Control of H_2 and D_2 dissociative ionization in the non-linear regime using EUV femtosecond pulses @FERMI

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Synopsis Two-photon ionization via selected vibrational levels in neutral H_2 and D_2 intermediate states is demonstrated to control the dissociative ionization rate with femtosecond FEL pulses.

H_2 and D_2 molecules were photoionized in a non-linear 2-photon process via resonantly excited intermediate states thanks to the unique properties FERMI free electron laser (FEL) providing bright, coherent and tunable EUV femtosecond (fs) pulses. Selective excitation of the vibrationally resolved H_2*($B\ ^1\Sigma_u^+$, v=8-17) and H_2*($C\ ^1\Pi_u$, v=2-4) states using 100 fs FEL pulses in the 12-15 eV range made it possible to investigate the influence of the nuclear degree of freedom (DOF) on the outcome of photoionization: depending on the intermediate state's internuclear distance expansion, absorption of the second photon leads to dissociative (DI) and/or non-dissociative ionization (NDI).

Time-of-flight (TOF) mass spectrometry, together with electron and ion velocity map imaging (VMI), were employed on the LDM beamline [1] to analyze the DI/NDI ratio, kinetic energy spectra and angular distributions of photoelectrons and H^+ fragment ions. DI was observed to be enhanced drastically by up to two orders of magnitude compared to one-photon ionization, as predicted by time-dependent Schrödinger equation calculations for 1-10 fs pulses and accounting for the DOF of electrons and nuclei [2,3]. Excitation of isolated H_2*($B\ ^1\Sigma_u^+$,v) levels even leads to situations where DI dominates NDI, which is very unusual for valence shell ionization. For such cases, clear oscillations in the ion fragment energy spectra are discerned, which reflect a projection of the intermediate state vibrational wave function onto the ionic $H_2^+(2p\sigma_u)$ dissociative state. The photoelectron spectra feature the analogous structures for electrons correlated to DI. The latter spectra moreover yield access to the vibrational distribution of the $H_2^+(X\ ^2\Sigma_g)$ ground state for NDI. As the distribution is quite sensitive to the vibronic intermediate state, it provides an additional probe of the induced nuclear dynamics. New calculations based on time-dependent second order perturbation theory, well adapted to the 100 fs EUV pulses as utilized in the experiment, are in progress.

The remarkable anisotropies observed in the angular distributions complement the characterization of the photoionization dynamics. Furthermore, the electron angular distribution provides a sensitive means to characterize quantum interferences resulting from the coherent superposition of indistinguishable reaction paths. The study was extended to D_2 also and, beyond, a first experiment supports the feasibility for coherent control [4] of NDI involving two-EUV-photon resonant and one-XUV-photon non-resonant channels, relying on the variable controlled phase available at FERMI between the fundamental and second harmonic FEL pulses.

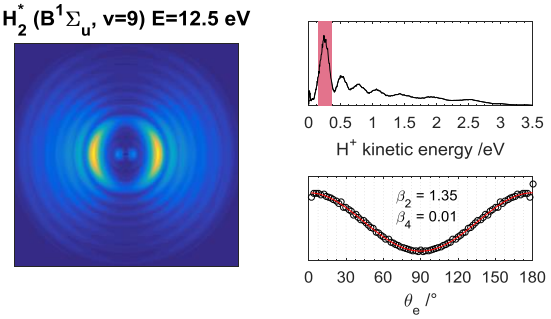

Figure 1. Inverted VMI, kinetic energy spectrum and angular distribution (highlighted band) for H^+ ions from 2-photon ionization via H_2^* ($B\ ^1\Sigma_u$, v=9).

References

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