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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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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}^{n+} (n = 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 n = 0,1,3. In the case of the ionization of C_{60}^{2+} (n = 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.

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 co-workers [2,3] and the 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. [6,7]). The various theoretical approaches (see also the more detailed discussion below) 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}^{n+} (n = 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.

Results and Discussion

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}^{n+} (n = 0-3), for which reliable experimental data have been reported [8,9]. Our new method is based on a modified additivity rule

$$\sigma_{tot}(X_m) = m^{2a} \cdot \sigma_{tot}(X) \tag{1}$$

and incorporates *ad hoc* two factors, a structure factor e^{-b} and an energy-dependent factor $F_{cage}(E)$.

$$\sigma_{tot}(X_m, E) = m^{2a} \cdot e^{-b} \cdot \sigma_{tot}(X, E^*) \cdot F_{cage}(E) \tag{2}$$

The influence of the low energy dependence F(E) on the cross sections can be realized by introducing an energy shift

$$E^* = E - \left[E_0 + \left(IP_{cluster-ion} - IP_{monomer}\right)\right] \tag{3}$$

where E^* denotes the effective energy of the primary electron within the cluster/fullerene, E is the kinetic energy of the primary electron, and E_0 the energy loss due to inelastic scattering. In the present work a constant value of 5 eV was used for E_0. The appropriate values for the ionization potentials (IPs) can be found in Refs. [9] and [10].

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}^{n+} (n= 0-3) reveals good agreement for n = 0,1,3, but indicates that additional indirect ionization processes are present in the case of n = 2 (see Figures 1-4).

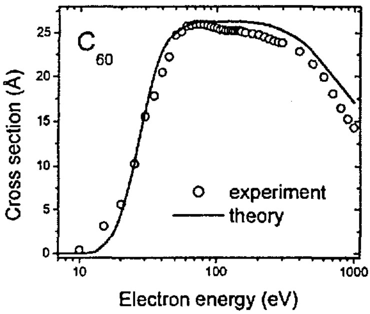

Fig. 1 Cross section for the formation of C_{60}^{+} ions following electron impact single ionization of C_{60}. The experimental data (open circles) are from Ref. [8], the solid line represents the present calculation.

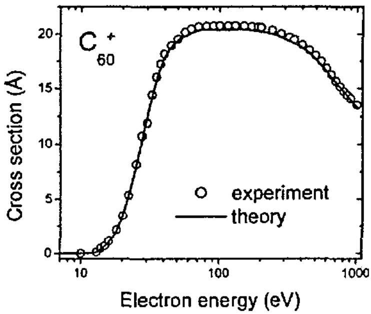

Fig. 2 Cross section for the formation of C_{60}^{2+} ions following electron impact single ionization of C_{60}^{+}. The experimental data (open circles) are from Ref. [9], the solid line represents the present calculation.

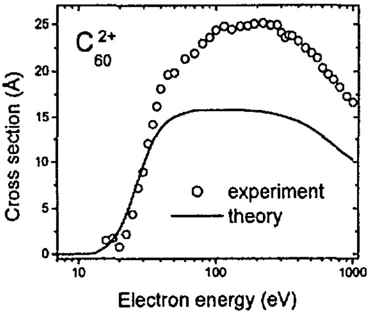

Fig. 3 Cross section for the formation of C_{60}^{3+} ions following electron impact single ionization of C_{60}^{2+}. The experimental data (open circles) are from Ref. [9], the solid line represents the present calculation.

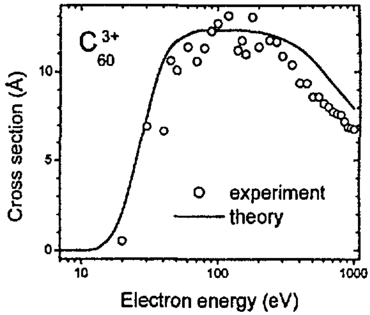

Fig. 4 Cross section for the formation of C_{60}^{4+} ions following electron impact single ionization of C_{60}^{3+}. The experimental data (open circles) are from Ref. [9], the solid line represents the present calculation.

Acknowledgments

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List of authors in alphabetical order

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