Sequential and direct two-photon double ionization of D$_2$ by extreme ultraviolet Radiation at FLASH


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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 ≈ 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 is plotted in Fig. 1. The experimental result is found to be well described by theoretical calculations in Fig. 1. The peak at E$_{KER}$=10 eV is populated by sequential DI after nuclear wave-packet motion in the D$_2^+$ (1s$^2\sigma$) 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$^2\sigma$), 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. 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$_2^+$.

The modulation of the KER spectra presents first experimental evidence on the interference between direct and sequential DI channels, where two photons are absorbed simultaneously and sequentially (with certain time-delay) within one pulse, respectively. This kind of interference was predicted very recently to occur with not too short (>10 fs) and intensive EUV-pulse [4]. Detailed evaluation is under preparation [5].
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References