NANOSCIENCE COLLOQUIUM

Ultrafast nano-imaging – probing structure, coupling, and dynamics of matter on its natural length and time scales

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ABSTRACT: Understanding and ultimately controlling the properties of matter, from molecular to quantum systems, requires imaging their elementary excitations on their natural time and length scales of femtoseconds and nanometers. In order to achieve this goal, we developed scanning probe microscopy with ultrafast and shaped laser pulses for multiscale spatio-temporal optical nano-imaging. In corresponding ultrafast movies, we resolve the fundamental quantum dynamics from the fastest few-femtosecond coherent to the nanosecond thermal transport regime (Fig. 1).

I will discuss specific examples visualizing in space and time the nanoscale heterogeneity of electronic and structural processes in different classes of functional materials. Specifically, in coherent nonlinear nanoimaging of graphene and 2D semiconductors, we resolve the competing dynamics of intra- and interlayer coupling underlying the mechanisms of the emergent quantum phenomena in 2D heterostructures [1,2]. In the extension to far-from equilibrium excitation with even simultaneous spatial, spectral, and temporal resolution we resolve electron-phonon, cation-lattice, and coupled polaron dynamics in photovoltaic perovskites obtaining a real-space and real-time view of their complex photophysical response [3,4].

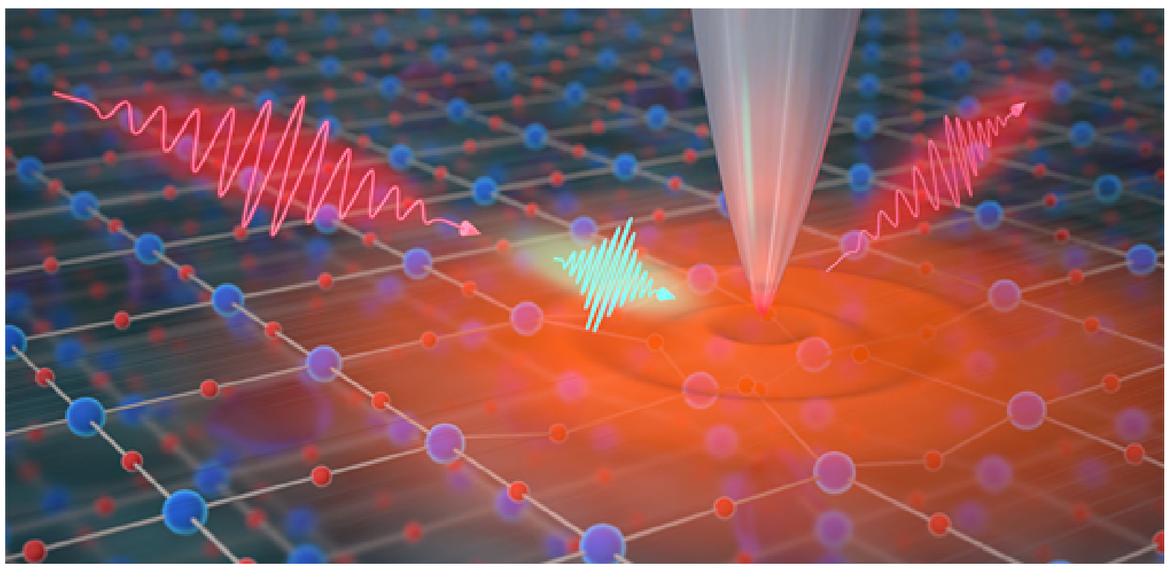

Fig. 1. Ultrafast optical nano-imaging with simultaneous nanometer spatial and femtosecond temporal resolution resolving from few-fs electronic coherence, to ps coupled electron-lattice, and ns thermal transport dynamics in molecular and quantum materials.

Lastly, we advanced nano-imaging also into the strong field, Purcell-enhanced, and strongly-coupled QED regime, with coherent superposition states for novel quantum-enhanced sensing and imaging [5,6]. Probing directly in the local electronic and molecular environments this will inform on the most fundamental level what limits coherence in solid-state quantum systems. I will then close with a perspective towards the ultimate goal of imaging and control, to systematically link coupled internal degrees of freedom to overcome relaxation and dissipation towards quantum materials with desired macroscopic performance.

[1] T. Jiang, et al., “Ultrafast coherent nonlinear nano-optics and nano-imaging of graphene”, Nature Nanotechnol. 14, 838 (2019).
[2] W. Luo, et al. “Nonlinear nano-imaging of interlayer coupling in 2D graphene-semiconductor heterostructures”, Small 2307345 (2024).
[3] J. Nishida, et al., “Ultrafast infrared nano-imaging of far-from-equilibrium carrier and vibrational dynamics”, Nature Commun. 13, 1083 (2022).
[4] J. Nishida, et al., “Nanoscale heterogeneity of ultrafast many-body carrier dynamics in triple cation perovskites”, Nature Commun. 13, 6582 (2022).
[5] R. Wilcken, et al. “Correlated nanoimaging of structure and dynamics of cation-polaron coupling in hybrid perovskites”, Science Advances 11, eads3706 (2025).
[6] R. Wilcken, et al. “Antenna-coupled infrared nano-spectroscopy of intramolecular vibrational interaction”, PNAS 120, e2220852120 (2023).

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