

# Advanced Ultrafast Probes of Symmetry Breaking in Quantum Materials

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The rapid evolution of advanced ultrafast techniques has enabled unique access to materials dynamics near the ground state and in transient phases far from equilibrium. I will discuss the development and utilization of ultrafast THz, electron, and X-ray probes to investigate transient structural and electronic dynamics across the Brillouin zone in quantum materials. In particular, atomic-scale stripes in correlated oxides and charge-density-waves (CDWs) in quasi-2D transition-metal dichalcogenides (TMDs) represent key platforms for dynamic studies of symmetry-broken phases. Using multi-THz spectroscopy, we captured transient vibrational symmetry breaking in an optically-driven correlated nickelate. The measurements provide evidence for a multi-component coupling between electronic and lattice order during stripe melting and formation [1]. Moreover, insight into non-equilibrium lattice distortions can be obtained via ultrafast electron diffraction (UED). I will discuss the first ultrafast investigation of TaTe<sub>2</sub>, which exhibits unique charge and lattice trimer order characterized by a transition upon cooling from stripe-like chains into a (3×3) superstructure of trimer clusters. Utilizing MeV-scale ultrafast electron diffraction with the LBNL HiRES beamline [2] we captured the photo-induced TaTe<sub>2</sub> structural dynamics. This reveals a rapid picosecond melting of its low-temperature ordered state followed by recovery into a hot cluster superstructure, with the initial quench triggered by intra-trimer Ta charge transfer [3]. Finally, I will outline the ultrafast accelerator-based X-ray light sources currently under development at ASU, which are designed to provide sophisticated femtosecond X-ray beams for imaging, crystallography, and ultrafast materials science in a compact footprint.

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## References:

- [1] G. Coslovich, A. F. Kemper, S. Behl, B. Huber, H. A. Bechtel, T. Sasagawa, M. C. Martin, A. Lanzara, and R. A. Kaindl, “Ultrafast Dynamics of Vibrational Symmetry Breaking in a Charge-ordered Nickelate,” *Science Advances* **3**, 1600735 (2017).
- [2] F. Ji, D. B. Durham, A. M. Minor, P. Musumeci, J. G. Navarro, and D. Filippetto, “Ultrafast Relativistic Electron Nanoprobes,” *Commun. Physics* **2**, 54 (2019).
- [3] K. M. Siddiqui, D. B. Durham, F. Cropp, C. Ophus, S. Rajpurohit, Y. Zhu, J. D. Carlström, C. Stavrakas, Z. Mao, A. Raja, P. Musumeci, L. Z. Tan, A. M. Minor, D. Filippetto, and R. A. Kaindl, “Ultrafast optical melting of trimer superstructure in layered 1T'-TaTe<sub>2</sub>,” *Commun. Physics* **4**, 152 (2021).