Intense multi-terahertz pulses: electric and magnetic nonlinearities on the sub-cycle scale

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Table-top sources of multi-THz pulses have provided phase-locked field transients covering the entire mid- to far-infrared spectrum [1]. Furthermore, peak electric and magnetic amplitudes beyond 10 GV/m and 33 Tesla have been attained with a hybrid laser source merging the stability of femtosecond Er:fiber lasers with the high power level of Ti:sapphire amplifiers [2]. In combination with field sensitive electro-optic detection, this toolbox offers an exciting pathway to nonlinear multi-THz optics with sub-cycle temporal resolution. We review latest high-field technology as well as nonlinearities driving the charge and spin degrees of freedom in solids: First, intense transients explore the nonlinear response of the bulk semiconductor InSb by amplitude- and phase-resolved four-wave mixing spectroscopy [3]. At amplitudes above 5 MV/cm the response exhibits a marked temporal substructure. Simulations based on a two-level quantum system demonstrate that in spite of the off-resonant character of the excitation the high-field pulses drive the interband resonances into a non-perturbative regime of Rabi flopping. A second study explores how spin order in pnictide compounds may be controlled via coherent THz phonons. Properly doped pnictides support unconventional superconductivity, but the microscopic pairing mechanism is unknown. Spin and lattice degrees of freedom as well as their interplay have moved into the focus of interest. We employ few-cycle multi-THz pulses to probe the evolution of a spin density wave gap in the parent compound BaFe2As2 after a femtosecond stimulus [4]. When starting in the low-temperature ground state, optical excitation melts the spin density order, followed by an ultrafast recovery. In contrast, a spin density wave gap is induced when we excite the normal state above the transition temperature. Surprisingly, the transient ordering quasi-adiabatically follows a coherent lattice oscillation at a frequency as high as 5.5 THz. Our results attest to a pronounced spin-phonon coupling in pnictides that fosters the emergence of a macroscopic order upon small vi! brational displacements. The THz electric field has been routinely exploited to couple to the charge degree of freedom. We complement this picture, by investigating the interaction of single-cycle THz pulses with an ordered spin system [5]: Antiferromagnetic NiO is exposed to a single-cycle THz transients exceeding peak magnetic fields of 0.1 Tesla. The induced dynamics is monitored via an 8-fs near infrared probe pulse. While the strong electric field component leaves no measurable trace in the solid, light-matter coupling proceeds exclusively via direct magnetic Zeeman interaction with the spins. A phase-locked sequence of THz magnetic transients allows us to switch a coherent 1-THz magnon oscillation on and off, on a single-cycle time scale. Further studies aiming at a complete spin reversal on the time scale of the oscillation period of THz radiation, are under way.

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