"Vibrational exciton and polaron nano-imaging: a molecular ruler to image structure, coupling, and disorder in functional molecular materials"

Properties and functions of molecular materials often emerge from intermolecular interactions and associated nanoscale structure and morphology. However, defects and disorder give rise to confinement and many-body localization of the associated wavefunction, disturbing the performance of, e.g., molecular electronic or photonic materials. However, conventional microscopy and even nanoscopy lacks spatio-spectral sensitivity to the low-energy and molecular length scales of intermolecular interactions, carrier-phonon coupling, and polaron formation, thus leaving a missing link between material structure and observed heterogeneity in the electronic or photonic response.

We address these outstanding problems in several novel combinations of spatio-spectral and spatio-temporal infrared nano-imaging. Through probing vibrational exciton formation as a molecular ruler, we resolve the evolution of defects in organic electronic materials and nano-domains in self-assembled monolayer on molecular length scales [1]. In the extension to probing both electronic and lattice degrees of freedom, in organic-inorganic perovskites we image the elementary processes of heterogeneous cation-lattice coupling that control the photovoltaic response [2]. We further demonstrate through Purcell-enhanced and strongly coupled light matter interaction the emergence of new hybrid light-matter state and their control for nano-metrology and -sensing [3]. As a perspective we show how electronic, vibrational, and polaron quantum state nano-spectroscopy in the low energy landscape of molecular matter provides for functional imaging as a new tool to guide the molecular device fabrication with improved performance.

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