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THE INTERACTIONS OF HIGH-ENERGY, HIGHLY-CHARGED IONS WITH FULLERENES

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In 1985, Robert Curl and Richard Smalley[1] discovered a new form of carbon, the fullerene, C_{60}, which consists of 60 carbon atoms in a closed cage resembling a soccer ball. In 1990, Krätschmer et al. were able to make macroscopic quantities of fullerenes[2]. This has generated intense activity to study the properties of fullerenes. One area of research involves collisions between fullerenes and atoms, ions or electrons[3,4]. In this paper we describe experiments involving interactions between fullerenes and highly charged ions in which the center-of-mass energies exceed those used in other 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[3]. Our results are discussed in terms of theoretical models analogous to those used in nuclear physics and this provides an interesting demonstration of the unity of physics.

In our experiments, 420-625 MeV beams of $^{136}Xe^{35+}$ or $^{136}Xe^{18+}$ ions from Argonne's ATLAS accelerator were incident on a C_{60} vapor target formed from 99.5% pure C_{60} heated to 475 ^{0}C in a two-stage stainless-steel oven. The target had a density of $\sim 10^{10}$molecules/cm^3 in the interaction region. A time-of-flight (TOF) spectrometer system was located at 90° to the ion beam. Grids around the target region were biased with voltages to extract positively charged fragments. The fragments passed through a 20-cm-long gridded flight tube to a micro-channel-plate detector. The total acceleration voltage was 6.9 kV. A "beam sweeper" allowed one 0.4 ns-wide beam pulse to reach the target every 10 μs. TOF spectra were obtained using a "multi-hit" time digitizer with the "start" signal coming from the detector and the "stop" signal from

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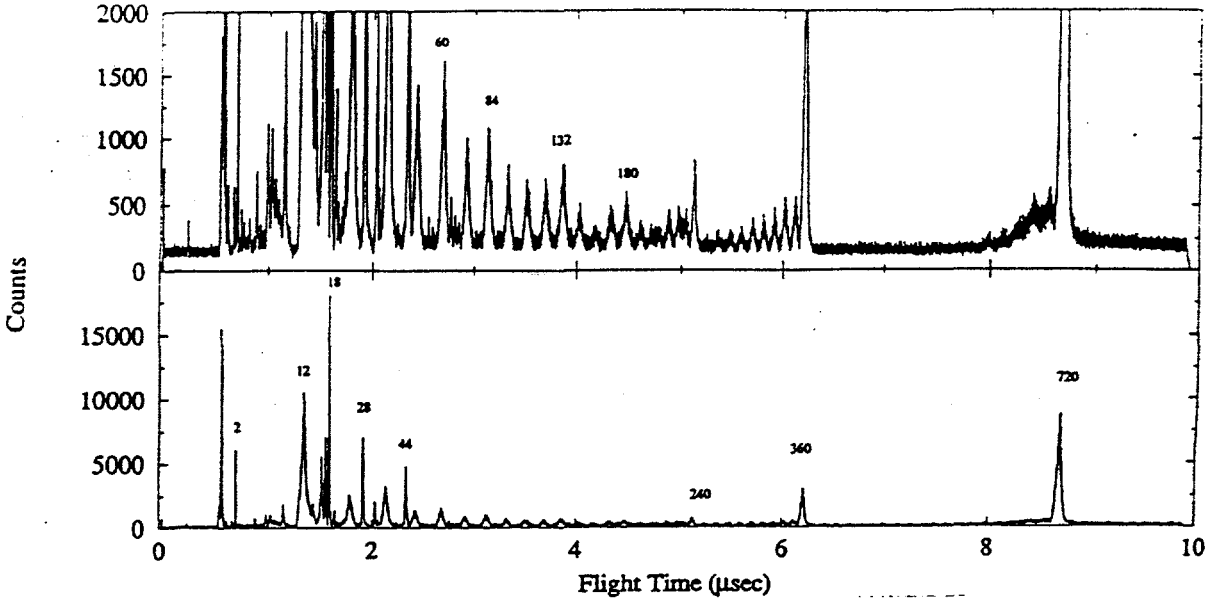

Figure 1. Time-of-flight spectrum for bombardment of C_{60} by 625 MeV $^{136}Xe^{35+}$ions. The numbers labeling the peaks give the calibration in terms of M/Q, the ratio of fragment mass (amu) to charge.

the accelerator's timing system.

Fig. 1 shows a TOF spectrum for bombardment of C_{60} by 625 MeV $^{136}Xe^{35+}$ions. The numbers labeling the peaks give the calibration in terms of M/Q, the ratio of fragment mass (in amu) to charge. This spectrum is the sum of all eight channels in the time digitizer and thus reflects all positive fragments detected. Clusters ranging from single carbon atoms up to C_{60}^+ are observed. The TOF spectrum also exhibits peaks attributable to light background gases such as H_2O, N_2, O_2, and CO_2.

The peaks in Fig. 1 that correspond to interactions of the projectiles with C_{60} fall into three categories:

(1) Peaks due to singly, doubly, triply, and (possibly) quadruply ionized C_{60}. These "parent" peaks decrease in relative intensity towards higher charge states. Their narrowness reflects the small kinetic energy releases involved in the generation of these ions.

(2) Even-numbered high mass peaks (ENHM) corresponding to the loss of even numbers of carbon atoms. Interestingly, these ENHM peaks (C_{58}^{q+}, C_{56}^{q+}, C_{54}^{q+}, etc.) are stronger relative to their parent peaks for the higher charge states.

(3) Peaks corresponding to the sequence of singly charged fragments C_n^+, with n assuming all values from 1 to at least 19 (higher values then become indistinguishable from the multiply charged pair-loss peaks). 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. These intensity variations mirror those seen in other studies[5,6]. We refer to this series of peaks, C_n^+ as the "multifragmentation" peaks since we believe (see below) that they arise predominantly from events in which there is a catastrophic disintegration of the 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 velocity v) can be expected to depend strongly on the impact parameter. The two principal distances of importance in discussing impact parameters are the mean radius, R (known[2] to be 3.55 Å) of the C_{60} "cage" on which are located the nuclei of the constituent carbon atoms, and the adiabatic distance[7] $b_0 = \gamma\hbar v/E$

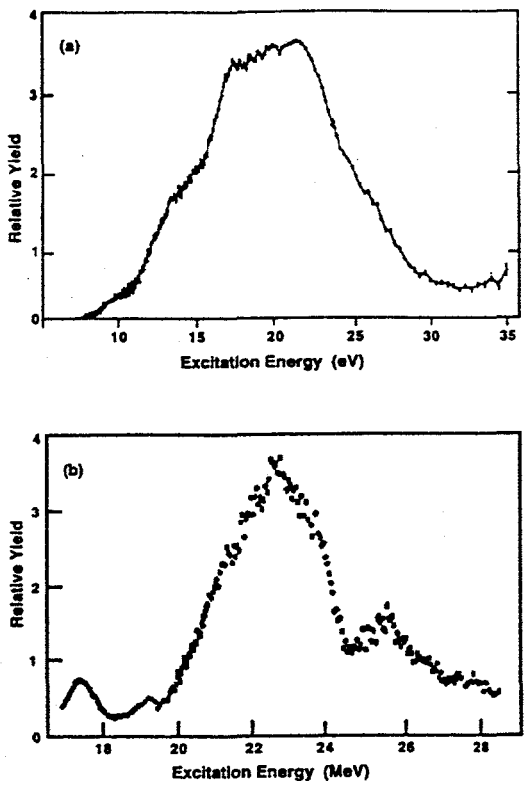

Figure 2. comparison of the giant dipole resonances observed in the C_{60} molecule[9] and in the ^{12}C nucleus[11].

(=10Å 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 C_{60} molecule has been predicted[8] and measured[9,10] to have an energy of 20 eV, and a FWHM of about 10 eV. An interesting connection to nuclear physics is shown in Fig. 2, which gives a comparison of the giant dipole resonances observed in the C_{60} molecule[9] and in the ^{12}C nucleus[11].

To estimate the total interaction cross section, we developed a quasi-classical model for the interaction[12] between the projectile and the fullerene that gives the total excitation and single-plasmon excitation probabilities as a function of impact parameter. These probabilities are presented in Fig 3(a). The total excitation probability reaches unity at an impact parameter of about 7 Å, still far outside the radius R. To determine the total interaction cross section, it is therefore not necessary to consider explicitly reactions at the smaller impact parameters where the xenon ion may interact with individual electrons. Using the model, we find that the total interaction cross section is 811 $Å^2$, whereas the single-plasmon cross section is 387 $Å^2$, i.e. 48% of the total.

Our model can be expected to be valid for single-plasmon excitation involving large impact parameters where the linear-response and dipole approximations are valid. It can be expected to break down at smaller impact parameters where multi-plasmon excitation occurs leading to multiple ionization, pair emission, and (at still smaller impact parameters) multifragmentation. The dominant decay mode of the single-plasmon excitation is via single electron emission[9,10,13]. We therefore compare the calculated single-plasmon cross section to our measured C_{60}^+ yield. The dependence on beam energy is illustrated in Fig. 3(b) for the projectile charge state $Z_p = 18$. The weak dependence on beam energy is reproduced by the calculation. Rough estimates of the experimental cross sections agree with the calculated values within a factor of two. The slope of the

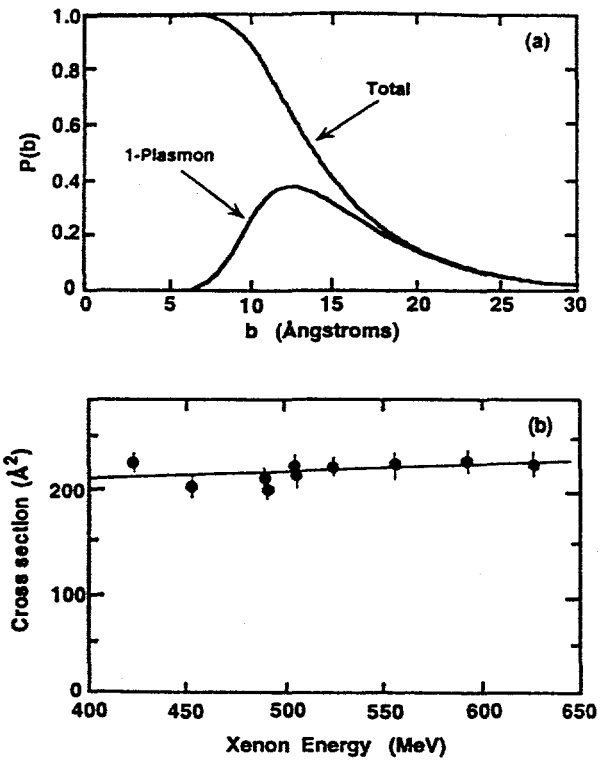

Figure 3. (a) Calculated probabilities for plasmon excitation vs impact parameter. (b) Calculated single-plasmon cross section compared to the measured yield of C_{60}^{+} from $C^{136}Xe^{18+}$ ions.

calculated curve is insensitive to small variations in the total oscillator strength. We also considered the role of electron capture by the Xe ions as a production mechanism for C_{60}^{+} ions but at the high velocities (11-14 a.u.) of our beams, the charge-capture cross sections[14] are negligible compared to the cross sections for plasmon excitation.

At impact parameters less than about 7Å where the energy deposition becomes large, essentially all projectile/target interactions can be expected to result in multifragmentation. We have constructed a bond-percolation model[12] to describe these fragmentation processes. C_{60} is represented 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.

After computing the breaking of the bonds by a given projectile, we employ a cluster recognition algorithm and identify the sites still connected via unbroken bonds as members of a cluster. We record the size of each cluster. This model is, except for considerations of the different reaction geometries involved, similar to a model of nuclear multi-fragmentation[15–17] used to explain production cross sections[18] for nuclear fragments emerging from heavy nuclei bombarded by protons with energies between 80 and 300 GeV.

Figure 4 shows the fragment-mass distribution calculated using the model. The calculation reproduces the overall shape of the measured fragment-mass spectrum. The calculation gives too little yield at high mass numbers because it does not take into account a) the contributions from evaporative processes following more gentle excita-

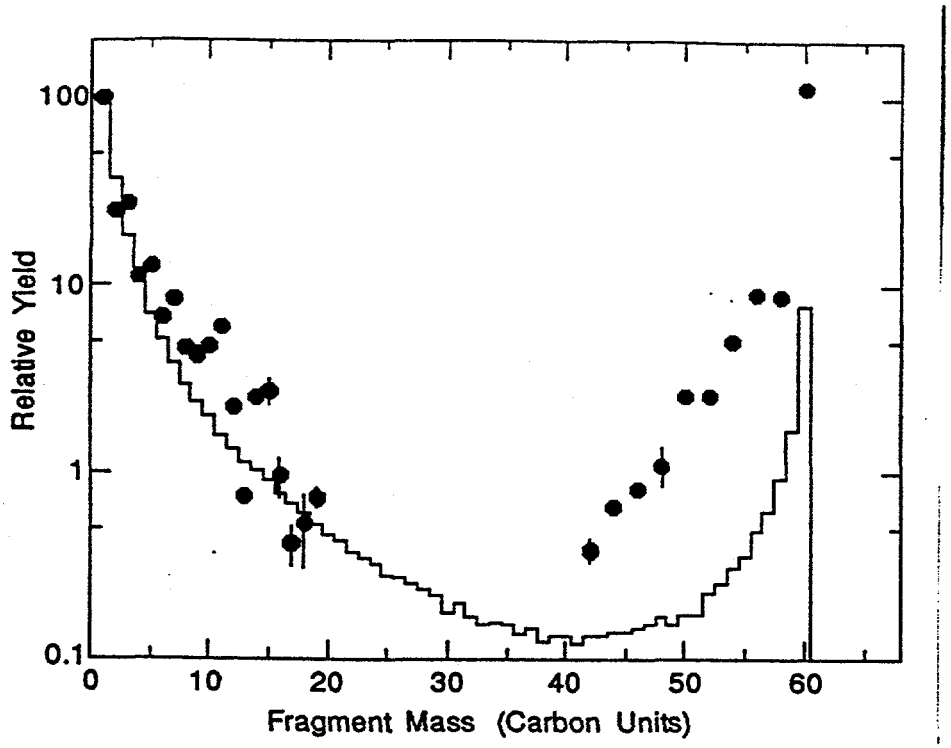

Figure 4. C_{60} bombarded by 625 MeV $^{136}Xe^{35+}$ ions. The histogram is the distribution calculated on the basis of a percolation-multifragmentation model (see text).

tions (eg. plasmon excitations), and b) the known instability of odd-numbered heavy fragments which decay rapidly into even-numbered ones.

Nuclear multifragmentation displays features similar to those observed in our data on fullerene disintegration, eg. the phenomenon of limiting fragmentation (the fragmentation yield does not change much above a certain beam energy). Another common feature is the U-shaped fragment-mass spectrum.

Both experiment and calculation display a power-law fall-off in the production cross sections for clusters of n carbon atoms, $\sigma(C_n) \propto n^{-\lambda}$ for $n \leq 20$. In experiment and calculation, $\lambda \approx 1.3$. This behavior is similar to the case of nuclear fragmentation[18] where one finds $\lambda \approx 2.6$ in inclusive (impact-parameter integrated) reactions. The power law is a consequence of the finiteness of the system and of the integration over different excitation energies in inclusive reactions[15]. The nuclear fragmentation data contain indications of a second-order phase transition in nuclear matter (albeit washed out due to finite-size effects), and at the critical point the apparent exponent λ has a minimum[16,17]. Our λ value is significantly lower than the critical exponent (τ=2.0) for two-dimensional infinite-size bond-percolation and can be attributed to the finiteness and the periodic boundary conditions of our fullerene. Similar observations[19] have been recorded for nuclear systems.

Acknowledgments

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