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Collisionally induced multifragmentation of C_{60}

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The inclusive yield distribution of C_n^+ fragments from 45 keV C_{60}^- collisions with Ar gas, exhibits comparable yields for fragments with n in the 10–25 range and for fullerene fragments with n in the 40–58 range. Single-collision two-fragment coincidence measurements show a high probablity for formation of two fragments with similar size in the $n=15$–25 range. Three C_n^+ fragments each with $n>2$, have been observed in triple coincidence measurements. An enhancement of the yield of the lightest fragments with an odd number of carbons is observed.

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I. INTRODUCTION

When C_{60} is excited to energies of several hundred eV, a broad spectrum of products are observed, including all even-n fragments from 32 to 58, and both even and odd fragments for $n=1$ to 30 [1–5]. Typically the yield of even-n fragments, falls off nearly monotonically as n decreases, reflecting largely the deposition energy spectrum for either collisional excitation or multiphoton absorption [6]. These even-n fragments for n larger than 30, result from a mixture of sequential C_2 and longer even-n C_n fragment emissions [7–9,5] and are thought to have a fullerene structure. Some of the light fragments in the $n=1$ to roughly $n=10$ range are either the primary even-n fragments from the fullerene breakup, or their sequential decay products. The dissociation energies for even-n fragments in this mass range favor sequential breakup into two odd-n fragments, accounting for the presence in the fragment distribution of odd-n fragments as partners to even-n fullerene residues. Much of the fragment yield in the $n=10$ to 30 size range, cannot be due to collisions where only two $n>2$ fragments, of which one is a fullerene, are produced. A likely mechanism for producing fragments in this size range is multifragmentation, where three or more $n>2$ fragments are produced. This process may be analogous to multifragmentation in nuclear reactions [10,11] at intermediate (on a nuclear energy scale) bombarding energies. Previous studies [12] of multifragmentation of C_{60} have shown that its competition with evaporation and asymmetrical fission increases with the charge state and the excitation energy. The results are presented, to our knowledge, for the first time, in this paper, where three coincident fragments, each with $n>2$, are observed.

Multifragmentation of H_{25}, at a projectile velocity much higher than that in the present experiment, has been studied by Farizon *et al*. [13,14]. The scaled results exhibit a pattern [13] very similar to a percolation model, which also provides a reasonable account of nuclear fragmentation of Au nuclei at 1 GeV/amu. The H_{25} fragmentation results have also been interpreted [13] as evidence for critical behavior reminiscent to a second-order phase transition in an infinite system.

II. EXPERIMENT

The experimental apparatus used in this present experiment is similar to that used in our previously described coincidence experiment [5]. Briefly, we use reverse kinematics, bombarding an argon gas target with a beam of C_{60}^- ions. Because of the heavy projectile incident on a light target, all of the reaction products are emitted in a narrow cone in the forward direction. The energy of a particular fragment is proportional to the number of carbons in the fragment. We analyze the positive ions in electrostatic deflectors with Channeltron (Burle 4839) detectors whose signals are observed in a pulse mode. To examine coincidences between fragments of similar size, we use a single deflector followed by two Channeltrons side by side in the horizontal plane of the deflector. The deflector plates are sufficiently far apart that fragments differing by one or two carbons can be transmitted simultaneously through the deflector. The apertures for the two channeltron detectors have to be displaced by the appropriate amount for the two sizes one wishes to study. This displacement is typically between 1 and 2 cm.

To study coincidences between three fragments, two of which are of comparable size, a second electrostatic deflector is inserted upstream of the deflector with dual detectors. The additional deflector is used to analyze fragments of appreciably smaller size (lower electricstatic rigidity) than the other two. It is operated at lower electrostatic fields and has an exit slot, which allows the larger, more electrostatically rigid, fragments to pass on to the second deflector. Fast timing signals are derived from each of the three Channeltron detectors. One of the signals from the dual detectors is used to start a time-to-amplitude converter, and the other (after appropriate delay) to stop the converter. Fragments from the same collision show up as a prompt peak in the time difference spectrum, while accidental coincidences are uniformly distributed in time. The correction for accidental coincidences between these two detectors was typically about 25%. The full width at half-maximum of the prompt peak is typically 0.2 μs. The ratio of the net area of the prompt peak to the number of pulses from the start detector is proportional to the probability that the fragment observed in the start detector is accompanied by a fragment of the size accepted by the stop detector. For coincidences between three fragments, a second time-to-amplitude converter is started by the third detector associated with the upstream deflector and stopped by one of the stop detectors of the dual deflector. A window is put on the prompt peak from this time difference spectrum. This is used as a gate on the time difference spectrum be-

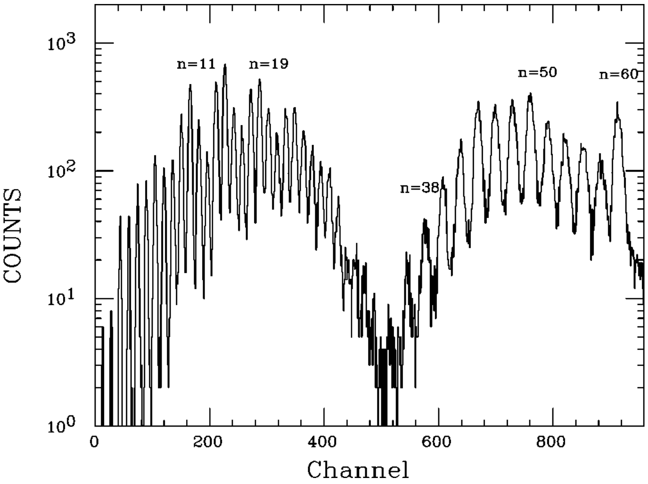

FIG. 1. Yield distribution of positively charged fragments for 45 keV C_{60} on Ar as a function of electrostatic rigidity. For singly charged ions, this is proportional to the number of atoms n in the cluster. Some typical n values for singly charged ions are indicated.

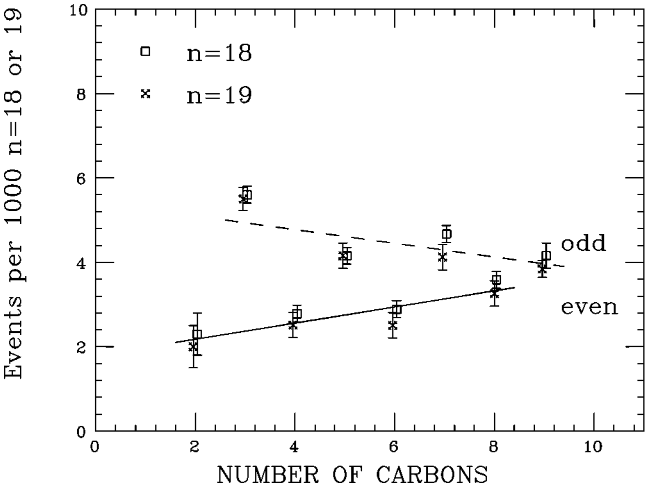

FIG. 2. Relative yield of a lighter coincident fragment when either an $n=18$ or $n=19$ fragment is detected.

tween the dual detectors. To determine the accidental rate the window is moved from the prompt peak to a region of the time difference spectrum containing only accidental coincidences. This accidental subtraction is typically less than 20%.

III. RESULTS

A. Inclusive singles distribution

We give an overview of the inclusive (singles) yields of positively charged ions in Fig. 1. Note the logarithmic scale. The yield distribution in Fig. 1 is in good agreement with that reported for the same system at 50 keV bombarding energy by Larsen *et al.* [15]. Comparison of the neutral yield distributions of Lykke [16] with previously reported distributions for positively charged species, suggests our positive-ion yield distribution is qualitatively representative of the total yield distribution. Our apparent relative yields for low n are affected by the detection efficiency [5]. The most important observation regarding the lighter fragments (below $n=30$) is that the yields of both even and odd-n fragments are comparable, in distinction to the results for the heavy fragments. This is indicative that these fragments do not have a fullerene structure.

The maximum energy available in the center-of-mass system in the present experiment is 2368 eV. It is interesting to contrast the yield distribution in the present experiment with that where the excitation energy is an order of magnitude lower [5]. At the lower bombarding energy the yield of even-n fullerene fragments fell off exponentially from the maximum yield for $n=58$. In the present experiment, the maximum yield for fullerene fragments was for $n=44$ to $n=50$. This shift to highest yields for lower n reflects the increase in the most probable energy deposition with increasing energy available in the center-of-mass system. At the lower bombarding energy the yield of fullerene fragments considerably exceeded the yield of fragments in the $n=10$ to 25 size range, whereas at the present higher energy the yield of fragments in the two size ranges are comparable.

B. Two-fragment coincidence (doubles) results

We first present the limited two-fragment coincidence results for the dual detector deflector. This deflector is only capable of transmitting pairs of fragments close in mass. For $n=19$, we find about a 1.5% probability for detecting a coincident $n=18$ fragment. For $n=23$, we find that the probability for detecting the adjacent mass $n=22$ fragment has dropped to 0.3%. For $n=27$, the probability for detecting a coincident $n=26$ fragment has dropped furthur to 0.07%.

We have more extensive two-fragment coincidence results for the probability of detecting a light fragment in coincidence with a heavy fragment. Some of these results are illustrated in Fig. 2, where we show the probability of detecting different light fragments in coincidence with $n=18$ or $n=19$ fragments. There is a kinematic effect that reduces the detection efficiency for the lightest fragments when using reverse kinematics, as in the present experiment. This effect is most easily recognized for binary breakup, where conservation of momentum requires the light fragment to have a higher velocity than its heavier partner. This leads to a larger angular spread of the lightest fragments and a reduced detection efficiency. This was modeled in our previous experiment and an efficiency given approximately by $(0.9\text{–}1.1/n)$ was found. It is not possible to model quantitatively the present experiment as typically more than three fragments are formed, some of which may result from prompt multifragmentation and some which may result from the sequential decay of binary fragmentations. We have, however, found that at the present bombarding energy, when two coincident fragments are detected, that the above efficiency is approximately correct if the multiplicity of the lightest fragments varies weakly with n. In the absence of better information, we have corrected both the twofold and threefold light-fragment yield by the aforementioned factor. The uncertainty in this correction does not compromise the odd-even dependence of the yields or the comparison between yields for

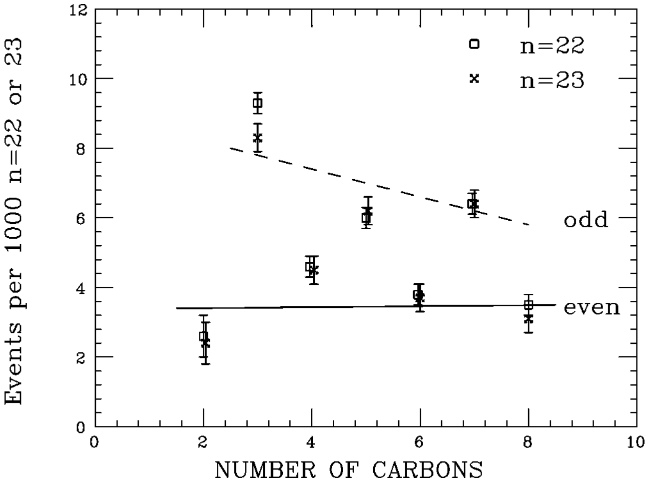

FIG. 3. Relative yield of a lighter coincident fragment when either an $n=22$ or $n=23$ fragment is detected.

different heavy-fragment partners.

The relative yields of light fragments per heavy fragment are nearly identical for $n=18$ and $n=19$. This result is independent of the detector efficiency. It is also found that it is more probable to emit a light fragment with an odd n value than an even n value. Similar results for the probability for observing a particular light fragment, in coincidence with $n=22$ or $n=23$ fragments, are shown in Fig. 3.

C. Three-fragment coincidence (triples) results

We now go on to present the results where three fragments from a single collision were observed. Figure 4 shows the relative yields of the lightest fragment observed in events in which both an $n=18$ and an $n=19$ fragment was detected. Figure 5 shows similar data for the case in which both an $n=22$ and an $n=23$ fragment was detected. Since the sum of the three n values never reaches 60, then for the triple events observed, there must have been at least a fourth fragment emitted.

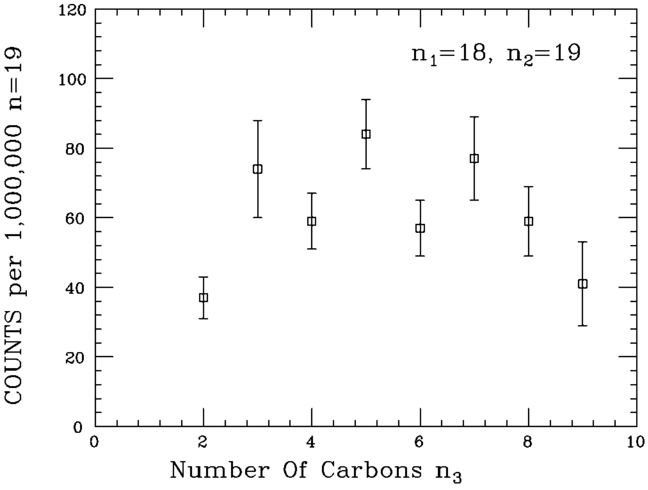

FIG. 4. Relative yield of a lighter third coincident fragment when both an $n=18$ and $n=19$ fragment has also been detected.

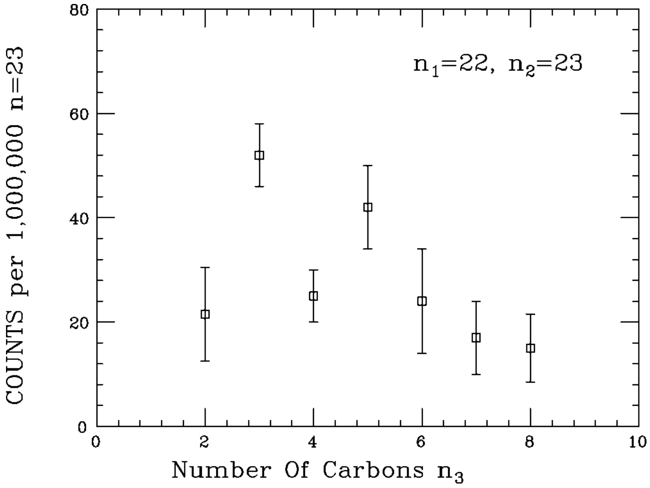

FIG. 5. Relative yield of a lighter third coincident fragment when both an $n=22$ and $n=23$ fragment has also been detected.

IV. DISCUSSION

Larsen *et al.* [15] have discussed the singles yield distribution of the fullerene products with $n>30$ in terms of a simple model of the sort used in stopping power calculations. The projectile undergoes elastic collisions with the individual carbon atoms of the target. Both prompt knockout and delayed emission of C_2 units are considered. No attempt to describe the yield of fragments with $n<30$ was attempted.

We turn now to a discussion of our coincidence results. The yields of light fragments in both the experiments where one or where two heavier partner fragments were detected, exhibit an enhancement of the odd number carbon clusters. This cannot arise from effects based on the conservation of the total number of carbons, as in no case does the sum of the observed number of carbons in all fragments reach that of the initial C_{60}. Furthurmore the effect in the doubles results is independent of whether the heavy partner is odd or even. The observed effect probably results from energetic considerations on the sequential decay of heavier primary fragments. Both for chains and rings in the $n=4$–10 range (and perhaps for larger n for which energetics data are not available) the energy required for binary fragmentation of even-n clusters favors breakup into two odd-n fragments [17,18]. In all cases the most energetically favored breakup split is the one in which a C_3 is formed.

A comparision of the n dependence of the light-fragment yield for the triples data shows a steeper falloff with n when an $n=22$ and $n=23$ pair (Fig. 5) is in coincidence as compared to an $n=18$ and $n=19$ pair (Fig. 4). This is to be expected; as for the latter pair, 8 less carbons are available for lighter fragments. There is some hint of a similar falloff for the doubles data, but less of an effect would be expected when less than half of the available carbons are detected.

If one assumes our results for fragments in the $n=18$–23 range are representative of fragments in the intermediate mass range between 15 and 25, one can estimate that the probability for a positively charged second fragment in this mass range is of the order of 25%. Similarly, for the three-particle results, one can estimate that when two positively charged fragments in the intermediate mass range are produced, there is a probability of about 5% that there will be a third light positively charged fragment with $n>2$ produced in the collision. Since it is likely that there are a significant number of events where one of the three fragments is neutral rather than positively charged, one can conclude that multifragmentation into three particles heavier than the carbon dimer is an important reaction channel at this collision energy.

The results we have obtained are consistent with the following scenario. For impact parameters which result in modest excitation energy deposition (50–200 eV) the excited fullerene evaporates C_2 (and sometimes longer chains such as C_4, C_6, ... [5]) giving rise to the even-*n* peaks for $n>32$ in Fig. 1. For larger energy deposition, the excited fullerene often multifragments into three or more fragments with $n>2$. The multifragmentation probability is expected to increase fairly rapidly with excitation energy [19], so that generally multifragmentation precedes C_2 evaporation. This is consistent with the absence of an unusually large contribution of C_2 in the lighter-fragment size distribution coincident with two heavier multifragmentation products. Often the multifragmentation primary products have considerable excitation energy and break up sequentially into still lighter fragments. This is consistent with the observation that the sum of the *n* values of the three observed coincident fragments is less than 60. The relative yields of the different breakup channels are probably governed by statistical considerations [6] as evidenced by the enhancement of energetically favored odd-*n* lighter fragments.

It would be interesting to have available molecular-dynamics calculations [20,21] for this system. In order to compare these with experiment, it would be necessary to consider the secondary decay [1,6] of the excited primary fragmentation products.

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