Signatures of Light-Induced Potential Energy Surfaces in H_2^+

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Synopsis Using theory and Cold Target Recoil Ion Momentum Spectroscopy we find signatures of light-induced molecular potential energy surfaces in the 3-dimensional proton momentum distributions of dissociating H_2^+.

H_2 continues to provide fundamental insights into the mechanisms of intense light-matter interactions [1]. Recently, there has been significant interest in so-called light-induced conical intersections (LICI) that arise from the angle dependence of the single-photon coupling of electronic states. Analogously to regular conical intersections, electronic and nuclear motions are strongly coupled in the vicinity of LICIs. As a signature of such rovibronic dynamics, weak modulations in the angular distribution of protons emitted from H_2^+ have been reported [2]. More generally speaking, infrared (IR) laser pulses couple the σ_g and σ_u electronic states of H_2^+ typically through several pathways involving an odd number of photons [1], which can produce complex light-induced potential energy landscapes, and consequently even richer dynamics.

Here, we report the observation of strongly structured proton angular distributions obtained from the dissociative ionization of H_2. Our experiment relies on the STIER (Sub-cycle Tracing of Ionization Enabled by infraRed) technique [3], where an intense few-cycle visible pulse ionizes H_2 to H_2^+. The cation subsequently dissociates under the influence of a moderately intense mid-IR (2300 nm) pulse, which is polarized perpendicularly to the visible pulse and does not cause ionization of H_2 on its own. The 3-dimensional momentum of the ejected protons were recorded using COLTRIMS (cold target recoil ion momentum spectroscopy). Figure 1 a) shows the proton distribution produced in a few-cycle, 800 nm laser pulse. In Figure 1b) a cross-polarized, multi-cycle, 2300 nm pulse is superimposed. Strikingly, the weak mid-IR field does not only add a contribution along its polarization axis, but also produces angular features closer to the polarization of the visible laser pulse. Using numerical solutions of the time-dependent Schrdinger equation, we explain the features of the measured momentum distribution as an angle-dependent competition of different dissociation pathways in a complex light-induced potential energy landscape [4].

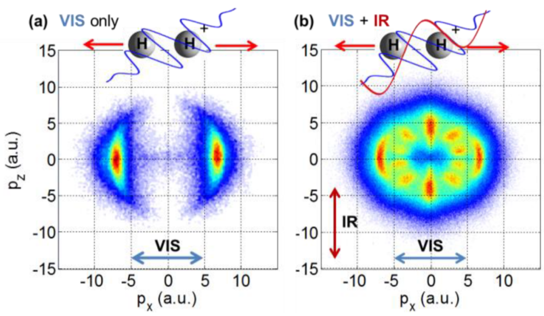

Figure 1. Measured proton momentum distributions produced by (a) a visible (730 nm) few-cycle pulse only and (b) and an additional mid-IR (2300 nm) dressing field. The arrows indicate the laser polarization.

References

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