Phase and population control of a vibrating wavepacket

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Synopsis We present experimental evidence of the manipulation of vibrational phase and population by light-induced dynamic potential surface deformation. A vibrational wavepacket is created in D_2^+ in a 10 fs NIR pump pulse; the evolution of the wavepacket is non-destructively modified by a control pulse. Depending on the pump-control delay, population is up- or down-shifted and the relative phase delayed or advanced with respect to the unperturbed motion.

Substantial efforts have been made to control the motion of vibrational wavepackets, with applications in quantum computation, chemical reactivity and actively steering molecular structure. The challenge in engineering the vibrational state of a molecule is two-fold: not to destroy the quantum system during manipulation and to quantify the evolution of the system.

Three NIR few-cycle (10 fs) laser pulses of moderate intensity generate, control and image a vibrational wavepacket in deuterium. First, a 'pump' pulse of intensity $I \simeq 10^{14}$ Wcm^{-2} causes ionization $D_2 \rightarrow D_2^+$, populating a range of vibrational states. The ionization step launches the vibrational wavepacket on the D_2^+ potential energy surface (PES). The propagation of a wavepacket on such a PES has been observed by the authors and other groups [1, 2], and is well understood theoretically [3, 4].

The 'control' pulse ($I \simeq 3 \times 10^{13}$ Wcm^{-2}) then dynamically distorts the PES through the Stark effect and the dipole transition; depending on the intensity and time of application of the control pulse, the varying potential gradient causes vibrational components to phase shift without destroying the coherence of the wavepacket. Population transfer is also possible: the deformation chirps the oscillation frequencies, analogous to a Raman interaction induced by the applied field. Changing the pump-control separation phase shifts different components, and selectively transfers population up or down the available vibrational states.

The final 'probe' pulse has an intensity $I \simeq 3 \times 10^{14}$ Wcm^{-2}; the influence of the control pulse is experimentally observed by recording the dissociation yield as the pump-probe delay is scanned for a range of pump-control delays.

At the conference we will present a quantitative comparison between observations of control-moderated wavepacket motion and a novel quasi-classical model (QCM). The wavepacket is represented by an ensemble of classical trajectories propagating on the PES; the dynamic deformation of the molecular bond by the control pulse is readily incorporated. Comparisons between the QCM and quantum mechanical solutions [3] for identical optical conditions indicate the QCM is a valid description of the system.

The agreement between the experimental observations and the QCM has implications for the dynamic control of molecular structure. The QCM is readily extended to complex chemically and biologically significant systems, intractable to more widely applied theoretical methods. Furthermore, the NIR pump could be replaced by a UV attosecond pulse, and multiple control pulses employed. The QCM could easily be adapted to such conditions, offering a route to exploring non-BOA molecular dynamics.

References

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