SIMULTANEOUS TRACKING OF TWO COHERENTLY LAUNCHED WAVE PACKETS IN REAL TIME USING SHORT INTENSE LASER PULSES

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The availability of extremely short intense laser pulses makes it now possible to follow in real time the heavy particle dynamics of even the simplest and lightest molecules. A non-stationary wave packet can be launched onto a potential curve of the molecule through ionization of the neutral, and the subsequent motion of this time-dependent wave packet can be followed as it coherently couples to other potential curves in the ionized system.

We have used intense laser pulses in a pump-probe configuration to track the vibrational motion of the wave packet launched onto the ground state potential curve of the H_2^+ molecule from neutral H_2. A short "weak" ($<1x10^{14}$ W/cm^2, 8-12 fs) ionizing pulse launches the packet onto the $1s\sigma_g$ potential curve of H_2^+. After a time delay ranging from 0 to 100 fs, a second "strong" pulse removes the remaining electron, causing the system to "Coulomb explode" into two protons. These protons are detected in time coincidence, and their momenta are measured, using well established COLTRIMS techniques.

Figure 1 shows the kinetic energy release of the two protons (KER) plotted as a function of the time delay between the pump and probe pulses. Two "trajectories" of the wave packet are seen. The first is an oscillatory one producing a KER near 10 eV, and is due to that part of the vibrational wave packet which remains bound on the $1s\sigma_g$ potential curve. The period of oscillation is characteristic of motion in that well, approximately 15 fs. The second trajectory shows a monotonically descending KER as a function of time and represents the wave packet which follows the $2p\sigma_u$ dissociating potential curve. The population of this trajectory occurs primarily near the crossing between the single-photon-promoted $1s\sigma_g$ potential curve and the $2p\sigma_u$ curve, which occurs near an internuclear distance of 4.6 a.u., and gives rise to the well known bond-softening KER in dissociation of H_2^+.

A quantum-mechanical model of this process has been constructed by launching a wave packet characteristic of the H_2 ground state onto the $1s\sigma_g$ potential curve and propagating this wave packet coherently on the (coupled) $1s\sigma_g$ and $2p\sigma_u$ curves of the H_2^+. Excellent agreement between the experiment and model is found. In particular, non-classical behavior of the shapes of the wave packets is seen in several spectra (not presented here) and is in agreement with model calculations.

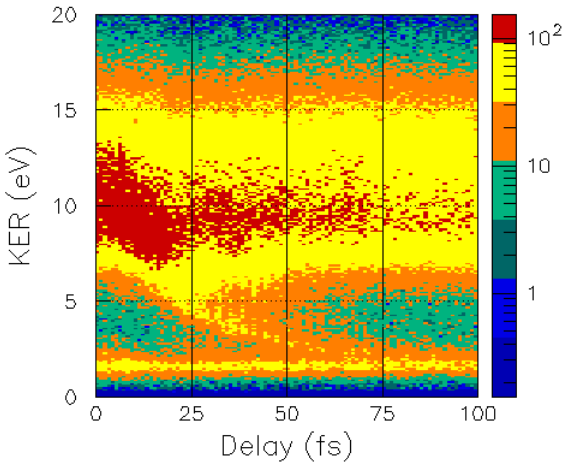

Figure 1. Plot of KER from H_2^+ as a function of pump-probe time separation.