



The interactions of high-energy, highly charged Xe ions with fullerenes

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Abstract

Ionization and fragmentation have been measured for C_{60} molecules bombarded in the vapor phase by Xe³⁵⁺ and Xe¹⁸⁺ ions with energies in the range 420–625 MeV. The CM energies exceeded those used in previous studies by several orders of magnitude. We present the observed mass distribution of positively charged fragments together with a theoretical model indicating that the total interaction cross section contains roughly equal contributions from (a) excitation of the giant plasmon resonance, and (b) large-energy-transfer processes that lead to multiple fragmentation of the molecule.

In recent experiments performed at Argonne's ATLAS heavy-ion linear accelerator, the interactions of high-energy (up to 625 MeV), highly charged (up to 35 +) Xe ions with C₆₀ have been studied. The center-of-mass energies exceeded those used in previous work by several orders of magnitude. The high values of projectile velocity and charge state result in excitation and decay processes differing significantly from those seen in studies at lower energies [1,2].

The C_{60} vapor target was formed from 99.5% pure material heated in an oven to 475°C. A time-of-flight (TOF) spectrometer system (see Fig. 1) was located at 90° to the incident beam. Grids around the target region were biased with voltages to extract positively charged fragments and to inject them into a 20-cm-long gridded flight tube and thence into a micro-channel-plate detector. A "beam sweeper" at ATLAS was employed so as to allow one 0.4-ns-wide beam pulse to reach the target every 10 μ s. TOF spectra were obtained using a "multi-hit" time digitizer with the "start" signal coming from the detector and the "stop" signal from the accelerator's timing system.

Fig. 2 shows the TOF spectrum and its equivalent calibration in terms of M/Q, the ratio of fragment mass to charge. The peaks in Fig. 2 that correspond to interactions of the projectiles with C₆₀ fall into three categories:

1) Peaks due to singly, doubly, triply, and (possibly) quadruply ionized C_{60} .

2) Peaks corresponding to the losses of even-numbered neutral carbon fragments.

3) Peaks corresponding to the sequence of singly charged fragments C_n^+ , with n assuming all values from 1 to at least 19. These peaks alternate in intensity up to around n = 9 with the odd-numbered peaks being more intense than the even-numbered. Above n = 9, the most intense peaks appear to be n = 11, 15, and probably 19. The intensity variations mirror those seen in other studies and indicate the relative stabilities of linear chain ($n \leq 9$) and cyclic ($n \geq 9$) structures [3]. We refer to this series of peaks, C_n^+ , as the "multifragmentation" peaks since they arise predominantly from events in which there is a catastrophic disintegration of the C_{60} molecule into many small fragments.

The manner in which energy is coupled into the C_{60} system from the passage of a highly charged fast ion (of



Fig. 1. Schematic diagram of experimental arrangement.

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Fig. 2. Time-of-flight spectrum for positive fragments arising from bombardment of C_{60} by 625-MeV ¹³⁶Xe³⁵⁺ ions. The numbers given above some of the peaks are the ratios (M/Q) of fragment mass (amu) to charge.

velocity, v) depends strongly on the impact parameter. The two principal distances of importance are the mean radius, \overline{R} , (3.55 Å) of the C₆₀ "cage" on which are located the nuclei of the constituent carbon atoms, and the adiabatic distance, $b_0 = \gamma hv / E$ (= 10 Å for E = 20 eV), for the excitation of the giant dipole plasmon resonance of energy *E*. This collective excitation of the 240 valence electrons of the C₆₀ molecule has been predicted [4] and measured [5,6] to have an energy of 20 eV and a FWHM of about 10



Fig. 3. (a) Calculated probabilities [7] for the total interaction and for single-plasmon excitation as functions of the impact parameter, *b*, for 625-MeV 136 Xe³⁵⁺ ions. (b) Calculated single-plasmon cross section [7] (shown as a line) compared to the measured yields of C⁺₆₀ from C₆₀ bombarded by 136 Xe¹⁸⁺ ions in the energy range 420–625 MeV.

eV. We have developed a quasi-classical mode [7] for the interaction between projectile and buckyball that gives the total excitation and single-plasmon excitation probabilities as a function of impact parameter. For 625-MeV ¹³⁶Xe³⁵⁺ ions, the corresponding cross sections are $\sigma_{tot} = 811$ Å² and $\sigma_{1pl} = 387$ Å² (about 50% of the total cross section). The dominant decay mode of the single-plasmon excitation is thought to be via electron emission [8]. We therefore compare the calculated single-plasmon cross section to our measured C⁺₆₀ yield and find that the calculations reproduce well the (weak) energy dependance displayed by the measurements (Fig. 3).

At impact parameters less than about 7 Å where the energy deposition becomes large, essentially all projectile/target interactions will result in multifragmentation. We have constructed a bond-percolation model [7] to describe these fragmentation processes. C_{60} is represented



Fig. 4. Measured mass distribution (solid points) for positive fragments arising from C_{60} bombarded by 625-MeV $^{136}Xe^{35+}$ ions. The histogram is the distribution calculated on the basis of a percolation-multifragmentation model [7].

as a collection of lattice sites located at the positions of the carbon atoms. Each site is connected to its three nearest neighbors via bonds. We assume that each xenon ion deposits excitation energy in proportion to its pathlength through the hollow fullerene structure. The energy is then rapidly distributed in a uniform manner over the whole C_{60} -cluster. This leads to the breaking of individual bonds with a probability proportional to the total energy deposition, which in turn, is dependent on the impact parameter. The fragment-mass distribution calculated using this model compares well with the measured fragment mass spectrum (Fig. 4).

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References

- For reviews see, for example, E.E.B. Campbell et al, in: Nuclear Physics Concepts in Atomic Cluster Physics (Springer, Berlin, 1992); Proc. XVIII Int. Conf. on the Physics of Electronic and Atomic Collisions, Aarhus, July 21, 1993.
- [2] B. Walch, C.L. Cocke, R. Voelpel and E. Salzborn, Phys. Rev. Lett. 721 (1994) 1439.
- [3] See, for example, G. von Helden, M.-T. Hsu, P.R. Kemper and M.T. Bowers, J. Chem. Phys. 95 (1991) 3835.
- [4] G.F. Bertsch, A. Bulgac, D. Tomanek and Y. Wang, Phys. Rev. Lett. 67 (1991) 2690.
- [5] R.K. Yoo, B. Ruscic and J. Berkowitz, J. Chem. Phys. 96 (1992) 911.
- [6] I.V. Hertel, H. Steger, J. de Vries, B. Weisser, C. Menzel, B. Kamke and W. Kamke, Phys. Rev. Lett. 68 (1992) 784.
- [7] T. LeBrun, H.G. Berry, S. Cheng, R.W. Dunford, H. Esbensen, D.S. Gemmell and E.P. Kanter, Phys. Rev. Lett. 72 (1994) 3965.
- [8] T. Drewello, W. Krätschmer, M. Fieber-Erdman and A. Ding, Int. J. Mass Spectrosc. Ion Process. 124 (1993) R1.