

A detailed comparison of calculated and measured electron-impact ionization cross sections of atoms using the Deutsch–Märk (DM) formalism

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Abstract

We present a comprehensive comparison between the results of the recently revised DM formula, which now exhibits the quantum mechanically correct high-energy behavior, and measured atomic ionization cross sections for selected atoms covering a wide range of targets of different electronic structure as well as elements along columns of the periodic table. Specifically, we selected the following elements (listed in order of increasing atomic number Z): Na, Si, S, Cl, K, Ca, Fe, Ga, Br, In, Cs, Hg, Bi, and U. The main objective of this study is to compare the results of the revised DM formula for these elements in both the low- and high-energy regime with available experimental data and to extend the formalism to targets with higher atomic number Z , where contributions to the ionization cross section from f -electrons have to be considered. In cases, where several, sometimes conflicting experimental data sets have been reported, an attempt is made to provide guidance as to the reliability of various measured cross sections. In addition, we also calculated ionization cross sections for the technically important species Cr, Mn, and W, for which no experimental data are available.

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1. Introduction

The ionization of an atom by electron impact is one of the most basic electron-driven processes both in terms of its importance in fundamental collision physics and from an application-motivated viewpoint in environments such as: discharges and plasmas; planetary and cometary atmospheres; mass spectrometry; and chemical analysis. The removal of an electron from a neutral atom as the consequence of the impact of another electron is the simplest electron collision process resulting in the formation of three charged particles: the incident and ejected negatively charged electron (which are indistinguishable) and a positive ion. The

complexity of the interactions of the three charged particles in the exit channel of this collision process renders a rigorous quantum mechanical description impossible even for the simplest target, atomic hydrogen. Approximations have been incorporated quite successfully into quantum mechanical descriptions of electron-impact ionization processes of H [1] and quasi one- and two-electron atoms such as the alkalis [2] and comparatively low collision energies (up to a few times the ionization energy). At higher impact energies (above approximately 20 times the ionization energy), the Born–Bethe theory provides an acceptable description of atomic electron-impact ionization cross sections. Semi-rigorous methods were found to be useful to provide convenient and easy-to-use formulas to calculate atomic ionization cross sections for the entire range of impact energies from threshold to the high-energy regime. Among those are the Deutsch–Märk (DM)

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formalism [3,4] and the Binary-Encounter-Dipole (BED) method of Kim and Rudd [5]. DM calculations have been carried out for all atoms for which experimentally determined ionization cross sections are available (H, He, Li, C, N, O, F, Ne, Na, Mg, Al, Si, P, S, Cl, Ar, K, Ca, Ti, V, Fe, Ni, Cu, Ga, Ge, As, Se, Br, Kr, Rb, Sr, Ag, In, Sn, Sb, Te, I, Xe, Cs, Ba, Yb, Hg, Pb, Bi, and U) [4]. As noted in previous publications [3,6], the DM formula was originally developed for use in plasma modelling applications and thus was aimed at a description of atomic ionization cross sections in the energy range from threshold to at most 100 eV. Its energy dependence was described in terms of a Gryzinski-type expression, which unlike, e.g. the BED formula, does not yield cross section shapes that show the quantum mechanically “correct” $\ln(E)/E$ behavior in the regime of high impact energies E [5,6].

Recently, a modified DM formula was introduced [6], where the Gryzinsky-type energy dependence was replaced by a “scaled” $\ln(E)/E$ energy dependence. The revised DM formula reproduces the $\ln(E)/E$ dependence in the limit of high impact energies and slightly improves the previously reported DM cross sections at lower impact energies. The cross sections obtained with the revised DM formalism when compared to the DM cross sections reported in Ref. [4], show a very minor difference in the position and absolute value of the cross section maximum and a cross section shape above about 200 eV that is in better agreement with experimentally determined cross section functions.

In this paper, we present a comprehensive comparison between the results of the revised DM formula and measured atomic ionization cross sections for selected elements covering a wide range of targets of different electronic structure as well as elements along columns of the period table (alkalis, halogens). Specifically, we selected the following targets (listed in order of increasing atomic number Z): Na ($Z=11$), Si ($Z=14$), S ($Z=16$), Cl ($Z=17$), K ($Z=19$), Ca ($Z=20$), Fe ($Z=26$), Ga ($Z=31$), Br ($Z=35$), In ($Z=49$), Cs ($Z=55$), Hg ($Z=80$), Bi ($Z=83$), and U ($Z=92$). The objective is: (1) to test the validity of the revised DM formula for these elements in both the low- and high-energy regime; (2) to extend the method to targets with higher atomic numbers Z (where f-electrons become important); and (3) to provide guidance as to the reliability of measured cross sections in cases where several experimental investigations reported drastically different absolute cross sections. Furthermore, we also calculated ionization cross sections for the elements Cr, Mn, and W, which are technically important, e.g. as impurities in fusion plasma devices and for which no experimental data are available.

2. Theoretical background

The physical foundation of the original DM method was first given by Deutsch and Märk in Ref. [3], and a more recent in-depth discussion can be found in the review by Deutsch et al. [7]. In Ref. [6], the revised DM formalism was

introduced, which expresses the total single electron-impact ionization cross section σ of an atom as

$$\sigma(u) = \sum_{n,l} g_{nl} \pi r_{nl}^2 \xi_{nl} b_{nl}^{(q)}(u) \left[\frac{\ln(c_{nl}u)}{u} \right] \quad (1)$$

where r_{nl} is the radius of maximum radial density of the atomic sub-shell characterized by quantum numbers n and l (as listed in column 1 in the tables of Desclaux [8]); and ξ_{nl} the number of electrons in that sub-shell. The sum extends over all atomic sub-shells labelled by n and l . The g_{nl} are the weighting factors, which were originally determined from a fitting procedure [3,4] using reliable experimental cross section data for the rare gases and uranium. The quantity u refers to the “reduced” energy $u = E/E_{nl}$, where E denotes the incident energy of the electrons and E_{nl} is the ionization energy in the (n, l) sub-shell. The energy-dependent function $b_{nl}^{(q)}(u)$ has the form

$$b_{nl}^{(q)}(u) = \frac{A_1 - A_2}{[1 + (u/A_3)^p] + A_2} \quad (2)$$

and the four quantities A_1 , A_2 , A_3 , and p are constants that were determined from reliable measured cross sections for the various values of n and l [6]. The superscript “ q ” refers to the number of electrons in the (n, l) sub-shell. The constant c_{nl} in Eq. (1) was found to be close to one except for d-electrons. For the convenience of the reader, we summarize the pertinent parameters needed to use Eq. (2) in Table 1. We note that the constants in the function $b_{nl}^{(q)}(u)$ for the ns and np ($n \geq 3$) electrons have been slightly modified compared to those given in the original publication [6] and constants for the f-electrons in the case of $n > 3$ have now also been determined (last row of Table 1). The slight revision of the constants for the ns and np ($n \geq 3$) electrons was necessary in an effort to avoid a negative value of the parameter A_2 for the ns electrons as reported originally [6], which has the potential to lead to an unphysical result under rare circumstances. The constraint placed on the fitting routine by this condition also

Table 1

Summary of parameters A_1 , A_2 , A_3 and p that characterize the energy-dependent function $b_{nl}^{(q)}(u)$ of Eq. (2)

Function $b_{nl}^{(q)}(u)$	A_1	A_2	A_3	p	c_{nl}
$b_{ns}^{(1)}(u), n = 1, 2, 3, \dots$	0.31	0.87	2.32	1.95	1.00
$b_{ns}^{(2)}(u), n = 1, 2$	0.23	0.86	3.67	2.08	1.01
$b_{np}^{(q)}(u), n=2; q=1, 2$	0.33	0.88	2.38	1.98	0.99
$b_{np}^{(q)}(u), n=2; q=3-6$	-0.15	1.17	4.05	1.31	1.01
$b_{ns}^{(2)}(u), n = 3, 4, 5, 6, \dots$	1.04	0.03	21.00	0.34	0.96
$b_{np}^{(q)}(u), n = 3, 4, 5, 6, \dots; q=1-6$	0.59	1.01	8.42	2.12	0.95
$b_{nd}^{(q)}(u), n = 3, 4, 5, 6; q=1-10$	0.44	1.55	4.38	1.87	1.42
$b_{nf}^{(q)}(u), n = 4, 5, 6; q=1, 2, 3, \dots$	-3.43	0.92	0.08	0.65	1.00

These parameters were determined from reliable measured cross sections for the single ionization of the atoms H, He, C, Ne, Mg, Al, and Ag. Also included are the values of the constant c_{nl} .

resulted in slightly changed constants for the np electrons. The change in the cross section curves reported in Ref. [6] resulting from the revised constants is less than the line width of the cross section curves shown in various figures in Ref. [6].

3. Results and discussion

The revised DM formula was introduced in our previous paper [6] by using the well-known ionization cross sections of the elements H, He, C, Ne, Mg, Al, and Ag to determine the value of c_{nl} in Eq. (1) and the constants in Eq. (2) for the various quantum numbers n and l and occupation number q . Subsequently, the DM cross section of Eq. (1) was tested for the targets O, F, P, Ar, Ge, Kr, and Xe by comparing the DM cross sections with experimentally determined reliable cross sections and excellent agreement between calculated and measured cross sections was found in all cases. Here, we extend the comparison to a much wider range of other targets of different electronic structure as well as to elements along columns of the periodic table of elements such as the alkalis and the halogens. While the choice of 17 elements studied in the present work, does not include all species for which experimental data are available, it constitutes a representative cross section of elements for which (i) three or more independent experimental data sets are available, (ii) two independent data sets have been reported, (iii) a single experiment has been carried out, and (iv) no experimental data have been reported till now. A critical analysis of experimental data is attempted in cases where more than a single experimental result has been reported. We note that there are no rigorous, statistically supported quantitative criteria describing the level of agreement between calculated and measured cross sections. Therefore, we follow the established tradition of using the position and absolute value of the cross section maximum, the overall energy dependence of the cross section function, and (to the extent applicable) any unusual structures in the cross section shape in conjunction with the uncertainty margins of carefully assessed experimental data to characterize the level of agreement between measured and calculated cross sections.

3.1. The alkalis Na, K, and Cs

Na, K, and Cs are quasi one-electron atoms whose ionization cross sections have been measured by several groups since the 1960s [9–18]. In the case of Na (Fig. 1a), the data of Zapesochnyi and Aleksakhin [11] are significantly higher than all other measured data and the calculated DM cross section. The DM cross section agrees best in terms of the absolute cross section value with the data of Johnston and Burrow [13], although it reaches its maximum value at a slightly higher impact energy. There is also excellent agreement between the DM cross section and the data of McFarland and Kinney [10], whose measurements were limited to higher impact energies above 50 eV. The experimental data of Tan et al. [14] and Fujii and Srivastava [12] are slightly lower than the DM cross section and the cross section curve of Ref. [12],

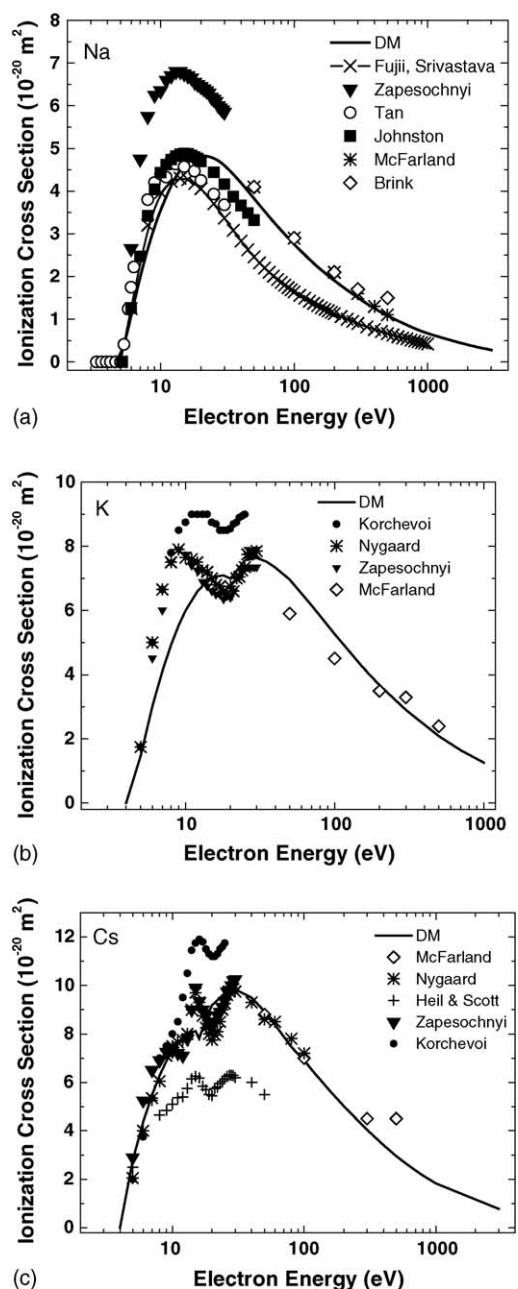


Fig. 1. (a) Calculated DM ionization cross section as a function of electron energy for Na (solid line) in comparison with the experimental data of Ref. [12] (crosses), Ref. [11] (inverted triangles), Ref. [14] (open circles), Ref. [13] (filled squares), Ref. [10] (stars), and Ref. [9] (open diamonds); (b) same as (a) for K with the experimental data from Ref. [15] (filled circles), Ref. [16,17] (stars), Ref. [11] (inverted triangles), and Ref. [10] (open diamonds); (c) same as (a) for Cs with the experimental data from Ref. [10] (open diamonds), Ref. [16,17] (stars), Ref. [18] (Heil and Scott), Ref. [11] (inverted triangles), and Ref. [15] (filled circles).

which is the only experimental data that covers the energy from threshold to 1000 eV, shows a high-energy dependence that is different from that of the DM cross section. We note that the calculated Na ionization cross section data of Bray [2] have not been included in Fig. 1a, since these calculations are limited to low energies up to 30 eV. In that energy regime,

the calculated data of Bray [2] lie below all experimental data.

The K ionization cross sections of Nygaard [16,17] and Zapesochnyi and Aleksakhin [11], which are in excellent agreement with one another, show a pronounced double maximum structure with a first maximum around 9 eV and a second maximum around 30 eV as shown in Fig. 1b. It is tempting to attribute the first maximum around 9 eV to autoionization. However, Nygaard [17] lists the energies of the two lowest autoionizing state in K as 18.72 eV and 18.98 eV, respectively. The DM cross section reproduces the second maximum very well, but underestimates the first maximum. This is not surprising, if the first maximum can be attributed to indirect ionization processes as the DM formalism describes only the direct ionization. In the high-energy regime, the DM cross section agrees well with the data of McFarland and Kinney [10], whose data cover the energy range from 50 to 500 eV. The experimental data of Korchevoi and Przhonski [15] are significantly above all other cross sections. The situation is similar in the case of Cs (Fig. 1c), where the DM cross section agrees well with the second maximum in the data of Nygaard [16,17] and Zapesochnyi and Aleksakhin [11] and the high-energy data of McFarland and Kinney [10]. As in K, the DM calculation does not reproduce the first maximum well. The data of Korchevoi and Przhonski [15] are significantly above these cross sections, whereas the cross section of Heil and Scott [18] is significantly lower than the DM cross section and the data of Refs. [11], [14], and [17]. In contrast to K, the first maximum in the Cs ionization cross section coincides with the energies of the two lowest autoionizing states [17].

3.2. The halogens Cl and Br

The only measured cross sections for the halogen atoms are those of Freund and co-workers [19]. Fig. 2 shows the DM cross sections for Cl and Br in comparison with the measured data. The agreement is very good in both the cases. The slight shift in energy between the calculated and measured cross sections in the low-energy range up to about 40 eV is exaggerated by the choice of a logarithmic energy scale. The situation is similar for the halogen atom F, which was discussed in Ref. [6] and for iodine (I), which is not shown here.

3.3. The elements Si, S, and Ca

Freund et al. [20] measured ionization cross sections for Si and S using the fast-beam method. In the case of Si, no other experimental data have been reported to date. The DM cross section shows excellent agreement with the measured data as shown in Fig. 3a, except perhaps for a minor discrepancy in the region of the cross section maximum (which is, however, well within the stated experimental uncertainty). In the case of S (Fig. 3b), the measured ionization cross section of Ziegler et al. [21] is about 25% smaller than that of Ref. [20], but shows a similar cross section shape with an unusually flat

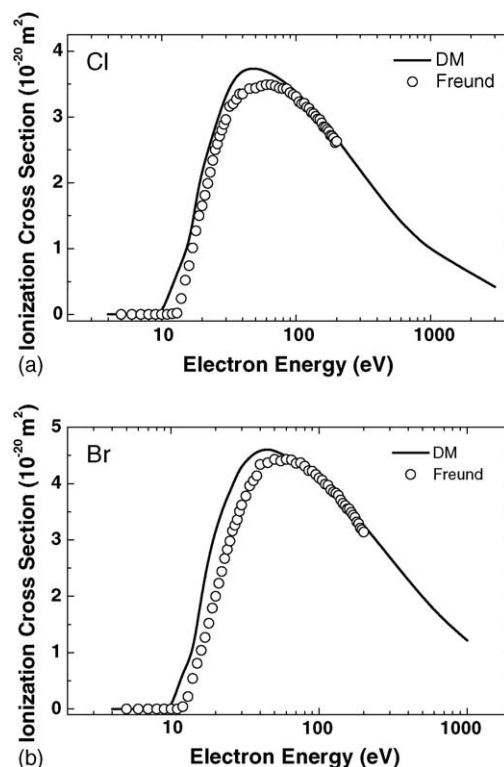


Fig. 2. (a) Calculated DM ionization cross section as a function of electron energy for Cl (solid line) in comparison with the experimental data of Ref. [19] (open circles); (b) same as (a) for Br.

maximum ranging from 30 to almost 100 eV. The DM cross section agrees fairly well with the data of Freund et al. [20] up to about 20 eV and above 100 eV, but does not reproduce the observed unusual cross section maximum structure.

In contrast to Si and S, many experimental studies have measured the Ca ionization cross section [22–26] and McGuire [27] also reported a calculated Ca ionization cross section. As shown in Fig. 4, there is very good agreement between the various experimental data sets except for the data of McFarland [22], which exceed most other measured data by a factor of 2. Both calculated cross section curves do not describe the experimental data very well. Neither curve reproduces the structure in the measured data around 20 eV. Both calculated cross sections peak at a lower impact energy compared to the measured data and exceed the measured maximum cross section value by respectively 10% (present calculation) and 15% (Ref. [27]). Overall, the DM cross section provides a somewhat better description of the measured data at higher impact energies above about 100 eV.

3.4. The elements Fe, Ga and In

Ionization cross sections for the atoms Fe, Ga, and In were measured by Freund and co-workers [20,28]. The Fe ionization cross section was also measured by Shah et al. [29] and their results are about 25% below the data of Ref. [20]. It should be noted that this discrepancy is still within the

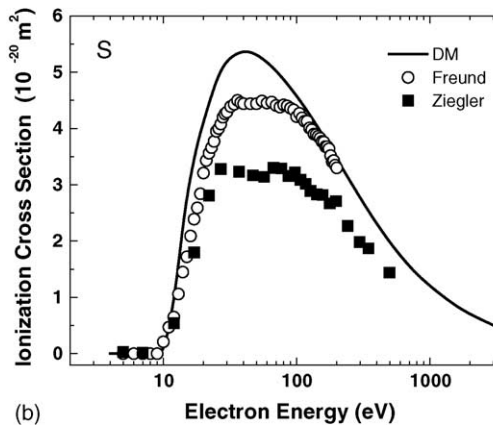
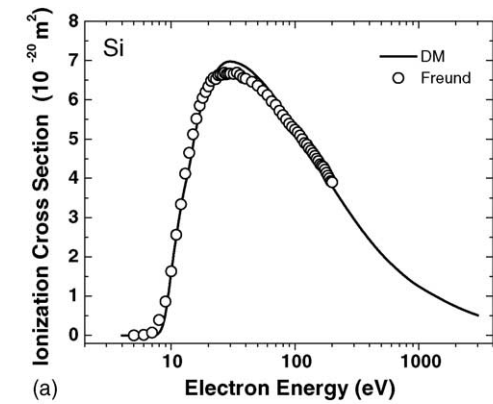


Fig. 3. Calculated DM ionization cross section as a function of electron energy for Si (solid line) in comparison with the experimental data of Ref. [20] (open circles); (b) same as (a) for S.

combined uncertainty of the two experiments. As shown in Fig. 5a, the DM cross section is closer to the data of Shah et al. [29] and, in fact, reproduces their data quite well above about 100 eV. Measured ionization cross sections for Ga were also reported by Patton et al. [30] and Vainshtein et al. [31].

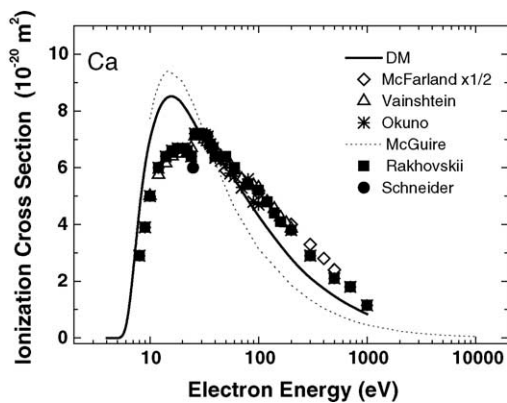


Fig. 4. Calculated DM ionization cross section as a function of electron energy for Ca (solid line) in comparison with the calculated cross section of Ref. [27] (dashed line) and the experimental data of Ref. [22] (open diamonds), Ref. [25] (open triangles), Ref. [24] (stars), Ref. [23] (filled squares), and Ref. [25] (filled circles).

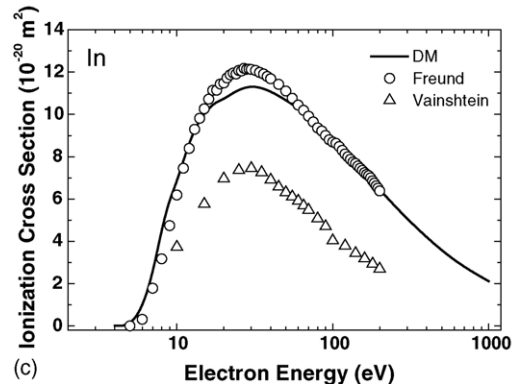
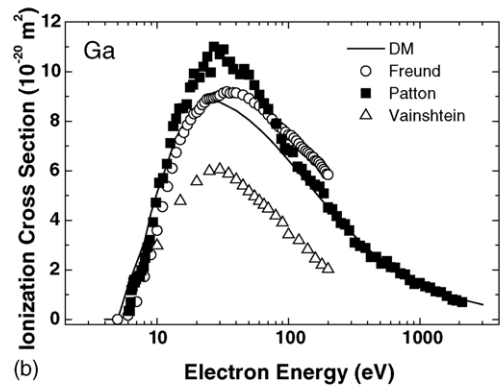
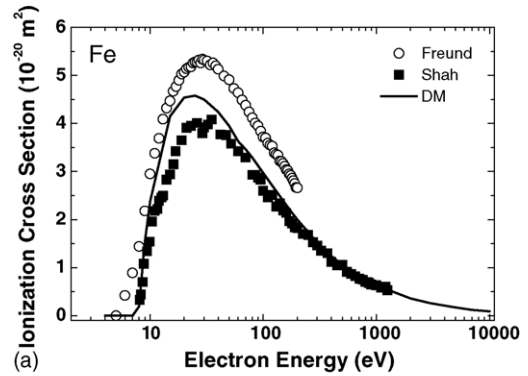


Fig. 5. (a) Calculated DM ionization cross section as a function of electron energy for Fe (solid line) in comparison with the experimental data of Ref. [20] (open circles) and Ref. [29] (filled squares); (b) same as (a) for Ga with the experimental data of Ref. [28] (open circles), Ref. [30] (filled squares), and Ref. [31] (open triangles); (c) same as (a) for In with the experimental data of Ref. [28] (open circles) and Ref. [31] (open triangles).

While the data from Refs. [28] and [30] are in reasonably good agreement (within their combined stated uncertainty), the cross sections of Ref. [31] are lower by almost a factor of 2 (Fig. 5b). The DM cross section agrees reasonably well with the data of Ref. [28] and is in fair agreement – except for the region near the cross section maximum – with data of Patton et al. [30]. Fig. 5c shows the two measured In ionization cross sections of Refs. [28] and [31]. The DM cross section is in good agreement with data of Ref. [28] and both cross section curves are about 60% higher than the data of Vainshtein et al. [31].

3.5. The elements Hg, Bi, and U

To the best of our knowledge, there has only been one experimental determination of the Hg ionization cross section, the paper by Bleakney [32], which dates back to 1930. This is quite surprising given the importance of Hg in the lighting industry. Fig. 6a shows the data of Ref. [32] in comparison with the DM cross section. The agreement in both absolute value and cross section shape is rather poor, which underscores the need for a renewed measurement of this ionization

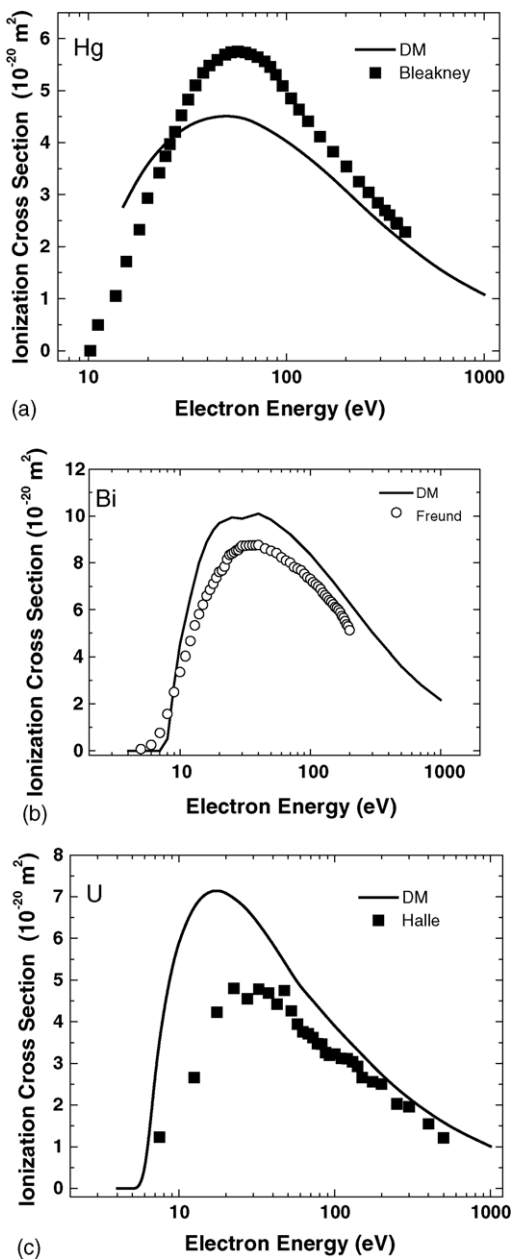


Fig. 6. (a) Calculated DM ionization cross section as a function of electron energy for Hg (solid line) in comparison with the experimental data of Ref. [32] (filled squares); (b) same as (a) for Bi with the experimental data of Ref. [28]; (c) same as (a) for U with the experimental data of Ref. [33]).

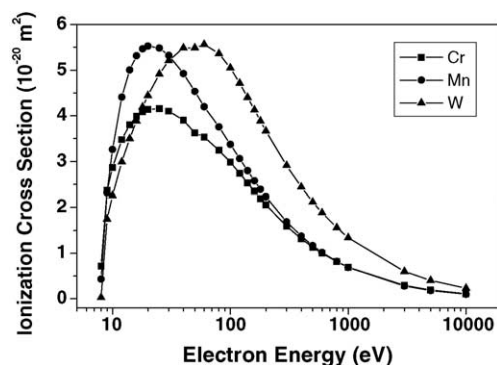


Fig. 7. Calculated DM ionization cross section as a function of electron energy for Cr (filled squares), Mn (filled circles), and W (filled triangles). No experimental data are available for comparison.

cross section. Likewise, there has only been one experimental determination of the Bi ionization cross section [20]. However, the data by Freund et al. [20] are generally considered among the most reliable experimentally determined ionization cross sections. A comparison of their data with the DM cross section (Fig. 6b) shows a reasonably good agreement, even though the experimental data lie about 15% below the calculation, but this is just within the stated experimental uncertainty. In the case of U, the only available experimental data of Halle et al. [33] are in poor agreement with the DM cross section (Fig. 6c). The calculated cross section lies above the measured data at all impact energies and exceeds the measured maximum cross section value, which is reached at a lower electron energy compared to the measured data, by more than 30%.

3.6. Calculations for Cr, Mn, and W

The atoms Cr, Mn, and W are of technical importance, e.g. as impurities in fusion plasma devices. To the best of our knowledge, there are no experimental ionization cross section data available for these atoms. We applied the DM formalism to these three atoms. The results are shown in Fig. 7. The maximum cross section values for these three atoms range between $(4 \text{ and } 5.5) \times 10^{-20} \text{ m}^2$. The Mn and Cr ionization cross sections peak at a comparatively low impact energies (between 20 and 30 eV), while the W cross section does not reach its maximum value until about 60 eV. Measurements of these important ionization cross sections would be highly desirable.

4. Conclusions

Here we present the results of the application of the recently revised DM formalism for the calculation of atomic ionization cross sections to the following atoms (listed in order of increasing atomic number Z): Na, Si, S, Cl, K, Ca, Fe, Ga, Br, In, Cs, Hg, Bi, and U. The objective was: (i) to test the

validity of the revised DM formula for these atoms in both the low- and high-energy regime; (ii) to extend the method to atoms with higher atomic numbers Z (where the contribution of f -electrons to the calculated ionization cross section has to be included); (iii) to provide guidance as to the reliability of measured cross sections in cases where several experimental investigations reported drastically different absolute cross sections; and (iv) to provide reliable cross sections data in cases where no experimental data are available.

The comparison between calculated and measured ionization cross sections leads to the following observations:

- (1) There are five atoms for which three or more independent sets of experimental data are available (Na, K, Cs, Ca, Ga). In cases where the available measurements tend to converge towards an ‘accepted’ ionization cross section (Na, K, Ca, Ga), the DM calculation tends to support the ‘accepted’ cross section. Exceptions are features that may be attributed to indirect processes such as e.g. autoionization (K, Cs, Ca), which are not included in the DM formalism. In Cs, where the different experimental data sets do not converge towards an ‘accepted’ cross section, the DM calculation provides guidance as to which measurements yield the more reliable data.
- (2) There are three atoms for which two independent sets of experimental data are available (S, Fe, In). In all three cases, the DM calculation may provide guidance as to which measurements yield the more reliable data, particularly in the case of S and In. However, it must also be noted that the DM calculation cannot reproduce the unusual energy dependence of the S ionization cross section, which was prominent on both experimental data sets.
- (3) There are four atoms (Cl, B, Si, Bi) for which a single measured data set is available. The data for all four atoms were obtained by the same group using the same experimental technique (Freund and co-workers, whose cross section data are considered to be among the most reliable data [34]). In the four cases, the DM calculation agrees well with the measured data with the level of agreement between measured and calculated ionization cross section ranging from excellent (Cl, B, Si) to good (Bi).
- (4) There are two atoms (Hg, U), for which a single measured data set is available. Each data set was obtained by a different group. Neither group has published the result of more than the one ionization cross section measurement to the best of our knowledge, so that the reliability of their ionization cross section data has not been solidly established. In both cases, the DM calculation shows poor agreement with the measured data.
- (5) Lastly, we also calculated ionization cross sections for the atoms Cr, Mn, and W, which are technically important, e.g. as impurities in fusion plasma devices. This was done in an effort to provide guidance to modellers, since no experimental data are available for these atoms.

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