



A Semi-Empirical Concept for the Calculation of Electron-Impact Ionization Cross Sections of Neutral and Ionized Fullerenes

H. Deutsch¹, P. Scheier², K. Becker³, and T.D. Märk²

¹ Institut für Physik, Ernst-Moritz-Arndt Universität, D-17487 Greifswald, Germany

² Institut für Ionenphysik, Leopold-Franzens Universität, A-6020 Innsbruck, Austria

³ Department of Physics, Stevens Institute of Technology, Hoboken, NJ 07030, USA

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^n , 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

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

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

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

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

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

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).

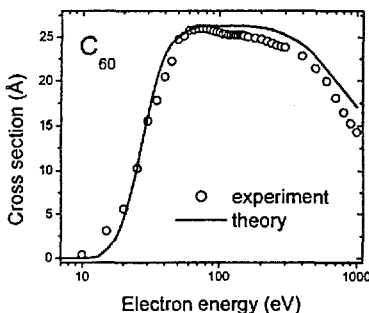


Fig. 1 Cross section for the formation of C_{60}^{+} ions following electron impact single ionization of C_{60} . The experimental data (open circles) are from Ref. [8], the solid line represents the present calculation.

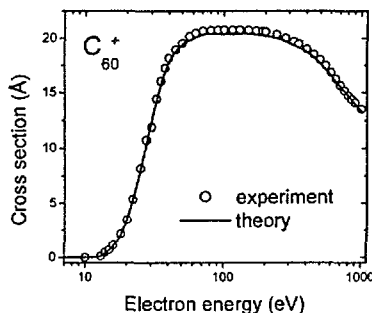


Fig. 2 Cross section for the formation of C_{60}^{2+} ions following electron impact single ionization of C_{60}^{+} . The experimental data (open circles) are from Ref. [9], the solid line represents the present calculation.

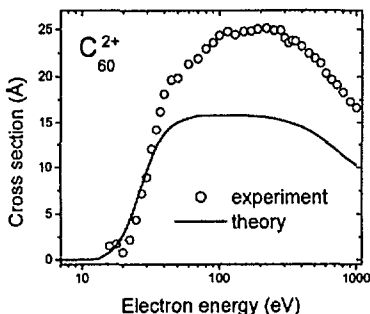


Fig. 3 Cross section for the formation of C_{60}^{3+} ions following electron impact single ionization of C_{60}^{2+} . The experimental data (open circles) are from Ref. [9], the solid line represents the present calculation.

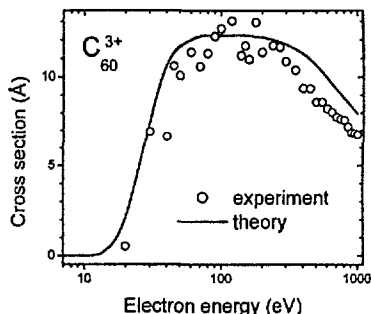


Fig. 4 Cross section for the formation of C_{60}^{4+} ions following electron impact single ionization of C_{60}^{3+} . The experimental data (open circles) are from Ref. [9], the solid line represents the present calculation.

Acknowledgments

This work was partially supported by the Österreichischer Fonds zur Förderung der Wissenschaftlichen Forschung, Wien, Austria and the EU Commission network programme, Brussels. One of us (K.B.) wishes to acknowledge partial financial support from the U.S. National Aeronautics and Space Administration (NASA) through award NAG5-8971.

References

- [1] K. Becker, "Electron-Driven Ionization and Dissociation: From Experiments under Single Collision Conditions to Processes in High-Pressure Discharge Plasmas", in Proc. XXII. International Conference on Photonic, Electronic, and Atomic Collisions (ICPEAC), Santa Fe, NM, USA (2001), AIP Press, Woodbury, NY (2002), in press
- [2] Y.-K. Kim and M.E. Rudd, *Phys. Rev. A* **50**, 3954 (1994)
- [3] W. Hwang Y.-K. Kim, and M.E. Rudd, *J. Chem. Phys.* **104**, 2956 (1996)
- [4] H. Deutsch, K. Becker, S. Matt, and T.D. Märk, *Int. J. Mass Spectrom.* **197**, 37 (2000)
- [5] W.M. Huo, V. Tarnovsky, and K. Becker, *Chem. Phys. Letters*, submitted for publication
- [6] H. Deutsch, K. Becker, J. Pittner, V. Bonacic-Koutecky, S. Matt and T.D. Märk, *J. Phys. B* **29** (1996) 5175
- [7] S. Matt, P. Scheier, T.D. Märk, and K. Becker, "Positive and Negative Ion Formation in Electron Collisions with Fullerenes", in "Novel Aspects of Electron-Molecule Collisions", edited by K. Becker, World Scientific Publishing, Seoul, South Korea (1998), p.1-69
- [8] B. Dünser, M. Lezius, P. Scheier, H. Deutsch and T.D. Märk, *Phys. Rev. Lett.* **74** (1995) 3364
- [9] R. Völpel, G. Hofmann, M. Steidl, M. Stenke, M. Schlapp, R. Trauss and E. Salzborn, *Phys. Rev. Lett.* **71** (1993) 3439
- [10] E. Brook, M.F.A. Harrison and A.C.H. Smith, *J. Phys. B* **11** (1978) 3115

List of authors in alphabetical order

Abdoul-Carime, H.	92	Brüning, F.	92
Abramzon, N.	192, 317	Buchet, J.P.	124
Albert, S.	238	Campbell, E.E.B.	86
Albu, M.	242	Cant, N.	208
Alcaraz, C.	249	Capozza, G.	265
Alexander, M.	67, 275	Caraiman, D.	262
Ascenzi, D.	71	Carré, M.	28, 124, 274
Asmis, K.R.	81	Cartechini, L.	265
Atterbury, C.	254	Casavecchia, P.	265
Aumayr, F.	101, 235, 242	Catone, D.	321
Bacchus-Montabonel, M.C.	26, 257	Cernusca, S.	101, 235
Bakunin, O.G.	260	Chandezon, F.	107
Balaj, O.P.	261	Cho, H.	159
Baloitcha, E.	26, 257	Christodoulatos, C.	170, 317
Balteanu, I.	261	Cicman, P.	269
Balzer, M.	218	Claas, P.	273
Barlow, S.	49	Clark, R.E.H.	135
Basner, R.	130	Corkum, P.	217
Bassi, D.	71	Coupier, B.	28, 274
Becker, K.	126, 130, 170, 192, 281, 300, 317, 344	Critchley, A.D.J.	254
Bergeat, A.	265	Crowe, R.	170, 317
Beyer, M.K.	261	D'Alessio, L.	223
Bohme, D.K.	55, 262	D'Amico, G.	285
Boissel, P.	163, 328	D'Ettole, A.	321
Bondybey, V.E.	261	Dalla Vedova L.	205
Buon, G.	275	Dawes, A.	141, 159
Boscaini, E.	67	de Castro Faria, N.V.	28, 274
Boudon, V.	238	de Lange, M.	275
Brabec, T.	217	Defrance, P.	138
Brand, W.A.	145	Deng, R.	82
Browning, R.	212	Denifl, S.	279, 300, 304
		Denisov, E.	49

Desouter-Lecomte, M.	26, 257	Glosik, J.	50
Deutsch, H.	130, 344	Gluch, K.	289
Dolejšek, Z.	105	Gobet, F.	28, 124
Donges, J.	120	Gohlke, S.	92
Drabbels, M.	275	Goldan, P. D.	237
Drescher, M.	217	Grabmer, W.	61, 293
Drexel, H.	38, 178, 201	Graus, M.	61, 64, 295
Duf, D.	107	Griffiths, P.	275
Dunn, K.F.	212	Grill, V.	201
Dutuit, O.	249	Gstir, B.	38, 201, 279, 300, 304
Echt, O.	82, 174	Guenther, A.	329
Eden, S.	159	Guet, C.	107
El Dakroury, A.	281	Gupta, M.C.	281
Erents, S.K.	208	Haberland, H.	120
Fall, R.	237, 309	Hagelberg, F.	75
Farizon, B.	28, 124, 274, 279, 304	Hanel, G.	28, 38, 201, 274, 279, 300, 304
Farizon, M.	28, 124, 274, 279, 304	Hansel, A.	61, 237, 293
Fedor, J.	78, 157, 201	Hansen, K.	231
Fehsenfeld, F.	309	Harper, S.M.	306
Fehsenfeld, F. C.	237	Heeren, R.M.A.	146
Fehsenfeld, F.C.	293	Heijnsbergen, D.	116
Feketeova, L.	300	Heinzmann, U.	217
Ferguson, E.	58	Helden, G.	116
Fiegele, T.	300	Heninger, M.	163, 249, 325
Filippi, A.	205, 321	Hentschel, M.	217
Flannery, M. R.	93	Herlert, A.	231
Foltin, V.	33	Herman, Z.	105, 157, 185
Fox, B.S.	261	Hermansson, K.	78
Franceschi, P.	71	Hess, W.	185
Freegarde, T.G.M.	71	Hippler, T.	120
Futrell, J.	49	Hjorth, J.	340
Gaillard, M.J.	28, 124	Hoekstra, R.	42
Gianturco, F.A.	74	Hoffmann, S.	159
Giardini, A.	205, 223, 285, 321	Hofmann, D.	47

Holtom, P.	141	Latimer, C.J.	212
Holubčik, L.	153	Le Caër, S.	325
Hu, S.WP.	306	Lebius, H.	107
Huber, B.A.	107	Lechleitner, T.	245
Huber, D.	245	Leisner, T.	107
Hübler, G.	237	Lemaire, J.	163, 328
Huels, M.A.	92	Lezius, M.	28, 157
Illenberger, E.	92, 201	Lifshitz, C.	30, 89
Ippoly, I.	33	Limão Vieira, P.	159
Issendorff, B.	120	Lindinger, C.	329
Jalbert, G.	28, 274	Lindinger, W.	64, 67, 153, 295, 309, 340
Janssen, M.H.M	196	Lukáč, P.	153
Jehn, H.	218	Mach, P.	78
Jensen, N. R.	340	Mackie, R.A.	212
Jobson, T.	309	Maier, J.P.	150
Jordan, A.	64, 295, 329	Mair C.	185
Jungwirth, P.	148	Mair, C.	157, 208, 333
Kaltsoyannis, N.	152	Maitre, P.	163
Karl, T.	309, 329	Manil, B.	107
Kendall, P.	159	Margesin, R.	166
Kennedy, R.A.	254, 314	Märk, T.	67
Kienberger, R.	217	Märk, T.D.	28, 38, 78, 124, 157, 166, 174, 178, 185, 201, 208, 269, 274, 279, 289, 300, 304, 333, 344
Kitajima, M.	159	Marotta, V.	285
Korfiatis, G.	317	Martus, K.	281
Koyanagi, G.K.	55	Martus, K.E.	192
Krausz, F.	217	Mason, N.J.	141, 159, 197, 201
Kudrna, P.	50	Matejčík, Š.	33
Kunhardt, E.	317	Matt, S.	174
Kuruczí, P.	281	Matthews, G.F.	208
Kuster, B.	309	Matt-Leubner, S.	289
Laganà, A.	321	Mauclair, G.	163
Lambert, N.	152	Mayhew, C.A.	164, 254, 314
Laroussi, M.	281		
Laskin, J.	49		

Mayr, D.	166	Quack, M.	111, 181, 188, 238
Meijer, G.	116	Raynor, T.	192
Mestdagh, H.	249, 325	Reider, G.A.	217
Meyer, M.	218	Ricatto, P.J.	170
Milosevic, N.	217	Roberts, J.M.	293
Misharin, A.S.	227, 336	Roeterdink, W.	196
Misiano, C.	218	Roithová, J.	105
Morgenstern, R.	42	Rosa, A.	92
Morva, I.	153	Rozum, I.	197
Moskwinski, L.	317	Ruckerbauer, F.	329
Mrazek, L.	242	Rümmele, M.	300
Ohanessian, G.	328	Sailer, W.	38, 178, 201
Okamoto, M.	159	Samukawa, S.	159
Orlando, S.	285	Sanche, L.	92
Ouaskit, S.	28, 274	Santagata, A.	223
Paduraru, S.	170	Satta, M.	205
Paladini, A.	205, 285, 321	Scheier, P.	28, 38, 124, 174, 178, 185, 201, 208, 274, 279, 289, 300, 304, 333, 344
Panikov, N.S.	170	Schinner, F.	166
Parajuli, R.	174, 289	Schlathölter, T.	42
Paretzke, H.	37, 329	Schlichtherle, S.	218
Parisi, G. P.	285	Schmidt, M.	120, 130
Pathak, S.	159	Schulz, C.P.	273
Pelc, A.	38, 178, 201	Schumacher, D.	273
Piccirillo, S.	205	Schustereder, W.	185, 208, 333
Plašil, R.	50	Schwarz, H.	48
Pochert, J.	181	Schweikhard, L.	231
Pollien, P.	64	Seifone, E.	74
Poterya, V.	50	Seuderi, D.	205
Price, S. D.	152	Scully, S.W.J.	212
Price, S.D.	306	Sebastianelli, F.	74
Probst, M.	78, 201	Seyfang, G.	181
Pulker, H.K.	218, 245	Shukla, A.	49
Pysanenko, A.	50		
Qayyum, A.	185, 208, 333		

Šiffalovič, P.	217	Tobias, D.J.	148
Silipo, V.	218	Tosi, P.	71
Skalny, J.D.	201, 269	Tschiersch, J.	329
Speranza, M.	205, 321	Urban, J.	78
Spielmann, Ch.	217	Vaeck, N.	26, 257
Stamatovic, A.	38, 174, 289	Vince, J.	208
Stano, M.	33, 300	Vinogradov, P.S.	227, 336
Stienkemeier, F.	273	Vogel, M.	231
Stöckl, H.	235	Volpi, G.G.	265
Stohner, J.	188	Vries, J.	42
Stolte, S.	275	Vrinceanu, D.	93
Strauss, G.N.	218	Watts, P.	164, 314
Stroud, C.	293	Wieserman, L.	317
Taatjes, C.	275	Williams, E.	309
Tanaka, H.	159	Winter, HP.	101, 235, 242
Tegeder, P.	141, 159	Winterhalter, R.	340
Teghil, R.	223	Wisse, M.	275
Tennyson, J.	197	Wisthaler, A.	61, 237, 293, 340
Tepnual, T.	333	Yan, J.	281
Tergiman, Y.S.	26, 257	Yeretzian, C.	64, 295
Thissen, R.	249	Žabka, J.	105
Thomas, R.	164, 314	Zakouřil, P.	50
Tinkle, M.	49		

Sun	Mon	Tue	Wed	Thu	Fri
RADAB Symposium 8 ⁴⁵ Opening 9 ⁰⁰ Scheier 9 ³⁰ Matejck 10 ⁰⁰ Lifshitz 10 ³⁰ Farizon 11 ⁰⁰ Coffee 11 ³⁰ Schlathöller 12 ⁰⁰ Bacchus 12 ³⁰ Paretzke Chair: Heinzmann	9 ⁰⁰ Schwarz 9 ⁴⁰ Futrell 10 ¹⁰ Glosik 10 ⁴⁰ Böhme Chair: Mayhew 16 ³⁰ Ferguson 17 ⁰⁰ Hansel 17 ³⁰ Yeretzian 18 ⁰⁰ Alexander Chair: de Puy 18 ⁴⁵ Dinner 20 ¹⁵ Poster A Chair: Stolte	9 ⁰⁰ Bassi 9 ⁴⁰ Gianturco 10 ¹⁰ Hagelberg 10 ⁴⁰ Probst Chair: Casavecchia 16 ³⁰ Asmis 17 ⁰⁰ Echt 17 ³⁰ Campbell 18 ⁰⁰ Lifshitz Chair: Matt 18 ⁴⁵ Dinner 20 ⁰⁰ Illenberger 20 ³⁰ Flannery Chair: Latimer	9 ⁰⁰ Schuch 9 ³⁰ Aumayr 10 ⁰⁰ Herman 10 ³⁰ Huber Chair: Winter 16 ³⁰ Quack 17 ¹⁰ Helden 17 ⁴⁰ Haberland 18 ¹⁰ Farizon Chair: Bieske 18 ⁴⁵ Dinner 20 ¹⁵ Poster B Chair: Dotan	9 ⁰⁰ Becker 9 ⁴⁰ Basner 10 ¹⁰ Clark 10 ⁴⁰ Defrance Chair: Giardini 16 ³⁰ Mason 17 ¹⁰ Brand 17 ⁴⁰ Coffee 18 ⁰⁰ Heeren 18 ³⁰ Jungwirth 19 ⁰⁰ Mair Chair: Price 20 ⁰⁰ Conference Dinner	Breakfast Departure
Informal discussions					

