The onset of asymmetry in electron angular distributions for dissociative photoionization of H_2 by ultrashort xuv laser pulses

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Synopsis A method to compute angular distributions for electrons ejected from fixed-in-space molecular hydrogen when exposed to ultrashort xuv laser pulses is proposed based on the ab initio solution of the time-dependent Schrödinger equation. This method allows for a temporal picture of the origin of the asymmetry in the electron angular ejection that arise due to the interference of the two symmetric dissociative ionization channels ($1s\sigma_g$ and $2p\sigma_u$) in which molecular autoionization states decay after photon absorption.

The recent arrival of COLTRIMS reaction microscopes [1] allows for kinematically complete experiments in which the momentum of all ejected photofragments is measured in coincidence after the breakup caused by a laser pulse. For the dissociative photoionization of molecular hydrogen ($H_2+\hbar\omega \rightarrow H^+ + H + e^-$), COLTRIMS gives access to measure proton kinetic energy distributions (KEDs) and photoelectron angular distributions (PADs) associated to a given molecular orientation with respect to the laser polarization direction. This technique is abel to detect the direction along the molecular axis in which the proton H^+ is emitted. This means that by detecting the localization of the ejected H^+, COLTRIMS does not respect the inversion symmetry of the molecule. To satisfy the conditions imposed by the experiment, the aymptotic behaviour of the molecular wavefunction must be modified, in particular when two ionization channels (for instance H_2^+ $1s\sigma_g$ and $2p\sigma_u$) are degenerated at $R\rightarrow\infty$. In our time-dependent method [2], the H_2 wavefunction is expanded in terms of vibronic eigenstates including the continuum for the two H_2^+ ionization channels $1s\sigma_g$ and $2p\sigma_u$. To satisfy the localization condition for the proton emission, the final wavepacket at $t\rightarrow\infty$ (long after the laser pulse of duration T is ended) must be projected onto the correct asymptotic function which results into a entanglement of the amplitudes coming from $1s\sigma_g$ and $2p\sigma_u$ ionization channels and ultimately producing asymmetries in the PADs with respect to nuclei inversion.

Figure 1 shows proton KEDs and PADs for protons emitted at 90^0 with respect to the laser polarization vector for some proton KEDs. The symmetry breaking of PADs emerges along the pulse duration for proton KEDs at ~ 4 eV where the $1s\sigma_g$ and $2p\sigma_u$ contributions interfere, in agreement with stationary results [3].

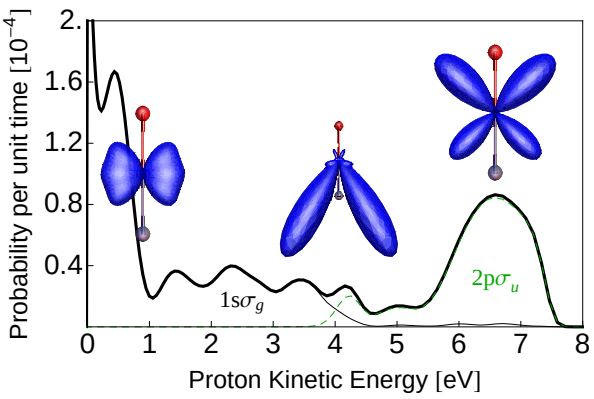

Fig. 1. H_2 exposed to a xuv laser pulse with photon energy E=33eV, intensity I=10^{12} W/cm^2 and duration T=10 fs. Thick solid line: Total ionization probability, thin solid line: contribution from $1s\sigma_g$ ionization channel, thin dashed line: from $2p\sigma_u$ ionization channel. Insets: PADs (in blue) for protons (upper red spheres) measured at 90^0 with respect to polarization axis corresponding to proton kinetic energies 0.9, 4.1 and 6.6 eV.

References

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