PROBING CHANGES IN THE SOLVENT MOTIONS DURING CHEMICAL DYNAMICS IN SOLUTION

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The dynamics of the local solvent environment are studied during chemically relevant events using the second-generation of resonant pump third-order Raman probe spectroscopy (RaPTORS). This two dimensional spectroscopy employs a nonresonant third-order Raman probe to track the changes in the complete intermolecular response as a function of time after photo-initiated dynamics. Several improvements have been made to the RaPTORS experiment in the current iteration. Although a diffractive optic (DO) is used to create a phase locked local oscillator (LO), the LO has been spatially separated from the beam crossing. This eliminates any unwanted pump-probe signals resulting from interactions between the LO and any of the other four interacting pulses. The non-collinear LO and signal are combined on a second DO, where the relative phase is controlled by translation of the DO normal to the LO. This method of phase selection creates a down-gearing of the phase, with a 2.5 cm translation of the DO rotating the relative phase at 800 nm by 2π, and provides stability of $> \frac{\lambda}{200}$ over the course of several hours. Signals are collected via a balanced detection scheme that provides single shot subtraction of two phase-cycled signals.