Atomic Photoionization and Molecular Dissociation Studies at FLASH Using Combined XUV Free Electron and Visible Laser Fields

M. Meyer
LIXAM, CNRS-UMR8624, Université Paris-Sud, Bât. 350, F-91405 Orsay, France

Ionization and dissociation are the dominant relaxation processes of atoms and molecules after interaction with photons of short wavelength. New possibilities to obtain dynamical information about these extremely fast processes were opened up in the last years due to the development of the XUV Free Electron Laser in Hamburg (FLASH) with its unprecedented characteristics, especially the short temporal width (about 20fs) and the extremely high number of photons per pulse (about 10^{12}-10^{13} photons/pulse).

In a series of experiments, we have used a two-color pump-probe set-up combining the intense and coherent XUV radiation from FLASH and strong synchronized IR/Visible laser pulses. In a first step, the Above Threshold Ionization (ATI) in rare gases was studied to characterize the FEL pulses, in particular the temporal resolution and stability of the set-up [1,2]. For the average mode, i.e. after integration of many pulses, an overall temporal resolution of about 600fs was determined, which is mainly caused by the jitter between the FEL and IR pulses. Using single shot analysis, experiments with a temporal resolution of better than 50fs are feasible [3].

By taking advantage of the monochromaticity and the short temporal width of the FLASH pulses, the two-color ATI could be investigated for the first time in a regime free from unwanted interference effects [4]. The polarization dependence of the sideband structures in the electron yields detailed insights into the photoionization dynamics, in particular into the distribution of angular momenta for the outgoing electrons. This can be achieved comparing the experimental results with theoretical treatments of the process employing second-order perturbation theory and the "soft-photon" approximation. Changes in the photoionization dynamics were followed systematically from the two-photon (low dressing field) to the multi-photon ATI regime (high dressing field).

In addition, the photodissociation of molecular hydrogen was investigated in a proof-of-principle experiment. Here, the FEL photons dissociate the molecules and the optical laser serves to probe excited atomic fragments via photoionization. By measuring the kinetic energy of the photoelectron as a function of the temporal delay between the XUV and optical pulses, the internal energy of the fragments, namely the production of atomic hydrogen with an excited electron in n=2 or n=3 orbitals, and their appearance time was determined. These first results demonstrate the capability of the two-color pump-probe set-up to perform time-resolved studies of molecular dissociation and represent the starting point for future work on more complex molecules.

References:

[1] M. Meyer, D. Cubaynes, P. O'Keeffe, H. Luna, P. Yeates, E. T. Kennedy, J. T. Costello, P. Orr, R. Taïeb, A. Maquet, S. Düsterer, P. Radcliffe, H. Redlin, A. Azima, E. Plönjes, and J. Feldhaus Phys. Rev. A 74, 011401(R) (2006).
[2] P. Radcliffe, S. Düsterer, A. Azima, W.B. Li, E. Plönjes, H. Redlin, J. Feldhaus, P. Nicolosi, L. Poletto, J. Dardis, J. P. Gutierrez, P. Hough, K. D. Kavanagh, E. T. Kennedy, H. Luna, P. Yeates, J. T. Costello, A. Delyseries, C. L. S. Lewis, D. Glijer, D. Cubaynes, and M. Meyer, Nucl. Instrum. Meth. A 583, 516 (2007).
[3] P. Radcliffe et al., Appl. Phys. Lett. 90, 131108 (2007).
[4] M. Meyer et al., submitted for publication (2008).