MONDAY MORNING, /03/20/10 –

Session A15. DCP: Multidimensional Spectroscopies I

Monday morning, 08:00, 205B, MCC

08:00 A15.001 Plyler Prize Talk MICHAEL D. FAYER, Department of Chemistry, Stanford University — A variety of ultrafast optical and infrared non-linear experiments used to study dynamics in condensed matter systems are described. The methods include transient grating techniques, photon echo and related pulse sequences, optical Kerr effect experiments, and infrared vibrational echo methods. Past and current investigations will be discussed. Transient grating methods have been applied to the study of acoustic wave dynamics in crystals, liquids, and thin films; transport processes such as thermal diffusion in solids, films, and chemical species in gases and flames; and electronic excited state dynamics in solids, liquids and gases. Ultrafast photon echo methods and related pulse sequences were developed and applied to the study of dynamics in crystals and glasses. Ultrafast non-linear optical Kerr effect techniques are used to study dynamics in complex liquids such as liquid crystals and supercooled liquids. Ultrafast infrared pulses from a free electron laser and from optical parametric amplifier systems are being used to perform vibrational echo studies of dynamics in liquids glasses and proteins. The vibrational echo, the vibrational equivalent of the NMR spin echo and the electronic excited state photon echo, is also used in a type of two dimensional spectroscopy.

08:36 A15.002 Inertial and Diffusive Dynamics in Liquids by Ultrafast Nonlinear Spectroscopy , MARK A. BERG, Department of Chemistry and Biochemistry, University of South Carolina, Columbia, SC 29208 — The differences and relationships between the inertial and diffusive components of liquid dynamics are proving to be essential for understanding a multitude of processes in solution. Simple viscoelastic models treat these two components in a unified fashion and can provide qualitative insight into and even quantitative interpretations of nonlinear experiments. Experiments on vibrational dephasing in supercooled liquids will be presented and interpreted with a viscoelastic model. Interaction of the vibration with diffusive motion of the solvent is found to play a major role in vibrational dephasing. In addition, connections between vibrational and electronic dephasing are predicted by viscoelastic models. Experimental tests of these predictions will be presented.

09:12 A15.003 The Vibrational Echo in Liquid Solution , ROGER F. LORING, Department of Chemistry and Chemical Biology, Cornell University, Ithaca, NY 14853 — The vibrational echo is a nonlinear optical analog of the spin echo of magnetic resonance that probes molecular vibrational dynamics. In solids, the measurement has the capacity to distinguish between static and dynamic line broadening mechanisms. In liquids, with the absence of a sharp separation of time scales among line broadening mechanisms, the measurement can discriminate among dynamical processes according to their time scales. We present an algorithm for calculating the vibrational echo in a system governed by classical mechanics. This procedure is applied to the dephasing dynamics of molecular vibrations in liquid solution.

09:48 A15.004 Diffractive Optics Based Six-Wave Mixing Studies of Liquid Dynamics , R. J. DWAYNE MILLER[1], Departments of Chemistry and Physics,University of Toronto — Femtosecond Two-Dimensional Raman Spectroscopy harbors great promise in revealing important details related to the intermolecular potential of the liquid state. The lowest order nonlinear response capable of accessing this information is fifth order and is down several orders of magnitude in signal amplitude relative to third order processes. The small signal and competing lower order cascaded signals make this experiment difficult to implement. This talk will discuss the application of diffractive optics to provide a single optic approach to generate the relatively complex beam geometries for this class of six-wave mixing experiments. This approach provides automatic phase matching for ultrafast pulses over a broad bandwidth and passive phase locking of all six interacting fields for implementing heterodyne detection for signal enhancement. To provide greater background suppression, a diffractive optic was designed to enable 2-color pump and probe pulse pairs. The fifth order response of liquid CS_2 was studied by comparing two different phase matching geometries with different degrees of supression of lower order responses. The experiments demonstrate the utility of diffractive optics in this application and the possibility of advancing this method to the study of more structured liquids.

[1]V. Astinov, K. J. Kubarych, C. J. Milne, and R. J. D. Miller

10:24 A15.005 'Direct and Cascaded Events in Six-Wave Mixing with Femtosecond and with Incoherent Light.' , ANDREAS C. ALBRECHT, Cornell University — This abstract not available.