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Mass spectroscopic confirmation of the presence of C_{60} in laboratory-produced carbon dust

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Krätschmer, Fostiropoulos and Huffman recently suggested that substantial quantities of C_{60} are present in carbon deposits made by evaporating graphite under 100 Torr He quenching gas. We have directly confirmed the presence of fullerenes in such deposits using a surface-analytical mass-spectroscopic detection technique.

In a recent Letter, Krätschmer, Fostiropoulos and Huffman [1] presented UV-visible and infrared spectra of carbon deposits made by evaporating graphite under 100 Torr of He. The IR spectrum of the deposit showed a set of four strikingly sharp lines at 1429, 1183, 577 and 528 cm^{-1} with widths between 3 and 10 cm^{-1} , in addition to broad absorption features characteristic of graphite dust. Spectra obtained with samples made from 99% pure ^{13}C showed a corresponding set of four lines, with frequencies reduced from those in the ^{12}C spectrum by the factor expected for pure carbon clusters. The agreement in number and frequency of the four sharp, strong lines with calculations for the proposed Buckminsterfullerene molecule [2-5], led Krätschmer et al. to believe that the deposited material contained significant quantities ($\approx 1\%$) of C_{60} . To date, however, no direct mass-spectroscopic evidence for the presence of C_{60} in such deposits has been presented.

We have now directly confirmed the presence of fullerenes (C_{2n} , $n \geq 16$) in deposits made as described by Krätschmer et al. [1] using a surface-analytical mass-spectroscopic detection technique. In this technique, molecules originally present on the surface are brought intact into the gas-phase using laser desorption, and are entrained in a pulsed supersonic gas expansion. This cools the molecules and transports them into a high-vacuum region where

they are photoionized between the extraction plates of a linear time-of-flight mass spectrometer. The spectra are displayed on a digital oscilloscope and read out by a PC. A mass resolution $M/\Delta M = 300$ is obtained. The apparatus used has been described fully in recent publications [6,7]. The mass spectrum obtained from carbon condensate deposited on a tungsten foil is shown in fig. 1. A KrF excimer laser (248 nm) was used for desorption (60 μJ in a 0.25 mm diameter spot). Argon with 8 atm backing pressure was used as the carrier gas, and ionization was performed with an ArF excimer laser (193 nm, 200 μJ

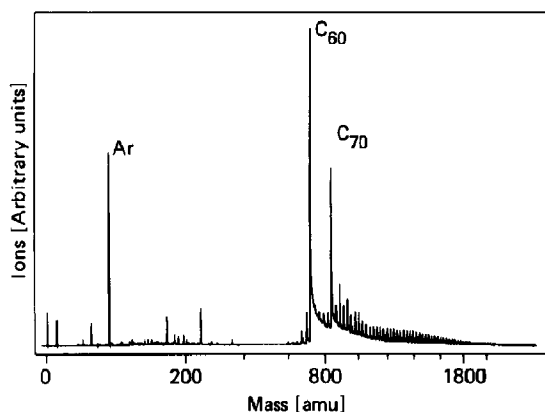


Fig. 1. Time-of-flight mass spectrum from carbon condensate on a tungsten foil, taken using the laser desorption jet-cooling detection apparatus. The horizontal axis is linear in time-of-flight and the corresponding (nonlinear) mass scale is indicated.

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in a 1.5 mm diameter spot). The overwhelmingly dominant peaks in the center of the spectrum are due to C_{60} and C_{70} . Smaller quantities of other even-numbered pure carbon clusters ranging from about C_{30} to C_{160} are also present. The sensitivity to high-mass clusters is reduced by the falling mass spectrometer transmission and the decreasing sensitivity of the channel-plate detector. The peaks at lower masses are due to contaminants unrelated to the carbon deposit.

Using the same detection technique, we have recently demonstrated that fullerenes can be deposited on surfaces by laser ablation of graphite under an inert gas atmosphere [7]. Mass spectra taken of samples prepared by this alternative technique were found to be remarkably similar to that shown in fig. 1, despite the drastically different time scales for the production of the carbon vapor.

An important question that should be addressed concerns the possibility that fullerenes might be produced in the desorption step of the detection process. That this is *not* the case, and that the detected clusters were indeed present on the surface, has been directly demonstrated using an isotope scrambling experiment [7]. Briefly, deposition of carbon clusters on a sample surface was performed by alternately laser vaporizing pure ^{12}C and pure ^{13}C . The sample material on the surface thus had comparable amounts of both isotopes. Nevertheless, the mass

spectrum of this material clearly shows isotopically pure clusters of both types, proving that they must have been produced in the original deposition process rather than in the desorption plume in the detection apparatus. This is also consistent with the observation that no fullerenes are observed in the mass spectrum if a pure graphite surface is used as a sample.

Our direct mass spectroscopic data thus clearly demonstrate that fullerenes can be deposited on surfaces. Furthermore, the apparent abundance of C_{60} in these samples strongly supports Krätschmer, Fostiropoulos and Huffman's identification of the strong infrared bands with the Buckminsterfullerene molecule.

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