Nonresonant signals in the two-dimensional spectroscopy

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Abstract:
Coherent electronic two-dimensional spectroscopy is mainly used to monitor the time evolution of the studied sample after its resonant optical excitation. However, the experimental experience shows that even nonresonant interactions can provide detectable spectral contributions. These are often present as a weak parasitic signals originating in the solvent and/or cuvette walls underlying the resonant spectrum of the actual sample and as such they are usually discarded from the analysis. In this work, we adapt the formalism of double-sided Feynman diagrams for the needs of coherent two-dimensional spectroscopy in the nonresonant regime. We analytically calculate the third-order polarization of a two-level and several variants of three-level systems. As a result, we demonstrate the typical appearance of the optical Kerr-effect, cross-phase modulation, excited-state coherence, two-photon absorption and stimulated Raman scattering in the 2D spectrum.