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Clocking Ultrafast Wave Packet Dynamics in Molecules through UV-induced Symmetry Breaking

ALICIA PALACIOS, Universidad Autónoma de Madrid

A UV pump - UV probe scheme is used to trace the evolution of nuclear wave packets in excited molecular states. 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 nuclear wave packet 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. The hydrogen molecule is used as benchmark target, which is represented within the Born-Oppenheimer approximation including all electronic, including correlation, 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 states 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 s g) and its first excited state (2p s u). As expected, the proton kinetic energy release (KER) distributions vary with the time-delay between the pulses, but any signature of the pumped wave packet is hardly visible. However, the superposition of the gerade (1s s g) and ungerade (2p s u) electronic states induces an asymmetry in the angular distributions, whose dependence with time delay leads to a sharp image of the time evolution of the wave packet.