Multidimensional ultrafast spectroscopy

Robin M. Hochstrasser*

Department of Chemistry, University of Pennsylvania, Philadelphia, PA 19104-6323

The development and applications of methods that can help us to visualize the evolution of structural changes of complex assemblies far from equilibrium or of ultrafast structure evolution close to equilibrium are exciting frontiers of research with particularly important examples in chemistry, physics, and biology. Such efforts are expected to complement the vast knowledge of average structures that continue to be discovered by established methods.

This Special Feature on Multidimensional Ultrafast Spectroscopy exemplifies the considerable progress toward meeting the challenges of developing and applying multidimensional methods based on optical and IR rather than radio frequencies. The contents of this feature, which are representative of a much larger and ever-increasing literature, demonstrate that these new approaches hold promise for advances in liquid-state dynamics and structure, energy transport in biological assemblies such as those in photosynthesis, structural and nonequilibrium dynamics of folding proteins, transmembrane proteins and peptide aggregates, new materials properties, and widely applicable fundamental physical processes.

The special feature contains contributions on theory and experiments in multidimensional optical and IR spectroscopy. First is a Perspective (1) that describes some of the challenges that can be addressed by these new techniques. New knowledge regarding energy transfer in complex systems is obtained from the coherent evolution of excited molecular electronic states (2) or electron hole–pair excitations by 2D optical methods (3). Essential to progress in 2D IR are the theory and computation of the spectral and dynamical responses; therefore, theoretical contributions in the important areas of water structure (4) and amyloid fibrils (5) are featured to exemplify progress in these areas. In nonequilibrium experiments, the 2D IR of the relaxation of myoglobin after ligand photodissociation is shown to expose new information about protein substates (6), and the transition states in protein folding are deduced from multidimensional spectra by combining temperature jumps with 2D IR (7). The ultrafast nature of the 2D IR experiments renders 2D IR suitable for unique studies of solvent solute interactions (8) and vibrational energy transfer (9). Advances in pulse control are key to the evolution of this field, so new results on automation and phase control of IR pulses in 2D and 3D IR spectroscopy are also presented (10).

For several years, PNAS has published special feature issues on many cutting-edge research topics. Some of the themes of past special features include: Tissue Engineering, Social and Behavioral Sciences, Asymmetric Catalysis, Science and Technology for Sustainable Development, Long-Range Electron Transfer, Cluster Chemistry and Dynamics, Interstellar Chemistry, Nitrogen Fixation, Eukaryotic Transposable Elements, Coordination Chemistry of Saturated Molecules, and, most recently, High-Pressure Geoscience, Single-Molecule Chemistry and Biology, and Economics of Health and Mortality. Scheduled for future issues of the journal are special features on Going Beyond Panaceas as well as Poverty and Hunger. One objective of these special features is to advance the journal's ongoing initiative to expand its coverage of the physical and social sciences and mathematics. PNAS continues to encourage and welcome research articles in all areas of the natural and social sciences and mathematics.

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*E-mail: hochstra@sas.upenn.edu.

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