Observation of Molecular Fusion and Deep Inelastic Scattering in $C_{60}^+ + C_{60}$ Collisions

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Direct experimental evidence is obtained for the occurrence of fusion in collisions between two C_{60} molecules at collision energies of 200 to 500 eV in the center-of-mass frame of reference. The C_{120}^+ formed at 200 and 250 eV collision energy has a lifetime of more than 70 μs with high internal energy. Results are in good qualitative agreement with recent molecular dynamics simulations combined with density functional theory.

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Buckminsterfullerene (C_{60}) has proved to be a fascinating object of research since the discovery by Krätschmer *et al.* of a method of easily producing macroscopic amounts of a mixture of C_{60} and C_{70} [1]. It has properties which very conveniently "bridge the gap" between the molecular gas phase and the solid state—one of the main aims of cluster science—which makes it an ideal object for cluster research. For the first time large amounts of a *neutral mass-selected atomic cluster with a well-defined structure* are available opening up the possibility of new kinds of cluster experiments. One exciting new development in cluster physics is the field of cluster collisions [2]. Recent molecular dynamics calculations combined with density functional theory have been used to simulate collisions between small alkali metal clusters [3–5]. The main reaction channels were found to be fusion, deep inelastic and quasielastic scattering in close analogy to nuclear heavy-ion collisions. It would thus seem that not only can similar concepts be used to describe the structure of atomic clusters and atomic nuclei as, e.g., in the shell model [6] but also that similar concepts may be applied for understanding dynamical features. Investigations of such processes should provide a valuable stimulus for both cluster and nuclear physics and uncover interesting new phenomena. The problem with the first system chosen for the calculations ($Na_x + Na_x$) is that it is not yet experimentally possible to produce two mass-selected beams of metal clusters with sufficient intensity to carry out collision experiments with. A comparison with the calculations will therefore have to wait until the experimental techniques have advanced sufficiently to make such experiments feasible. The perfect clusters for testing these concepts are the fullerenes with which it is possible to have high intensities of projectile and target and where the structures are well known. Such experiments with the fullerenes have the additional attraction of being able to deliver some insight into the growth mechanisms of the larger fullerenes. In the present Letter we report direct experimental evidence for the occurrence of fusion in cluster-cluster collisions. We also show that the concept of deep inelastic collisions can explain the structure observed in the mass spectra. These are collisions in which two deformed and highly excited fragments, which preserve the identity of the target and projectile, are seen in the exit channel. The experimental results are in good qualitative agreement with molecular dynamics simulations which have very recently become available for this system [3,4].

A schematic diagram of the experiment is shown in Fig. 1. A C_{60}^+ beam is produced by low fluence uv laser desorption of commercially available C_{60}/C_{70} deposited on a rotating metal plate. Positively charged C_{60}^+ and C_{70}^+ ions are produced by the laser with very little fragmentation occurring [7] and are accelerated away from the surface by a constant electric field. C_{60}^+ is mass selected, by using a pulsed electric field to remove C_{70}^+ and any other ions that may be present from the flight path, and enters the collision chamber. This is an oven containing the C_{60}/C_{70} commercial fullerene extract which can be heated to a temperature of about 600 °C. After the collision the C_{60}^+ and product ions are mass selected in a reflectron time-of-flight mass spectrometer and detected by two channel plates operated in the Chevron configuration. The signal is read into a transient recorder and signal averager with transfer to a computer via a CAMAC interface. The laboratory collision energies in the experiments reported here were from 400 to 1000 eV with an energy spread of $\approx 1\%$. A retarding field energy analyzer placed immediately before the channel plates can be used to determine the energy of parent and product ions.

A mass spectrum produced at a collision energy of 200

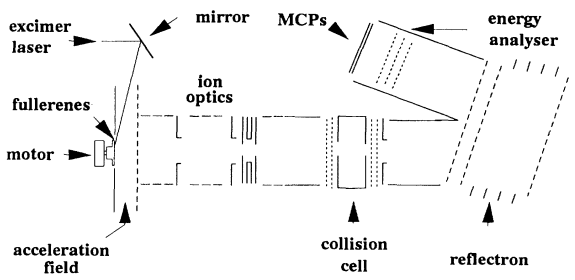

FIG. 1. Schematic diagram of apparatus.

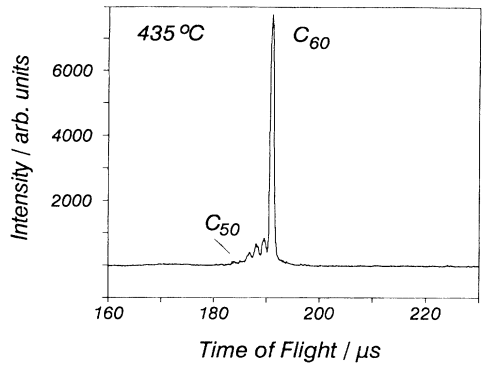

FIG. 2. Mass spectrum produced in $C_{60}^+ + C_{60}$ collisions at 200 eV c.m. collision energy. Target pressure ca. 7×10^{-5} mbar.

eV center of mass (c.m.) is shown in Fig. 2. The temperature of the collision chamber was 435 °C which corresponds to a C_{60} vapor pressure of approximately 7×10^{-5} mbar [8] and is in the range where single collision conditions are expected. The fragmentation observed is much less than that seen for collisions with rare gases or small molecules at equivalent collision energies [9] which is presumably mainly due to the much larger scattering angles to be expected in the present system. The reflectron was adjusted to detect product ions with laboratory scattering angles in the range $0° \pm 3°$. Another possible reason for the increased fragmentation observed with rare gases is that penetration of the carbon cage by the rare gas target facilitates fission of the C_{60}, i.e., breakup into a small number of large fragments rather than statistical evaporation of C_2 molecules [10].

The same mass spectrum is shown with an expanded intensity scale in Fig. 3(a). Here it can be clearly seen that species with masses greater than C_{60} are present. In order to interpret the mass spectrum, simulations of the flight times of different product ions were carried out. The lines at the foot of the spectrum give the calculated arrival times assuming collision occurs in the center of the collision cell. The two prominent peaks that appear at the longest flight times (> 220 μs) are seen to be due to fusion of C_{60}^+ with either C_{60} or C_{70} which is also present in the collision cell. The flight time was calculated assuming that the collision is completely inelastic, i.e., that all the available center-of-mass energy is converted into internal energy or rotational energy of the product, as would be expected from considering conservation of linear momentum. These species survive at least until entry into the reflectron which gives a minimum lifetime of ca. 70 μs. The peaks for C_{120}^+ and C_{130}^+ are of approximately equal intensity although the vapor pressure of C_{70} in the collision cell is about a factor of 2 less than that of C_{60} [8] which indicates that the cross section is considerably larger for fusion with C_{70} at this collision energy. The other lines lying between the C_{60}^+ parent and C_{120}^+

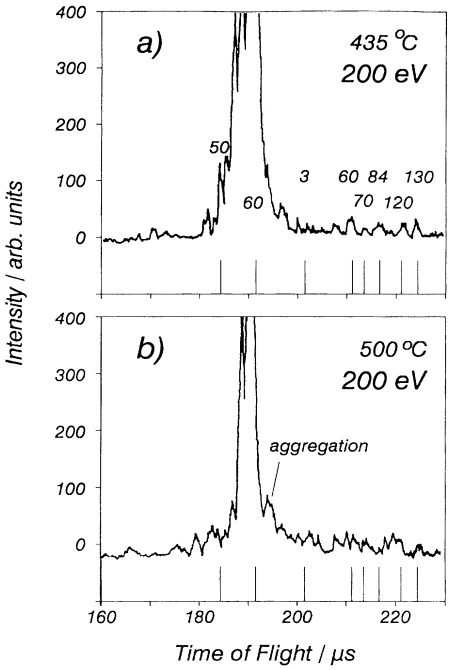

FIG. 3. (a) Spectrum shown in Fig. 2 with expanded intensity scale. The numbers n and lines refer to simulated flight times of C_n^+ species (see text for details). (b) Mass spectrum obtained with a target pressure of 7×10^{-4} mbar, showing the effect of multiple collisions and aggregation of C_{60}^+ with small neutral fragments.

are positions calculated for fragments from excited C_{120}^+ which decays in the field free region between the collision cell and the reflectron. Peaks corresponding to C_{60}^+ and C_{70}^+ are clearly seen. The structure appearing at C_{84}^+ is more probably due to fragmentation to species around C_{60}^+ and C_{70}^+ from C_{130}^+.

Molecular dynamics simulations combined with density functional theory have recently been carried out for this system at collision energies of 150 and 500 eV c.m. and impact parameter $b = 0$ [3,4]. They show that practically all of the incident c.m. kinetic energy is lost and transferred into potential (or deformation) energy and intrinsic "heat" energy (deep inelastic collisions). At 150 eV the deformed clusters rebound from the collision retaining the sixty carbon atoms and then undergo shape relaxation which will gradually increase the internal energy leading to sequential evaporation of C_2. At 500 eV the fusion barrier can be overcome and the system evolves towards a common large cluster, completely losing the geometry of the original clusters. The C_{120} then deexcites by the emission of small species, e.g., C_2 and C_4. Further molecular dynamics simulations using phenomenological two- and three-body forces [11] have shown that at these high energies the C_{120}^+ cluster can also fragment into two large fragments which essentially retain the atoms present in the target and projectile, respectively (although

not the geometry), and some small fragments containing atoms from both collision partners.

The experimental results show that the fusion barrier must lie below 200 eV and give strong evidence for the process in which two large fragments are formed from the highly excited large cluster. The mass spectrum shown in Fig. 3(b) was obtained with a temperature of 500 °C in the collision cell corresponding to a vapor pressure of approximately 7×10^{-4} mbar [8] where multiple collisions will play an important role. The fragments from C_{60}^+, which arrive at shorter times than the parent peak, have been to a large extent scattered out of the beam path by further collisions and the larger masses from the fusion process have been "smeared out" and can no longer be resolved. Under such conditions collisions between C_{60}^+ and small neutral fragments can occur in the collision chamber leading to aggregation and growth of larger fullerenes from C_{60}^+. Such aggregation processes have been observed in laser desorption experiments but only with C_{70} not with C_{60} [7]. One may have to overcome a barrier for growth of C_{60} and there is not sufficient relative kinetic energy in the laser desorption experiments for this to be possible. Alternatively, the internal energy of the relatively unstable products (C_{61}^+, C_{62}^+, etc.) may be too high for them to survive long enough to be detected in the mass spectrometer. In very nice collision experiments carried out by Anderson and his group at energies of a few eV, pickup of C^+ by C_{60} can be observed to form C_{61}^+ [12]. In recent work reported by Yeretzian *et al.* [13] the laser desorption experiments have been extended by using a high laser fluence to desorb pure C_{60} under a supersonic helium flow. Under these conditions beautiful mass spectra are obtained showing coalescence of C_{60} to produce a broad distribution of carbon clusters with peaks at C_{118}, C_{178}, and C_{236}. The conditions in the experiment of Yeretzian *et al.* are considerably different from those in the single collision experiments described in this Letter. A pulse of very hot, dense vapor is produced by the laser desorption of the fullerene film and confined in a flow channel thus increasing the frequency of collisions between the highly internally excited fullerenes. The He gas serves to rapidly cool the products of these multiple collisions on expansion into the vacuum. The fragmentation observed, which looks very like successive statistical evaporation of C_2 molecules, is strongly dependent on the internal energy of the fullerenes. In the experiments presented here which are single collisions carried out under well-defined conditions the products do not depend so much on the internal energy of the species involved but are mainly determined by the dynamics of the collision process. Our predominant fragmentation channel is thus seen to be fission of C_{120}^+ (or C_{130}^+) although statistical evaporation will also be occurring to a certain extent and is likely to become more important with increasing collision energy.

Figure 4 shows mass spectra taken at collision energies of 250 and 500 eV c.m. At 250 eV [Fig. 4(a)] the results

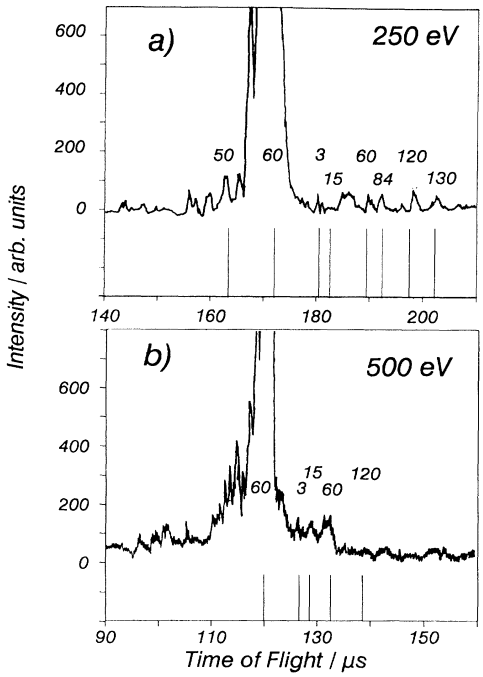

FIG. 4. As in Fig. 3(a), single collision conditions but for (a) 250 eV and (b) 500 eV c.m.

are similar to 200 eV with clear peaks corresponding to C_{120}^+ and C_{130}^+ and also fragments at around C_{60}^+. In addition, smaller, unresolved fragments can be observed between C_{30}^+ and C_{60}^+, with possible contributions from doubly charged species, and there is evidence for a C_3^+ fragment. At a collision energy of 500 eV [Fig. 4(b)] there is no longer any convincing evidence for stable C_{120}^+ or C_{130}^+. The internal energy is too high and they rapidly fragment, in agreement with the molecular dynamics simulations. A strong fragment peak is seen for masses in the range C_{50}^+ to C_{70}^+ with additional fragments around C_{15}^+ (strongly reminiscent of the bimodal fragment distribution seen in the collision fragmentation [9] and photofragmentation [14] of C_{60}^+) and C_3^+.

These first experiments in the field of cluster-cluster collisions have shown that the concepts developed for an understanding of the dynamics of nuclear collisions can be applied to cluster collisions. They also show that such experiments have potential for obtaining an understanding of the growth mechanisms of the fullerenes. More detailed investigations involving the collision energy and scattering angle dependence of the products are underway.

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