Ion acceleration and molecular recombination from methane cluster explosions

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Experiments were conducted at the FLASH facility at DESY in July 2009. Throughout 4 days of beamtime clusters were irradiated with intense (10^{13}-10^{15} W/cm^2), 15fs long soft X-ray pulses at Beamline 2. FLASH operated at a wavelength of 13.5nm (92eV) and had a very stable, average pulse intensity of 45uJ. The scope of targets included methane (CH_4) and deuterated methane (CD_4) clusters of varying sizes that were at times doped with Xenon to create mixed structures. The main motivation was to get a deeper understanding of light ion acceleration mechanisms in cluster explosion as well as completion of our data sets from earlier experiments in 2008 by increasing the range of achievable clusters sizes and the amount of Xenon. Those extensions to the experimental plan should lead to higher kinetic energies for the light ions.

Clusters were created by supersonic expansion of gas through a nozzle of a helium-cooled cluster source. The maximum cluster size was increased to 2.5×10^6 molecules per cluster and doping with Xenon was performed in a doping cell attached to the main vacuum chamber. The doping process was controlled by choosing different backpressures for Xenon to allow a fixed amount of it to condensate onto the clusters. The explosion products – positively charged ions – created through irradiation of clusters with soft X-rays pulses were recorded with a multi-channel plate detector (MCP) situated in a time-of-flight tube (TOF). Each measurement contains up to 40,000 shots with the FEL and the resulting TOF spectra were analyzed. Figure 1 shows the experimental setup used throughout all the experiments.

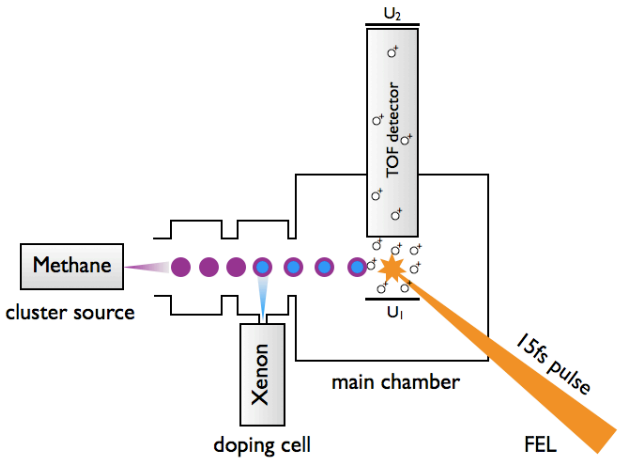
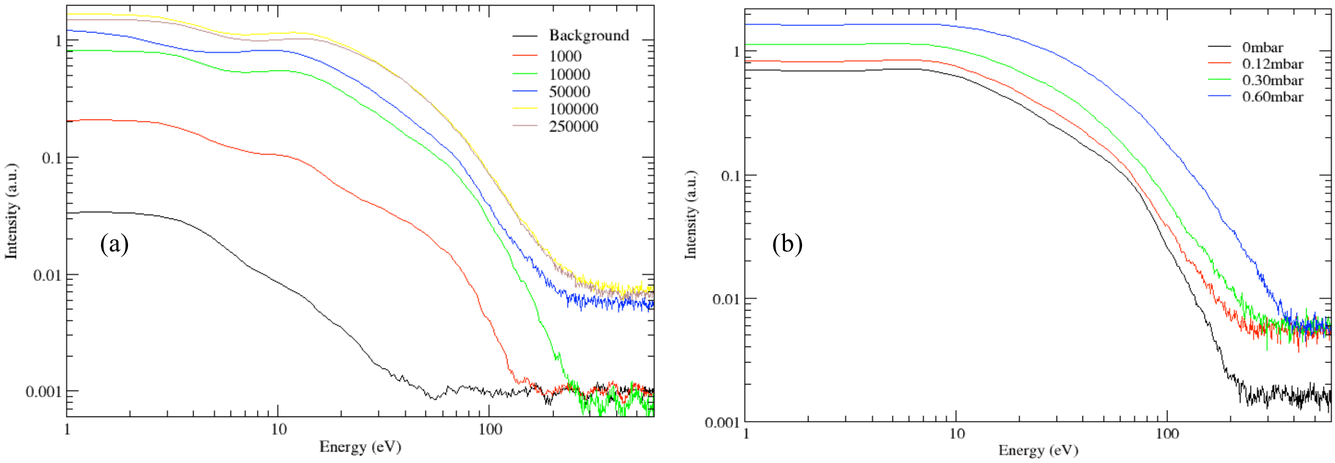


Figure 1: Experimental setup used during the FLASH beatime in July 2009. Methane clusters were injected into the vacuum chamber and could be mixed with Xenon in a doping cell. The clusters then traveled on towards the interaction region where they were interjected by 15fs long ultra-intense soft X-ray pulses. These pulses ionized the matter leading to a violent explosion of the clusters. Positively charged ions where then accelerated into the TOF tube with help of a repeller plate at $U_1 = 1000$V. Their impact on the MCP, held at $U_2 = -2600$V, was recorded with the data acquisition system and analyzed off-line.

The kinetic energy distribution for each ion peak in the spectrum allows to draw conclusions about the mean charge state, mean ionization energy and the number of absorbed photons per cluster atom. That is done through analyzing the resulting TOF spectra with help of the ion flight simulation package SIMION. Simulated ion trajectories help to find the corresponding kinetic energies distributions of the recorded ion peaks and get information about the underlying dynamics of the explosion process. The extracted information will act as a basis for planned MD simulations on cluster formation and explosion.

In agreement with earlier results and with data from the optical regime a strong dependency of the kinetic energies with cluster size, cluster composition and pulse intensity has been found in the new data set as can be seen in Figure 2.



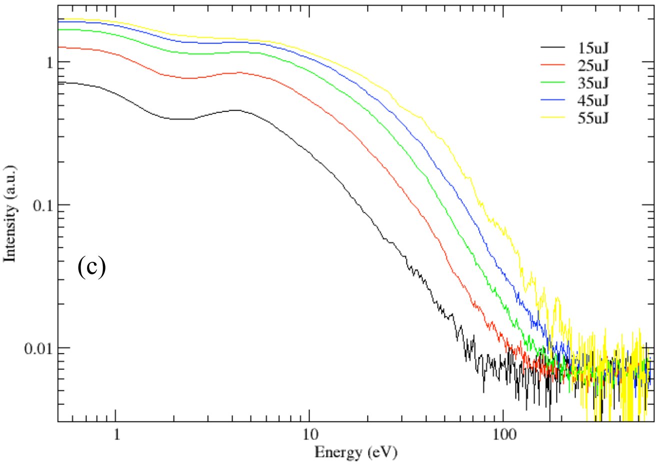


Figure 2: Energy distributions for hydrogen ions from FEL induced cluster explosion. The plot in 2a shows the dependency of the proton kinetic energy on cluster size. The difference in baseline is due to a different sampling voltage during data taking. The effect of Xenon doping is visible in 2b where a change in the energy distribution occurs with varying amounts of Xenon. Here, the cluster size has been fixed to 20,000 atoms. In 2c, at a cluster size of 250,000 atoms, the response to changes in pulse intensity can be seen.

Additionally, signals from protonated methane CH_5^+ were observed in the spectra. CH_5^+ is a carbocation commonly studied in astrophysics and its formation in FEL induced cluster explosions posts a lot of questions. As can be seen in Figure 3, CH_5^+ shows a non-linear behavior with cluster size increase. Going to bigger cluster sizes seems to break down the formation process of the molecule. CH_5^+ does not show a very strong energy dependency on pulse intensity (Figure 3b). Apart from a slight change in peak height, the energy gain when going to more intense pulses is in the range of only a few (1-2) eV. The low kinetic energy of the molecule, ranging from 1-10eV, indicates that it was formed after the violent explosion of the cluster, most likely through collisional recombination of hydrogen with CH_4^+. Quantum chemical calculations will shed light on the exact formation process of protonated methane.

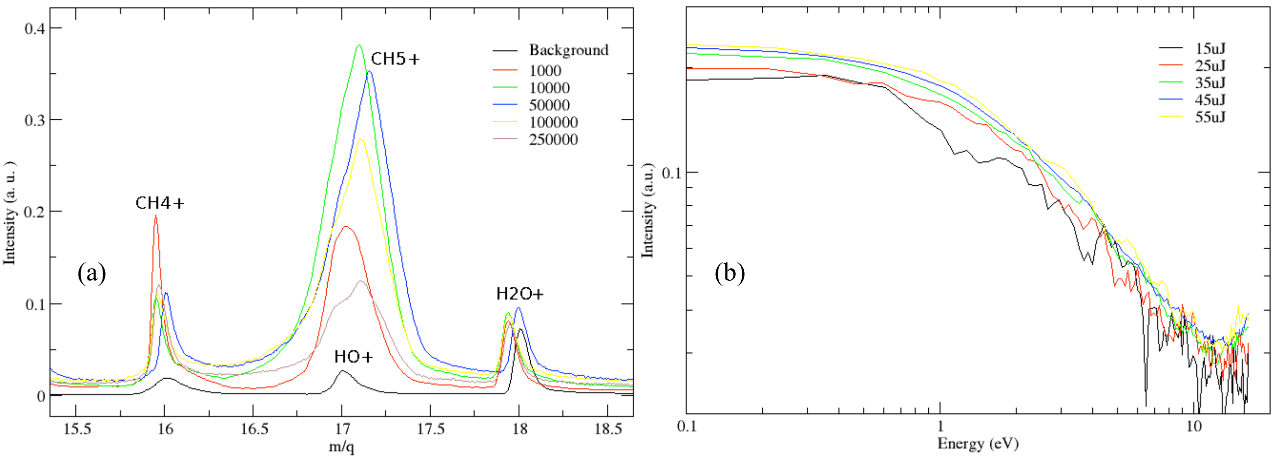


Figure 3: Ion TOF spectra of CH_5^+ molecule (17 m/q). The background trace in 3a shows signal from HO^+ which is overwhelmed by CH_5^+ when increasing the cluster size. The plot in 3b shows the energy distribution for the CH_5^+ peak as a function of incoming pulse intensity.

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