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Using ultrashort xuv laser pulses to investigate symmetry breaking in one-photon single-ionization of H₂.

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Synopsis We have evaluated photoelectron angular distributions from fixed-in-space molecular hydrogen exposed to ultrashort xuv laser pulses. The theoretical method is based on the solution of the time-dependent Schrödinger equation in a basis of stationary states that include all electronic and vibrational degrees of freedom. We conclude that the origin of the asymmetry in these angular distributions is the interference of the two dissociative ionization channels ($1s\sigma_g$ and $2p\sigma_u$) due to delayed ionization from the H₂ doubly excited states.

Kinematically complete experiments [1] have recently been performed to study dissociative photoionization of molecular hydrogen ($\text{H}_2 + \hbar\omega \rightarrow \text{H}^+ + \text{H} + e^-$) by using synchrotron radiation [2]. In particular, proton kinetic energy distributions (KEDs) and photoelectron angular distributions (PADs) associated to a given molecular orientation (molecular frame) with respect to the laser polarization direction (laboratory frame) have been measured. An interesting finding in the experiment reported in [2] is that electron angular distributions are asymmetric when the photon energy is large enough to populate the H₂ doubly excited states. A subtle point in these experiments is that, by determining the direction of the emitted protons, the molecular inversion symmetry is not respected in the measurement. However, in the absence of doubly excited states, the angular distributions are always symmetric.

We have considered a xuv pulse of 33 eV and durations between 1 and 10 fs. The TDSE is solved by expanding the time-dependent wave function $\Psi(\mathbf{r}_1, \mathbf{r}_2, R, t)$ in a basis of H₂ vibronic eigenstates that include bound states, doubly excited states and the continuum states associated with the $1s\sigma_g$ and $2p\sigma_u$ ionization channels [3]. To impose the boundary conditions of the experiment, we have projected $\Psi(\mathbf{r}_1, \mathbf{r}_2, R, t)$ onto stationary continuum states that localize the remaining electron in given proton. At $t \rightarrow \infty$, the latter states are simply a linear combinations of of the H₂⁺ states associated with the $1s\sigma_g$ and $2p\sigma_u$ ionization channels.

Figure 1 shows proton KEDs and PADs for a time $t > T = 10$ fs associated to protons emitted at 90° with respect to the laser polarization vector. In agreement with reference [2], the angular distributions are very asymmetric in a wide

range of proton kinetic energies. The analysis of these results with time show that the angular distributions are totally symmetric at short times and only become asymmetric when autoionization starts a few fs later. Autoionization leads to electrons with equal energy in both the $1s\sigma_g$ and $2p\sigma_u$ channels, which then interfere thus leading to the asymmetry of the angular distributions.

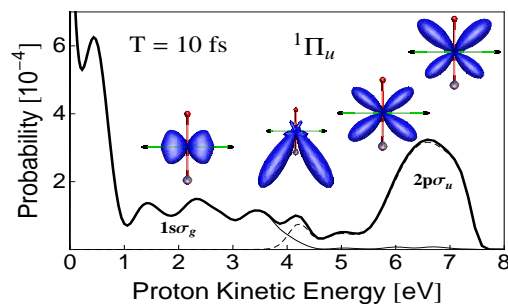


Fig. 1. H₂ exposed to a xuv laser pulse with photon energy $E=33$ eV, intensity $I=10^{12}$ W/cm² and duration $T=10$ fs. Thick solid line: Total ionization probability, thin solid line: contribution from $1s\sigma_g$ ionization channel, thin dashed line: from $2p\sigma_u$ ionization channel. Insets: PADs (in blue) for protons (upper red spheres) measured at 90° with respect to polarization axis (green arrows) corresponding to proton kinetic energies 2.10, 4.15, 5.65 and 7.10 eV.

References

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