

Molecular interferometer using XUV attosecond pulses to unravel electron and nuclear dynamics

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Synopsis Two identical XUV attosecond pulses interact with the hydrogen molecule creating an interferometer resulting from the direct and sequential two-photon absorption paths reaching the same final ionized states. The dependence of the ionization yields with the time delay between the pulses allows to reconstruct the pumped vibronic (electronic and vibrational) wave packet created in the singly excited states of the molecule. The use of XUV pulses avoids a laser-induced distortion of the molecular potential, ensuring the characterization of the intrinsic behaviour of the system.

Monitoring and controlling electron dynamics in atoms and molecules requires the use of short and intense enough pulses. Significant progresses have been made using XUV-pump/IR-probe schemes, whose applications range from creating interferometers in atoms to reconstruct an electron wave packet [1], to capturing time-resolved images of hole migration in molecules [2]. Despite these unquestionable achievements, the use of IR fields usually distorts the molecular potential and shorter-wavelength pulses are desirable to avoid the modification of the intrinsic behaviour of the system under study.

In the present theoretical work, we propose the use of a XUV-pump/XUV-probe scheme in the hydrogen molecule that allows for a full characterization of the pumped vibronic wave packet [3, 4]. The molecule is illuminated with identical XUV pulses with a given delay, creating an interferometric signal due to the electron ejection through both two-photon direct and sequential absorption processes reaching the same final state. We demonstrate that such a scheme allows us to unravel the coupled electron and nuclear dynamics, extracting both amplitudes and phases of the pumped vibronic wave packet. Figure 1 shows the dissociative ionization probability as a function of the time delay between two identical pulses of 2 fs duration and centered at 14 eV energy. A short-time Fourier transform of the ionization signal, with a proper choice of the time window to obtain a good balance in energy (y-axis) and time (x-axis) resolution, allows to disentangle two well defined components in the pumped wave packet. The probability localizes around 12 eV and around 14 eV of total absorbed energy revealing two different electronic states, each containing a nuclear wave packet (NWP). The averaged vibrational periods associated to each NWP (around 20 and 30 fs, respectively)

are also captured. Finally, we also show that while total ionization yields reveal the entangled electronic and nuclear dynamics associated to the singly excited states, doubly differential yields (differential in both electronic and nuclear energies) exhibit in addition the dynamics of autoionization, i.e., of electron correlation in the ionization continuum.

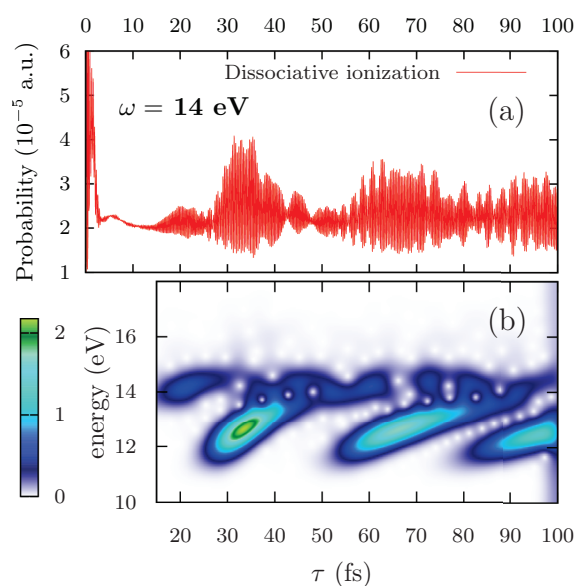


Figure 1. (A) Dissociative ionization probability as a function of the time delay between two XUV pulses. (B) Short-time Fourier transform of the ionization probability in A.

References

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