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Single Pulse Time Resolved Four Wave Mixing YEHIAM PRIOR, YURI PASKOVER, ILYA SH. AVERBUKH, Department of Chemical Physics, Weizmann Institute of Science, Rehovot, Israel 76100 — We present a new experimental technique for single-shot time resolved ultrafast Coherent Anti-Stokes Raman Spectroscopy (CARS), where we use the arrival time of pulses at the intersection of broad beams as controlled time delays. The three dimensional (Boxcars) phase-matching configuration allows unique mapping of two independent time delays (pump-Stokes and pump-probe) onto the geometrical axes of the interaction region. The signal emitted from each point of the beams' intersection carries information on the molecular state at the particular time delay after the excitation, and thus the spatial profile of the CARS beam provides time resolved trace of the intra-molecular vibrational dynamics. We show that our technique allows for capturing of a few picoseconds of vibrational evolution by means of a single ultrashort pulse. Moreover, the ability to resolve two time delays between pulses enables us to study vibrational dynamics on the ground and the excited electronic states, as well as the correlation between the nuclear motions within these different vibrational potentials. The ability to record vibrational dynamics while exposing the molecules to a single optical pulse allows for characterization of short living and unstable chemical species as transitional complexes of chemical reactions.

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