TWO-DIMENSIONAL INFRARED SPECTROSCOPY: REVEALING MOLECULAR STRUCTURE AND DYNAMICS IN SOLUTION

ANDREI TOKMAKOFF, *Department of Chemistry, Massachusetts Institute of Technology, Cambridge, MA 02139*.

I will describe the development of two-dimensional infrared spectroscopy as a tool for following molecular dynamics in solution. Fourier transform 2D IR spectra are obtained by observing the response of a sample to a sequence of femtosecond mid-infrared pulses. These two-dimensional spectra can be used to capture transient molecular conformation in solution, characterize the dynamics of these structures, observe dynamics of intra- and inter-molecular interactions, and characterize relaxation and spectral broadening mechanisms. Experiments on model systems show that the splitting and amplitude of cross peaks in a 2D IR spectrum encodes the coupling and relative orientation of vibrational states. A local mode description of the coupled vibrations leads directly to information on the molecular structure. Analysis of the line shapes of the peaks reveals information on distributions of structure. By observing the time-evolution of the 2D spectrum as a function of a mixing period, the solvation, relaxation and structural dynamics of the system can be described.