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Journal of Physics: Conference Series **194** (2009) 032057 doi:10.1088/1742-6596/194/3/032057

Sequential and direct two-photon double ionization of D_2 at FLASH

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Synopsis Sequential and direct two-photon double ionization (DI) of D_2 molecule is studied experimentally and theoretically at a photon energy of 38.8 eV. Experimental and theoretical kinetic energy releases 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.

Two-photon double ionization (DI) of H_2 or D_2 is of fundamental interest for studies of electron-electron and electron-ion correlations [1, 2]. Free electron lasers, delivering coherent pulses of EUV-photons with femtosecond durations at unprecedented intensities, in combination with advanced multi-particle detection systems − a reaction microscope [3] – open a new era exploring the dynamics of molecular ionization, dissociation, alignment, and nuclear wave packet propagation.

The measurements were performed at photon energies of 38.8±0.5 eV, with pulse durations of ≅30 fs, and intensities of $I \cong 10^{13}$ W/cm^2 at FLASH. Ionic fragments produced by the interaction with the light-pulse are projected by means of an electric field onto time- and position-sensitive MCP detectors. From the measured time-of-flights and positions of each individual fragment the initial 3D momentum vectors are reconstructed.

We have performed two model calculations in the framework of the Franck-Condon approximation in which (i) the two electrons are directly ionized (direct mechanism) and (ii) the two electrons are treated as independent particles and the two photons are absorbed sequentially by each electron (sequential mechanism). The probabilities resulting from both calculations have been added incoherently.

The kinetic energy release (KER) spectrum for coincident D^++ D^+ fragments as well as the angular distribution of the fragments (cosθ) as a function of KERs are plotted in Fig. 1(a) and (b), respectively. The experimental result is found to be well described by theoretical calculations in Fig. 1(b). The peak at E_{KER}=10 eV is populated by sequential DI after nuclear wave-packet motion in the $D_2^+(1s\sigma_g)$ state initiated by the first photon. Since the FEL pulse duration (~30 fs) is about four times longer than the vibration period (7~8 fs) of $D_2^+(1s\sigma_g)$, populated by absorption of the first photon the nuclear wave packet is able to move to the outer classical turning points, where the second photon is absorbed with larger probability. Two different ionization processes, namely sequential DI occurring in the FC regime, when the nuclear wave packet had not moved significantly, and direct DI via a virtual state, both are responsible for forming the second peak at E_{KER}=17.5 eV.

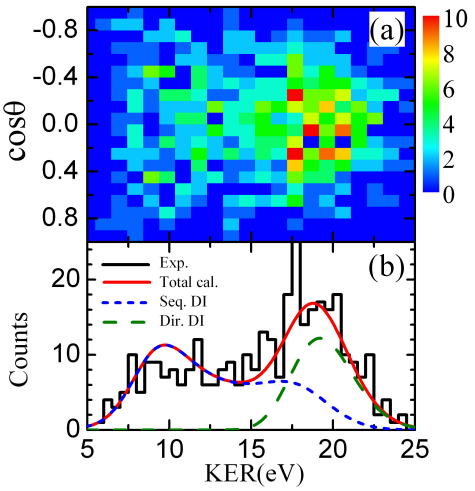

Fig.1. (a) angular distribution for the coincident fragments D^++D^+ and (b) KER spectra. θ is angle between the molecular axis and the light polarization.

Surprisingly, neither the doubly excited states of D_2 nor excited states of D_2^+ play a significant role. The reason is that more than 95% of one-photon ionization probability leads to the ground state of D_2^+.

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