IOP Publishing Journal of Physics B: Atomic, Molecular and Optical Physics

J. Phys. B: At. Mol. Opt. Phys. **40** (2007) 3835–3841 doi:10.1088/0953-4075/40/19/004

Analytical expression for *K*- and *L*-shell cross sections of neutral atoms near ionization threshold by electron impact

C S Campos[1], M A Z Vasconcellos[2], J C Trincavelli[3,5] and S Segui[4,5]

[1] Instituto de Geociências, Centro de Pesquisa em Geologia e Geofísica, Universidade Federal da Bahia (UFBA), 40170-290 Salvador, BA, Brazil
[2] Instituto de Física, Universidade Federal do Rio Grande do Sul (UFRGS), 91501-970 Porto Alegre, RS, Brazil
[3] Facultad de Matemática, Astronomía y Física, Universidad Nacional de Córdoba, Ciudad Universitaria, 5000, Córdoba, Argentina
[4] Centro Atómico Bariloche, Comisión Nacional de Energía Atómica, 8400 San Carlos de Bariloche, Río Negro, Argentina

E-mail: segui@cab.cnea.gov.ar

Received 15 June 2007, in final form 14 August 2007
Published 19 September 2007
Online at stacks.iop.org/JPhysB/40/3835

Abstract

An analytical expression is proposed to describe the *K*- and *L*-shell ionization cross sections of neutral atoms by electron impact over a wide range of atomic numbers ($4 \leqslant Z \leqslant 79$) and over voltages $U < 10$. This study is based on the analysis of a calculated ionization cross section database using the distorted-wave first-order Born approximation (DWBA). The expression proposed for cross sections relative to their maximum height involves only two parameters for each atomic shell, with no dependence on the atomic number. On the other hand, it is verified that these parameters exhibit a monotonic behaviour with the atomic number for the absolute ionization cross sections, which allows us to obtain analytical expressions for the latter.

1. Introduction

Theoretical and experimental research has been carried out during the last few decades to determine the cross section associated with inner-shell ionization of neutral atoms due to electron impact. Knowledge of this function is of basic importance in elucidating physical mechanisms related to inelastic electron–atom interactions. In addition, ionization cross section data and their dependence with the atomic number and electron energy, particularly near the ionization threshold, are required in radiation physics, bulk, particle, surface and thin film analysis using electron probe microanalysis (EPMA), Auger electron spectroscopy (AES), electron energy-loss spectroscopy (EELS) and transmission electron microscopy (TEM). In

[5] Also member of the Consejo Nacional de Investigaciones Científicas y Técnicas (CONICET), Argentina.

particular, the models used for the ionization cross section in the conventional EPMA yield correct values except for a multiplicative factor different for each element, which is cancelled out when performing the ratio between the sample and the standard. Nevertheless, this cancellation cannot be performed in standardless applications and in these cases the need for absolute values for cross sections becomes a must.

Reviews concerning measurements and theoretical calculations of inner-shell ionization cross sections have been published by Powell [1, 2]. According to these reviews, it appears that both measurements and calculations of this quantity have been performed mostly for the *K*-shell and for incident electrons with high energies, whereas there are relatively few measurements or calculations for the *L*- and *M*-shells, and energies near the ionization threshold. In a pioneering work, Bethe [3] developed a formula to compute inner-shell ionization cross sections, based on the first Born approximation for high-energy particles which can be accurately described by plane waves. This expression was later modified to take into account relativistic effects [4], but it fails in describing exchange effects and the distortion of the projectile wavefunction due to the atomic field for electrons with kinetic energy comparable to the ionization threshold. More sophisticated calculations based on the plane wave Born approximation (PWBA) [5, 6] yield accurate cross section values only when the kinetic energy of the incident projectile is much larger than the ionization energy of the active electron shell. In order to account for the effects arising at energies close to the ionization threshold, semiempirical modifications of the PWBA have been proposed [7, 8]. Other theoretical models to obtain cross section data include classical and semiclassical formulations, such as that proposed by Gryzinski [9]. Finally, a large number of empirical and semiempirical analytical expressions have been proposed to describe the ionization cross section shape as a function of energy [1, 2, 10, 11]. However, these formulae are based on the available experimental data, which are limited both in number and accuracy [1, 2].

The purpose of this work is to provide analytical expressions for the ionization cross sections corresponding to *K*- and *L*-shells of elements with atomic numbers between 4 and 79, and within the energy range usual in EPMA and related techniques. The proposed model, inspired on the functional form given by Bethe in the high-energy limit, is a parameterization of the calculated data obtained with the distorted-wave Born approximation (DWBA), which has proved to be highly accurate [12].

2. Theoretical frame

Nowadays, the DWBA is one of the most sophisticated models for studying inner-shell excitations and ionizations, and allows us to evaluate cross sections systematically within certain energy limits. In that approximation, the wavefunctions for the initial and final states of the projectile are solutions of the Dirac equation for a distorting potential. This potential is set equal to the Dirac–Fock–Slater self-consistent atomic potential. Hence, the distorting effect on the wavefunctions of the projectile is accounted for 'exactly'. When the projectile is an electron, the DWBA also allows us to include electron exchange effects in a consistent way by antisymmetrizing the initial and final states of the system. This formulation has been coded in a FORTRAN 77 code named DWION (see [12]), which calculates energy-loss differential cross sections and total (integrated) cross sections for ionization of inner shells of neutral atoms and ions, by impact of electrons or positrons. This code uses the subroutine package RADIAL [13] to solve the Dirac radial equations and calculates vector coupling coefficients using high-accuracy arithmetics.

The reliability of DWBA calculations presented in [12] has been established for the *K*- and *L*-shells, by comparing with measurements of ionization and x-ray production cross

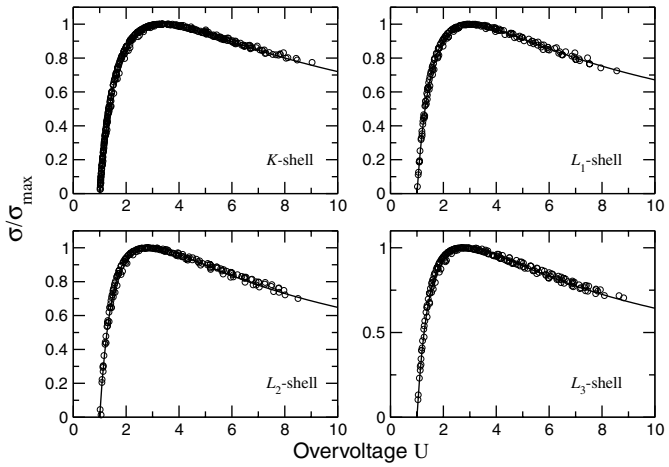

Figure 1. Overvoltage dependence of the calculated ionization cross sections by the DWBA method [12], normalized to the maximum height, for (a) K, (b) L_1, (c) L_2 and (d) L_3 atomic shells. The continuous curve is the result from the analytical expression proposed in this work. The symbols represent DWBA calculations of Be, B, C, O, F, Al, Si, S, K, Ti Cr, Mn, Fe, Ni, Cu, Ge and Zr for the K-shell, and of Ni, Cu, Ge, Zr, Ag, Te, W, Pt and Au for the $L_{1,2,3}$-subshells.

sections, respectively (see [14–18]). These comparisons show a satisfactory agreement within the energy range of interest in EPMA and other related techniques, i.e. up to about 8–10 times the ionization energy. For incident energies much larger than the ionization threshold, the convergence of the partial-wave series involved in the DWBA becomes very slow and the calculation is impractical. Moreover, since the code DWION calculates up to 50 000 radial integrals (or even more) to ensure the convergence of the partial-wave series, the computation is very slow, not only because of the number of radial integrals, but also due to the need of calculating them with a high precision to prevent the accumulation of numerical errors. Therefore, a simple model to determine reliable ionization cross section values based on these calculations would be of great interest for the application in the aforementioned analytical techniques.

In order to obtain an analytical expression to predict ionization cross section values with DWBA accuracy, a large database of theoretical values was generated with the DWION code to study the dependence of the cross section with the incident energy (E), the atomic number (Z) and the ionization threshold (E_c). The following sets of elements were considered: Be, B, C, O, F, Al, Si, S, K, Ti, Cr, Mn, Fe, Ni, Cu, Ge and Zr associated with the K-shell; and Ni, Cu, Ge, Zr, Ag, Te, W, Pt and Au associated with $L_{1,2,3}$-subshells. In figure 1, the relative ionization cross section values (normalized to the maximum height σ_{max}) are displayed as functions of the overvoltage $U = E/E_c$, for the K- and $L_{1,2,3}$-shells. The data calculated with the DWION are displayed with the same symbol for all the considered elements since, for each particular shell, no dependence could be observed on the atomic number Z. To select an adequate analytical form to fit the DWBA values, we recall that, as suggested by Bethe [3], the cross section is a function of U, and is asymptotically proportional to $\ln(U)/U$. This asymptotic dependence was also shown to describe adequately the ionization cross section of ions [19]. We thus propose the following expression for the normalized ionization cross section:

$$\frac{\sigma(U)}{\sigma_{max}} = \frac{a_n}{b_n + U}\ln(U), \tag{1}$$

Table 1. Expressions obtained for the parameters A_n and B_n as functions of the atomic number Z, for the K-shell and the $L_{1,2,3}$-subshells.

Shell	Parameter A_n	Parameter B_n
K	$3.135 \times 10^9 Z^{-4.3434}$	$\exp[0.665 - 0.614 \ln Z + 0.0810(\ln Z)^2 - 0.000\,05(\ln Z)^3]$
L_1	$2.203 \times 10^{12} Z^{-5.109}$	$12.909 Z^{-1.006}$
L_2	$7.5231 \times 10^{12} Z^{-5.3305}$	$\exp[4.4243 - 2.0777 \ln Z + 0.2039(\ln Z)^2] - 0.5$
L_3	$6.599 \times 10^{12} Z^{-5.0797}$	$4.8642 Z^{-0.5645} - 0.5$

where a_n and b_n are fitting parameters, and depend on the specific shell considered. This expression keeps the asymptotic behaviour and avoids the deviations found in the Bethe equation for overvoltages approaching unity, where the first Born approximation fails.

3. Results and discussion

Equation (1) was fitted for each shell, and is represented in figure 1 by the continuous curves. As can be seen, the proposed function describes very well the shape of the calculated data for all elements and shells, with relative differences below 4%. The values of the parameter a_n for the ionization cross sections normalized to the maximum height, associated with each atomic shell studied, are the following: $a_K = 3.339\,41$, $a_{L_1} = 3.000\,74$, $a_{L_2} = 2.844\,04$ and $a_{L_3} = 2.813\,76$; whereas the b_n values for each atomic shell studied are: $b_K = 0.6763$, $b_{L_1} = 0.2935$, $b_{L_2} = 0.098\,33$ and $b_{L_3} = 0.0710$.

Though relative cross section values may be useful in particular cases, such as quantitative analysis using standards, the absolute ionization cross section is a fundamental parameter to describe the combined transport of electron and photons. Detailed knowledge of this quantity is required for a number of applications such as radiotherapy, design of radiation detectors or the standardless quantitative electron probe microanalysis. In order to provide a simple and accurate expression to obtain absolute cross section values, we propose the following expression, in barns, based on equation (1),

$$\sigma(U) = \frac{A_n(Z)}{B_n(Z) + U} \ln(U), \tag{2}$$

where now the parameters $A_n(Z)$ and $B_n(Z)$ for the K- and L-shells were fitted for each particular atomic number Z. The behaviour of these parameters as a function of Z is presented in figure 2. For the K-shell, the parameters A and B exhibit a monotonic decrease with Z, as can be seen in figures 2(a) and (b). The same behaviour is also observed for the parameters corresponding to $L_{1,2,3}$-subshells (see figures 2(c) and (d)). Since it would be desirable to count with ionization cross section values for any element through the periodic table, and bearing in mind the smooth trends shown by the parameters, analytical expressions were fitted for them as a function of the atomic number. The obtained expressions are shown in table 1, and are illustrated in figure 2. As can be seen, there is a very good agreement between the parameters obtained from the DWION and the function proposed to fit them.

The results of absolute ionization cross sections calculated from equation (2) are plotted in figure 3 for the K-shell of iron and the L_3-subshell of gold. The obtained values are compared with experimental data [15, 20, 21] and with calculated values from the DWBA method [12]. As can be seen, the function proposed in the present method gives a good description of

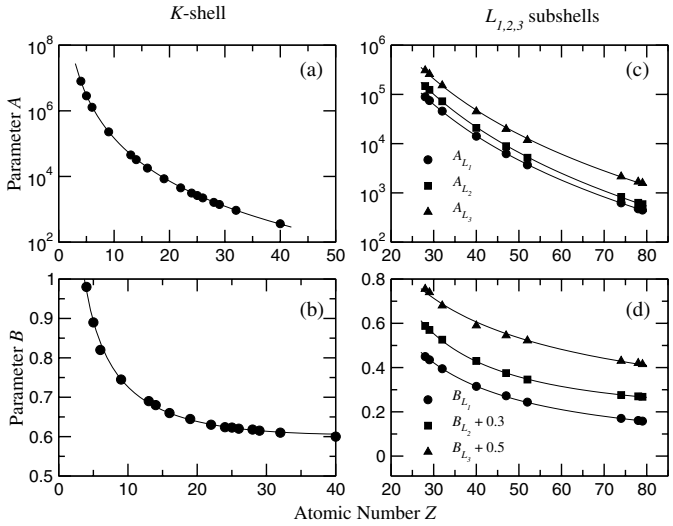

Figure 2. Variation of the parameters A and B with atomic number, given in equation (2) to describe the absolute ionization cross section values obtained by the DWBA method [12]. (a) Parameter A for K-shell. (b) Parameter B for K-shell. (c) Parameter A for L_1, L_2 and L_3-subshells. (d) Parameter B for L_1, L_2 and L_3-subshells. The continuous lines correspond to the functions fitted for each parameter given in table 1.

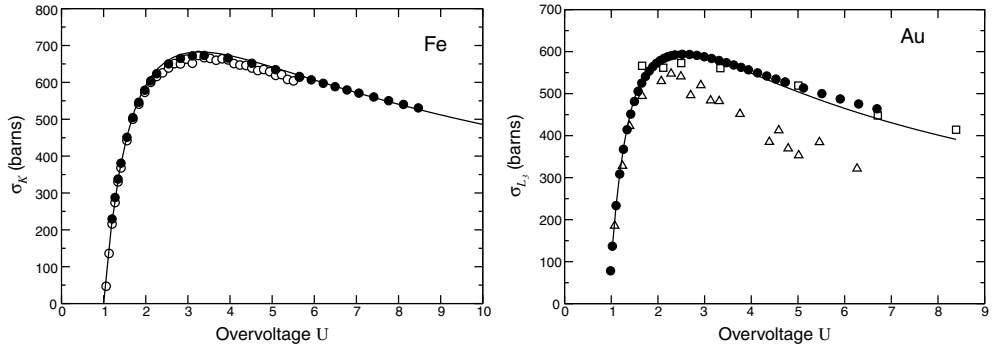

Figure 3. Total cross sections for ionization of the K-shell of iron and the L_3-shell of gold. The solid line represents the results obtained by equation (2) proposed in this work. The DWBA results are represented with solid circles [12]; the open symbols represent experimental data: circles [15], squares [20], triangles [21].

the theoretical data and reproduces the experimental values very satisfactorily. In particular, for iron, the experimental and predicted values almost coincide. In the case of gold, there are important discrepancies (up to 40%) among experimental data. The good agreement of DWBA calculations with data measured by Davis *et al* [19] suggests a systematical error in the data measured by Schneider *et al* [20]. On the other hand, equation (2) produces a slight underestimation for DWBA data (around 5% for the less favourable overvoltage and the atomic number considered).

A further comparison is presented in figure 4 for nickel K-shell ionization. In this figure, besides the comparison with experimental data [14], and DWBA calculations, the values

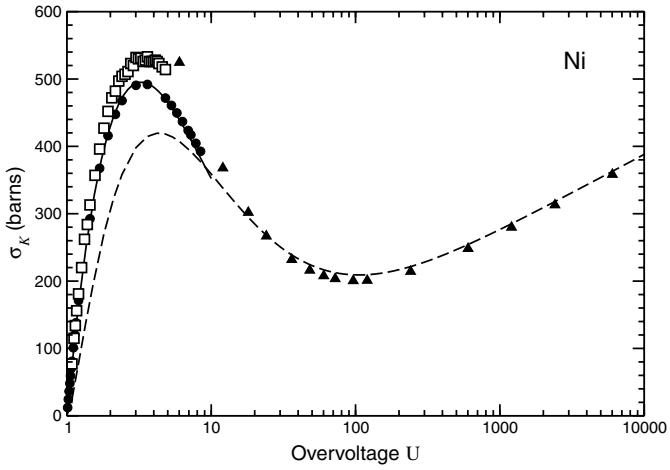

Figure 4. Total cross section for ionization of the *K*-shell of nickel obtained using the DWBA method (solid circles) [12], the model proposed by Mayol and Salvat [7] (dashed line) and calculations by Scofield [6] (triangles). The open symbols are experimental data [14]. The solid line represents the results obtained with equation (2) proposed in this work.

obtained using the model proposed by Mayol and Salvat [7] are included. This model is based on a representation of the generalized oscillator strength (GOS), and includes corrections to take into account exchange and Coulomb effects. As can be seen, the results obtained with the DWBA, and hence the fitting function proposed in this work, describe more accurately the shape of the experimental data near the ionization threshold ($U < 5$); differences in absolute values are systematic and can be easily overrided by rescaling with a constant factor [12]. For incident energies above ten times the ionization energy, the GOS-based model [7] generates cross section values which are in close agreement with more sophisticated calculations, such as those provided by Scofield [6]. Finally, it is worth remarking that the low-energy DWBA cross sections can be extended to intermediate energies by means of the analytical expressions proposed in this work, to merge with the high-energy models [6, 7], as is suggested by the plot.

4. Conclusion

The analytical expression proposed in this work represents a convenient approach to generate absolute ionization cross sections for any element and energy near the threshold ($U < 10$) in a straightforward way, interpolating the discrete values calculated with the DWBA. It is worth mentioning that DWION results have already been used to generate a database for the PENELOPE simulation code [22]; the adoption of the present model may reduce the computation time keeping the accuracy of the simulated x-ray spectra of different elements in such calculations [23].

As has been mentioned before, one of the motivations of this work was the lack of reliable ionization cross section values, needed in numerous analytical techniques such as EPMA. We think that the proposed expression will cover these needs for *K*- and *L*-shells, at least within the range of validity of the fit. Moreover, though the DWION code generates cross sections for a limited range of energies, the present model can be extrapolated to intermediate energies to be combined with high-energy models, covering a wider range of incident energies.

Acknowledgment

CSC would like to thank financial support from FAPESB (Fundação de Amparo à Pesquisa no Estado da Bahia).

References

[1] Powell C J 1985 *Electron Impact Ionization* ed T D Mark and D H Dunn (Berlin: Springer)
[2] Powell C J 1990 *Microbeam Analysis* ed J R Michael and P Ingram (San Francisco, CA: San Francisco Press)
[3] Bethe H 1930 *Ann. Phys.* **5** 325
[4] Bethe H 1932 *Z. Phys.* **76** 293
[5] Fano U 1963 *Ann. Rev. Nucl. Sci.* **13** 1
[6] Scofield J H 1978 *Phys. Rev.* A **18** 963
[7] Mayol R and Salvat F 1990 *J. Phys. B: At. Mol. Opt. Phys.* **23** 2117
[8] Hippler R 1990 *Phys. Lett.* A **144** 81
[9] Gryzinski M 1965 *Phys. Rev.* A **138** 305
[10] Casnati E, Tartari A and Baraldi C 1982 *J. Phys. B: At. Mol. Phys.* **15** 155
[11] Hombourger C 1998 *J. Phys. B: At. Mol. Opt. Phys.* **31** 3693
[12] Segui S, Dingfelder M and Salvat F 2003 *Phys. Rev.* A **67** 062710
[13] Salvat F, Fernández-Varea J M and Williamson W Jr 1995 *Comput. Phys. Commun.* **90** 151
[14] Llovet X, Merlet C and Salvat F 2000 *J. Phys. B: At. Mol. Opt. Phys.* **33** 3761
[15] Llovet X, Merlet C and Salvat F 2002 *J. Phys. B: At. Mol. Opt. Phys.* **35** 973
[16] Llovet X, Merlet C, Fernández-Varea J M and Salvat F 2000 *Mikrochim. Acta* **132** 163
[17] Campos C S, Vasconcellos M A Z, Llovet X and Salvat F 2002 *Phys. Rev.* A **66** 012719
[18] Merlet C, Llovet X and Fernández-Varea J M 2006 *Phys. Rev.* A **73** 062719
[19] Bernshtam V A, Ralchenko Yu V and Maron Y 2000 *J. Phys. B: At. Mol. Opt. Phys.* **33** 5025
[20] Davis D V, Mistry V D and Quarles C A 1972 *Phys. Lett.* **38A** 169
[21] Schneider H, Tobehn I, Ebel F and Hippler R 1993 *Phys. Rev. Lett.* **71** 2707
[22] Salvat F, Fernández-Varea J M and Sempau J 2003 PENELOPE—a code system for Monte Carlo simulation of electron and photon transport. France: OECD/NEA Data Bank, Issy-les-Moulineaux, France
[23] Llovet X, Sorbier L, Campos C S, Acosta E and Salvat F 2003 *J. Appl. Phys.* **93** 3844