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Electron detachment and fragmentation in collisions between 1.25 keV/carbon C_8^- and C_{60}^- clusters and H_2

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Abstract. The destruction cross-section for 22.5 and 50 keV C^{1-} , for 10 and 50 keV ${C_8}^{1-}$ and for 50 and 75 keV ${C_{60}}^{1-}$ clusters in collisions with H_2 has been measured by an attenuation method. The destruction of the cluster anions is dominated by electron detachment rather than fragmentation and is of the order of the geometric cross-section. The cross-sections vary little with bombarding energy.

PACS. 34.70.+e Charge transfer – 36.40.-c Atomic and molecular clusters – 36.40.Qv Stability and fragmentation of clusters

1 Introduction

Relatively little is known about charge exchange cross-sections for clusters. It is of interest to understand how they scale with cluster size, and to what extent they depend on ionization potentials. For anions one might expect the extent of the geometrical delocalization to play a role. Shen *et al.* [1] have reported carbon anion destruction cross-sections in collisions with H_2 at a fixed bombarding energy for a large range of cluster sizes. They relate the dependence on cluster size to geometrical effects. Charge exchange cross-sections are also of interest with regard to the interpretation of fragmentation cross-sections, and to what extent such cross-sections are biased by which charge state one is observing. We report here an experiment where we compare destruction cross-sections for ${C_8}^{1-}$ and ${C_{60}}^{1-}$ at the same velocity (same bombarding energy per carbon atom). We have chosen an energy of 1.25 keV/carbon since we have studied [2] fragmentation cross-sections of ${C_{60}}^{1-}$ at this velocity. We have also made some measurements at other energies and with other ions in order to compare with previous work.

2 Experimental

${C_8}^{1-}$ ions were produced in a sputter ion source by bombarding graphite with 3–4 keV cesium ions and extracting the negative ions with a 10 keV electrostatic potential. After extraction the negative ions were mass-analyzed using a 90° bending magnet. The ${C_{60}}^{1-}$ ions were produced in a different source by attaching electrons to neutral molecules exiting an oven through a capillary tube. The oven was typically heated to about 500 °C and the electron beam had an energy of 6 eV. The negative ions are pre-accelerated to 6 keV, mass-analyzed with a 90° bend magnet, and then further accelerated to 75 keV by an additional electrostatic potential. The ${C_8}^{1-}$ and ${C_{60}}^{1-}$ ions were transported several meters to a gas cell. This cell consisted of an entrance arm with a 0.067 cm^2 aperture on a 1.27 cm long, 0.476 cm diameter tube and an exit arm with a 0.075 cm^2 aperture on a 0.95 cm long, 0.476 cm diameter tube. These arms were attached to a central gas feed region 1.91 cm long. The gas pressure in the central region was measured with a VRC Pirani gauge [3] and with a Hastings thin film Pirani gauge [4]. The pressure profile in the entrance and exit tubes was calculated from the conductance of the tubes and the knife-edge apertures. This information was used to determine the total number of gas molecules in the cell. Typical pressures in the central region of the gas cell were between 0.04 and 9 mTorr. The chamber was pumped with both a turbomolecular pump and a cryogenic pump. Pressures downstream from the cell were typically less than 10 microTorr. The destruction cross-section was determined by attenuation. Two types of measurements were made initially. The first was to measure the attenuation of the negative ion current in a Faraday cup downstream from the gas cell. The second method exploited an electrostatic analyzer to deflect the unreacted negative ions to a Channeltron [5] detector. The first method suffers somewhat from a contribution of positive ions, either from double charge exchange or from fragmentation. This leads to an attenuation curve for the Faraday cup method which for ${C_8}^{1-}$ was typically 20% steeper than that obtained by the electrostatic an-

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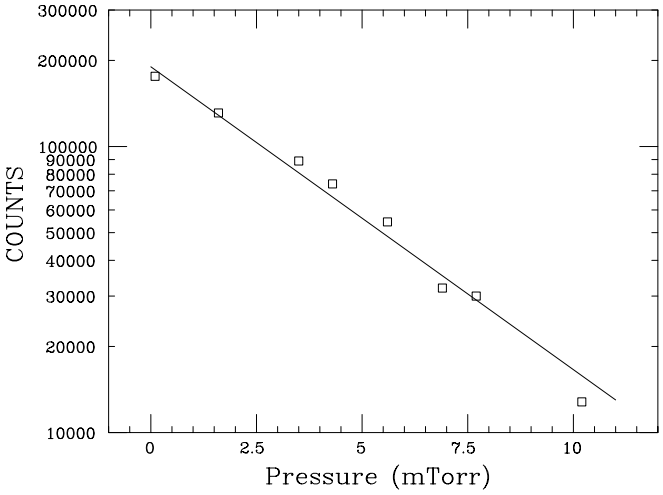

Fig. 1. Attenuation curve for 10 keV $C_8{}^{1-}$ on H_2.

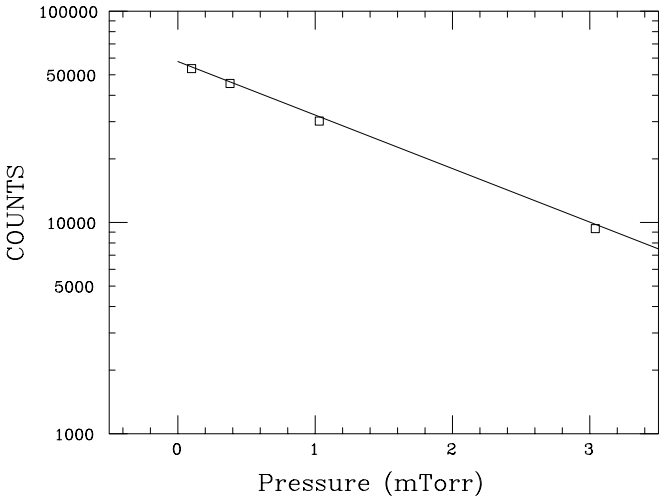

Fig. 2. Attenuation curve for 75 keV $C_{60}{}^{1-}$ on H_2.

alyzer. The destruction cross-sections reported here were obtained by the more accurate second method. An example of an attenuation curve for $C_8{}^{1-}$ is shown in Figure 1.

A similar curve for C_{60} is shown in Figure 2.

3 Results and discussion

The destruction cross-section is obtained from the average of the slopes of several attenuation curves such as those illustrated and from the total number of target molecules from a modeling of the pressure profile based on the conductance of various segments of the gas cell. We estimate an absolute uncertainty in each cross-section of approximately 20%, primarily from an uncertainty in the total number of target molecules. The relative cross-sections for different species is determined with an uncertainty of less than 10%. Our results are summarized in Table 1, along with other values from the literature where available. The cross-section for H^{1-} on H_2 was measured as a check on our procedures. This system has been studied extensively previously [6,7]. Our results are in good agreement with these results, and also for O^{1-} on H_2. We obtained a cross-section for C^{1-} on Ar which is a factor of two larger than that of a previous measurement [8].

Table 1. Anion destruction cross-sections

All cross-sections are in units of 10^{-16} cm^2.				
Reaction	Energy (keV)	This work	Previous work	Ref.
$H^- + H_2$	10	11.3	12	[7]
	22.0	8.6	8.5	[6]
	50	5.8	5.9	[6]
$C^- + H_2$	22.5	10.9		
	50	10.7	13	[1]
$C_8{}^- + H_2$	10	20		
	50	23	34	[1]
$C_{60}{}^- + H_2$	50	47	90	[1]
	75	50		
$C^- + Ar$	30	13	6.5	[8]
$O^- + H_2$	50	6.7	7.2	[6]

For 10 keV $C_8{}^{1-}$ we obtain a cross-section of 20×10^{-16} cm^2. This destruction cross-section corresponds to the sum of several possible reaction channels. The dominant channel is expected to be the

$$C_8{}^{1-} \rightarrow C_8 + e^-$$

channel. Other possible channels are the

$$C_8{}^{1-} \rightarrow C_{n<8}{}^{1-} + X \text{ and}$$

$C_8{}^{1-} \rightarrow C_{n<8} + e^- + X$ channels. The last channel is not directly observable in our experiment, but the second channel is observable. We have scanned the negative ion spectrum at several cell pressures, and find that the cross-section for the second channel is very small. At a pressure of 1 mT, only 10% of the $C_8{}^{1-}$ ions destroyed have appeared in a $C_{n<8}{}^{1-}$ channel. Of this 10%, most are in the $C_5{}^{1-}$ channel. This is to be expected, since the $C_3 + C_5$ fragmentation channel is the most energetically favored channel [9]. The observation that the electron appears on the C_5 rather on the C_3 is consistent with the fact that the electron affinity for C_3 is 1.995 whereas that for C_5 is 2.839 eV [10]. The cluster products of the first and third channels can lose a second electron to make positive ions. We find that the positive ion spectrum is dominated by unfragmented $C_8{}^{1+}$, indicating that the first channel above (electron detachment) is stronger than the third channel (fragmentation with loss of an electron).

For 75 keV $C_{60}{}^{1-}$ we obtain a cross-section of 50×10^{-16} cm^2. As in the case of $C_8{}^{1-}$, the electron detachment cross-section is about an order of magnitude larger than the fragmentation cross-section. This and our other result for $C_8{}^{1-}$ at 1.25 keV/carbon are appreciably less

than those given in a very recent report by Shen *et al.* [1] of the cross-sections at 50 keV. In order to make a more direct comparison, we also measured the cross-sections at 50 keV total energy. As can be seen from the table, our results at this energy are still somewhat smaller than their results. We have also measured the destruction cross-section of C^- at two bombarding energies. At 50 keV, the energy studied by Shen *et al.*, we obtain a cross-section of 10.7×10^{-16} cm^2, in reasonable agreement with their result of 13×10^{-16} cm^2.

Ion chromatography experiments show that carbon cluster cations in the light-fragment size range are predominantly chains for n less than 8, that chains and rings coexist for $n = 8$ through $n = 10$, and that for $n > 10$ rings predominate [11,12]. The dominance of the even-n over the odd-n carbon anions in the cluster spectrum from our sputtering source [13] indicates that the $C_8{}^{1-}$ ions in our beam are primarily chains. We have used the geometry calculations of Watts and Bartlett [14] to estimate the geometrical cross-section of the $C_8{}^{1-}$ ions. We have treated the linear chain as a cylinder with spherical endcaps. The diameter of the cylinder represents the effective extent of the electron density distribution. The length of the cylinder from their calculations is 9.0 Å, and we have estimated the diameter of the cylinder to be equal to the average C-C distance and the radius of the endcaps to be half this distance. After averaging over the random orientations of the chain, we find an average cross-sectional area of 11×10^{-16} cm^2, somewhat smaller than our measured values. A similar calculation for $C_{60}{}^{1-}$ gives a cross-section of 55×10^{-16} cm^2, very similar to our measured cross-section. The qualitative conclusion we draw from these very crude estimates is that the absolute values of our cross-sections are reasonable. The reason for the observed cross-section for $C_8{}^{1-}$ relative to $C_{60}{}^{1-}$ being larger than the geometrical estimate may be related to the fact that the extra electron in $C_8{}^{1-}$ is considerably less delocalized than in $C_{60}{}^{1-}$. If we extend the simple geometrical argument above to C^- and use the endcap radius of 0.64 from the $C_8{}^{1-}$ calculation we obtain an estimate of 1.3×10^{-16} cm^2 for C^-, considerably smaller than the measured value. This may again be related to the still smaller delocalization of the extra electron in C^-. It may also reflect the fact that the electron affinity of C is only 1.26 eV, whereas for C_8 it is 4.4 eV [10] and for C_{60} it is 2.6 eV [15].

In summary, we have measured the destruction cross-sections for $C_8{}^{1-}$ and $C_{60}{}^{1-}$ clusters at the same velocity as well as at the same bombarding energy (50 keV). Although the ratio of the cross-sections for the two clusters at the same velocity is similar to that obtained in an earlier study comparing at the same bombarding energy, our absolute cross-sections are somewhat smaller. In order to verify the absolute values of our cross-sections we have also measured cross-sections for the well-studied H^{1-} on H_2 reaction, obtaining good agreement with previous results. The carbon cluster cross-sections are in qualitative agreement with simple geometrical considerations. The cross-sections vary little with bombarding energy.

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