Clocking ultrafast wave packet dynamics in H_2 by using UV pump - UV probe schemes

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A UV pump - UV probe scheme is used to trace the evolution of nuclear wave packets (NWPs) in excited molecular states by measuring the time-delay dependence of ionization probabilities. We present theoretical calculations performed in H_2, which allows a full dimensional treatment including all electronic and nuclear degrees of freedom. Two identical UV pulses of a few fs of duration constitute a typical UV pump - UV probe scheme. The photon energy is chosen such that the pump pulse excites the system in the lowest single excited state of the neutral and the probe pulse will ionize system. Two-photon ionization is the major channel leaving the ion in both its ground ($1s\sigma_g$) and its first excited state ($2p\sigma_u$). As expected, the proton kinetic energy release distributions vary with the time-delay between the pulses, but any signature of the pumped wave packet is hardly visible. However, a direct map is obtained by analyzing the asymmetry of the electron angular distributions resulting from dissociative ionization. The asymmetry results from the coherent superposition of gerade and ungerade states of the remaining molecular ion in the region where the pumped NWP is located. The variation of this asymmetry with the time delay between the pump and the probe pulses thus parallels that of the moving wave packet and, consequently, can be used to clock its field-free evolution [1].

We seek to use similar UV pump - UV probe schemes to extract information on the autoionization processes on H_2. These are triggered upon populating optically forbidden doubly excited states, accessible through two-photon transitions from the ground [2]. The ionization probabilities differential in both electron and nuclear kinetic energies reveal complex patterns that result from the interferences between several two-photon ionization paths: time-delayed replicas of direct ionization and autoionization upon two-photon absorption from the ground state, and one-photon ionization triggered from the time-evolved NWP in the excited state. We demonstrate that despite the complexity of these patterns, most of the contributions from different ionization paths can be easily disentangled from a fine scan in time-delays by using simple analytical expressions.

References:

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