Revealing vibronic coupling in the ultrafast dynamics of photoexcited materials for optoelectronics and solar energy conversion

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Photoexcitation drives functional materials far from equilibrium. During the first few tens to hundreds of femtoseconds, coupled electronic and structural dynamics influence how energy and charge redistribute, relax and eventually localize. These processes involve the quantum dynamics of coupled electronic and nuclear degrees of freedom, with nonadiabatic couplings and quantum coherence playing a central role. Identifying couplings that control these dynamics is essential for understanding and ultimately manipulating energy and charge flow in photoexcited matter, as well as for and guiding the rational design of materials for energy conversion and emerging quantum technologies. To probe these interactions experimentally, requires methods that can resolve ultrafast dynamics with high-time resolution and reveal couplings between excitations. In this talk, I will present some of our recent results obtained using ultrafast two-dimensional electronic spectroscopy with sub-10-fs time resolution to reveal vibration-mediated excited state dynamics. Specifically, I will discuss quadrupolar molecules undergoing excited-state symmetry breaking and ultrafast wavepacket motion through conical intersections in their molecular aggregates [1-3]. Finally, in halide perovskite crystals, I will present the observation of exciton population oscillations driven by coherent phonon fields [4], demonstrating the importance of nonadiabatic coupling for coherent exciton dynamics in these materials.

[1] Nature Nanotechnol. 16, 63-68 (2021)
[2] Nature Chem. 17, 1742 (2025)
[3] J. Phys. Chem. Lett. 7, 976-983 (2026)
[4] Nature Commun. 14, 1047 (2023)