Knockout fragmentation and endohedral formation in collisions between C_{60}^+ and noble gas atoms

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Synopsis We have investigated the non-statistical, knockout-driven fragmentation of C_{60}^+ in collisions with noble gas atoms at center-of-mass energies between 30 and 1500 eV. For Ne, Ar, and Xe, small yields of C_{59}^+ were measured, while for He, endohedral $He@C_{60}^+$ and $He@C_{58}^+$ were observed.

In the past five years, the fullerenes C_{60} and C_{70} have been observed in an ever-increasing variety of interstellar environments. Recently, it was confirmed that C_{60}^+ is responsible for at least four of the Diffuse Interstellar Bands [1], a group of broad absorption features in interstellar clouds whose origin has been a mystery for nearly a century. These discoveries have brought about intense study of the formation and destruction mechanisms of fullerenes and other large molecules such as Polycyclic Aromatic Hydrocarbons (PAHs) in the interstellar medium (ISM).

One important processing mechanism for both fullerenes and PAHs is collisions with fast atoms or ions, particularly H and He. Such collisions, with center-of-mass energies in the 10-1000 eV range, should be especially important when shockwaves from exploding supernovae pass through interstellar clouds, or when solar winds penetrate atmospheres such as that of Titan. We have shown that non-statistical fragmentation - the direct, prompt (fs) knockout of single carbon atoms - is an important mechanism for fullerene/PAH destruction under these conditions [2,3]. Such single-carbon loss processes are highly disfavored in statistical fragmentation, in which the excitation energy is first distributed over all internal degrees of freedom and the channels with the lowest dissociation energy (C_2-loss for fullerenes or C_2H_2-loss for PAHs) usually dominate strongly.

We have investigated non-statistical knockout fragmentation of C_{60}^+ ions in collisions with noble gas atoms at center-of-mass energies from 30 to 1500 eV. For Ne, Ar, and Xe, knockout is definitively identified by the detection of C_{59}^+ as shown in Figure 1. Interestingly, the yield of C_{59}^+ is much lower than the analogous non-statistical fragments of PAHs. This is presumably due to the non-planar structure of C_{60}, and higher probability for secondary statistical fragmentation.

No C_{59}^+ was observed in collisions with He at center-of-mass energies up to 60 eV, although endohedral $He@C_{60}^+$ and $He@C_{58}^+$ were formed.

The products of non-statistical fragmentation tend to be highly reactive, with important consequences for interstellar chemistry. We have observed such collision-induced reactivity in earlier experiments studying collisions between atomic ions and clusters of fullerenes and/or PAHs conducted at the ARIBE facility in Caen, France. In one experiment, for example, C_{59}^+ ions formed by single carbon knockout reacted with nearby C_{60} molecules inside the cluster, forming dumbbell-shaped C_{119}^+ [4]. Such reactions may play a role in the growth of large molecular species in the ISM.

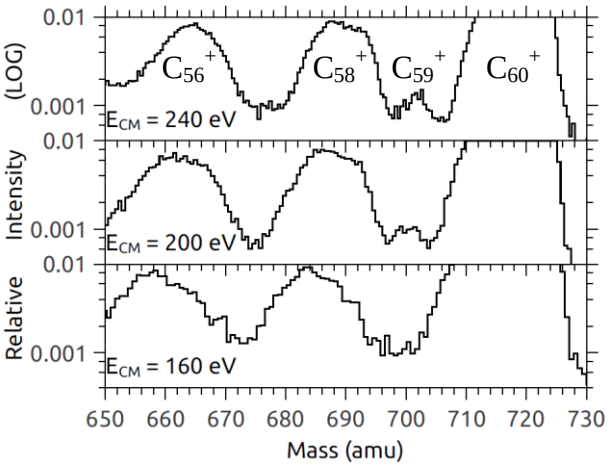

Figure 1. Mass spectra showing formation of C_{59}^+ following collisions between C_{60}^+ and Ne.

References

[1] E. K. Campbell *et al.* 2015 *Nature* **523** 322.
[2] M. H. Stockett *et al.* 2014 *Phys. Rev. A* **89** 032701.
[3] M. Gatchell *et al.* 2014 *Int. J. Mass Spectrom.* **365** 260.
[4] H. Zettergren *et al.* 2013 *Phys. Rev. Lett.* **110** 185501

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