Historical perspective on: Negative carbon cluster ion beams: New evidence for the special nature of C60 [Volume 126, Issue 2, 2 May 1986, Pages 215–217]
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A good colleague at Rice recently remarked to me that he had occasion to read our 1985 Letter to Nature announcing finding C_{60}, and was surprised that our Letter had not been rejected because of our claim that the molecule had the structure of a truncated icosahedron. All the evidence for the proposed structure that we had was that the C_{60} species did not appear to be susceptible to chemical attack, and we could not imagine a more attractive structure to propose.

After publication of the 1985 Nature Letter, we should not have been surprised that others believed that there might be possible alternative explanations for the prominence of the C_{60} mass peak in our positive ion mass spectra. The alternative idea that initially gained the most traction was that this C_{60} species, whatever its actual structure, was prominent because the ionization laser had an energy of only 6.4 eV; therefore, C_{60} just had a lower ionization energy than its neighbors. This alternative soon seemed to be unlikely, because it did not explain why the C_{60} was not prominent under other conditions.

Objections to our seemingly hasty structure conclusion then shifted to alternative suggestions; that perhaps C_{60}^{+} was a favoured product of the fragmentation of larger structures in the laser ionization process, and these larger structures were not present under the experimental conditions where low C_{60}^{+} peaks were observed. Alternatively, the prominence of the C_{60}^{+} peak might arise because the positive ion was perhaps favoured for some other reason.

We had the good fortune to be in the position where we could carry out experiments that could test these alternatives and our own proposal. Doing such experiments would either confirm us or, if (unthinkable) we were wrong, we would at least be the ones correcting ourselves.

This Chemical Physics Letters paper was the first such structural test experiment. Its point was to test the notion that maybe the prominence of the C_{60}^{+} peak arose from some special nature of the positive ion. We decided to look at the negative ion mass spectrum with the aim of, if possible, finding conditions under which

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**Fig. 1.** Distribution of negative and positive carbon cluster ions of cluster size between 46 and 74 atoms. (a) depicts the distribution of negative cluster ions formed when the cluster pulse is intercepted just in front of the 4 mm expansion tube by KrF excimer laser (248 nm) radiation (fluence $\geq 1$ mJ/cm$^2$). In (b) the distribution of positive ions produced by F$_2$ laser (157 nm) ionization in the source of the time-of-flight mass spectrometer 1 m downstream is shown. (c) shows the positive ion distribution produced by ArF excimer laser (193 nm) photoionization in the molecular beam at the same point downstream.
the negative ion spectrum would show a prominent \( \text{C}_{60} \) peak. The negative ions were produced in a plasma just outside the expansion orifice by a high intensity UV pulse at 248 nm from a KrF excimer laser. In Figure 1a of this Letter, the resulting negative ion mass spectrum exhibits a convincingly prominent \( \text{C}_{60} \) peak, thereby negating the argument that the prominent peak was somehow a property of the positive ion.

However, it did little to undermine the suggestion that \( \text{C}_{60} \) might be a special fragmentation product. Later in 1986, we carried out another set of experiments (Chem. Phys. Lett. 132, 99–102 (1986)), over a wide range of ionizing laser fluences that showed that the relative prominence of the \( \text{C}_{60}^+ \) actually decreased as the ionizing laser fluence increased. This is strong evidence that prominent \( \text{C}_{60} \) is not a fragmentation product.