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Memory in 2D FT Spectra of Quantum Dots SAMUEL PARK, DMITRY BARANOV, BYUNGMOON CHO, TREVOR COURTNEY, DAVID JONAS, Univ of Colorado - Boulder — We have used the first femtosecond 2DFT spectrometer in the short-wave infrared to record 2DFT spectra of the polar dye IR26 in dichloroethane. The 2DFT spectra of IR26 at early mixing times shows a diagonally elongated positive peak, which reflects the strong correlation between excitation and detection frequencies. The peak also has a slight shift above the diagonal and an off-diagonal negative region that is indicative of vibrational and solvent frequency memory (the finite timescale for frequency shifts from inertial solvation). Nearly all correlation is gone at long mixing times and the 2D spectra approach a product lineshape. We also measured the first 2DFT spectra of oleate-capped colloidal PbSe quantum dots in tetrachloroethylene in the short-wave infrared region. These measure a bi-exciton binding energy that is consistent with prior spectrally resolved pump-probe experiments. Most interestingly, certain similarities between the 2DFT spectra of IR26 and quantum dots at early mixing times points towards coherent phonon/solvent memory; calculations by Prezhdo and co-workers both predicted phonon memory in quantum dots and indicated that it affects carrier relaxation. The analysis and implications of these results will be discussed.

Samuel Park
Univ of Colorado - Boulder

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