Attosecond photoionization of a coherent superposition of bound and dissociative molecular states: effect of nuclear motion

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Synopsis We study numerically the possibility for monitoring electron motion in a dissociating molecule using an attosecond XUV probe pulse which photoionizes a coherent superposition of two nuclear wave packets prepared using a femtosecond pump pulse. We present the photoelectron spectra and forward-backward asymmetries in these spectra obtained from a numerical solution of the time-dependent Schrödinger equation.

Ultrashort flashes of light allow us to take snapshots of microscopic structures and let us to reconstruct their motion. Recently, pulses as short as 80 attoseconds have been obtained [1]. At such time scale monitoring the electron motion becomes in principle possible. A simplest scheme allowing to watch the electron motion inside the atom using two laser pulses [2, 3] relies on the preparation of the coherent two bound electronic states (e.g. 1s+2s or 1s+$2p_0$ states of the hydrogen atom) followed by an attosecond pulse which photoionizes the atom. Measuring the photoionization signal as function of the time-delay between two pulses allows to "watch" the oscillating electron.

Recently, we have investigated such schemes for monitoring the electron motion in H_2^+ at a fixed internuclear distance R [4, 5]. We found that in the case when both states have the opposite parity (like 1s and $2p_0$ states in the hydrogen atom) a particularly convenient way for watching those structures is via the measurement of the photoelectron asymmetries as function of the pump-probe delay time, which is a normalized difference between the photoelectron signal for the momentum $\mathbf{p}$ and $-\mathbf{p}$.

All previous theoretical studies (except [6]) of attosecond photoionization of a coherent superposition in molecules were restricted to the molecules fixed at a internuclear distance R and the preparation stage using pump pulse was not included in the dynamics at all. We include the nuclear motion in both stages, i.e. in the preparation of the superposition and in photoionization, by solving numerically the time-dependent Schrödinger equation (TDSE) for a molecular ion H_2^+ in 1-D interacting with both the pump and the probe pulse. More specifically, we consider two cases of coherent superpositions, case (I): initial vibrationial bound state superposed on a dissociative upper state prepared with a resonant pump pulse and case (II): a superposition of two dissociating wave packets prepared by two pump pulses. We show that the asymmetries in photoelectron spectra in both cases exhibit oscillations as function of the time delay between XUV probe and the pump pulse, as expected from the model with fixed nuclei. In case (I), the oscillations disappear when the dissociating wave packet looses the overlap with the initial state. In case (II), if one achieves the synchronization of the movement of two dissociating overlapping wave packets (using two laser pump pulse), the oscillations persist for much longer time delays than in case (I) despite the fast nuclear motion in the H_2^+ molecule.

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