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Interaction of Terahertz Radiation with Ferroelectrics

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Ferroelectric crystals have long been used as acoustic transducers and receivers. An extensive toolset has been developed for MHz-frequency acoustic wave generation, control, guidance, and readout. In recent years, an analogous toolset has been developed for terahertz wave transduction and detection. Femtosecond optical pulses irradiate ferroelectric crystals to generate responses in the 0.1-5 THz frequency range that are admixtures of electromagnetic and polar lattice vibrational excitations called phonon-polaritons. Spatiotemporal femtosecond pulse shaping may be used to generate additional optical pulses that arrive at specified times and sample locations for control and manipulation of the THz waves. Femtosecond laser machining may be used for fabrication of waveguides, resonators, and other structures that are integrated into the ferroelectric host crystal. Finally, real-space imaging of the THz fields can be executed with variably delayed femtosecond probe pulses, permitting direct visualization of THz wave spatial and temporal evolution. This "polaritonics" toolset enables multiplexed generation of arbitrary THz waveforms and use of the waveforms within the ferroelectric host crystal or after projection into free space or an adjacent medium. The polaritonics platform will be reviewed and several new developments and applications will be presented. These include spectroscopy of relaxor ferroelectrics, whose temperature-dependent dielectric responses in the GHz-THz regime reveal complex polarization dynamics on well separated fast and slow time scales; direct measurement of phonon-polariton lattice vibrational displacements through femtosecond time-resolved x-ray diffraction; generation of high polariton field amplitudes and pulse energies; use of large-amplitude polariton waves to drive nonlinear lattice vibrational responses; and enhancement of optical-to-THz conversion efficiency through a pseudo-phase-matching approach that circumvents the very large disparity between refractive index values at optical and THz frequencies.