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Projectile charge-state dependence of methane fragmentation

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The projectile charge-state dependence of methane fragmentation is studied at a constant projectile (Ar^{q+}, $1 \leq q \leq 7$) velocity of 0.42 a.u. using the technique of time-of-flight mass spectroscopy. The relative cross sections for fragment production are reported as a function of the incident projectile charge state. The fragments appearing as molecular ions show a decrease in cross sections with increasing projectile charge state as opposed to the carbon ions which show an increase with increasing projectile charge state.

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I. INTRODUCTION

Studies on molecular dissociation provide us valuable information about the breakup dynamics of multiply charged molecules. Such studies are important from the fundamental aspect, as a test for various theoretical models, and also for practical areas such as plasma physics and planetary atmospheric chemistry. Molecular dissociation can be induced by particle impact ionization using electrons or charged particle beams and also by photon impact using lasers or synchrotron radiation. Studying molecular dissociation using charged particle beams provides us with the unique opportunity of varying the field strength for constant interaction times.

Small alkane molecules, the smallest being methane, are prototypes for polyatomic organic molecules and are important in many research fields in chemical physics. They are also interesting in astrophysics since they are important constituents in the planetary and cometary atmospheres [1]. Dissociation of methane has been studied with particle impact [2,3] as well as with lasers [4]. The velocity dependence of the ionization and fragmentation of methane caused by fast proton impact has also been studied [5]. The fragmentation has been shown to be sensitive to the projectile characteristics [6]. In the present paper we report the relative cross sections of the fragments resulting from methane dissociation caused by impact of Ar^{q+} ($1 \leq q \leq 7$) ions for a constant interaction time of around 750 attoseconds. The effect of projectile charge state on the population of the various fragments is discussed.

II. EXPERIMENT

The experiment was carried out at the Low Energy Ion Beam Facility (LEIBF) of the Inter-University Accelerator Centre (IUAC), New Delhi, India. Low energy Ar^{q+} ions ($1 \leq q \leq 7$) are produced from a 10 GHz Nanogan ECR ion source placed on a 200 kV high voltage platform along with its extraction system [7]. The ions are mass analyzed by a 90° bending magnet and transported to the collision chamber at the center of which the projectile ions interact with methane molecules effusing from a grounded needle located at right angles to the beam direction. The dissociated fragments are extracted from the interaction zone in a linear time-of-flight mass spectrometer (TOFMS) by applying a uniform electric field perpendicular to both the ion beam and the gas jet. At the end of the TOFMS, the dissociation products are detected by a microchannel plate (MCP) detector. The ejected electrons are extracted in the opposite direction to TOFMS and detected by a channel electron multiplier which gives the trigger for starting the data acquisition. The TOF spectrum was acquired by a time-to-digital converter (TDC) interfaced to a computer where the fragment ions were recorded. The details of the setup and data acquisition have been published elsewhere [8]. The number of counts under each peak was determined by fitting these with Gaussian curves and a linear or quadratic background. The relative cross sections for fragment production were then determined as a fraction of the total. The error in determination of relative cross section (constrained to error in fitting) is less than 3% in all cases. We expect the detection efficiencies not to vary much over the mass ranges involved, as expected from [9].

III. RESULTS AND DISCUSSIONS

The typical time-of-flight mass spectra for methane dissociation is shown in Fig. 1. The spectra shows peaks corresponding to the undissociated molecular fragments $\mathrm{CH_4}^+$, $\mathrm{CH_3}^+$, $\mathrm{CH_2}^+$, and CH^+ formed due to loss of one to three H

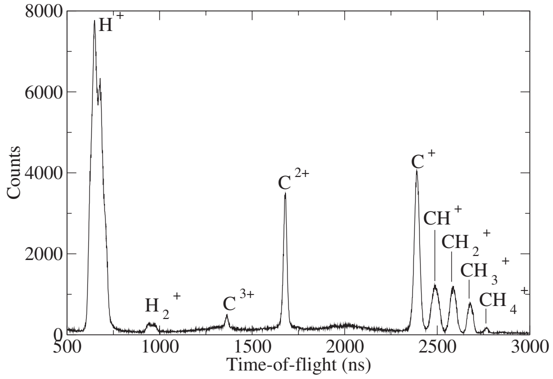

FIG. 1. A typical TOF spectra for methane dissociation induced by the impact of Ar^{7+} ions at a velocity of 0.42 a.u.

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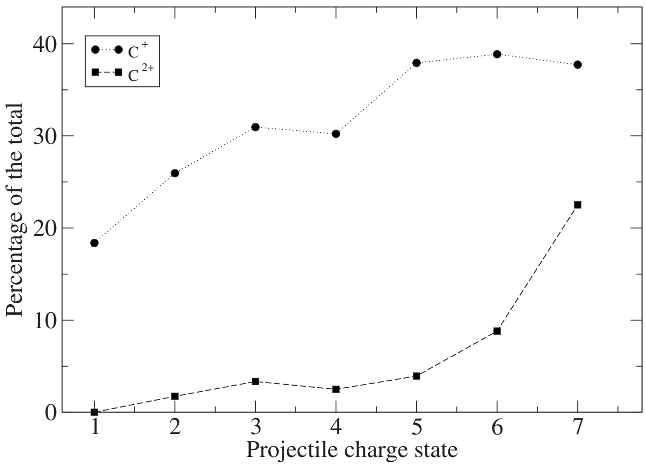

FIG. 2. Relative cross sections (%) for carbon ions as a function of projectile charge. The total does not include the counts under the H$^+$ and the H$_2$$^+$ peaks. Lines are drawn to guide the eye.

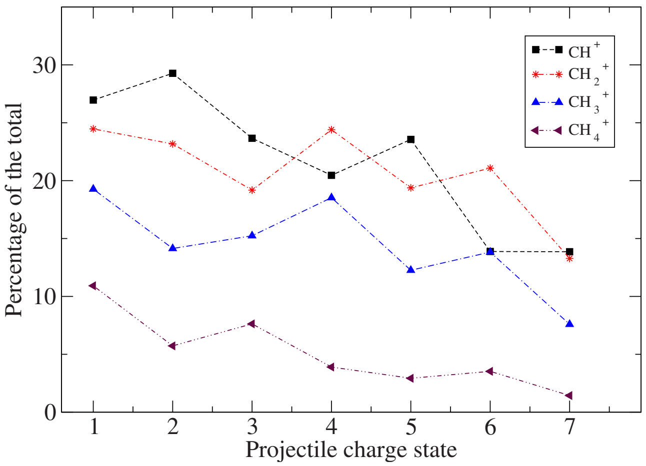

FIG. 3. (Color online) Relative cross sections (%) for the molecular ionic fragments as a function of projectile charge. The total does not include the counts under the H$^+$ and the H$_2$$^+$ peaks. Lines are drawn to guide the eye.

atoms and also peaks corresponding to the atomic fragments H$^+$, C$^+$, and C^{2+}. A small peak corresponding to the formation of H$_2$$^+$ molecular ion is also visible. We do not see any peak corresponding to C^{3+} in the methane TOF for projectile charge state less than seven.

The variation of the relative cross section of the fragments as a function of incident projectile charge are shown in Figs. 2 and 3. The fragments appearing as molecular ions show a decrease in cross sections with increasing projectile charge state. The probability of single hydrogen loss and triple hydrogen loss show a complimentarity to each other in the sense that a higher cross section for the CH$_3$$^+$ fragment always shows up with a relatively lower cross section of the CH$^+$ counterpart and vice versa (see Fig. 3). The carbon ionic fragments show an increase in their relative cross sections with increasing projectile charge. This is expected because as we increase the projectile charge more and more electrons will be captured by the projectile [10], giving the transient methane molecular ion a higher and higher charge state. The relative cross sections for the formation of highly charged transient methane molecular ions thus goes up with increasing projectile charge. Such transient molecular ions then Coulomb explode resulting in carbon and hydrogen fragments.

IV. CONCLUSION

The effect of the charge state of the incident Ar projectile on the relative cross sections of the CH$_4$ fragments produced due to ion-induced dissociation of methane has been studied. The projectile target interaction time is kept constant by keeping the projectile velocity fixed at 0.42 a.u. The fragments appearing as molecular ions show a decrease in cross sections with increasing projectile charge states. The relative cross section of the carbon ions on the other hand increase with increasing projectile charge. The probability of single hydrogen loss and triple hydrogen loss show a complimentary behavior to each other. The decrease in molecular fragments and increase in atomic fragments is attributed to the increase in multielectron capture probability with increasing projectile charge.

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