MAR12-2011-008940

Abstract for an Invited Paper
for the MAR12 Meeting of
the American Physical Society

Control over coherent light fields enables multidimensional coherent spectroscopy and multispectral coherent control

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Using a combination of spatial and temporal shaping of optical laser fields, fully coherent spectroscopy and coherent control can be carried out to high order from optical to THz spectral ranges. A single beam with a single femtosecond pulse can be transformed into multiple beams and multiple pulses, reconfigurably under computer control with no human alignment needed, retaining full phase coherence among all the noncollinear fields. This enables multiple-quantum 2D and 3D Fourier transform optical spectroscopy of excitons and exciton-polaritons in inorganic quantum wells and microcavities, in organic J-aggregate films, and in inorganic/organic hybrid structures, the results of which will be discussed. Spatiotemporal shaping also enables coherent control over THz phonon-polariton waves in ferroelectric crystals. The THz waves can be coherently superposed to reach extremely large field amplitudes both in the host crystals and in free space, and the fields can be further enhanced in dipolar antenna and metamaterial structures, enabling highly nonlinear coherent spectroscopy and coherent control in the THz regime. Results from solid, liquid, and gas phases, including multiple-quantum rotational coherences in molecular gases and THz-induced phase transitions in crystalline solids, will be presented. Prospects for further generalization of the approach all the way to the hard x-ray regime will be discussed.