INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS B: ATOMIC, MOLECULAR AND OPTICAL PHYSICS

J. Phys. B: At. Mol. Opt. Phys. **39** (2006) 1499–1516 doi:10.1088/0953-4075/39/6/018

Double ionization of heavy positive ions by electron impact: empirical formula and fitting parameters for ionization cross sections

V P Shevelko[1], H Tawara[2], I Yu Tolstikhina[1], F Scheuermann[3], B Fabian[3,4], A Müller[3] and E Salzborn[3]

[1] P N Lebedev Physical Institute, 119991 Moscow, Russia
[2] Max-Planck Institut für Kernphysik, D-69117 Heidelberg, Germany
[3] Institut für Atom- und Molekülphysik, Justus-Liebig-Universität Giessen, D-35392 Giessen, Germany

E-mail: shev@sci.lebedev.ru

Received 24 November 2005, in final form 10 February 2006
Published 6 March 2006
Online at stacks.iop.org/JPhysB/39/1499

Abstract

Electron-impact double-ionization processes of multi-electron heavy ions from Ti up to Bi (nuclear charge from $Z = 22$ up to $Z = 83$) are investigated for incident electron energies $E < 50\ I_{\rm th}$ where $I_{\rm th}$ is the threshold energy for double ionization. The following ions are considered: Ti^{q+} ($q = 1$–6), Fe^{q+} ($q = 1, 3$–6), Ni^{q+} ($q = 1$–6), Ga^{q+} ($q = 1$–6), Kr^{q+} ($q = 1$–4), Mo^{q+} ($q = 1$–6), Pr^{q+} ($q = 1$–4), Sm^{q+} ($q = 1$–6), W^{q+} ($q = 1$–6), Pb^{q+} ($q = 1$–9) and Bi^{q+} ($q = 1$–10, 12). On the basis of experimental data, mostly obtained at an electron–ion crossed-beams set-up in Giessen, and quantum-mechanical considerations, a simple semi-empirical formula with six fitting parameters is developed taking into account the contribution of direct double ionization of two outer-shell electrons of the ions and also of single inner-shell ionization processes followed by autoionization with additional ejection of an electron as was suggested in our previous paper (Shevelko *et al* 2005). The formula obtained there was found to describe well the available experimental double-ionization cross sections within an accuracy of 20–30%. However, for multi-electron, very heavy ions significant deviations of that formula from experiment are found in the low-energy region. These deviations are most likely caused by higher order processes, including inner-shell excitation and subsequent double autoionization (EDA). The tabulated parameters can be used for easy analytical representation of the double-ionization cross sections of heavy positive ions in the modelling of laboratory and astrophysical plasmas.

[4] Present address: Aston Laboratory for Mass Spectrometry, Purdue University, West Lafayette, IN 47907-2084, USA.

1. Introduction

The ionization processes of positive ions by electron impact are of critical importance for many fundamental problems and in applications related to investigations of laboratory and astrophysical plasmas as well as radiation physics, ion source development, accelerator physics and tumour therapy physics (see, e.g., Currell (2002)).

The quantitative quantum-mechanical description of the ionization cross sections of multi-electron heavy ions is an extremely complicated problem even for electron-impact single-ionization processes because it requires accurate calculations of outer- and inner-shell excitation, and ionization as well as branching ratios (see, e.g., Loch *et al* (2005)). However, for many applications like those in tokamak or astrophysical plasmas, there is a need to represent the data in a closed analytical form using relatively simple formulae. Such formulae should be based upon quantum-mechanical treatment with a suitable description of the asymptotic high-energy behaviour of the cross sections so that they can be extrapolated to higher electron energies.

Electron-impact single ionization is usually dominant among various ionization processes, but in some cases the double electron ionization processes play a key role. In our previous paper (Shevelko *et al* 2005), double-ionization processes of relatively *light* positive ions with the atomic numbers $Z < 26$ under electron impact were systematically analysed and, on the basis of reliable experimental data and quantum-mechanical calculations, a simple semi-empirical formula for their cross sections was proposed and found to reproduce the data with an accuracy of 20–30%. The present work is a continuation of the previous work aiming to obtain a similar semi-empirical formula, in a closed analytical form, for electron-impact double-ionization cross sections of *heavy, multi-electron* ions in a wide range of atomic numbers (from $Z = 22$ up to $Z = 83$) with relatively low ionization charge states.

In the analytical formula proposed here, the double-ionization cross sections are presented as the sum of two contributing parts: (i) direct ionization cross sections σ_{dir} for simultaneous ionization of two outer-shell electrons in the target ion and (ii) the sum over single-ionization cross sections of inner-shell electrons leading to net double ionization via subsequent autoionization processes. The necessary input parameters in the proposed formula for double ionization are the threshold energy for the direct double ionization and binding energies of inner-shell electrons of the target ion, similar to the well-known Lotz formula for electron-impact single-ionization cross sections (Lotz 1970).

2. Fitting formula for electron-impact double-ionization cross sections

In the present work, the double-ionization cross sections are fitted in a way similar to the previous work (Shevelko *et al* 2005), i.e., with a formula consisting of two parts as given in equation (1). We consider the double ionization of multi-electron positive ions X^{q+},

$$e^- + X^{q+} \rightarrow X^{(q+2)+} + 3e^-$$

and represent its cross section σ_2 in the form

$$\sigma_2 = \sigma_{\mathrm{dir}} + \sigma_{\mathrm{indir}} \equiv \sigma_{\mathrm{dir}} + \sum_{\gamma} a_\gamma \sigma_\gamma, \qquad I_\gamma \geqslant I_{\mathrm{th}}. \tag{1}$$

The first term, σ_{dir}, denotes the *direct* double-ionization cross section for simultaneous ionization of two of the outermost electrons, and the second term, σ_{indir}, that for the so-called *indirect* processes related to single inner-shell ionization followed by autoionization. Here I_{th} denotes the threshold energy for double ionization which is the sum of the first (I_1) and the second (I_2) ionization potentials of the target ion X^{q+},

$$I_{\rm th} = I_1 + I_2, \tag{2}$$

and σ_γ denotes the electron-impact single-ionization cross section of the inner-shell labelled by γ, and a_γ is the corresponding branching ratio for autoionization processes given by

$$a_\gamma = \frac{A^a_\gamma}{A^a_\gamma + A^r_\gamma}. \tag{3}$$

Here A^a_γ and A^r_γ denote the autoionization and radiative decay rates, respectively, for the ion $X^{(q+1)+}$ having a vacancy in the shell γ. Equation (1) has to be summed over all possible inner shells for which the binding energy of an electron I_γ is equal to or larger than the threshold energy of the direct double ionization, i.e., $I_\gamma \geqslant I_{\rm th}$.

In the previous work (Shevelko *et al* 2005) it was found that in many cases the direct double-ionization cross section is well described by the semi-empirical formula

$$\sigma_{\rm dir}(E) = 10^{-13}\,{\rm cm}^2(1 - {\rm e}^{-3(u-1)})\frac{A_{\rm dir}}{I_{\rm th}^3}\frac{u-1}{(u+0.5)^2}, \qquad u = E/I_{\rm th}, \tag{4}$$

where $A_{\rm dir}$ denotes the fitting parameter (in units of eV^3), u is the scaled energy E of the incident electron over the threshold energy $I_{\rm th}$. We have empirically found that the direct part for double-ionization cross sections scales as ${I_{\rm th}}^{-3}$ against the scaled electron energy $E/I_{\rm th}$, in contrast to the single-ionization cross section scaling as ${I_{\rm th}}^{-2}$, thus leading to equation (4).

The factor $(1 - {\rm e}^{-3(u-1)})$ is introduced to provide the reasonable asymptotic behaviour of $\sigma_{\rm dir}(E)$ at a low electron energy range according to the Wannier law (Wannier 1955): $\sigma_{\rm dir}(E) \sim (E - I_{\rm th})^\alpha$, $\alpha \approx 2$, for the double ionization near $E \sim I_{\rm th}$. Strictly speaking, the power α is not exactly equal to 2 but slightly differs from this value as more exact calculations show (see, e.g., Klar and Schlecht 1976 and Grujić 1982). However, we use the value of $\alpha = 2$ which closely follows from the classical Wannier law and perturbation theory because we are interested in the energy region above the threshold energy and, thus, this choice of α is not very critical for our purpose to present the double-ionization cross sections over a wide electron energy range.

In relatively light target ions (Shevelko *et al* 2005), the contribution of ionization–autoionization can be described with the known branching ratios a_{1s} for the 1s shell vacancy. In heavy ions, one has to know a_γ for many inner-shell electrons contributing to the double ionization, but their calculation for L, M, N..., shells in multi-electron systems is not straightforward. Moreover, for heavy ions many other processes can be important: (i) excitation of the inner-shell electron followed by double autoionization (EDA) and (ii) resonant excitation–triple-autoionization where the incident electron is captured into the target ion, while one of the inner-shell electrons in this ion is excited. The resulting multiply excited state decays preferentially through sequential or simultaneous emissions of three electrons and thus contributes to net double ionization of the parent ion. Certainly, a more complete description of double-ionization cross sections is not an easy task.

In order to describe the double-ionization cross sections in a wide range of incident electron energies, logarithmic terms have to be included in the fitting formula. Thus, we finally fit the total double-ionization cross sections σ_2 in the following form,

$$\begin{aligned}\sigma_2(E) = 10^{-13}\,{\rm cm}^2 \times \Bigg[&(1 - {\rm e}^{-3(u-1)})\frac{A_{\rm dir}}{I_{\rm th}^3}\frac{u-1}{(u+0.5)^2}\,(1 + 0.1\ln(4u+1)) \\ &+ \sum_\gamma \frac{A_\gamma}{I_\gamma^2}\frac{x-1}{x(x+5.0)}\left(1 + \frac{0.3}{n}\ln(4x+1)\right)\Bigg] \\ &u = E/I_{\rm th} \geqslant 1, \quad x = E/I_\gamma \geqslant 1,\end{aligned} \tag{5}$$

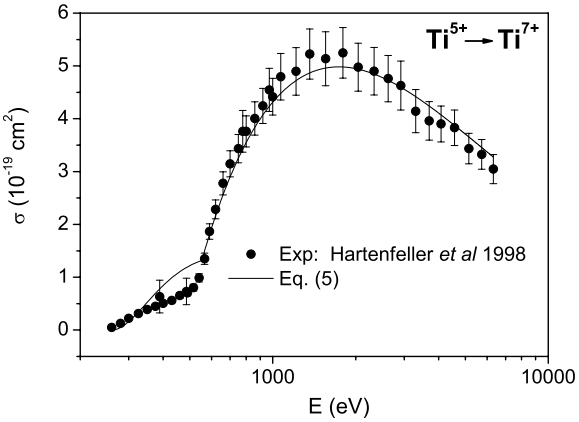

Figure 1. Electron-impact double-ionization cross section of Ti^{5+} ions. Full circles—experiment (Hartenfeller *et al* 1998); solid line—equation (5) with parameters given in table 1.

Table 1. Threshold energies I_{th}, binding energies I_γ (both in eV), and fitting parameters of equation (5) for double ionization of Ti^{q+} ions ($Z = 22$): A_{dir} (in eV^3) and A_γ (in eV^2). The electron energy range applicable to equation (5) is given in the last column.

Ion configuration	I_{th} A_{dir}	I_{3p} A_{3p}	I_{3s} A_{3s}	I_{2p} A_{2p}	I_{2s} A_{2s}	Energy range
$Ti^{1+}(3d^24s)$	41.07	48.83	80.82	473.4	581.4	$E < 3$ keV
	41.7	3.8	0.5	0.5	20.0	
$Ti^{2+}(3p^63d^2)$	70.76	58.20	90.23	483.2	591.1	$E < 3$ keV
	201.1	–	0.10	20.0	10.0	
$Ti^{3+}(3p^63d^1)$	142.49	75.81	108.6	503.2	611.7	$E < 3$ keV
	144.6	0.0	0.0	6.0	0.01	
$Ti^{4+}(3s^23p^6)$	218.6	97.79	129.2	527.9	635.6	$E < 3$ keV
	87.2	0.0	0.0	12.85	0.001	
$Ti^{5+}(3s^23p^5)$	260.2	117.9	146.5	552.6	662.1	$E < 10$ keV
	149.3	0.0	0.0	9.5	0.01	
$Ti^{6+}(3s^23p^4)$	309.3	139.0	164.7	580.5	690.2	$E < 3$ keV
	149.5	0.0	0.0	9.2	0.01	

where E denotes the incident electron energy, I_{th} is the lowest threshold energy of (outer-shell) double ionization, I_γ is the binding energy of the inner-shell electron (all the energies in eV), and n is the principal quantum number of the electron to be ionized. Here A_{dir} (in eV^3) and A_γ (in eV^2) are the fitting parameters. The logarithmic term is known to depend on the principal quantum number n of the electron to be ionized (Beigman *et al* 1995). The fitting parameters (A_{dir} and A_γ) strongly depend on the atomic structure of the target ion: ion charge, principal and orbital quantum numbers, availability of autoionization states and others. We add, for clarity, that each term in the sum occurring in equation (5) vanishes for $u, x \leqslant 1$.

3. Numerical calculations and comparison with experiment

The parameters A_{dir} and A_γ in equation (5) were determined by the least-square fitting the formula (5) to the experimental double-ionization cross sections for Ti^{q+} ($q = 1$–6), Fe^{q+} ($q = 1, 3$–6), Ni^{q+} ($q = 1$–6), Ga^{q+} ($q = 1$–6), Kr^{q+} ($q = 1$–4), Mo^{q+} ($q = 1$–6), Pr^{q+} ($q = 1$–4), Sm^{q+} ($q = 1$–6), W^{q+} ($q = 1$–6), Pb^{q+} ($q = 1$–9), Bi^{q+} ($q = 2$–4, 7–10, 12)

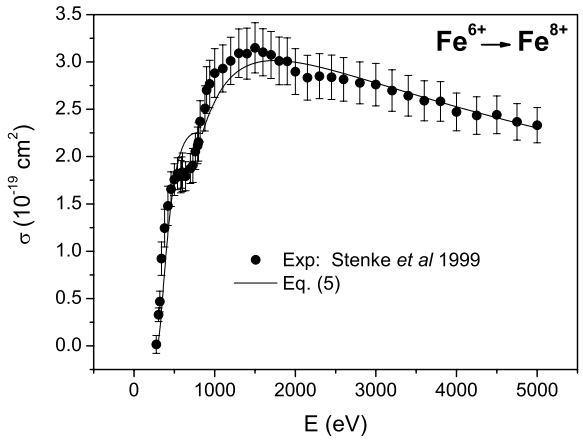

Figure 2. Electron-impact double-ionization cross section of Fe^{6+} ions. Full circles—experiment (Stenke *et al* 1999); solid line—equation (5) with parameters given in table 2.

Table 2. Threshold energies I_{th}, binding energies I_γ (in eV), and fitting parameters of equation (5) for double ionization of Fe^{q+} ions ($Z = 26$): A_{dir} (in eV^3) and A_γ (in eV^2). The electron energy range applicable to equation (5) is given in the last column.

Ion configuration	I_{th} A_{dir}	I_{3p} A_{3p}	I_{3s} A_{3s}	I_{2p} A_{2p}	I_{2s} A_{2s}	Energy range
$Fe^{1+}(3d^64s)$	43.54	69.58	112.85	727.64	868.24	$E < 3$ keV
	17.0	3.72	3.31	1.0	1.0	
$Fe^{2+}(3p^63d^6)$	80.60	80.45	123.74	738.60	879.18	
	–	–	–	–	–	–
$Fe^{3+}(3p^63d^5)$	129.8	99.7	142.1	760.7	900.5	$E < 3$ keV
	359.0	0.0	1.0	1.0	1.0	
$Fe^{4+}(3p^63d^4)$	174.1	120.5	165.7	786.2	927.1	$E < 5$ keV
	303.0	0.0	0.0	0.1	0.01	
$Fe^{5+}(3p^63d^3)$	222.3	145.4	191.1	815.9	956.4	$E < 10$ keV
	281.5	0.0	0.0	2.0	11.0	
$Fe^{6+}(3p^63d^2)$	286.0	171.5	218.2	847.2	988.6	$E < 10$ keV
	261.3	0.0	0.0	8.0	2.0	

ions. The binding energies I_1 and I_2 were either taken from tables (Lide 2000–2001) or calculated by the Flexible Atomic Code (FAC) (Gu 2003). The inner-shell binding energies (I_γ) were calculated using also the FAC. The binding and threshold energies, used in this paper, together with the final fitted parameters are given in tables 1–11. The last column of these tables indicates the maximum electron energy for which the formula can be applied. The upper limit of the energy was estimated to be approximately three times the binding energies of the next (deeper) inner-shell electrons where their single-electron ionization cross sections reach roughly a maximum.

The data of the Giessen group were obtained using the electron-impact ionization facility described in detail in the works of Tinschert *et al* (1989), Hofmann *et al* (1990) and Jacobi *et al* (2005). For all data except for Pb and Bi a 10 GHz ECR ion source (Liehr *et al* 1993) has been used. For production of Pb and Bi ions a 14 GHz ion source (Brötz 2000) has been installed. While Ti, Fe, Ni and Mo ions were produced by axially inserting bundles

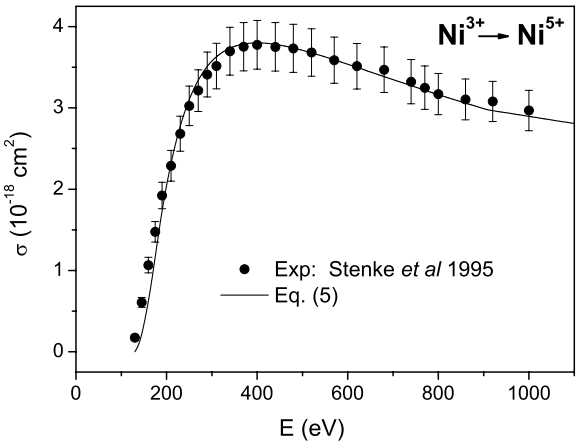

Figure 3. Electron-impact double-ionization cross section of Ni^{3+} ions. Full circles—experiment (Stenke *et al* 1995); solid line—equation (5) with parameters given in table 3.

Table 3. Threshold energies I_{th}, binding energies I_γ (in eV), and fitting parameters of equation (5) for double ionization of Ni^{q+} ions: A_{dir} (in eV^3) and A_γ (in eV^2). The electron energy range, for which equation (5) can be applied, is given in the last column.

Ion configuration	I_{th} A_{dir}	I_{3d} A_{3d}	I_{3p} A_{3p}	I_{3s} A_{3s}	I_{2p} A_{2p}	I_{2s} A_{2s}	Energy range
$Ni^{1+}(3d^84s)$	53.34	20.69	84.68	135.26	875.81	1050.4	$E < 3$ keV
	15.6	0.0	4.5	3.0	0.1	0.00	
$Ni^{2+}(3p^63d^8)$	90.1	32.18	96.20	146.78	887.42	1046.6	$E < 3$ keV
	58.12	0.0	4.0	3.0	0.1	0.0	
$Ni^{3+}(3p^63d^7)$	130.4	52.73	115.48	166.72	910.24	1069.7	$E < 3$ keV
	279.5	0.0	0.0	3.53	35.0	0.0	
$Ni^{4+}(3p^63d^6)$	183.5	74.09	137.87	188.74	936.67	1095.9	$E < 3$ keV
	376.6	0.0	0.0	0.93	0.1	0.00	
$Ni^{5+}(3p^63d^5)$	241.0	106.14	162.73	212.54	966.60	1124.9	$E < 3$ keV
	400.2	0.0	0.0	0.0	17.0	0.00	
$Ni^{6+}(3p^63d^4)$	295.0	131.84	188.14	240.84	998.62	1157.8	$E < 3$ keV
	360.0	0.0	0.0	0.0	0.1	0.0	

of thin wires of the desired metals near the edge of the ECR plasmas, all other elements (except for krypton) were evaporated into the ECR plasmas from an oven. The ions extracted at typically 10 kV were mass-to-charge selected using a magnet and, after collimation and beam formation, transported to the interaction region, where the ions crossed the electron beam at 90°. For electron energies up to 1 keV, a high-current electron gun (Becker *et al* 1985) was used, and for higher energies up to 6.5 keV a high-voltage electron gun (Stenke *et al* 1995) was employed. The primary ion beam and the product ions (with their charge state increased by two units) were separated by a second magnet. While the parent ion beam current was measured with a Faraday cup, the secondary ions were registered using a single particle detector. The absolute cross sections were obtained employing the dynamic crossed-beams technique (Müller *et al* 1985) where the electron gun is moved mechanically up and down across the ion beam at a fixed position. The total experimental uncertainties of the measured

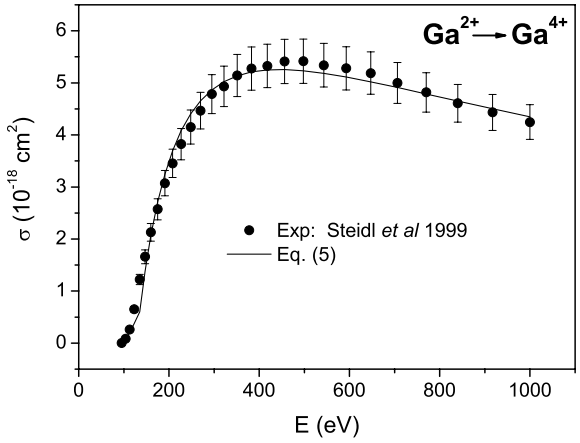

Figure 4. Electron-impact double-ionization cross section of Ga^{2+} ions. Full circles—experiment (Steidl *et al* 1999); solid line—equation (5) with parameters given in table 4.

Table 4. Threshold energies I_{th}, binding energies I_γ (in eV), and fitting parameters of equation (5) for double ionization of Ga^{q+} ions: A_{dir} (in eV^3) and A_γ (in eV^2). The electron energy range, for which equation (5) can be applied, is given in the last column.

Ion configuration	I_{th} A_{dir}	I_{3d} A_{3d}	I_{3p} A_{3p}	I_{3s} A_{3s}	I_{2p} A_{2p}	I_{2s} A_{2s}	Energy range
$Ga^{1+}(3d^{10}4s^2)$	51.22	33.14	122.18	179.02	1133.1	1322.1	$E < 4$ keV
	30.3	0.0	7.3	0.1	0.0	0.0	
$Ga^{2+}(3d^{10}4s)$	94.71	45.59	134.73	191.21	1145.9	1334.7	$E < 4$ keV
	48.4	0.0	7.17	0.10	0.01	0.00	
$Ga^{3+}(3p^63d^{10})$	142.5	59.62	148.85	205.29	1160.1	1348.9	$E < 4$ keV
	315.5	0.0	0.0	4.0	0.1	0.0	
$Ga^{4+}(3p^63d^9)$	226.0	82.87	168.41	229.32	1186.9	1376.9	$E < 4$ keV
	900.0	0.0	0.0	0.40	0.01	0.00	
$Ga^{5+}(3p^63d^8)$	294.7	110.0	191.84	255.17	1216.9	1407.7	$E < 10$ keV
	974.0	0.0	0.0	0.0	0.04	0.0	
$Ga^{6+}(3p^63d^7)$	323.3	139.2	218.67	282.68	1225.2	1441.6	$E < 10$ keV
	660.0	0.0	0.0	0.0	0.04	1.5	

cross sections are typically 8% at the maximum cross section resulting from the quadrature sum of the non-statistical errors of 7.8% and the statistical error at the 90% confidence level.

3.1. Ti^{q+} ions with $q = 1$–6 (atomic number $Z = 22$)

The calculated threshold energies I_{th}, binding energies I_γ and the best fitting parameters of equation (5) for double ionization of Ti^{q+} ions are given in table 1. In general, equation (5) with parameters in table 1 yields a quite good description of the experimental cross sections (Hartenfeller *et al* 1998). A typical example of the fitting is given in figure 1 for double ionization of Ti^{5+} ions. Only for the charge state $q = 5$ experimental data are available at energies beyond 1 keV, meanwhile in all the other cases 1 keV is the maximum energy covered by the experiments.

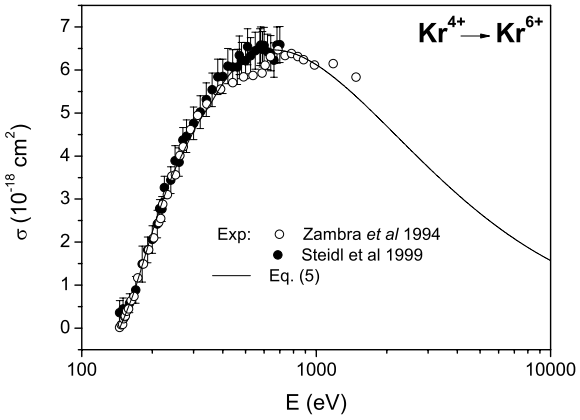

Figure 5. Electron-impact double-ionization cross section of Kr^{4+} ions. Open circles—experiment (Zambra *et al* 1994), full circles—experiment (Steidl *et al* 1999); solid line—equation (5) with parameters given in table 5.

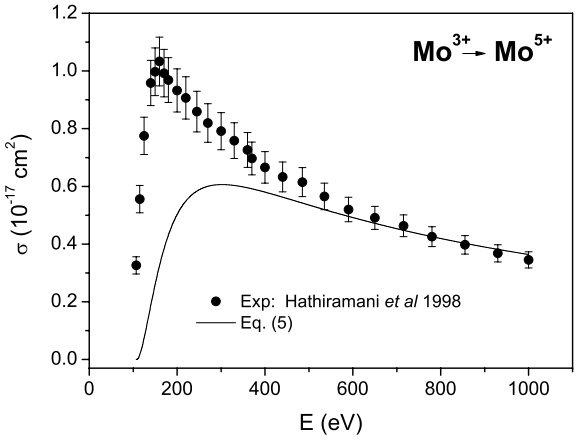

Figure 6. Electron-impact double-ionization cross section for Mo^{3+} ions. Full circles—experiment (Hathiramani *et al* 1998); solid line—equation (5) with parameters given in table 6.

Table 5. Threshold energies I_{th}, binding energies I_γ (both in eV), and fitting parameters of equation (5) for double ionization of Kr^{q+} ions: A_{dir} (in eV^3) and A_γ (in eV^2). The electron energy range, for which equation (5) can be applied, is given in the last column.

Ion configuration	I_{th} A_{dir}	I_{3d} A_{3d}	I_{3p} A_{3p}	I_{3s} A_{3s}	I_{2p} A_{2p}	I_{2s} A_{2s}	Energy range
$Kr^{1+}(4s^24p^5)$	61.309	103.77	228.09	307.52	1690.4	1942.5	$E < 5$ keV
	124.0	1.0	7.0	3.0	0.0	0.0	
$Kr^{2+}(4s^24p^4)$	89.45	118.0	242.0	321.76	1705.0	1957.1	$E < 5$ keV
	191.0	1.0	7.0	8.0	0.0	0.0	
$Kr^{3+}(4s^24p^3)$	117.2	133.76	258.0	337.27	1721.2	1973.2	$E < 5$ keV
	353.0	2.0	6.5	7.5	0.0	0.0	
$Kr^{4+}(4s^24p^2)$	143.2	150.36	274.78	354.27	1738.7	1990.6	$E < 7$ keV
	354.0	6.0	6.0	6.0	0.0	0.0	

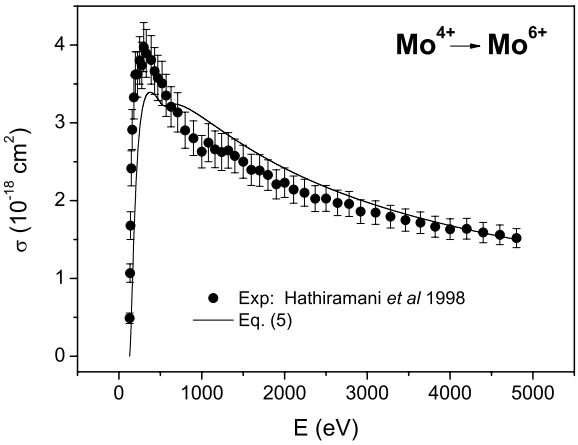

Figure 7. Electron-impact double-ionization cross section of Mo^{4+} ions. Full circles—experiment (Hathiramani *et al* 1998); solid line—equation (5) with parameters given in table 6.

Table 6. Threshold energies I_{th}, binding energies I_γ (in eV), and fitting parameters of equation (5) for double ionization of Mo^{q+} ions: A_{dir} (in eV^3) and A_γ (in eV^2). The electron energy range applicable to equation (5) is given in the last column.

Ion Configuration	I_{th} A_{dir}	I_{4p} A_{4p}	I_{4s} A_{4s}	I_{3d} A_{3d}	I_{3p} A_{3p}	I_{3s} A_{3s}	Energy range
$Mo^{1+}(4p^64d^5)$	43.31	47.48	79.86	240.89	410.12	522.02	$E < 2$ keV
	105.0	3.5	0.5	0.1	0.1	0.1	
$Mo^{2+}(4p^64d^4)$	73.56	59.21	92.72	254.10	423.37	535.38	$E < 2$ keV
	460.0	0.0	0.1	0.1	0.1	0.1	
$Mo^{3+}(4p^64d^3)$[a]	107.6	73.42	108.0	269.47	438.80	550.79	$E < 2$ keV
	350.0	0.0	0.5	0.1	0.1	0.1	
$Mo^{4+}(4p^64d^2)$[b]	129.2	88.38	130.0	286.46	455.66	567.93	$E < 10$ keV
	387.0	0.0	0.1	0.7	1.0	25.0	
$Mo^{5+}(4p^64d^1)$[c]	227.6	104.70	140.54	304.43	473.87	586.61	$E < 10$ keV
	700.0	0.0	0.0	7.0	8.0	7.0	
$Mo^{6+}(4s^24p^6)$[d]	290.0	124.21	158.81	325.67	494.76	606.69	$E < 10$ keV
	1500.0	0.0	0.0	1.5	2.0	1.0	

[a] For Mo^{3+} ion, the FAC gives the binding energy $I_{4s} = 107.32$ eV.
[b] For Mo^{4+} ion, the FAC gives the binding energy $I_{4s} = 123.34$ eV.
[c] For Mo^{5+}, 2s and 2p shells are also included with $I_{2s} = 2950$ eV, $A_{2s} = 70.0$ and $I_{2p} = 2600$ eV, $A_{2p} = 60.0$.
[d] For Mo^{6+}, 2s and 2p shells are also included with $I_{2s} = 2972$ eV, $A_{2s} = 30.0$ and $I_{2p} = 2621$ eV, $A_{2p} = 25.0$.

3.2. Fe^{q+} ions with $q = 1$–6 ($Z = 26$)

For Fe^{q+} ions with $q = 1$–6, the calculated threshold energies I_{th}, binding energies I_γ and fitting parameters of equation (5) for double ionization are given in table 2. Similar to Ti ions, equation (5) with parameters in table 2 gives quite a good description of the experimental cross sections (Stenke *et al* 1999). A typical example of fitting is given in figure 2 for double ionization of Fe^{6+} ions. For Fe^{2+} ions, the fitting parameters are absent because there are no experimental data.

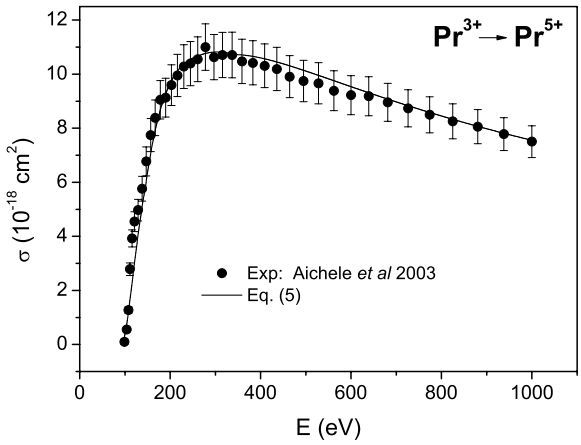

Figure 8. Electron-impact double-ionization cross section of Pr^{3+} ions. Full circles—experiment (Aichele *et al* 2003); solid line—equation (5) with parameters given in table 7.

Table 7. Threshold energies I_{th}, binding energies I_γ (in eV), and fitting parameters of equation (5) for double ionization of Pr^{q+} ions: A_{dir} (in eV^3) and A_γ (in eV^2). The electron energy range applicable to equation (5) is given in the last column.

Ion configuration	I_{th} A_{dir}	I_{5p} A_{5p}	I_{5s} A_{5s}	I_{4d} A_{4d}	I_{4p} A_{4p}	I_{4s} A_{4s}	I_{3d} A_{3d}	Energy range
$Pr^{1+}(5p^6 6s)$[a]	32.174	33.38	51.23	118.74	231.31	318.54	935.83	$E < 3$ keV
$Pr^{2+}(5s^2 5p^6)$[b]	75.0	3.5	1.3	1.0	3.0	1.0	1.0	
	60.604	35.69	61.00	126.23	238.82	326.06	943.43	$E < 3$ keV
	236.0	0.0	1.5	1.5	1.5	3.0	1.0	
$Pr^{3+}(5s^2 5p^5)$	96.51	49.25	73.96	144.76	254.47	347.02	964.29	$E < 3$ keV
	420.0	0.0	0.0	3.0	4.0	4.0	4.0	
$Pr^{4+}(5s^2 5p^4)$	137.09	64.30	90.44	165.88	281.22	370.35	987.87	$E < 2$ keV
	975.0	0.0	0.0	0.05	0.01	2.5	0.01	

[a] For Pr^{1+} ion, the FAC gives the binding energy $I_{5p} = 28.38$ eV.
[b] For Pr^{2+} ion, the FAC gives the binding energy $I_{5s} = 58.60$ eV.

3.3. Ni^{q+} ions with $q = 1$–6 ($Z = 28$)

Table 3 summarizes the calculated thresholds and the fitting parameters for Ni^{q+} ions with $q =$ 1–6. Equation (5) with parameters in table 3 gives quite a good description of the experimental cross sections. A typical example of fitting is shown in figure 3 for double ionization of Ni^{3+} ions (Stenke *et al* 1995).

3.4. Ga^{q+} ions with $q = 1$–6 ($Z = 31$)

The parameters for calculating double-ionization cross sections of Ga^{q+} ions with $q = 1$–6 are given in table 4. Equation (5) with parameters in table 4 provides a good description of all available experimental data. A typical example of fitting is given in figure 4 for double ionization of Ga^{2+} ions (Steidl *et al* 1999).

Table 8. Threshold energies I_{th}, binding energies I_γ (in eV), and fitting parameters of equation (5) for double ionization of Sm^{q+} ions: A_{dir} (in eV^3) and A_γ (in eV^2). The electron energy range, for which equation (5) can be applied, is given in the last column.

Ion Configuration	I_{th} A_{dir}	I_{5s} A_{5s}	I_{4d} A_{4d}	I_{4p} A_{4p}	I_{4s} A_{4s}	I_{3d} A_{3d}	Energy range
$Sm^{1+}(4f^6 6s)$	34.47	54.91	134.83	260.58	360.85	1085.6	$E < 4$ keV
	115.0	3.0	4.0	2.0	0.01	0.0	
$Sm^{2+}(5p^6 4f^6)$[a]	64.80	65.62	142.66	268.41	368.69	1093.4	$E < 4$ keV
	181.0	0.5	0.3	0.3	0.3	0.0	
$Sm^{3+}(5p^6 4f^5)$	101.55	78.67	162.19	289.27	390.03	1114.9	$E < 4$ keV
	446.0	0.0	1.0	2.5	3.0	0.0	
$Sm^{4+}(5p^6 4f^4)$	144.26	95.90	185.27	312.91	413.80	1139.6	$E < 4$ keV
	815.0	0.0	1.0	1.5	1.5	0.0	
$Sm^{5+}(5p^6 4f^3)$	185.21	114.13	209.68	337.28	439.63	1166.6	$E < 5$ keV
	1500.0	0.0	0.4	0.1	0.0	0.0	
$Sm^{6+}(5p^6 4f^2)$	219.51	133.26	234.15	363.21	467.26	1194.91	$E < 5$ keV
	2200.0	0.0	0.01	0.05	0.0	0.0	

[a] For Sm^{2+} ion, the FAC gives the binding energy $I_{5s} = 62.62$ eV.

3.5. Kr^{q+} ions with $q = 1$–4 ($Z = 36$)

Calculated binding energies and fitting parameters for Kr^{q+} ions with $q = 1$–4 are presented in table 5. Equation (5) with parameters in table 5 provides a good description of the experimental data. A typical example is given in figure 5 for double ionization of Kr^{4+} ions (Steidl *et al* 1999).

3.6. Mo^{q+} ions with $q = 1$–6 ($Z = 42$)

In table 6, calculated binding energies and fitting parameters for Mo^{q+} ions with $q = 1$–6 are given. Equation (5) with parameters in table 6 provides a good description of the experimental data except for Mo^{3+} ions for which the experimental data (Hathiramani *et al* 1998) have a very sharp maximum (figure 6). The steep rise in the cross sections for charge states $q = 1$–4 is most probably caused by the contribution of inner-shell mechanisms like excitation–double-autoionization (EDA) (Müller 1999). Another example of fitting the ionization cross sections is given in figure 7 for Mo^{4+} ions.

During the fitting procedure, it was found that, in some cases, the calculated binding energies of inner-shell electrons are very close to or even lower than the threshold energies, e.g., for Mo^{3+} ion, $I_{th} = 107.6$ eV while for the binding energy of 4s electrons the FAC gives the value of $I_{4s} = 107.32$ eV. In all such cases, we take into consideration all inner shells γ for which $|I_\gamma - I_{th}| \leqslant 5$ eV which accounts for the typical inaccuracies of the calculated binding and threshold energies; moreover, in the fits the energies I_γ are then set to a value slightly above the threshold energy. The corresponding energies are given in table 7.

3.7. Pr^{q+} ions with $q = 1$–4 ($Z = 59$)

Calculated data for Pr^{q+} ions with $q = 1$–4 are given in table 7. All available experimental data are described well using equation (5) with parameters in table 7. A typical example of fitting is given in figure 8 for double ionization of Pr^{3+} ions.

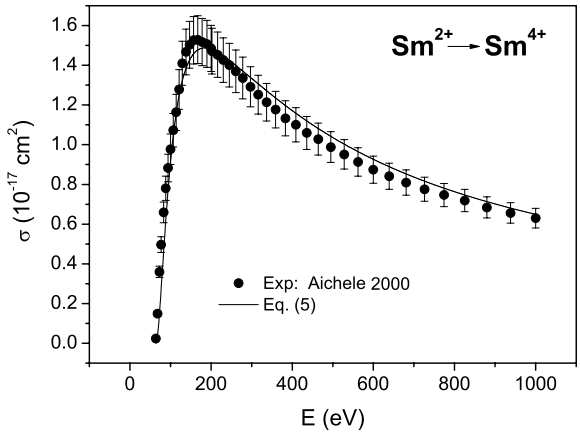

Figure 9. Electron-impact double-ionization cross section of Sm^{2+} ions. Full circles—experiment (Aichele 2000); solid line—equation (5) with parameters given in table 8.

Table 9. Threshold energies I_{th}, binding energies I_γ (in eV), and fitting parameters of equation (5) for double ionization of W^{q+} ions: A_{dir} (in eV^3) and A_γ (in eV^2). The electron energy range, for which equation (5) can be applied, is given in the last column.

Ion configuration	I_{th} A_{dir}	I_{5p} A_{5p}	I_{5s} A_{5s}	I_{4f} A_{4f}	I_{4d} A_{4d}	I_{4p} A_{4p}	I_{4s} A_{4s}	Energy range
$W^{1+}(5d^46s)$	39.83	50.87	95.17	44.25	260.2	442.4	614.3	$E < 2$ keV
	44.7	5.0	5.0	7.0	8.0	8.0	47.0	
$W^{2+}(5p^65d^4)$[a]	61.28	61.5	104.35	53.44	269.39	451.51	623.52	$E < 2$ keV
	350.0	1.0	1.0	0.0	10.0	7.0	0.1	
$W^{3+}(5p^65d^3)$	86.93	73.05	117.97	67.18	283.25	465.42	637.49	$E < 2$ keV
	450.0	0.0	0.5	0.0	0.5	0.5	0.0	
$W^{4+}(5p^65d^2)$	113.84	86.97	133.02	82.57	298.64	480.70	652.89	$E < 2.5$ eV
	450.0	0.0	1.5	0.0	1.5	0.5	0.0	
$W^{5+}(5p^65d^1)$	180.56	101.77	149.11	98.46	314.68	496.83	669.53	$E < 7$ keV
	780.0	0.0	0.0	0.0	3.0	8.0	4.0	
$W^{6+}(5s^25p^6)$[b]	257.73	119.94	166.21	116.93	333.13	515.25	687.25	$E < 7$ keV
	1250.0	0.0	0.0	0.0	5.5	1.5	4.5	

[a] For W^{2+}, the FAC gives $I_{5p} = 60.04$ eV.
[b] For W^{6+}, the contribution from the M shell should be included with $I_{3s} = 2911$ eV, $A_{3s} = 10.0$; $I_{3p} = 2369$ eV, $A_{3s} = 3.0$; $I_{3d} = 1894$ eV, $A_{3d} = 20.0$.

3.8. Sm^{q+} ions with $q = 1$–6 ($Z = 62$)

The calculated threshold energies I_{th}, binding energies I_γ and the best fitting parameters of equation (5) for double ionization of Sm^{q+} ions with $q = 1$–6 are given in table 8. With equation (5) these data provide a good description of all experimental results (Aichele 2000). Figure 9 shows a typical example of fitting in the case of Sm^{2+} ions. The steep rise from the onset of the ionization cross section of Sm^{6+}, shown in figure 10, appears to arise from 4d-EDA or 4d-IA processes (ionization with subsequent autoionization).

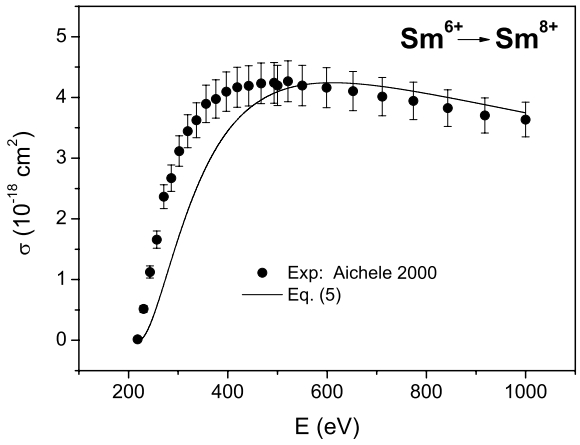

Figure 10. Electron-impact double-ionization cross section of Sm^{6+} ions. Full circles—experiment (Aichele 2000); solid line—equation (5) with parameters given in table 8.

Table 10. Threshold energies I_{th}, binding energies I_γ (in eV), and fitting parameters of equation (5) for double ionization of Pb^{q+} ions: A_{dir} (in eV^3) and A_γ (in eV^2). The electron energy range applicable to equation (5) is given in the last column.

Ion Configuration	I_{th} A_{dir}	I_{5p} A_{5p}	I_{5s} A_{5s}	I_{4f} A_{4f}	I_{4d} A_{4d}	I_{4p} A_{4p}	Energy range
$Pb^{1+}(6s^26p)$	46.97	99.45	168.13	149.29	428.77	661.23	$E < 3$ keV
	180.0	1.0	0.1	0.0	0.0	0.0	
$Pb^{2+}(5d^{10}6s^2)$	74.26	109.68	178.32	159.54	439.03	671.49	$E < 3$ keV
	450.0	0.1	0.1	0.0	0.0	0.0	
$Pb^{3+}(5d^{10}6s)$	111.12	121.56	190.03	171.82	451.31	683.73	$E < 3$ keV
	1000.0	0.7	0.7	0.0	0.0	0.0	
$Pb^{4+}(5p^65d^{10})$	151.05	134.53	203.04	184.96	464.47	696.88	$E < 3$ keV
	1500.0	0.0	0.5	0.3	0.7	0.0	
$Pb^{5+}(5p^65d^9)$	182.18	148.50	220.51	203.39	482.33	715.83	$E < 3$ keV
	1650.0	0.0	0.5	0.3	1.5	0.0	
$Pb^{6+}(5p^65d^8)$	218.81	164.62	238.74	223.63	502.65	736.09	$E < 3$ keV
	2403.0	0.0	0.5	0.3	2.0	0.0	
$Pb^{7+}(5p^65d^7)$	256.47	182.55	257.78	244.55	523.67	756.96	$E < 3$ keV
	1100.0	0.0	0.1	0.1	0.5	0.0	
$Pb^{8+}(5p^65d^6)$	295.75	201.71	277.38	266.10	545.31	778.82	E < 3.5 keV
	1120.0	0.0	0.0	0.0	0.5	0.1	
$Pb^{9+}(5p^65d^5)$	352.87	221.85	297.45	289.33	568.58	801.94	E < 3.5 keV
	1650.0	0.0	0.0	0.0	2.0	1.0	

3.9. W^{q+} ions with $q = 1$–6 ($Z = 74$)

The calculated threshold energies I_{th}, binding energies I_γ and the best fitting parameters of equation (5) for double ionization of W^{q+} ions with $q = 1$–6 are provided in table 9. Again, equation (5) with parameters in table 9 gives a quite good description of the experimental cross sections. A typical example of fitting is given in figure 11 for double ionization of W^{6+} ions.

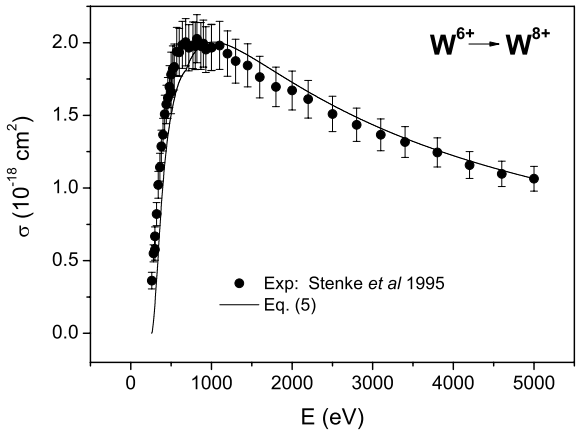

Figure 11. Electron-impact double-ionization cross section of W^{6+} ions. Full circles—experiment (Stenke *et al* 1995); solid line—equation (5) with parameters given in table 9.

Table 11. Threshold energies I_{th}, binding energies I_γ (in eV), and fitting parameters of equation (5) for double ionization of Bi^{q+} ions: A_{dir} (in eV^3) and A_γ (in eV^2). The electron energy range covered by equation (5) is given in the last column.

Ion configuration	I_{th} A_{dir}	I_{5p} A_{5p}	I_{5s} A_{5s}	I_{4f} A_{4f}	I_{4d} A_{4d}	I_{4p} A_{4p}	Energy range
$Bi^{1+}(6s^26p^2)$	41.78	109.74	182.72	169.63	457.21	696.55	$E < 3$ keV
	241.0	7.0	8.0	10.0	8.0	1.0	
$Bi^{2+}(6s^26p)$	70.86	120.04	193.05	180.16	467.70	707.03	$E < 3$ keV
	430.0	0.5	0.0	0.00	0.00	0.00	
$Bi^{3+}(5d^{10}6s^2)$	101.30	131.53	204.52	191.69	479.27	718.60	$E < 3$ keV
	850.0	0.5	0.0	0.0	0.0	0.0	
$Bi^{4+}(5d^{10}6s)$	144.32	144.40	217.21	205.12	492.71	731.99	$E < 3$ keV
	1400.0	0.5	1	0.1	1	0.0	
$Bi^{5+}(5p^65d^{10})$	181.11	158.33	231.22	219.31	506.92	746.20	$E < 3$ keV
	1550.0	0.0	0.5	1.0	5.0	0.0	
$Bi^{6+}(5p^65d^9)$	224.32	173.13	249.75	239.02	525.98	766.47	$E < 3$ keV
	1992.0	0.0	2.0	1.0	5.0	0.0	
$Bi^{7+}(5p^65d^8)$	263.06	190.14	269.01	260.54	547.57	788.00	$E < 3$ keV
	2000.0	0.0	2.0	1.0	0.0	0.0	
$Bi^{8+}(5p^65d^7)$	302.8	209.00	289.02	282.69	569.82	810.07	$E < 3$ keV
	1500.0	0.0	0.0	0.0	0.3	0.1	
$Bi^{9+}(5p^65d^6)$	343.76	229.06	309.57	305.39	592.59	833.08	E < 3.5 keV
	2000.0	0.0	0.0	0.0	0.1	0.0	
$Bi^{10+}(5p^65d^5)$	403.2	250.13	330.61	329.72	616.97	857.33	E < 3.5 keV
	3200.0	0.0	0.0	0.0	0.5	0.3	
$Bi^{11+}(5p^65d^4)$	436.29	273.81	354.84	354.68	641.99	882.39	E < 3.5 keV
	–	–	–	–	–	–	
$Bi^{12+}(5p^65d^3)$	494.4	295.60	377.84	379.65	667.09	907.65	E < 3.5 keV
	5500.0	0.0	0.0	0.0	0.5	0.0	

All the observed cross sections show no significant contribution from indirect, i.e., multi-step ionization processes.

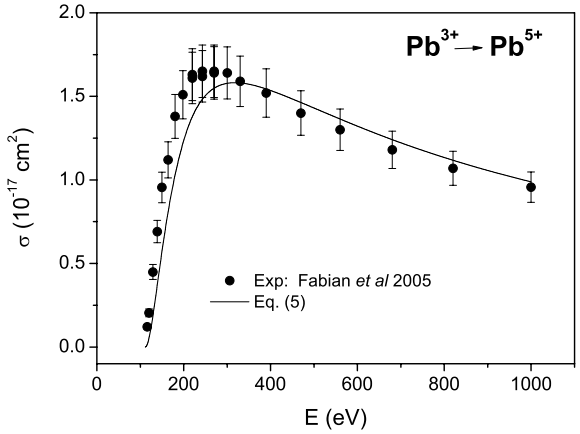

Figure 12. Electron-impact double-ionization cross section of Pb^{3+} ions. Full circles—experiment (Fabian *et al* 2005); solid line—equation (5) with parameters given in table 10.

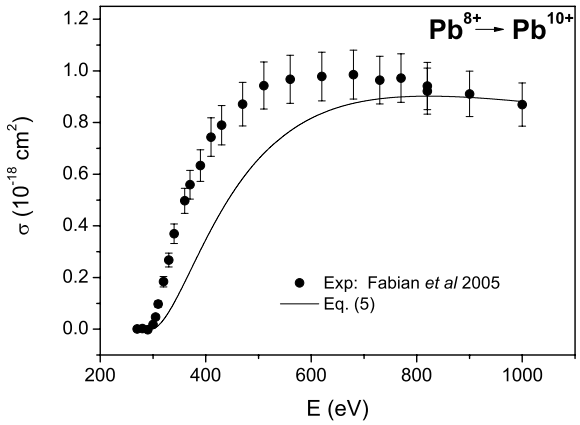

Figure 13. Electron-impact double-ionization cross section of Pb^{8+} ions. Full circles—experiment (Fabian *et al* 2005); solid line—equation (5) with parameters given in table 10.

3.10. Pb^{q+} ions with $q = 1$–9 ($Z = 82$)

Table 10 shows the calculated threshold energies $I_{\rm th}$, binding energies I_γ and the best fitting parameters of equation (5) for double ionization of Pb^{q+} ($q = 1$–9) ions. In figure 12 a typical result is shown for Pb^{3+} ions. Again, as observed with lighter ions, the contribution from inner-shell processes (in this case, the excitation–double-autoionization (EDA) from 5p-, 4f- and 5s-states leads to a discrepancy in the region from threshold to maximum). It can be observed that the contribution of indirect processes tends to increase as the ion charge gets higher (compared to W ions). In figure 13 experimental data of Pb^{8+} ions are compared to the calculated values of equation (5) using the values given in table 10. Here contributions of 4d-EDA are probably the reason for the excess double-ionization strength between the threshold and the maximum of the cross section. The calculated ionization threshold of the 5s shell ($I_{5s} = 277.9$ eV) is below the double-ionization threshold.

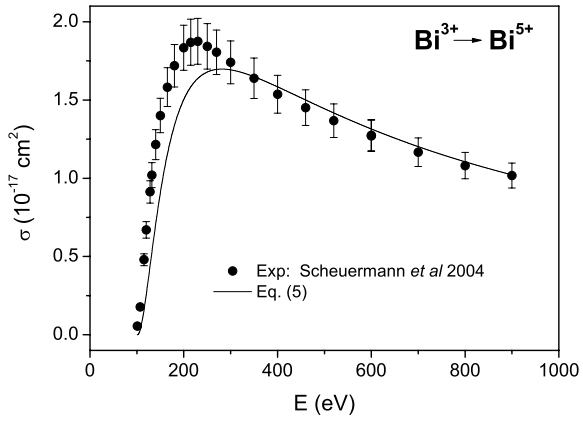

Figure 14. Electron-impact double-ionization cross section of Bi^{3+} ions. Full circles—experiment (Scheuermann *et al* 22); solid line—equation (5) with parameters given in table 11.

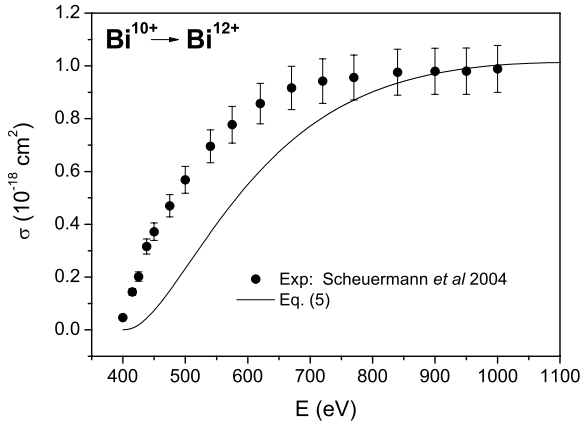

Figure 15. Electron-impact double-ionization cross section of Bi^{10+} ions. Full circles—experiment (Scheuermann *et al* 22); solid line—equation (5) with parameters given in table 11.

3.11. Bi^{q+} ions with $q = 1$–12 ($Z = 83$)

In table 11 all calculated threshold energies I_{th}, binding energies I_γ and the best fitting parameters of equation (5) for double ionization of Bi^{q+} ions with $q = 1$–10, 12 are given. Figure 14 shows the result for Bi^{3+} ions, figure 15 for Bi^{10+} ions. Like those observed in Pb^{8+} ions the contributions from indirect (inner-shell excitation) processes again lead to serious discrepancies between the predictor formula and the experiment in the region from threshold to the cross section maximum. For $q = 3$, 5p- and 4f-EDA and similarly for $q = 10$, 4d-EDA seem to contribute to the total cross sections. No contribution of ions in metastable states could be observed for double ionization of any charge state other than $q = 5$.

4. Conclusions

Double-ionization cross sections of multi-electron heavy ions by electron impact are described by a semi-empirical formula with six fitting parameters on the basis of experimental data and

quantum-mechanical formulae for simultaneous ionization of two target electrons and single ionization of inner-shell electrons contributing to double ionization. The required energy input parameters, i.e., the relevant threshold energies as well as the binding energies were taken from tables of recommended energies or were calculated by the FAC. The suggested formula was found to describe most of the available experimental data within an uncertainty of 30–40%. In some cases, however, discrepancies between experimental data and the proposed formula are noted, particularly in the low-energy region. This might be caused by higher order processes such as the excitation–double-autoionization (EDA) mechanism. These processes cannot be expected to follow simple predictor formulae and therefore are not explicitly included in the present formula. Nevertheless, the present semi-empirical formula, which is similar to the well-known Lotz formula for the calculation of direct *single* ionization, can be favourably used in a wide range of applications for modelling processes occurring in laboratory and astrophysical plasmas.

Acknowledgments

One of the authors (VPS) is grateful to DFG for financial support under grant no 436 RUS 17/77/05. This work was also supported in part by the Russian grant RFBR no 04-02-16309.

References

Aichele K 2000 *PhD Thesis* University Giessen (unpublished)
Aichele K, Arnold W, Bräuning H, Hathiramani D, Scheuermann F, Trassl R and Salzborn E 2003 *Nucl. Instrum. Methods Phys. Res.* B **205** 437
Becker R, Müller A, Achenbach C, Tinschert K and Salzborn E 1985 *Nucl. Instrum. Methods* B **9** 384
Beigman I L and Shevelko V P 1995 *Phys. Scr.* **51** 60
Brötz F 2000 *PhD Thesis* University Giessen (unpublished)
Currell F J (ed) 2003 *The Physics of Multiply and Highly Charged Ions* (Dordrecht: Kluwer)
Fabian B, Müller A, Bräuning H, Jacobi J, Scheuermann F A and Salzborn E 2005 *J. Phys. B: At. Mol. Opt. Phys.* **38** 2833
Grujić P 1982 *J. Phys. B: At. Mol. Phys.* **15** 1913
Gu M F 2003 *Astrophys. J.* **582** 1241
Lide D R (ed) 2000–2001 *Handbook of Chemistry and Physics* 81st edn (Boca-Raton, FL: CRC Press) (editor-in-chief)
Hartenfeller U, Aichele K, Hathiramani D, Schäfer V, Steidl M, Scheuermann F and Salzborn E 1998 *J. Phys. B: At. Mol. Opt. Phys.* **31** 3013
Hathiramani D, Hartenfeller U, Aichele K, Hofmann G, Steidl M, Stenke M, Völpel R and Salzborn E 1998 *J. Phys. B: At. Mol. Opt. Phys.* **31** 2101
Hofmann G, Müller A, Tinschert K and Salzborn E 1990 *Z. Phys.* D **16** 113
Jacobi J, Knopp H, Schippers S, Shi W and Müller A 2005 *J. Phys. B: At. Mol. Opt. Phys.* **38** 2015
Klar H and Schlecht W 1976 *J. Phys. B: At. Mol. Phys.* **9** 1699
Liehr M, Schlapp M, Trassl R, Hofmann G, Stenke M, Völpel R and Salzborn E 1993 *Nucl. Instrum. Methods* B **97** 697
Loch S D, Ludlow J A, Pindzola M S, Scheuermann F A, Kramer K, Fabian B, Huber K and Salzborn E 2005 *Phys. Rev.* A **72** 032713
Lotz W 1970 *J. Opt. Soc. Am.* **60** 206
Müller A 1999 *Atomic Physics with Heavy Ions* ed H F Beyer and V P Shevelko (Berlin: Springer) p 271
Müller A, Tinschert K, Achenbach C, Becker R and Salzborn E 1985 *J. Phys. B: At. Mol. Phys.* **18** 3011
Scheuermann F 2004 *PhD Thesis* University Giessen (unpublished)
Scheuermann F, Kramer K, Huber K and Salzborn E 2004 Abstract of contributed papers *12th Int. Conf. on the Physics of Highly Charged Ions (Vilnius, Lithuania, 6–11 Sept.)* ed Z Rudzikas pp 142
Shevelko V P, Tawara H, Scheuermann F, Fabian B, Müller A and Salzborn E 2005 *J. Phys. B: At. Mol. Opt. Phys.* **38** 525

Steidl M, Aichele K, Hartenfeller U, Hathiramani D, Scheuermann F, Westermann M and Salzborn E 1999 *Verh. Deutsch. Phys. Ges.* VI **34** 256

Stenke M, Aichele K, Hathiramani D, Hofmann G, Steidl M, Völpel R, Shevelko V P, Tawara H and Salzborn E 1995 *J. Phys. B: At. Mol. Opt. Phys.* **28** 4853

Stenke M, Hartenfeller U, Aichele K, Hathiramani D, Hofmann G, Steidl M and Salzborn E 1999 *J. Phys. B: At. Mol. Opt. Phys.* **32** 3641

Tinschert K, Müller A, Phaneuf R A, Hofmann G and Salzborn E 1989 *J. Phys. B: At. Mol. Opt. Phys.* **22** 1241

Wannier G H 1955 *Phys. Rev.* **100** 1180

Zambra M, Belić D, Defrance P and Yu D J 1994 *J. Phys. B: At. Mol. Opt. Phys.* **27** 2383