Focus on multidimensional optical spectroscopy and imaging

To cite this article: Steven T Cundiff and Harald F Kauffmann 2014 New J. Phys. 16 065006

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Focus on multidimensional optical spectroscopy and imaging

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Received 23 April 2014
Accepted for publication 23 April 2014
Published 12 June 2014

New Journal of Physics 16 (2014) 065006
doi:10.1088/1367-2630/16/6/065006

Abstract
Optical spectroscopy using ultrafast light pulses has undergone a revolution in the last decade due to the introduction and development of multidimensional coherent techniques. These methods build on established coherent spectroscopic techniques, such as photon echoes, but go beyond them by correlating the coherent dynamics during two time periods. The resulting multidimensional spectra have a number of advantages including disentangling congested spectra, revealing coupling between resonances, and removing the effects of inhomogeneous broadening. Similar ideas are currently being used for imaging. The papers in this ‘focus on’ collection document some of the current areas of activity in these fields.

Keywords: optical multidimensional coherent spectroscopy, optical two-dimensional Fourier transform spectroscopy, ultraviolet 2D-optical spectroscopy, higher-order exciton correlations, 2D-THz spectroscopy, double-quantum coherence, exciton correlations
Multidimensional spectroscopic methods were first developed in nuclear magnetic resonance over 30 years ago [1]. Implementing these methods in the optical domain was first proposed just over 20 years ago [2]. In the last 15 years, there have been extensive efforts to implement similar techniques [3] based on lasers in the infrared [4, 5], through visible [6, 7], and into the ultraviolet [8]. These efforts have borne fruit in the last few years [9], with a shift in emphasis from developing the methods to using them as a tool to gain new understanding into the underlying dynamical processes. Work in the infrared focuses on vibrational excitations, whereas working with visible or ultraviolet light accesses electronic transitions. Typically these experiments utilize nonlinear optical signals, and thus are closely related to established optical spectroscopic methods such as photon-echo spectroscopy or transient four-wave-mixing.

Pushing multidimensional spectroscopy into the ultraviolet is an important, and challenging, direction [8]. A diffractive optic-based 2D apparatus using UV generated in a hollow core waveguide allowed the light-induced ring opening in α-terpinine to be studied [10]. Two-dimensional spectra in the UV have also been generated using a pair of pump pulses generated by a pulse shaper and a supercontinuum probe [11]. Spectra are obtained for two representative molecules: pyrene and 2,2-diphenyl-5,6-benzo(2H)chromene. The spectra show well-resolved cross-peaks and reveal the excitation energy dependence of the relaxation processes. Using fluorescence detected 2D spectroscopy is an alternative route to the UV. It has been used to observe the conformation-dependent electronic coupling between the monomeric subunits of a dinucleotide of 2-aminopurine [12].

Multidimensional spectroscopy has proven powerful for studying many-body interactions in semiconductor quantum wells and quantum dots [13]. The many-body interactions results in coherent coupling between excitons that are spatially separated, as observed using 3D spectroscopy to study asymmetric quantum wells [14]. Disorder plays an important role by localizing excitons. Nevertheless, non-local coupling between excitons can be observed [15]. In the limit of strong disorder, localized quantum dot states can occur. The vectorial coherent response of the exciton–biexciton system can be studied using 2D spectroscopy [16]. Confining the light in a microcavity results in exciton polaritons; nevertheless, multiexciton correlations still play an important role [17]. Using terahertz frequencies, it is possible to perform two-dimensional coherent spectroscopy on inter-subband transitions in quantum wells, as well as on graphene [18].

The ability to observe coupling between states is a strength of two-dimensional spectroscopy. This ability is demonstrated in a study the the Soret band of a chiral porphyrin dimer [19]. Theoretically, the two-exciton wave function can be reconstructed by using localized double-quantum-coherence spectroscopy [20]. Coupling between pigments is critical in light harvesting. An optimized tight-binding electronhole model of the photosystem II reaction center captures the main experimental features observed in two-dimensional optical spectra [21].

Two-dimensional infrared spectroscopy has typically been used to study vibrations. Theory calculations show that it can also be extended to study molecular predissociation [22].

Multidimensional optical spectroscopy and imaging is clearly continuing to make significant progress and provide new insight into a range of problems. The papers in this ‘focus on’ collection document some of the current areas of activity and point the way toward future directions.
References


