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SEMINAR

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From four-wave to N-wave mixing: novel efficient computational schemes

Four-wave mixing signals arising from the third-order nonlinear polarization constitute the core of femtosecond nonlinear time-resolved spectroscopy. In perturbative nonlinear optics, the N-th order contribution to the polarization is expressed in terms of nonlinear response functions [1]. For complex material systems and/or high-order signals, however, the evaluation of the response functions becomes prohibitively cumbersome. In the alternative nonperturbative approach to N-wave-mixing spectroscopy, the laser-matter interaction is included in the system Hamiltonian and is treated numerically exactly. The different nonlinear signals must be then extracted from the total polarization via the appropriate phase-matching conditions [2].

Here we discuss novel efficient methods for the computation of four-wave and N-wave mixing signals. In the limit of weak laser fields, the equation-of-motion phase matching approach (EOM-PMA) yields the time evolution of the N-pulse-induced polarization in any of the phase-matching directions in terms of certain auxiliary density matrices [3]. Pulse-overlap effects are automatically included. In the case of N strong laser pulses which are well separated in time, the strong-pulse doorway-window approximation is a more appropriate tool [4]. To illustrate the performance of EOM-PMA, vibronic effects in electronic two-dimensional photon-echo signals are considered. Broadening and resolution issues are addressed.

[1] S. Mukamel, Principles of Nonlinear Optical Spectroscopy (Oxford University Press, Oxford, UK, 1995).
[2] L. Seidner, G. Stock and W. Domcke, J. Chem. Phys. 103, 3988 (1995).
[3] M. F. Gelin, D. Egorova and W. Domcke, Acc. Chem. Res. 42, 1290 (2009).
[4] M. F. Gelin, W. Domcke and D. Egorova, J. Phys. Chem. A, submitted.

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