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Calculation of cross sections and rate coefficients for the electron impact multiple ionization of beryllium, boron, carbon, and oxygen atoms

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Abstract

The semiclassical Deutsch-Märk (DM) formalism was used in a series of calculations of cross sections for the formation of Be^{m+} (m = 1–4), B^{m+} (m = 1–5), C^{m+} (m = 1–6), and O^{m+} (m = 1–8) ions by electron impact on the neutral atoms. Our calculated cross sections are compared with available experimental and other theoretical data and systematic trends in the data are highlighted. We also calculated a set of ionization rate coefficients on the basis of the calculated cross sections and Maxwell-Boltzmann electron energy distributions. In many applications, particularly applications relating to fusion plasmas, rate coefficients are more desirable than ionization cross sections. (Int J Mass Spectrom 192 (1999) 1–8) © 1999 Elsevier Science B.V.

Keywords: Cross section; Rate coefficient; Electron ionization; Multiply charged ions

1. Introduction

The simultaneous removal of several atomic electrons following the impact of a single electron on a neutral atom A (single-step multiple ionization)

$$A + e \rightarrow A^{m+} + (m + 1)e \quad (1)$$

is a collision process of fundamental interest. Ionization processes play an important role in many applications such as discharges and plasmas, gas lasers, planetary, cometary, and stellar atmospheres, radiation chemistry, mass spectrometry, and chemical analysis [1,2]. Even though cross sections for the single-step multiple ionization of an atom are significantly smaller than cross sections for single ionization [1], multiple ionization processes are important in fusion plasmas [2] and in other "high-temperature" environments.

Calculations of multiple atomic ionization processes using rigorous quantum mechanical methods are difficult for all but the simplest targets [1,3] because of—among other things—the need to consider two or more continuum electrons and their mutual interaction in the exit channel. Experimental data for the formation of highly charged atomic ions

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are scarce for most atoms because (1) of the fact that the cross section declines rapidly with increasing charge state of the final ion and (2) it is quite difficult for most substances to produce atomic target beams of known number density. Therefore, modelers and practitioners rely heavily on semiempirical and semiclassical methods to determine multiple ionization cross sections for modeling purposes and for other applications [1–4]. The semiclassical Deutsch-Märk (DM) formalism [5] has been applied successfully to the calculation of cross sections for the formation of multiply charged ions A^{m+} via reaction (1) for several atoms, primarily for high-Z atoms with a nuclear charge of Z of 10 and above, and various stages of ionization [6–8]. Recently, the DM formalism has been used to calculate cross sections for the formation of Ne and Si ions up to the fully stripped ion, i.e. for Ne^{m+} (m = 1–10) and Si^{m+} (m = 1–14) ions [9]. The DM approach was found to have—besides a better agreement with the available data sets (see discussions in [6–9]) the following advantages over the two other reported semiempirical methods [10,11]: (1) its application requires fewer semiempirical parameters which, in addition, can more easily be related to physical quantities and (2) the energy-dependent function (derived from classical considerations) is the same for all stages of ionization.

In this article, we present the results of the application of the DM formalism to the calculation of cross sections for the formation of multiply charged ions for several low-Z atoms, Be [beryllium, Z = 4, electron configuration $(1s)^2(2s)^2$], B [boron, Z = 5, $(1s)^2(2s)^2(2p)$], C [carbon, Z = 6, $(1s)^2(2s)^2(2p)^2$] and O [oxygen, Z = 8, $(1s)^2(2s)^2(2p)^4$] up to the formation of the fully stripped ions Be^{4+}, B^{5+}, C^{6+}, and O^{8+}. Where possible, we compare the present results obtained with the DM formalism with experimental data. Moreover, we also compare the present results with cross sections derived from one of the two semiempirical methods [11] (the other method [10] shows very large deviations from the data for higher charge states, see Fig. 5 in [8]). The targets are of possible importance in tokamak fusion edge plasmas [2]. The presence of carbon is the result of the breakup of hydrocarbon contaminants and a by-product of the deposition of protective SiC wall coating material. The other three atoms (beryllium, boron, and oxygen) are also among the possible more abundant low-Z trace contaminants in fusion edge plasmas under specific experimental conditions. Although electron temperatures in these plasma regions are relatively low, well below 1 keV, the high energy part of the energy distributions may after all lead to the production of more highly charged species and thus influence the plasma properties.

2. Theoretical background

The application of the DM formalism, which was originally developed for the calculation of cross sections for the single ionization of an atom [5,12], to the calculation of cross sections for the formation of multiply charged ions by electron impact on the neutral atom has been described in detail in previous publications [6–9]. The cross section σ^{m+} for the formation of an ion A^{m+} via reaction (1), which in principle is a product of m independent terms each describing the removal of a single electron, can be simplified to an expression of the form

$$\sigma^{m+} = g^m \sum_k \pi(r_k)^2 \xi_k f_k(U) \tag{2}$$

where the summation extends over the various atomic subshells with k = 1 referring to the outermost subshell, k = 2 to the second outermost subshell, etc. In Eq. (2), $(r_k)^2$ is the mean square radius of the atomic subshell labeled by k, ξ_k is the number of electrons in that subshell, and g^m are weighting factors (see [6–9] for further details). The functions $f_k(U)$ describe the energy dependence of the ionization cross section [9,12] (see also [13])

$$f_k(U) = (1/U)[(U-1)/U+1)]^a \times \{b + c[1-(2U)^{-1}]\ln[2.7+(U-1)^{1/2}]\} \tag{3}$$

Here U refers to the reduced impact energy, $U = E/E_m$, where E is the energy of the incident electron and E_m is the ionization energy required for the simultaneous removal of m electrons from atom A,

Table 1
Parameters a and b from Eq. (4) for the atoms Be, B, C, and O

Atom, nuclear charge Z	Parameter a	Parameter b
Be, 4	2.6×10^4	5.15
B, 5	1.3×10^4	4.95
C, 6	6.2×10^3	4.75
O, 8	1.5×10^3	4.30

which is larger than the binding energy E_k of electrons in the subshell labeled by k. A detailed discussion of the functions $f_k(U)$, whose exact form differs for s, p, d, and f electrons, is given in [12]. For instance, the parameters a, b, and c have the values $a = 7/4$, $b = 1$, and $c = 1$ for s electrons, $a = 2$, $b = 1$, and $c = 1$ for p electrons, $a = 3/2$, $b = 3$, $c = 2/3$ for d electrons and $a = 3/2$, $b = 1$, and $c = 2/3$ for f electrons. The weighting factors g^m ($m = 1$–3) were taken from Deutsch et al. [6] and the weighting factors g^m ($m > 3$) were determined from a fitting procedure (for details, see the previous paper by Deutsch et al. [8]) according to an exponential function of the form

$$g^m(Z) = a(Z) \exp[-b(Z)\, m] \tag{4}$$

where Z is the nuclear charge and $a(Z)$ and $b(Z)$ are two empirically determined functions (see [8,9] for details). The values of the parameters a and b for the four atoms studied in this article have been summarized in Table 1.

3. Results and discussion

Experimental data for the formation of highly charged ions A^{m+} (for $m > 3$) produced by electron impact on the neutral atom are available for only a few atoms, primarily for the noble gases Ne, Ar, Kr, and Xe [14–21]. Experimental data for the formation of singly and multiply charged ions of Be, B, C, and O are very scarce. To the best of our knowledge, the only experimental data available in the literature are cross sections for the formation of singly charged C^+ and O^+ ions [22,24] and doubly charged O^{2+} ions [23,24]. Fig. 1 shows the experimental data for the reaction

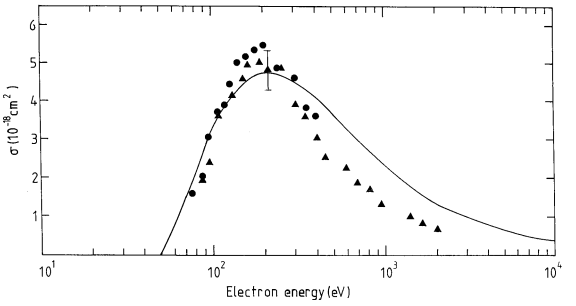

Fig. 1. Cross sections for the formation of O^{2+} ions by electron impact on O as a function of electron energy. The solid line represents the present calculation, the full dots are the experimental data of Ziegler et al. [23], and the full triangles are the experimental data of Thompson et al. [24].

$$O + e \rightarrow O^{2+} + 3e \tag{5}$$

from Ziegler et al. [23] and Thompson et al. [24], which are in good agreement with each other, in comparison with the present calculation. There is also good agreement between both experimental data sets and our calculation for energies below 100 eV. The peak in both measured cross section between 100 and

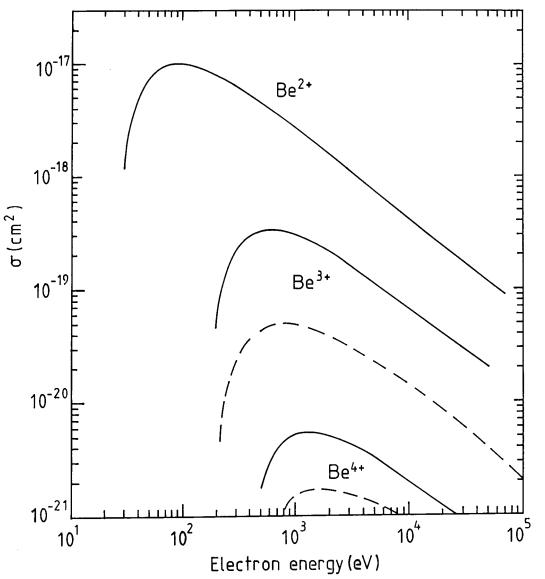

Fig. 2. Calculated cross sections for the formation of Be^{m+} ($m = 1$–4) ions as a function of electron energy using the DM formalism (solid line) and the method of Shevelko and Tawara [11] (dashed line).

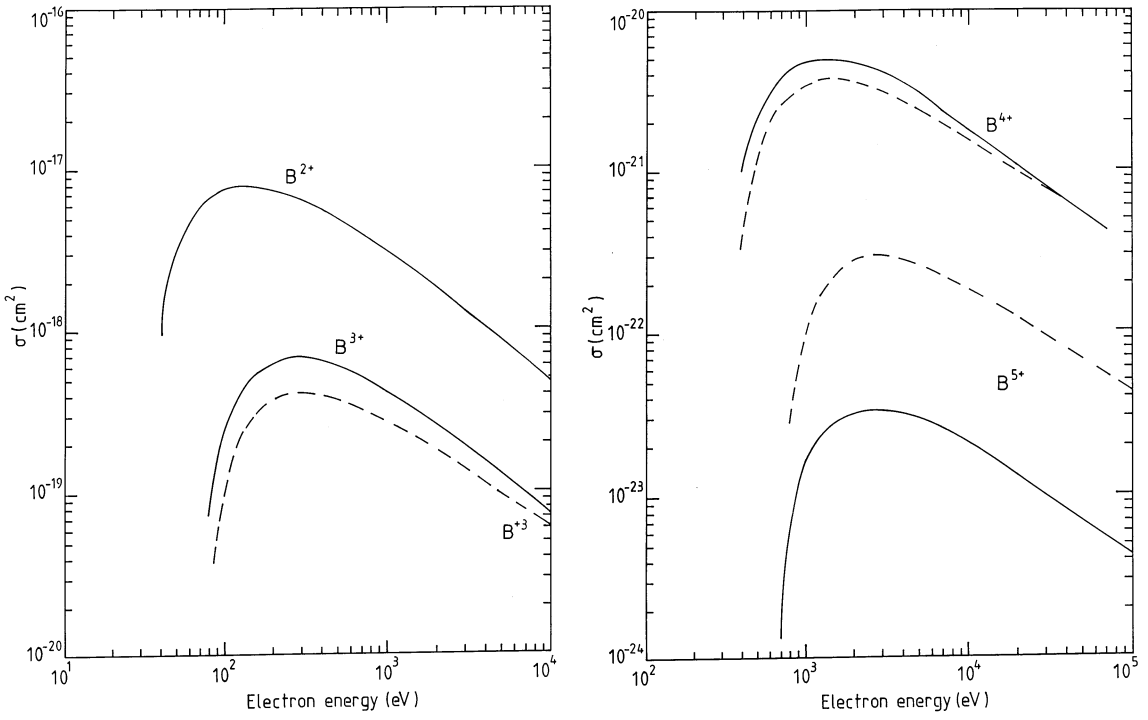

Fig. 3. (a) Calculated cross sections for the formation of B^{2+} and B^{3+} ions as a function of electron energy using the DM formalism (solid line) and the method of Shevelko and Tawara [11] (dashed line); (b) same as (a) for B^{4+} and B^{5+} ions.

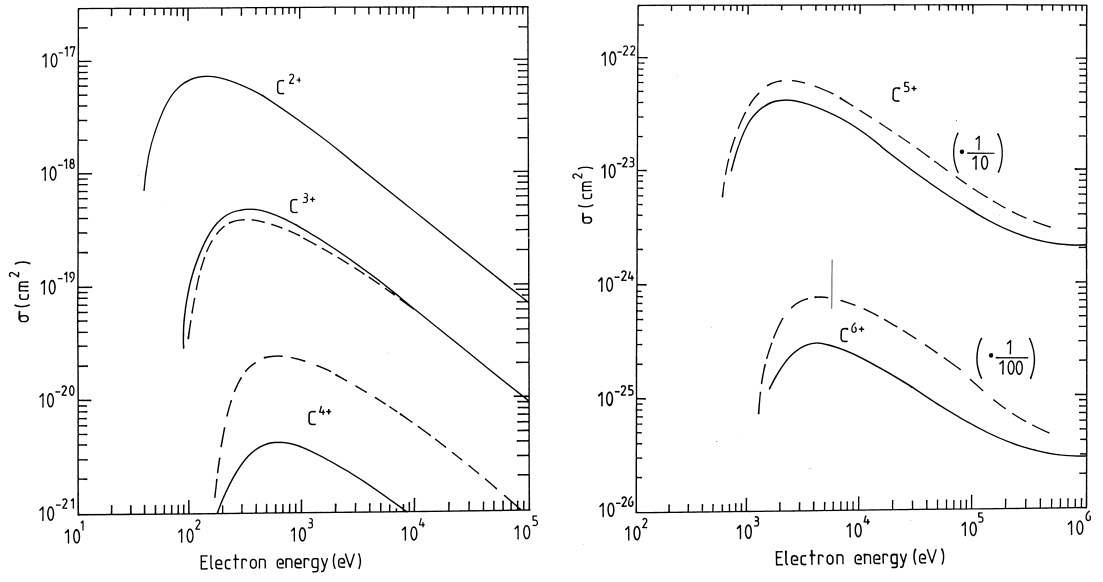

Fig. 4. (a) Calculated cross sections for the formation of C^{m+} ($m = 2$–4) ions as a function of electron energy using the DM formalism (solid line) and the method of Shevelko and Tawara [11] (dashed line); (b) same as (a) for C^{5+} and C^{6+} ions. For clarity of presentation, the cross sections calculated on the basis of the method of Shevelko and Tawara [11] have been multiplied by 0.1 (C^{5+}) and 0.01 (C^{6+}).

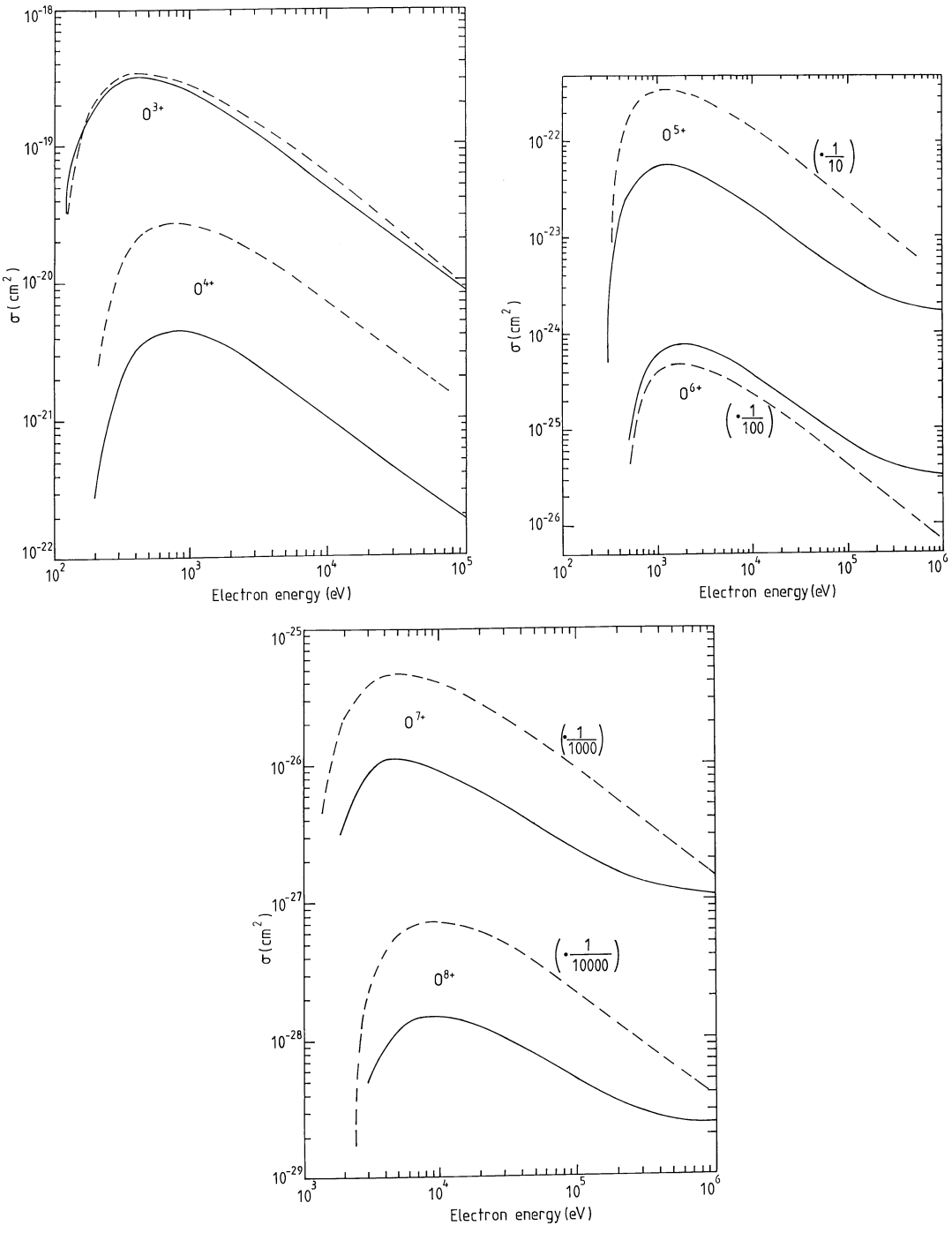

Fig. 5. (a) Calculated cross sections for the formation of O^{3+} and O^{4+} ions as a function of electron energy using the DM formalism (solid line) and the method of Shevelko and Tawara [11] (dashed line); (b) same as (a) for O^{5+} and O^{6+} ions. For clarity of presentation, the cross sections calculated on the basis of the method of Shevelko and Tawara [11] have been multiplied by 0.1 (O^{5+}) and 0.01 (O^{6+}); (c) same as (a) for O^{7+} and O^{8+} ions. For clarity of presentation, the cross sections calculated on the basis of the method of Shevelko and Tawara [11] have been multiplied by 0.001 (O^{7+}) and 0.0001 (O^{8+}).

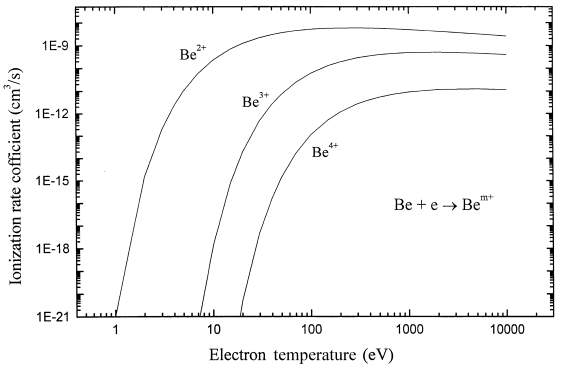

Fig. 6. Ionization rate coefficient as a function of the electron temperature for Be (see text for details).

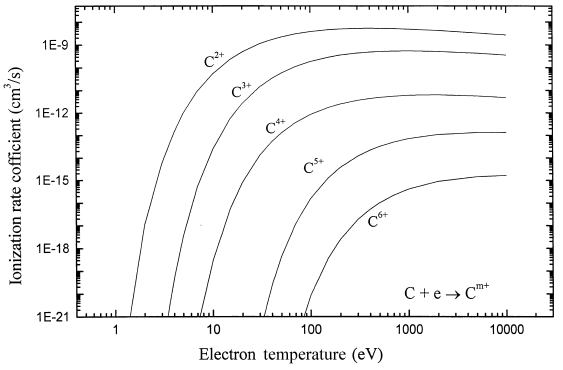

Fig. 8. Ionization rate coefficient as a function of the electron temperature for C (see text for details).

200 eV has been attributed to contributions from autoionization [23,24]. Since autoionization is a process that is not included in the present model, it is not surprising that our calculated cross sections lie systematically below the measured data in that energy regime. Nevertheless, taking into account the experimental error bars given there is a very good agreement of the peak cross sections between our calculated results and the experiments. At higher impact energies (above about 200 eV), our calculated cross sections decline less rapidly with increasing impact energy as both measured cross sections.

Figs. 2–5 summarize the results of the present calculations for the formation of Be^{m+} ions for m = 2–4, B^{m+} ions for m = 2–5, C^{m+} ions for m = 2–6, and O^{m+} ions for m = 3–8. For clarity of presentation, no curves are shown in Figs. 2–5 for the singly charged ions which have already been published and discussed earlier [12]. The O^{2+} cross section was omitted in Fig. 5, since it is shown separately in Fig. 1. In addition, each figure is presented in two or three parts, since the cross sections cover several orders of magnitude. It is apparent from Figs. 2–5 that the cross sections for the formation of multiply charged ions decrease rapidly with the degree of ionization m. This is to be expected, since the probability that a collision of an atom with a single electron results in the simultaneous, single-step removal of m electrons decreases rapidly with increasing m. Moreover, the position of the maximum of the cross section curves for the formation of the multiply charged ions shifts strongly

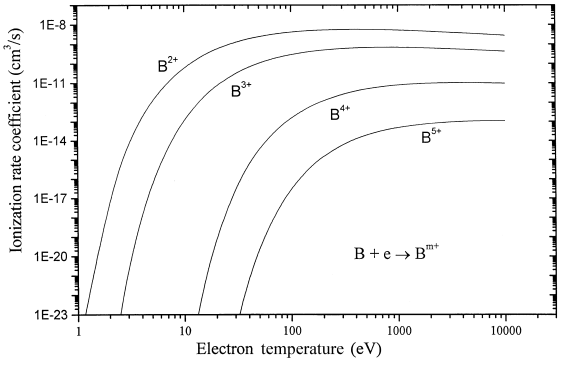

Fig. 7. Ionization rate coefficient as a function of the electron temperature for B (see text for details).

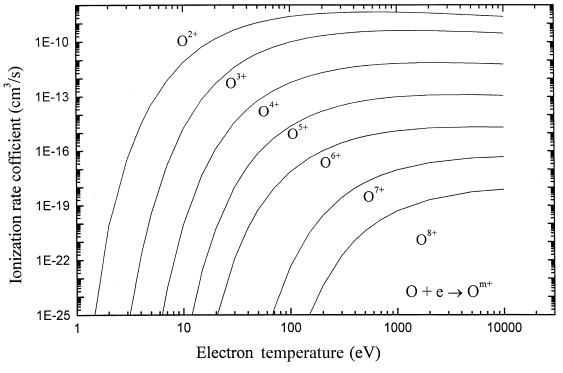

Fig. 9. Ionization rate coefficient as a function of the electron temperature for O (see text for details).

with the degree of ionization. This again is to be expected as the threshold for ionization increases strongly with the degree of ionization and thus the whole cross section curve is shifted to higher energies (see also discussion in [9]).

Also shown in Figs. 2–5 are the results of calculations using the method of Shevelko and Tawara [11] (referred to as ST cross sections hereafter) for $m > 2$. We note that the model of Shevelko and Tawara [11] is not applicable to the calculation of cross sections for the formation of singly and doubly charged ions. A comparison between the present calculations and ST cross sections leads to the following observations and reveals the following systematic trends.

(1) For $m = 3$, the ST cross sections are below our cross sections for Be and B by factors of 8 and 2, respectively, whereas the two calculated cross sections agree well for C and O. This indicates that the ST cross sections lie below our cross sections for small values of Z ($Z = 4,5$) and gradually approach our values as Z increases ($Z = 6,8$). This notion is supported by the fact that the ST cross sections were found to agree well with our cross sections and with experimental data for targets with $Z > 10$ as discussed in a previous publication [7].
(2) A similar trend is apparent for $m = 4$ except that the ST cross sections now exceed our cross sections for C and O by a factor of 6. A similar trend was also found in earlier data for $Z > 10$ [7], where the ST cross sections exceeded our cross sections significantly for $m = 4$ as well as for higher stages of ionization.
(3) For $m = 5$ and 6, the ST cross sections exceed our cross sections by a factor of 10 (for $m = 5$ in B, C, and O) and a factor of 100 (for $m = 6$ in C and O).
(4) In oxygen, the ST cross sections for $m = 7$ and 8 exceed our cross sections by, respectively, factors of 10^3 and 10^4.
(5) For all target atoms studied here, the ST cross section decreases much less rapidly with increasing stage of ionization m than our cross sections. On the other hand, previous results for atoms with $Z > 10$ [6–9,21] indicated that the trend exhibited by our calculated cross sections as a function of increasing m was in reasonable agreement with available experimental data.

Clearly, there are significant discrepancies between our calculated cross sections and the ST cross sections for the higher stages of ionization for all four atoms studied here. This should be viewed as a challenge to experimentalists to renew the emphasis on measuring cross sections for the multiple ionization of atoms as common and simple as carbon and oxygen as well as for other low-Z atoms.

From the perspective of applications, in particular applications to fusion plasmas, it is often more desirable to have ionization rate coefficients available rather than electron-impact ionization cross sections [2]. We derived a set of ionization rate coefficients for Be, B, C, and O as a function of electron temperature on the basis of our calculated ionization cross sections and Maxwell-Boltzmann energy distributions for the electrons. The results are shown in Figs. 6–9. We note that the electron energy in these figures is expressed in terms of an electron temperature (1 eV corresponds to 11 604 K).

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