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Two-dimensional Fourier transform electronic spectroscopy

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Sensitive interference detection of the electric field of femtosecond four-wave mixing signals (stimulated photon echoes) at their point of origin in the sample can be used to record two-dimensional (2D) Fourier transform electronic spectra. In direct analogy to 2D nuclear magnetic resonance, 2D Fourier transform spectra have nearly homogeneous linewidths in each frequency dimension and sort the signal spectrum according to the initial excitation frequency. The initial excitation frequency information is stored in a robust population grating, so 2D spectra can be used to study both coherent and incoherent processes, and have revealed coherent aspects of energy transfer processes. Femtosecond 2D spectra also have the advantage of "freezing out" vibrational motions as inhomogeneities, raising interesting questions about what kinds of broadening can be rephased in 2D spectra recorded with stimulated photon echo pulse sequences. This talk will focus on coherent aspects of non-adiabatic electronic curve crossing and their manifestation in 2D electronic spectra.