



Graduate School Event

Propagation-Resolved Nonlinear Terahertz Spectroscopy of Bulk Dielectrics

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It is a basic property of electromagnetic waves that they propagate. Yet the implications of this fact are often glossed over when it comes to nonlinear terahertz (THz) spectroscopy of bulk dielectrics, with propagation effects being given marginal attention. In thick samples, the THz fields, optical probe, and nonlinear polarization all propagate with different velocities, experiencing different dispersion, absorption and birefringence. This gives rise to complicated nonlinear optical interactions prone to misinterpretation in the absence of a comprehensive theoretical framework. This thesis combines two-dimensional THz Kerr-effect spectroscopy (2D-TKE), two-dimensional THz-field-induced second-harmonic generation (2D-TFISH), and propagation-resolved numerical modeling to interpret time-resolved THz-pump/optical-probe spectroscopies and track nonlinear THz polariton transport in bulk dielectrics. In birefringent LaAlO_3 , we use 2D-TKE data and four-wave-mixing simulations to separate propagation-induced electronic features from the intrinsic Raman-mediated Kerr response. We unambiguously identify the $0.86\text{~}\mathrm{THz}$ and $0.36\text{~}\mathrm{THz}$ oscillations in TKE as arising from phase-matched co- and counter-propagating THz-pump/optical-probe coupling, whereas the $1.1\text{~}\mathrm{THz}$ oscillation arises from the E_g Raman phonon in LaAlO_3 excited through THz two-photon absorption. In KTaO_3 , we identify the origin of the non-oscillatory part of the TFISH signal by combining single-pulse and 2D-TFISH measurements with simulations based on a Maxwell-Duffing model. We show that propagating soft-mode polaritons produce non-local transient polarization inside the sample. Spatial integration of the TFISH response at different locations acts to single out a polariton mode with a wavevector corresponding to the phase mismatch between the probe and its second harmonic. A Maxwell-Duffing model reproduces the temperature and field-strength dependence of both the single-pulse and 2D-TFISH signals, showing that the non-oscillatory component arises purely from nonlinear polariton propagation effects without invoking transient ferroelectricity. The 2D data also rule out the proposed mechanisms based on relaxational polar nanoregions or hot phonons. Finally, we develop a spatiotemporal method for tracking soft-mode phonon-polariton transport in bulk SrTiO_3 using 2D-TKE. The nonlinear Kerr signal is shown to probe the spatial overlap between two independent polaritonic wave packets. Then, using the less anharmonic A_{2u} polariton as a moving reference, we track the field-dependent evolution of E_u polaritons. We observe that anharmonic effects lead to the formation of additional polarization accumulation near the sample entrance and an approximately 30%

increase in the effective velocity of the leading peak of the E_u polariton. A minimal Maxwell--Duffing model captures many qualitative features of the 2D-TKE signal, including the entrance-layer response, but predicts a substantially weaker velocity renormalization than observed experimentally. These results establish 2D-TKE as a protocol to trace field-tunable polariton transport in an anharmonic medium, while revealing behavior beyond the Maxwell--Duffing description. Together, these results establish propagation as a crucial part of understanding nonlinear THz spectroscopy of bulk dielectrics. Propagation effects can mimic intrinsic material response but, when identified, become a sensitive probe of nonlinear lattice dynamics and transport in quantum materials.

Monday, November 9, 2026 10:30am - 11:30am

Sunstone Bldg / Ground floor / Big Seminar Room A (I23.EG.102)



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