XUV two-pulse correlation of the desorption of ionic species from ice with FLASH pulses


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The interaction of high-energy photons in the soft x-ray regime with surface systems may lead to new and unexpected results due to the large number of accessible product states. In the past this photon energy regime has mainly been investigated using synchrotron radiation sources. The development of the Free Electron Laser at Hamburg (FLASH) now opens a new area in this spectral range. It provides pulsed radiation in the photon energy range from 20 to 200 eV, with a pulse energy in the tens of microjoule regime, and a pulse duration of ~ 30 fs [1 - 3].

The chemistry of hydrogen on graphitic and ice surfaces is of central importance for the generation of molecular hydrogen and hydrogen containing molecules in interstellar molecular clouds. In the dilute regions of these clouds the interaction of the surfaces with the light from new-born hot stars is of great importance. The interaction of the soft x-ray radiation with the ice surfaces is possibly a source of larger hydrocarbon molecules. Due to the lack of soft x-ray source only a few studies at high photon energies have been devoted to investigate this process[4,5].

In first experiments in 2005 we observed different ionic species directly formed by the FEL radiation on the surface and directly emitted from the ice surface adsorbed on graphite at cryogenic temperatures. Kinetic energies of desorbing H\(^+\), O\(^+\) and O\(_2^+\) of 1.8 eV, 559 meV and 390 meV, respectively were observed. Additionally, we determined the desorption yield of H\(^+\), O\(^+\), and O\(_2^+\). For H\(^+\) and O\(^+\) a linear desorption yield is observed. Surprisingly the desorption yield for the ionic molecular oxygen is highly nonlinear, see Figure 1 [6].

![Desorption yield of H\(^+\) and O\(^+\) at 38 eV with a H\(_2\)O covered surface. The presented data for the O\(_2^+\) desorption yield were measured for a D\(_2\)O covered surface and with a photon energy of 58 eV. Similar measurements on H\(_2\)O ice and energies of 38 eV also show a nonlinear yield with n ~ 3, but occasionally even up to n ~ 10 [6].](image)

The nonlinear behaviour of desorbing molecular oxygen ions opens the way to nonlinear correlation measurements with an autocorrelator [7] using two FEL pulses synchronized on a femtosecond time scale. Highly oriented pyrolytic graphite (HOPG) surfaces covered with D\(_2\)O (~300 ML) serve as analoga for ice-covered interstellar dust particles. The graphite sample is mounted in UHV and cooled to 95 K. The UHV chamber was placed behind the beam splitter and delay unit at BL 3. It provides two beams delayed in time and overlapped in space. The focus was produced by a toroidal
mirror and had an ellipsional spotsize of 50 x 130 µm on the surface, due to an incident angle of 67.5° to the surface normal. The surface is irradiated with FLASH pulses and desorbing products are analyzed with a time-of-flight mass spectrometer. FLASH operated at $h\nu = 38.8$ eV with an average pulse energy of about $10 \pm 4 \, \mu J$. The pulse duration was about 30 fs and the FEL repetition rate 5 Hz. For different time delays the signal of the desorbing ions is recorded.

Figure 2: First time resolved desorption experiment with soft x-ray radiation at 32 nm (38 eV).

In figure 2 the dots show the signal of the desorbing ionic oxygen molecules as a function of delay time between the pump and the probe beam. Thereby the data represents the averaged data of several delay runs with 500 FLASH pulses per adjusted delay time. The continuous curve represents an preliminary estimate asymmetric Gaussian. The asymmetric Gaussian was fitted with two different parameter. For the rising edge a time constant of $\tau_{g1} = 370$ fs and for the falling slope a time constant of $\tau_{g2} = 2$ ps is used.

Due to the ~300 ML thick ice layer the FLASH pulses do not penetrate the graphite substrate. Therefore the reaction process occurs in the ice surface. The steep increasing edge points to an electronic processes initiating the reaction. The long decay time of the $O_2^-$ signal suggests a motion of molecular constituents during the reaction.

References