

L A S E R C O L L O Q U I U M

Development of on-chip nonlinear two-dimensional terahertz spectroscopy

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ABSTRACT

On-chip THz spectroscopy enables the study of samples smaller than the free-space THz diffraction limit by confining the light fields. The THz polaritonics platform utilizes slab waveguides composed of LiNbO₃ or LiTaO₃ for compact THz generation, interaction, and detection.[1] However, THz fields generated by optical rectification within the waveguides are typically broadband and suffer from low electric field amplitudes, limiting experiments to the linear light-matter regime. To expand into the nonlinear regime, we have developed a method for generating tunable, high-amplitude narrowband THz fields in LiNbO₃ slab waveguides to resonantly drive material modes.

Narrowband THz fields are generated by matching the femtosecond optical pulse group velocity to a THz waveguide mode phase velocity, in the lateral excitation geometry (Fig. 1a). Yet, as the optical and THz group velocities are not equal, the optical pulse slides from one THz field lobe to another as it propagates, limiting the THz generation efficiency. Instead, we use a sequence of optical pulses which pump multiple THz lobes as they propagate through the waveguide, yielding higher THz generation efficiencies. THz pulses centered at 0.30 THz were produced with peak fields up to 40 kV/cm and spectral full-width-half-maxima of 36 GHz (11% of peak central frequency).[2] Frequency tunability was achieved by tuning the waveguide dispersion by varying the distance between a conductive film and the waveguide.

Despite the moderate 2 mJ laser power, the on-chip narrowband THz fields have comparable spectral brightness to our free-space propagating THz fields generated by tilted pulse front pumping of bulk LiNbO₃ using 6 mJ laser pulses. This opens the door for on-chip nonlinear

THz spectroscopy and as a first demonstration, we successfully reproduced the magnon dynamics in YFeO₃ using on-chip 2D-THz spectroscopy. This work advances waveguide-based THz spectroscopy into the nonlinear regime, which offers exciting possibilities for measuring microscopic samples and in space-constrained experimental geometries. THz field enhancement structures deposited on the waveguides may enhance the field sufficiently to even enable far-from-equilibrium excursions.

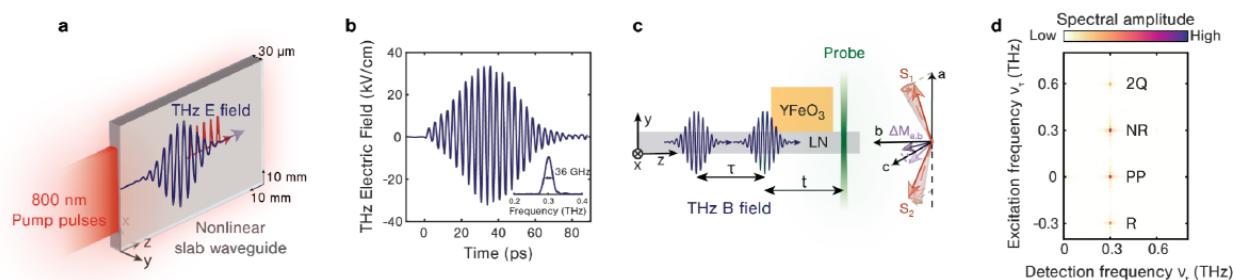


Fig. 1 (a) Narrowband THz generation by lateral excitation method in a LiNbO₃ slab waveguide and (b) the measured THz field with inset showing corresponding frequency spectrum. (c) Schematic of waveguide-based 2D-THz spectroscopy of the qFM magnon mode in YFeO₃ and (d) measured qFM mode spectrum featuring nonlinear rephasing (R), pump-probe (PP), non-rephasing (NR), and 2-quantum signal (2Q) peaks.

References

- [1] T. Feurer, N. S. Stoyanov, D. W. Ward, J. C. Vaughan, E. R. Statz, and K. A. Nelson, "Terahertz polaritonics," *Annu. Rev. Mater. Res.* 37, 317–350 (2007).
- [2] N. Berndt, D. Rohrbach, X. Zhang, and K. A. Nelson, "Generation of spectrally bright THz fields for nonlinear spectroscopy in LiNbO₃ slab waveguides," *Opt. Express* 33, 50280-50391 (2025)

ABOUT THE SPEAKER

Nadia Berndt is a PhD candidate in Prof. Keith Nelson's group at the Massachusetts Institute of Technology in the Department of Chemistry. Prior to that, she completed her Bachelors degree in Chemistry at the University of California Berkeley, working in Prof. Michael Zuerch's group. Her research uses on-chip terahertz spectroscopy to investigate light-matter interactions.