Time-Resolved Molecular Fragmentation at FLASH

Y.H. Jiang[1,*], A. Rudenko[2], M. Kurka[1], K.U. Kühnel[1], M. Toppin[1], O. Herrwerth[3], M. Lezius[3], M. Kling[3], L. Foucar[4], M. Schöffler[4], K. Cole[4], B. Ullrich[4], N. Neumann[4], R. Dörner[4], J. Pérez-Torres[5], E. Plésiat[5], F. Martín[5], A. Cassimi[6], S. Düsterer[7], R. Treusch[7], M. Gensch[7], C. D. Schröter[1], R. Moshammer[1], J. Ullrich[1,2]

[1] Max-Planck-Institut für Kernphysik, 69117 Heidelberg, Germany; [2] Max-Planck Advanced Study Group at CFEL, 22607 Hamburg, Germany; [3] Max-Planck-Institut für Quantenoptik, 85748 Garching, Germany; [4] Institut für Kernphysik, Universität Frankfurt, 60486 Frankfurt, Germany; [5]Departamento de Química C-9, Universidad Autónoma de Madrid, 28049, Spain; [6] CIRIL at Ganil, Caen, France; [7] DESY, 22607 Hamburg, Germany

Synopsis: Multiple ionization and fragmentation of D_2 and N_2 induced by absorption of few photons have been explored in pump-probe measurements at the free electron laser at Hamburg. Photoionization of aligned N_2^+ ions, produced in sequential step, is explored by inspecting the kinetic energy releases and the fragment-ion angular distributions as function of the pump-probe delay time.

Multiple ionization of molecules induced by absorption of few extreme ultra-violet (EUV) photons represents one of the most fundamental non-linear molecular processes. Free electron lasers, delivering coherent pulses of EUV-photons with femtosecond durations at unprecedented intensities, in combination with advanced multi-particle detection systems − the Heidelberg reaction microscope [1] – open a new era in molecular physics. Here we present first results of few photon induced fragmentation of D_2 and N_2, and EUV-pump/EUV-probe experiment.

The measurements were performed at photon energies of 38, 44 and 46 eV, with pulse durations of ≅30 fs, and intensities of $I \cong 10^{13}$ W/cm^2 at FLASH (the free-electron laser at Hamburg).

Sequential and direct two-photon double ionization (DI) of D_2 molecule is studied experimentally and theoretically at 38 eV [2]. Experimental and theoretical kinetic energy releases (KER) of D^++D^+ fragments, consisting of the contributions of sequential DI via the $D_2^+(1s\sigma_g)$ state and direct DI via a virtual state, agree well with each other. The modulation of KER spectra presents first experimental evidence on nuclear interference between direct and sequential DIs, where two photons are absorbed simultaneously and sequentially (with certain time-delay) within one pulse, respectively.

At 44 eV we studied multiple ionization of N_2 induced by absorption of up to 5 photons leading to dominant sequential ionization mechanisms [3]. For various intermediate charge states N_2^{n+} we find a considerable excess of photons absorbed compared to the minimum number that would energetically be required. Rich-structured patterns in the KER-dependent fragment ion angular distributions highlighting alignment-dependent multi-photon absorption and ionization dynamics allowing us to trace SI pathways in details for some of the channels.

Very recently, we succeeded in performing a pump-probe experiment on multiple ionization of N_2 using a multilayer spilt-mirror setup. In Fig. 1 the kinetic energies of N^{2+} ions, resulting from two dissociation channels, namely $N_2^{2+}\rightarrow N^{2+}+N$ (E_{KER}<6 eV) and $N_2^{3+}\rightarrow N^{2+}+N^+$ (E_{KER}>6 eV), are plotted vs. the pump-probe delay time (identical pulses, 46 eV). A very sharp structure (~10 fs FWHM) in the time trace for E_{KER}>10 eV at zero delay (Fig. 1b) might be due to highly nonlinear ionization. Further results and interpretations will be presented.

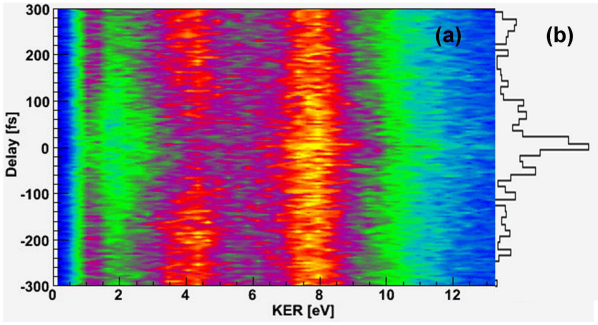

Fig. 1: **(**a) Kinetic Energies of N^{2+} fragments as function of the time delay between pump and probe pulse. (b) Time trace for events with E_{KER}>10 eV.

YHJ thanks for support from DFG project no JI 110/2-1. Production of multi-layer mirrors by the group of M. Vrakking is acknowledged.

References

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* E-mail: yuhai.jiang@mpi-hd.mpg.de