
01 Jan 2009

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Recommended Citation

"Sequential and Direct Two-Photon Double Ionization of D₂ at Flash," *Journal of Physics: Conference Series*, vol. 194, no. 3, Institute of Physics - IOP Publishing, Jan 2009.

The definitive version is available at <https://doi.org/10.1088/1742-6596/194/3/032057>

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To cite this article: Y H Jiang *et al* 2009 *J. Phys.: Conf. Ser.* **194** 032057

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Sequential and direct two-photon double ionization of D₂ at FLASH

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Synopsis Sequential and direct two-photon double ionization (DI) of D₂ 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 D₂⁺(1sσ_g) state and direct DI via a virtual state, agree well with each other.

Two-photon double ionization (DI) of H₂ or D₂ 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 ≃ 10¹³ W/cm² 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 D₂⁺(1sσ_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 D₂⁺(1sσ_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.

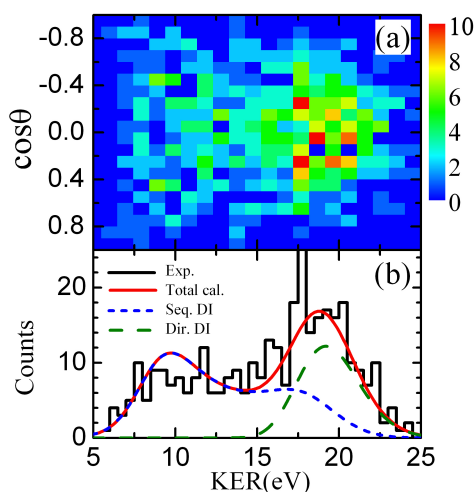


Fig.1. (a) angular distribution for the coincident fragments D⁺+D⁺ and (b) KER spectra. θ is angle between the molecular axis and the light polarization.

Surprisingly, neither the doubly excited states of D₂ nor excited states of D₂⁺ play a significant role. The reason is that more than 95% of one-photon ionization probability leads to the ground state of D₂⁺.

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