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Abstract for an Invited Paper
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Earle K. Plyler Prize for Molecular Spectroscopy Talk: Coherent Ultrafast Multidimensional Spectroscopy of Molecules; From NMR to X-rays

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Multidimensional spectroscopic techniques which originated with NMR in the 1970s have been extended over the past 15 years to the optical regime. NMR spectroscopists have developed methods for the design of pulse sequences that resolve otherwise congested spectra, enhance selected spectral features and reveal desired dynamical events. The major experimental and computational advances required for extending these ideas to study electronic and vibrational motions on the femtosecond timescale will be surveyed. The response of complex molecules and semiconductor nanostructures to sequences of optical pulses provides snapshots of their structure and dynamical processes. Two-dimensional correlation plots of the signals show characteristic cross-peak patterns which carry information about hydrogen bonding, secondary structure fluctuations of proteins and amyloid fibrils, and coherent and incoherent energy and charge transfer in photosynthetic complexes. Double quantum coherence signals that are induced by correlations among electrons or excitons allow the visualization of correlated wavefunctions. Future extensions to the attosecond regime using xray pulses will be discussed. Since core excitations are highly localized at selected atoms, such signals can monitor the motions of valence electron wavepackets in real space with atomic spatial resolution. Common principles underlying coherent spectroscopy techniques for spins, valence electrons, and core electronic excitations, spanning frequencies from radiowaves, infrared, ultraviolet all the way to hard X-rays will be discussed.

[1] “Coherent Multidimensional Optical Probes for Electronic Correlations and Exciton Dynamics; from NMR to X-rays”, S. Mukamel, D. Abramavicius, L. Yang, W.Zhuang, I.V. Schweigert and D. Voronine. *Acct.Chem.Res.* **42**, 553-562 (2009).

[2] “Coherent Multidimensional Optical Spectroscopy Excitons in Molecular Aggregates; Quasiparticle vs. Supermolecule Perspectives”, D. Abramavicius, B. Palmieri, D. Voronine, F. Sanda and S. Mukamel, *Chem. Rev.* **109**, 2350-2408 (2009).