroutine library. The model adopted in PENELOPE to describe ISII is highly detailed, as explained by Bote and Salvat (2008). All the secondary electrons were followed along their multiple-scattering until their energy become lower of a threshold value. All photons produced by the electrons at every stage were accumulated. Polarization was not considered at this time.

The angular, spatial and energy behavior of ISII was investigated using the KERNEL code. The polar angular distribution of IISI photons was simulated for different elements, with a monochromatic photon source of 100 keV. The distribution was confirmed to be isotropic. Then, the spatial distribution of the photon emission sites was analyzed in units of the Bethe range $R(E_0)$ (defined in an infinite medium as the average path-length travelled by a particle of kinetic energy E_0 in the course of its slowing down process, i.e., before being absorbed). Since in photon transport codes the length scale for the transport process is the photon mean free path mfp_p, the spatial distribution can be ignored when $\tau_s = R(E)/\mathrm{mfp}_p << 1$. The value of τ_s was evaluated using the program tables.f comprised in the PENELOPE package (Salvat et al., 2008). It was verified that τ_s never exceeds 10^{-1}. As electrons travel through the material following very tortuous paths, it is more appropriate to consider the effective range (the average longitudinal penetration length) instead of the Bethe range. The effective range is always smaller than the Bethe range (for gold, for example, is four times smaller), confirming the negligibility of the spatial distribution for the more conservative choice of the Bethe range.

Finally, the angular distribution of the characteristic photons due to ISII can be safely considered as isotropic, and the source of photons from electron interactions is well represented as a point source centered at the place of the photon collision.

To quantify the correction in terms of energy, PENELOPE calculations were performed for all the lines of the elements with $Z=11$–92 in the range 1–150 keV. To parameterize the correction for a generic energy, the whole energy interval was divided into five regions delimited by the K, L1, L2 and L3 absorption edges (see Fig. 1). A simple parametric expression was defined in every region. The whole model requires 20 parameters for each line.

To avoid data-base differences between PENELOPE and other transport codes, the electron correction $f_{\mathrm{ISII}}(\lambda')$ is computed as a relative one, in units of the photon contribution $Q_{\lambda_i}(\lambda')$. Then, the corrective term can be assembled as (Fernández and Scot, 2012):

$$\Delta k_{P_{\lambda_i}}\left(\vec{\omega},\lambda,\vec{\omega}',\lambda'\right)\big|_{\mathrm{electron}} = \frac{1}{4\pi} f_{\mathrm{ISII}}(\lambda') Q_{\lambda_i}(\lambda')\delta(\lambda'-\lambda_i)\ [1-U(\lambda'-\lambda_{e_i})],$$

where

$Q_{\lambda_i}(\lambda') = \tau_s(\lambda') g_{e_i}(\lambda') \Gamma_{\lambda_i}$ describes the uncorrected line probability emission in [cm^2/g], and

$$f_{\mathrm{ISII}}(\lambda') = \frac{\Delta Q_{\lambda_i}(\lambda')\big|_{\mathrm{electron}}}{Q_{\lambda_i}(\lambda')}.$$

$\tau_s(\lambda')$ represents the photoelectric mass attenuation coefficient [cm^2/g] of the emitter element s for the corresponding subshell, g_{e_i} denotes the radiative fraction for a given series of transitions, and Γ_{λ_i} the line emission probability of the line centered at λ_i into its own spectral series.

Thus, the corrected kernel comprising the electron contributions for a single line has the expression:

$$k_{P_{\lambda_i}}\left(\vec{\omega},\lambda,\vec{\omega}',\lambda'\right) = \frac{1}{4\pi} Q_{\lambda_i}(\lambda')(1+f_{\mathrm{ISII}}(\lambda'))\delta(\lambda'-\lambda_i)\ [1-U(\lambda'-\lambda_{e_i})]$$

The relative electron correction is parameterized by means of the best fit coefficients α_{kR} (for each one of the R regions defined in Fig. 1) as:

$$f_{\mathrm{ISII}}(\lambda) = \sum_{R=1}^{5} H_R(\lambda) \exp\left(\sum_{k=0}^{3} \alpha_{kR} \ln(E(\lambda))^k\right)$$

with

$$H_R(\lambda) = \begin{cases} 1, & \text{if } \lambda \text{ belongs to region } R \\ 0, & \text{otherwise} \end{cases}$$

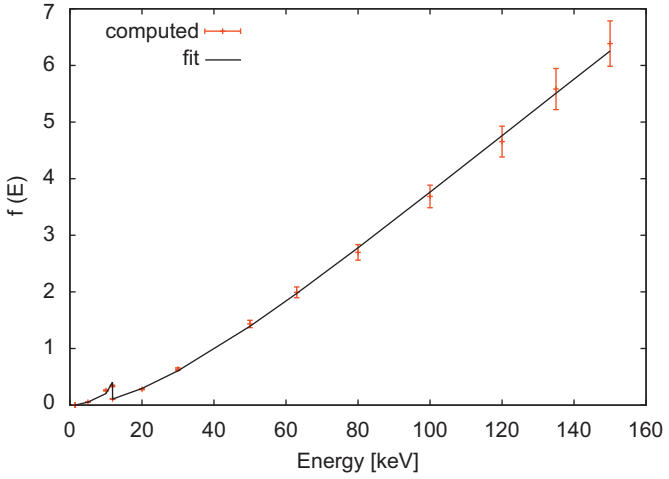

Fig. 2. Electron correction for As $L_{\alpha 1}$ (1.282 keV). For the higher energies, the correction is six times and more the value due to photons only. Notice the presence of a discontinuity due to the K absorption edge.

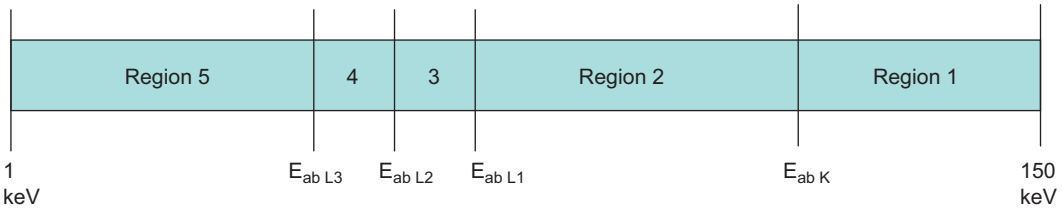

Fig. 1. Schematic representation of the division of the energy interval.