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Calculated cross sections for the single ionization of fullerenes by electron impact

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Abstract

A spherically symmetric model is constructed in order to obtain one-electron wavefunctions for the valence electrons of the Buckminsterfullerene. Using these wavefunctions, the cross sections for the ionization of the fullerene ion and of the neutral fullerene by electron impact are calculated within the distorted-wave Born approximation. The obtained results are compared with the experimental data.

1. Introduction

The unique properties of the C_{60} cluster (Buckminsterfullerene) have induced several theoretical and experimental investigations. Ab initio calculations have been performed for the study of the electronic structure of the molecule [1–4], and there have been developed simplified models [5] to describe its main features.

The ionization and fragmentation of the fullerenes in collision with different particles have been studied experimentally by several groups [6–10]. Electron–fullerene ion collisions have been realized by Völpel et al. [7], electron–neutral C_{60} collisions by Dünser et al. [10]. They have measured the cross sections for single and multiple ionization of the cluster, and have also counted the main fragment ions produced by the collisions. The single ionization cross section of the C_{60}^+ as a function of the projectile energy shows an unusual wide maximum between 50 and 250 eV. No similar behaviour was found in case of the ionization cross section of the neutral C_{60}.

Previously we have performed a semiclassical calculation for the ionization of the C_{60}^+ ion by charged particle impact [11]. This type of calculation is able to give good results for electron impact ionization only above 500 eV projectile energy. In the present paper we report our results for the ionization of the C_{60}^+ and of the neutral C_{60} by electron impact, obtained with the distorted-wave Born approximation (DWBA), more appropriate to lower energies. Calculations were made using a spherical model of the fullerene, and these results have been compared with those obtained by the additivity rule.

2. Theory

We have constructed a spherically symmetric model of the C_{60} molecule [11]. The one-electron wavefunctions of the valence electrons are calculated in the Coulomb potential created by a positive background charge. The positive charge is localized on a hollow spherical shell with the radius of $6.7a_0$, which is the radius of the C_{60} molecule. In order to keep the electrons in the vicinity of the shell of the sphere (near to the carbon nuclei), we have introduced a pseudopotential (as in [5]), with the minimum value at $R = 6.7a_0$. The pseudopotential is taken spherically symmetric and of Gaussian shape.

The 180 σ electrons (with one node in the radial wavefunction, in the origin) occupy the angular momentum states up to $l = 9$ (with 18 electrons at the highest level), and the 60 π electrons (two nodes in the radial wavefunction) occupy the angular momentum states up to $l = 5$ (remaining 10 electrons for this level). The parameters of the pseudopotential are adjusted to obtain the energies of electronic states in accordance with more elaborate models and the experimental data [1,5].

The continuum wavefunctions of the incident and scattered electron are calculated numerically in the field of the target, whereas the wavefunction for the ejected electron is calculated in the field of the residual ion.

For the ionization of the fullerene we have applied a DWBA computer code developed previously for the study of the ionization of different atomic ions by low-energy electron impact [12]. Each continuum wavefunction is expanded into the partial waves. Summation is made for the incident and scattered electron up to $l = 22$, and for the ejected electron up to $l = 16$. The sum over the partial waves was found to be convergent only for projectile energies be-

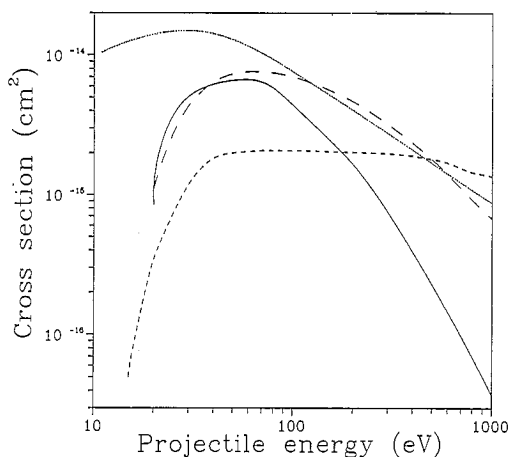---

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Fig. 1. Single ionization cross sections of the C_{60}^+ ion by electron impact as a function of the projectile energy. The solid line stands for our results got with a spherical model of the cluster and using the DWBA. Cross sections obtained with the additivity rule are represented by long-dashed line, and dotted line represents our previous semiclassical calculations [11]. The short-dashed line stands for a fit of the experimental data of Völpel et al. [7].

low 200 eV.

We have integrated over the energy of the ejected electron using a five-point Gauss–Legendre formula. The ionization cross sections from each bound state have been calculated separately, have been multiplied by the number of occupancy, and have been summed up for all states.

3. Results and discussion

Cross sections for the single ionization of the C_{60}^+ ion by electron impact as a function of the projectile energy are plotted on Fig. 1. The solid line represents the DWBA cross sections obtained with the spherical model of the fullerene, while the long-dashed line stands for the cross sections obtained by the additivity rule and the DWBA. We have plotted also the cross sections calculated previously [11] within a semiclassical approximation (dotted line), and the experimental data of Völpel et al. [7] (short-dashed line).

As it was expected, at low collision energies the DWBA cross sections are lower, and closer to the experimental data, than those got with the semiclassical approximation. At projectile energies up to 100 eV, cross sections obtained with the spherical model of the fullerene and those obtained with the additivity rule are close to each other, and overestimate about 3 times the experimental data.

Above 150 eV projectile energy the semiclassical, and the DWBA cross sections calculated with the additivity rule, have close values. Above this energy the semiclassical approximation becomes to be valid, and the DWBA cross sections for the individual carbon atoms can be calculated with a good approximation, because in this case the partial-wave
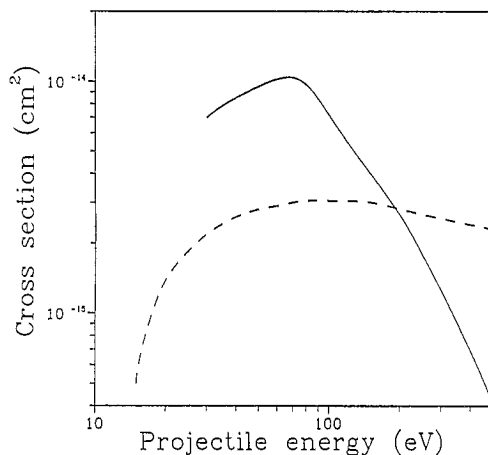

Fig. 2. Single ionization cross sections of the neutral C_{60} by electron impact as a function of the projectile energy. The results of our calculations (solid line) are compared to the experimental data of Dünser et al. [10] (dashed line).

series is fastly convergent. This is not the case when using the DWBA for the spherical model of the C_{60}^+. While at 100 eV the truncation of the partial-wave series at $l = 22$ for the incident and the scattered electron introduces an error of 20%, at 200 eV this increases to a factor of two. At 1000 eV the inclusion of the partial waves up to $l = 36$ increases the cross section by ten times, and no convergence was found.

It is clear, that our DWBA using the spherical model is valid only at low impact energies, below 150 eV. Above this energy the semiclassical approximation is more appropriate. Cross sections obtained with the additivity rule are reliable in the whole energy range.

The discrepancy between our theoretical results and the experimental data at low impact energies may have several causes. First, our calculations were made within the framework of the independent-electron approximation (we have used one-electron wavefunctions). In such a complex target, electron correlation during the collision may have decisive importance. Second, the deviation from sphericity of the real C_{60}^+ cluster modifies considerably the ground-state wavefunctions, especially those with higher angular momentum.

Third, the comparison with the experiment is not very accurate. Völpel et al. have measured the cross sections for the production of the C_{60}^{2+} ion, so the published cross sections for the single ionization are exclusive. We have calculated the cross sections for the ejection of an electron from the C_{60}^+, whatsoever happens with the residual ion. Our cross sections are inclusive, include multiple ionization and fragmentation-ionization cross sections, too. Völpel et al. have published results also for the production of the C_{60}^{3+}, C_{56}^+ and C_{56}^{2+} ions. Adding these to the production of the C_{60}^{2+}, the cross section is increased by 50% at 100 eV impact energy. While the production of the C_{58}^+ and C_{58}^{2+} ions might be very important, the inclusive cross section for the ejection of an electron is probably more than twice the cross section for the production

of the C_{60}^{2+} ions.

In case of the ionization of the neutral C_{60} we have made calculations with the spherical model of the cluster using plane waves for the description of the incident and scattered electron. The results are shown on Fig. 2 (solid line), in comparison with the total counting ionization cross sections measured by Dünser et al. [10] (dashed lines). The situation is similar to that for the C_{60}^+ ion. Our calculated cross sections overestimate the experimental data by a factor of 3, up to 80 eV projectile energy. Above 150 eV, our calculations become inaccurate, because of the truncation of the partial-wave series.

The total counting cross sections of Dünser et al. include single and multiple ionization of the C_{60}, and the production of heavy singly and multiply charged ions. The production of light charged fragments (like C_2^+, C_4^+) may be important, too, leaving the heavy fragments (C_{58} and C_{56}) neutral. This kind of ionization-fragmentation could increase the experimental total ionization cross sections.

4. Conclusions

We have presented DWBA calculations for the ionization of the C_{60}^+ ion and of the neutral C_{60} by electron impact, using a simple spherical model of the cluster. Due to the bad convergence of the partial-wave series at high projectile energies, our calculations are correct only below 150 eV. At these low projectile energies, our results overestimate the experimental data by a factor of 3. This discrepancy may be caused by the inaccuracies of the model, and by the fact that the calculations give the total cross sections for the ejection of an electron, while the experimentalist measure the rate of the production of some ions, but not for all fragments.

Acknowledgements

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