Time-Resolved Imaging of Strong-Field-Induced Nuclear Wave Packets in Halomethane Molecules

A Rudenko

1 J.R. Macdonald Laboratory, Department of Physics, 116 Cardwell Hall 66506, Manhattan KS, USA.
Contact Phone: +16502848076
Contact Email: rudenko@phys.ksu.edu

Light-driven nuclear wave packets play an important role in molecular imaging and coherent control applications. Currently, there is significant interest in extending efficient wave packet measurement schemes from diatomics to simple polyatomic molecules [1]. In particular, nuclear dynamics in halomethanes have attracted considerable attention since they often serve as prototype systems for ultrafast x-ray studies [2,3], laser-controlled chemistry (e.g., selective bond breaking, concerted elimination reactions or light-induced conical intersections) [4,5], and are important objects in atmospheric chemistry. Here, we combine a femtosecond laser pump-probe setup with coincident 3D ion momentum imaging apparatus to study strong-field induced nuclear dynamics in several halomethane molecules (CH3I, CH2I2, CHIICl).

We apply a time-resolved Coulomb explosion imaging technique to map the nuclear motion on both, bound and continuum potential surfaces, disentangle different fragmentation pathways and observe clear signatures of vibrational wave packets in neutral and ionic states. Channel-selective and kinetic-energy resolved Fourier analysis of these data allows for unique identification of different electronic states and vibrational modes responsible for a particular structure. For iodomethane (CH3I), we observe the signatures of vibrational motion in the ground state of the neutral molecule, as well as in the ground and the first excited states of the cation. We found the degeneration of the oscillatory structures from the cationic states within ∼2 ps, analyze most likely reasons for such behavior (dephasing, coupling to different vibrational modes), and discuss possible mechanisms of vibrational excitation in the neutral molecule. For diiodomethane (CH2I2) we observe signatures of both, bending and stretching vibrations, and reveal correlation between bending motion (the I-C-I “scissors” mode) and different fragmentation pathways, including I2+ elimination. Furthermore, using three-body breakup, we disentangle different fragmentation pathways and trace the time evolution of both, bond lengths and angles for major reaction channels.

References