Fakultät für Mathematik und Physik
Albert-Ludwigs-Universität Freiburg

Physikalisches Institut

IRTG-Seminar

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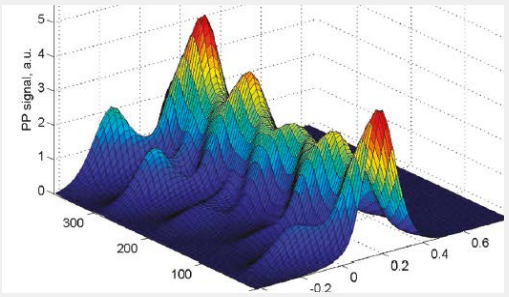
“Time- and frequency-resolved spectroscopy beyond the third-order polarization: what strong laser pulses can tell us”

The spectroscopic signals arising from the third-order nonlinear polarization represent the core of femtosecond time-resolved optical and infrared spectroscopy. In perturbative nonlinear optics, the *N*-th order contribution to the polarization is expressed in terms of nonlinear dipole response functions [1]. For complex material systems and/or high-order signals, however, the evaluation of the nonlinear response functions becomes prohibitively cumber-some. In the alternative nonperturbative approach to *N*-wave-mixing spectroscopy, the laser-matter interaction is included in the system Hamiltonian and the equation of motion for the wave function or the density matrix is solved numerically exactly. The nonlinear signals of interest are extracted from the total polarization via the appropriate phase-matching conditions [2].

In this lecture, a comprehensive outline of fully or partially nonperturbative descriptions of four-wave-mixing spectroscopies is given, considering pump-probe spectroscopy [3], double-pump coherence spectroscopy [4] and femtosecond stimulated resonance Raman spectroscopy [5] as specific examples. The motivation for employing (moderately) strong laser pulses is to induce enhanced responses of the molecular system. By tuning the strength and/or duration of the pulses, highly nonequilibrium vibrational wave packets may be prepared in excited electronic states and in the electronic ground state. By explicit calculations of pump-probe responses for selected model systems and by simulations of selected experiments, we show that strong pulses can be applied to enhance weak transitions, to provide time resolution beyond the pulse duration, to manipulate electronic and vibrational coherences and to control the dynamics of vibrational wave packets. It is shown that strong-pulse (nonperturbative) time-resolved spectroscopy allows us to extract more information from complex material systems than is accessible with weak laser pulses.



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Tuesday, May 2, 2017, 6:00 p.m., HS II, Physics high rise, Hermann-Herder-Str. 3

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