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A semi-empirical concept for the calculation of electron-impact ionization cross-sections of neutral and ionized fullerenes

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In memoriam of Werner Lindinger.

Abstract

We have developed a semi-empirical approach to the calculation of cross-section functions (absolute value and energy dependence) for the electron-impact ionization of several neutral and ionized fullerenes $C_{60}{}^{q+}$ ($q = 0$–3), for which reliable experimental data have been reported. In particular, we propose a modification of the simplistic assumption that the ionization cross-section of a cluster/fullerene is given as the product of the monomer ionization cross-section and a factor m^a, where "m" is the number of monomers in the ensemble and "a" is a constant. A comparison between our calculations and the available experimental data reveals good agreement for $q = 0$, 1, and 3. In the case of the ionization of $C_{60}{}^{2+}$ ($q = 2$), our calculation lies significantly below the measured cross-section which we interpret as an indication that additional indirect ionization processes are present for this charge state. (Int J Mass Spectrom 223–224 (2003) 1–8)
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1. Introduction

Electron-impact ionization cross-section functions have been measured for nearly 100 molecules including free radicals, clusters, and fullerenes [1]. Calculation of absolute total single ionization cross-sections for most molecules and their energy dependence using semi-rigorous methods such as the BEB method of Kim and coworkers [2,3] and the Deutsch–Märk (DM) formalism [4] reveal reasonably good agreement (to within 20%) between measured and calculated data for the vast majority of molecules. Even in cases such as the fluorine-containing free radicals CF_x and NF_x ($x = 1$–3), where earlier calculations revealed a significant disagreement between measured and calculated cross-sections of up to a factor 2, the same 20% level of agreement has been achieved recently using a modified BEB approach [5].

The situation is distinctly different for clusters and fullerenes. Several theoretical models have been applied to the quantitative characterization of the ionization properties of clusters and fullerenes, i.e., to the calculation of their absolute ionization cross-sections (see e.g., Deutsch et al. [4,6] and Matt et al. [7]). The various theoretical approaches (for more see also

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the following detailed discussion) result in different absolute ionization cross-sections both in terms of the absolute value and the cross-section shape as a function of electron-impact energy for a given target. However, none of the methods appears to succeed in reproducing the experimental data that are available for a few targets. In this context, we will not distinguish between "clusters" and "fullerenes". Both will be considered "ensembles of monomers" encompassing a large number of constituents and geometrical structures ranging from a hard sphere packing arrangement to a hollow cage structure.

In this paper, we introduce a semi-empirical approach to the calculation of cross-section functions (absolute value and energy dependence) for the electron-impact ionization of several neutral and ionized fullerenes $C_{60}{}^{q+}$ ($q = 0$–3), for which reliable experimental data have been reported. In particular, we want to test for these selected cases to what extent, if at all, the assumption holds that the ionization cross-section of a cluster/fullerene is given as the product of the monomer ionization cross-section and a factor m^a, where "m" is the number of monomers in the ensemble and "a" is a constant.

2. Background

In the past, the following theoretical approaches have been applied to the calculation of ionization cross-sections of clusters/fullerenes:

1. The simple Additivity Rule (AR)—based on the AR for molecules [8–10]—has been applied to the calculation of total ionization cross-sections of *small* clusters X_m consisting of m monomers. The cluster cross-section $\sigma_{\text{tot}}(X_m)$ is expressed in terms of the monomer cross-section $\sigma_{\text{tot}}(X)$ as

$$\sigma_{\text{tot}}(X_m) = m\sigma_{\text{tot}}(X) \tag{1}$$

and both cross-sections are functions of the electron energy E and obviously have the same energy dependence. While Eq. (1) holds for total ionization cross-sections, similar considerations also apply to partial ionization cross-sections. Eq. (1) was sometimes applied without taking into account possible fragmentation and cascading effects (see Ref. [11] for a discussion of potential pitfalls resulting from this approach). Even for dimers and trimers Eq. (1) has to be modified according to a "defect concept" to

$$\sigma_{\text{tot}}(X_m) = m^{(1-\Delta)}\sigma_{\text{tot}}(X) \tag{1a}$$

with Δ depending on the proton number of the cluster constituent. With $0.35 < \Delta < 0.1$, all experimentally determined cross-sections can be reproduced quite nicely [12].

2. A modified additivity rule (MAR) based on geometrical concepts and particularly applicable to large clusters was proposed which introduced an exponent "α" ($\alpha < 1$) in the following way:

$$\sigma_{\text{tot}}(X_m) = m^{\alpha}\sigma_{\text{tot}}(X) \tag{2}$$

Simple geometrical considerations lead to a prediction of $\alpha = 2/3$ [13] in the limit of large cluster sizes. The same formula was later proposed by Deutsch et al. [14] using a simplified version of the DM formalism [15,16] (see also the following discussion) in which the radius of the cluster, R_{cluster}, replaced the atomic radius of the outermost shell of the target which is a critical quantity in the DM formula. In this case, the value $\alpha = 2/3$ represents the lower limit of the exponent "α" and corresponds to some kind of hard sphere packing approximation. These authors used an empirically determined exponent $\alpha = 0.84$ in their calculations of ionization cross-sections for H_2 and CO_2 clusters ranging in size range from a few hundred to 100,000 monomers. We note that Eq. (2) is sometimes written in the form

$$\sigma_{\text{tot}}(X_m) = m^{2a}\sigma_{\text{tot}}(X) \tag{2a}$$

where the radius of the cluster, R_{cluster}, is related to the radius of the monomer, R_{monomer}, via

$$R_{\text{cluster}} = m^{a} R_{\text{monomer}} \tag{2b}$$

As was the case in Eq. (1), the cross-sections in Eqs. (2) and (2a) are functions of the electron energy *E* and have the same dependence on the electron energy [14].

3. The DM formalism, which was originally developed as a semi-classical approach to the calculation of atomic single ionization cross-sections, has been extended to molecules, fullerenes, and clusters [4,6] by employing a Mulliken or other atomic population analysis that expresses the fullerene orbitals in terms of the atomic orbitals of the constitutent atoms. In the case of C_{60}, a Mulliken population analysis of the outermost 120 C_{60} orbitals was carried out in terms of the C(2s) and C(2p) atomic carbon orbitals, whereas contributions from the tightly bound C(1s) orbitals were neglected. In this approach, the 60 carbon atoms are considered an ensemble of 60 independent atoms and no allowance is made for the cage structure of C_{60}. The application of this variant of the DM formula to the calculation of the ionization cross-section of C_{60} resulted in an overestimation of the cross-section maximum by at least a factor of 2 compared to the most reliable experimental results [17–20]. Deutsch et al. [6] speculated that this discrepancy may be due to the fact that this variant of the DM formalism did not take into account the special geometrical arrangement of the C_{60} cage. Moreover, the DM formula was also not capable of reproducing the unusual energy dependence of the measured C_{60} ionization cross-section, which is not surprising, as the DM formalism uses a Gryzinski-type energy dependence for the atomic ionization cross-sections [4] which does not change when the formula is applied to molecules and clusters/fullerenes.
4. The jellium model [21] incorporates a quantum mechanical calculation of the integrated cross-section for the ionization of C_{60} by fast electrons (kinetic energies in excess of 100 eV) based on the plane-wave Born approximation (PWBA) and used the spherical jellium model to represent the bound and continuum states of the target. The spherical jellium model completely disregards the ionic substructure of C_{60}, which is just the opposite of the DM approach presented earlier that treats the C_{60} as an ensemble of 60 independent carbon atoms, thus, neglecting the cage structure of C_{60} completely. Given the overestimation resulting from the application of the DM formula to C_{60}, Keller and Engel [21] argued that the fullerene geometry is an important factor in ionization cross-section calculations. It is thus instructive to consider an ionization cross-section calculation for C_{60} based on the other extreme representation of C_{60}, namely the jellium model. Keller and Engel [21] demonstrated that their model represents the experimental data quite well for impact energies above about 500 eV, but significantly overestimates the measured cross-section at lower impact energies. These authors attribute the failure of the PWBA–jellium model at lower energies to the general tendency of the PWBA to overestimate cross-sections at lower impact energies rather than to the breakdown of the jellium shell model.
5. Bottiglioni and Coutant [22] were the first to propose a calculation scheme for relative ionization cross-sections of clusters that included (i) the energy loss inside the cluster of the primary electron, and (ii) the fact that secondary electrons are created which will subsequently suffer further energy losses and have an escape probability from the cluster of less than unity, i.e., they only escape from the cluster if their energy exceeds a certain minimum, which depends on the position inside the cluster where they are initially formed. This resulted in a cross-section shape that was markedly different from the shape predicted by the other methods and was found to agree better with the unusual shape of the C_{60} ionization cross-section. However, these authors limited their calculations to cross-sections shapes and did not calculate absolute cross-section values.

3. Present approach for the calculation of partial ionization cross-sections

In the present paper, we start from the MAR discussed earlier in Section 2, and we attempt to incorporate ad hoc the apparent advantages of the other approaches discussed before, that is, achieve improvements in the ability to predict the absolute cross-section value and the cross-section shape that were determined in reliable experiments. It is important to note that the theoretical approaches mentioned earlier (Section 2, approaches 1, 2, and 3) are methods to calculate absolute total single ionization cross-sections, whereas here we attempt to derive a method to calculate absolute partial ionization cross-sections. Our goal is to do this in a fashion that is consistent and transparent and that reflects the underlying physical principles. In a first step, we limit our discussion to the following singe ionization processes:

$$e^- + C_{60} \rightarrow {C_{60}}^{+} + 2e^- \tag{3a}$$

$$e^- + {C_{60}}^{+} \rightarrow {C_{60}}^{2+} + 2e^- \tag{3b}$$

$$e^- + {C_{60}}^{2+} \rightarrow {C_{60}}^{3+} + 2e^- \tag{3c}$$

$$e^- + {C_{60}}^{3+} \rightarrow {C_{60}}^{4+} + 2e^- \tag{3d}$$

for which reliable experimental cross-section data are available [17–20,23]. On the basis of the fact that Eq. (2a) yields a cross-section shape that differs from the experimentally determined shape, in particular at higher impact energies and that overestimates the absolute cross-section value significantly, we modify Eq. (2a) in the following way:

$$\sigma_{\mathrm{tot}}(X_m, E) = m^{2a} e^{-b} \sigma_{\mathrm{tot}}(X, E) F(E) \tag{4}$$

where the two factors "e^{-b}" and "$F(E)$" serve the following purpose.

3.1. The factor e^{-b}

This factor reflects a reduction in the cross-section which arises from the fact that multiple single ionization processes can occur when a fullerene is ionized by an incident electron which will reduce the cross-section for the particular channel under consideration; the exponent "b" is a function of the cluster size "m" and of the charge state "q". The feasibility of the application of such a function of the charge state has been demonstrated successfully for instance in Ref. [24]. For the single ionization processes (3a) through (3d), it is advantageous to write the exponent "b" in the form $b = b_1 + qb_2$, where q refers to the charge state of the target prior to the ionization, i.e., $q = 0$ refers to C_{60}, $q = 1$ refers to ${C_{60}}^{+}$, $q = 2$ refers to ${C_{60}}^{2+}$, etc. The constant b_1 reflects the effective reduction of the ionization cross-section of the target because of the fact that clusters/fullerenes cannot be considered simply ensembles of independent monomers. The factor "$\exp(-b_1)$" may be viewed as a "structure factor". If we use the data of Salzborn and coworkers for the processes (3b) and (3d), we can plot the exponent "b" as a function of q and obtain the two data points shown in Fig. 1 as full circles for $q = 1$ and 3. A linear extrapolation yields the two points in Fig. 1 shown as open circles which correspond to $q = 0$ and 2, respectively. The intercept of the straight line in Fig. 1 with the b-axis yields the value of the constant b_1 as $b_1 = 0.79$. The constant b_2 has a value of $b_2 = 0.255$. It is interesting to note that the work of Keller and Engel [21] suggests a value close to the present factor of $e^{-0.79} = 0.454$ for the case of C_{60} ($q = 0$). Keller and Engel [21] calculate a value for the C_{60} cross-section at 1000 eV of $35 \times 10^{-16}\,\mathrm{cm}^2$

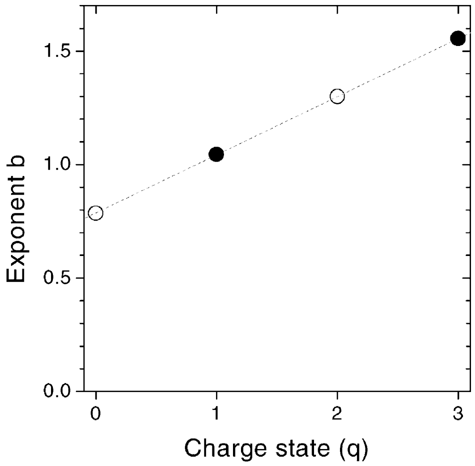

Fig. 1. Plot of the exponent "b" vs. the charge state of the target. The filled circles (●) for $q = 1$ and 3 were obtained from an analysis of the experimental data of Ref. [23]. The open circles (○) represent the two data points for $q = 0$ and 2 that are obtained by linear extrapolation.

using their PWBA method assuming 60 independent C atoms, whereas the measurements and the PWBA calculations based on the spherical jellium model yield a value of about $16 \times 10^{-16}\,\mathrm{cm}^2$ at the same impact energy. This suggests a reduction in the cross-section as a consequence of the assumptions of the jellium model (i.e., completely disregarding the ionic substructure of C_{60}) of the order of $16/35 = 0.46$ which is essentially identical to our factor $e^{-0.79} = 0.454$ in this case.

3.2. The energy dependence F(E)

This function describes the deviation of the cross-section shape of the cluster/fullerene X_m from the cross-section shape of the monomer X. The function $F(E)$ is defined as:

$$F(E) = \frac{\sigma_{X_m}(E)/\sigma_{X_m,\max}(E)}{\sigma_X(E)/\sigma_{X,\max}(E)} \tag{5}$$

and has a similar shape for both clusters (see [22,25]) and fullerenes (see [18–20], if $F(E)$ is verified with the data reported in these references. For both clusters and fullerenes, the function $F(E)$ consists of two parts which are separated by a characteristic energy E_C. For energies between the ionization threshold E_{thr} and E_C, $F(E)$ starts from 0 and approaches 1 which is due to an effective shift of the energy scale caused by inelastic energy losses of the primary electrons inside the cluster/fullerene [22,26]. For energies above E_C, $F(E)$ increases essentially linearly from the value 1 at $E = E_C$ and represents the fact that secondary electrons created in the initial ionization process can leave the cluster/fullerene only, if their energy exceeds a certain minimum energy and the probability for escape increases with increasing energy E. The function $F(E)$ can be explicitly determined theoretically [22] or experimentally based on measurements of the cross-sections $\sigma_{X_m}(E)$ and $\sigma_X(E)$. Table 1 shows the values of $F(E)$ for selected electron energies from 20 to 1000 eV obtained empirically from the data of Ref [23] for the process (3b). The resulting energy dependence $F(E)$ is also shown in Fig. 2 (here E_C has a value of about 70 eV). The resulting cross-section formula has the form of Eq. (4).

Table 1
Explicit form of the functions $F(E)$ and $F_{cage}(E)$ obtained empirically from the measured data of Ref. [23] for the single ionization of $C_{60}{}^{+}$ (process (3b), see text)

Energy (eV)	$F(E)$	$F_{cage}(E)$ including energy shift
20	0.374	1
30	0.772	1
40	0.922	1
50	0.969	1
70	0.967	1
100	1.122	1.122
150	1.292	1.292
200	1.442	1.442
300	1.730	1.730
400	1.889	1.889
500	2.040	2.040
700	2.340	2.340
900	2.500	2.500

Alternatively, the influence of the low energy dependence $F_A(E)$ on the cross-sections can be realized by introducing an energy shift

$$E^* = E - [E_0 + (\mathrm{IP}_{\text{cluster-ion}} - \mathrm{IP}_{\text{monomer}})] \tag{6}$$

where E^* denotes the effective energy of the primary electron within the cluster/fullerene, E the kinetic energy of the primary electron, and E_0 the energy loss due to inelastic scattering. In the present work, a

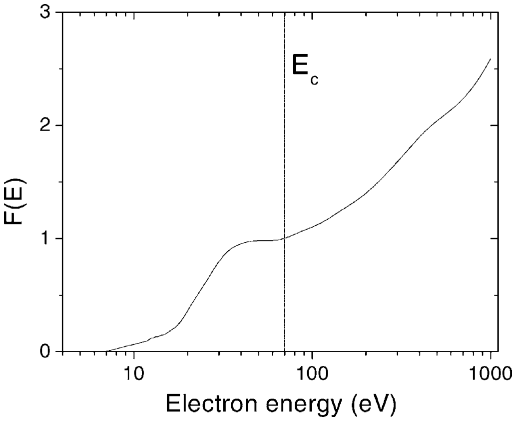

Fig. 2. Energy dependence $F(E)$ as defined in Eq. (5) in the text for the specific case of the formation of $C_{60}{}^{2+}$ ions following electron-impact single ionization of $C_{60}{}^{+}$. The curve was obtained empirically from the data reported in Ref. [23] (see also text and Table 1).

constant value of 5 eV was used for E_0. The appropriate values for the ionization potentials (IPs) can be found in Refs. [23,27]. This leads to a cross-section formula of the form

$$\sigma_{\text{tot}}(X_m, E) = m^{2\alpha} \mathrm{e}^{-b} \sigma_{\text{tot}}(X, E^*) F_{\text{cage}}(E) \tag{7}$$

where $F_{\text{cage}}(E)$ is taken from column 3 of Table 1. We further note that in the expression of E^* in Eq. (6), the term in parenthesis is applicable only for ionic targets. For the four cases presented in this paper (see Section 4), the above cross-section formulas (4) and (7) yield essentially the same energy dependence of the cross-sections.

4. Results and discussion

In this section, we develop explicit functional forms that allow us to calculate the absolute ionization cross-sections for the processes (3a) through (3d) listed earlier. We either use previously determined values for some of the constants in formula (4) for the various cases or derive values from the experimental data of Ref. [23] for process (3c) and use them accordingly for the other processes. For reasons of consistence, we use the energy dependence as described in formula (8) throughout.

4.1. $e^- + C_{60}{}^+ \rightarrow C_{60}{}^{2+} + 2e^-$

We use the exponent $2a = 0.786$ from Deutsch et al. [14] and the experimentally determined atomic carbon ionization cross-section $\sigma_{\mathrm{C}}(E)$ of Brook et al. [27] and find the following expression for the absolute ionization cross-section for C_{60} as:

$$\sigma_{60^+}(E) = 60^{0.786} \mathrm{e}^{-1.045} \sigma_{\mathrm{C}}(E^*) F_{\text{cage}}(E) \tag{8}$$

The constant "b" has the value $b = 0.79 + 1 \times 0.255 = 1.045$. Since the experimentally determined ionization cross-section for this process was used to determine the constant "b" and the function $F_{\text{cage}}(E)$, it is obvious that the cross-section of Eq. (8) represents the experimental data very well over the entire energy range (see Fig. 3).

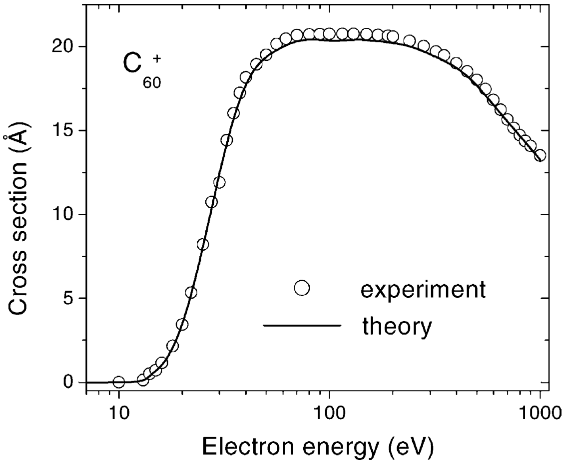

Fig. 3. Cross-section for the formation of $C_{60}{}^{2+}$ ions following electron-impact single ionization of $C_{60}{}^+$. The experimental data (○) are from Ref. [23], the solid line represents the present calculation.

4.2. $e^- + C_{60}{}^{3+} \rightarrow C_{60}{}^{4+} + 2e^-$

In this case, the constant "b" has the value $b = 0.79 + 3 \times 0.255 = 1.555$ and the ionization cross-section has the form

$$\sigma_{60^{3+}}(E) = 60^{0.786} \mathrm{e}^{-1.555} \sigma_{\mathrm{C}}(E^*) F_{\text{cage}}(E) \tag{9}$$

where we use the same function $F_{\text{cage}}(E)$ from before for the single ionization of $C_{60}{}^+$. As can be seen in Fig. 4, the overall agreement between the prediction of Eq. (9) and the experimentally determined ionization of Ref. [23] is quite good over the entire range of impact energies.

4.3. $e^- + C_{60}{}^{2+} \rightarrow C_{60}{}^{3+} + 2e^-$

The constant "b" has the value $b = 0.79 + 2 \times 0.255 = 1.300$ and the ionization cross-section has the form

$$\sigma_{60^{2+}}(E) = 60^{0.786} \mathrm{e}^{-1.3} \sigma_{\mathrm{C}}(E^*) F_{\text{cage}}(E) \tag{10}$$

where we again use the same function $F_{\text{cage}}(E)$ from before for the single ionization of $C_{60}{}^+$. As can be seen in Fig. 5, the overall agreement between the prediction of Eq. (10) and the experimental data of Ref. [23] is good only for very low energies up to about

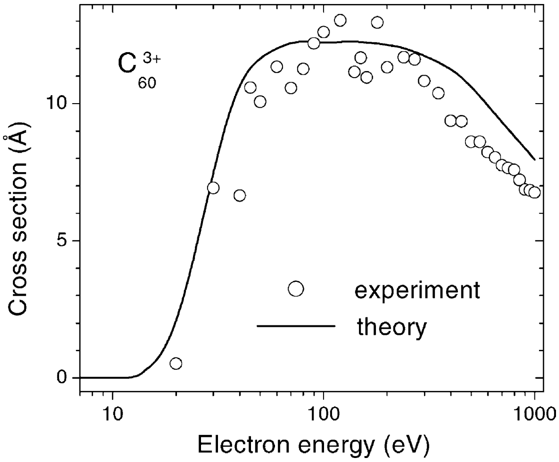

Fig. 4. Cross-section for the formation of $C_{60}{}^{4+}$ ions following electron-impact single ionization of $C_{60}{}^{3+}$. The experimental data (○) are from Ref. [23], the solid line represents the present calculation.

35 eV. For higher energies, there appears to be an additional process contributing to the measured data. For fullerene ions this effect may be caused by autoionization processes [23].

4.4. $e^- + C_{60} \rightarrow C_{60}{}^+ + 2e^-$

This is a particularly interesting case which in some sense triggered the discussion leading up to the present paper. There have been two independent and reliable experimental determinations of the C_{60} ionization cross-section which both revealed a very unusual cross-section shape and an absolute cross-section value with a maximum value which amounted to only about half the cross-section value predicted by various standard theoretical approaches which worked quite well for many molecules including rather complex molecules [6,18–20]. On the basis of the approach described here, we find a constant "b" with a value $b = 0.79 + 0 \times 0.255 = 0.79$ and the ionization cross-section for C_{60} has the form

$$\sigma_{60}(E) = 60^{0.786} e^{-0.79} \sigma_C(E^*) F_{\text{cage}}(E) \qquad (11)$$

where we again use the same function $F_{\text{cage}}(E)$ from before for the single ionization of $C_{60}{}^+$. As can be seen in Fig. 6, Eq. (11) now provides a description of the C_{60} ionization cross-section that agrees very well with the measured cross-section over the entire range of impact energies from threshold to 1000 eV.

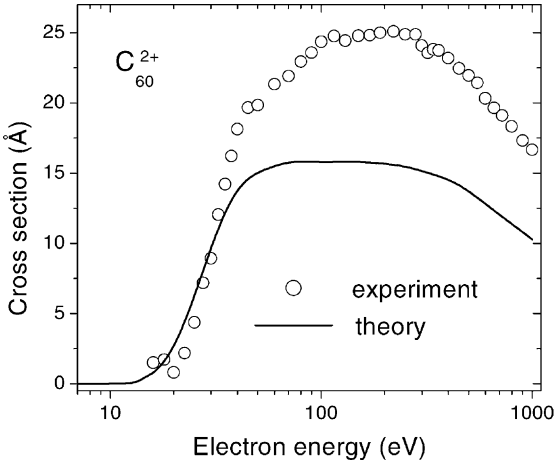

Fig. 5. Cross-section for the formation of $C_{60}{}^{3+}$ ions following electron-impact single ionization of $C_{60}{}^{2+}$. The experimental data (○) are from Ref. [23], the solid line represents the present calculation.

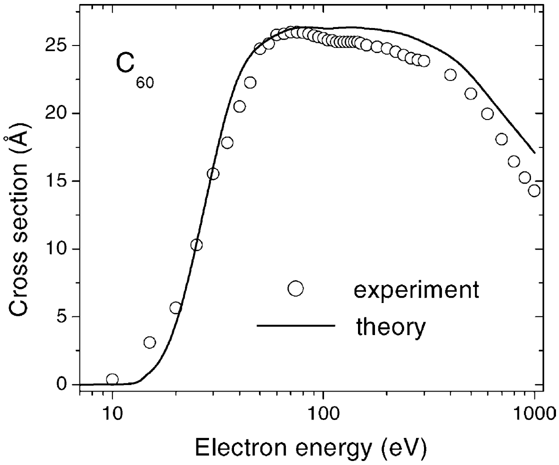

Fig. 6. Cross-section for the formation of $C_{60}{}^+$ ions following electron-impact single ionization of C_{60}. The experimental data (○) are from Ref. [18], the solid line represents the present calculation.

5. Conclusions

We derived a semi-empirical approach to the calculation of cross-section functions (absolute value and energy dependence) for the electron-impact ionization of several neutral and ionized fullerenes $C_{60}{}^{q+}$ ($q = 0$–3), for which reliable experimental data have been reported. Our new method is based on an MAR and incorporates ad hoc two factors, a structure factor and an energy-dependent factor. Our model allows us to predict the experimentally determined absolute cross-section values and the cross-section shapes for a variety of fullerene ionization processes. A comparison between our calculations and the available experimental data for $C_{60}{}^{q+}$ ($q = 0$–3) reveals good agreement for $q = 0$, 1, and 3, but indicates that additional indirect ionization processes are present in the case of $q = 2$.

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