Ultrafast exciton-phonon dynamics and conical intersections in optoelectronic materials probed by two-dimensional electronic spectroscopy.

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The dynamics of non-equilibrium excitations upon light-matter interaction in photoactive materials are governed by the details of the electronic structure, by its coupling to the vibrational degrees of freedom, and by the specific structural arrangement in the condensed phase. All these microscopic properties ultimately determine the ultrafast flow of energy and motion of charges upon photoexcitation. They are therefore important for the efficient application of such materials in optoelectronic applications, such as e.g., light-harvesting and energy conversion, sensing, light-emission. In this talk, I will discuss some of our most recent results probing strongly coupled exciton-phonon dynamics in technologically relevant materials for energy conversion and how two-dimensional electronic spectroscopy (2DES) can provide detailed new insight into these dynamics and underlying couplings [1-3]. Specifically, using 2DES we reveal wave packet motion through intermolecular conical intersections in molecular aggregates and their potential role for steering coherent energy transport in these materials, and phonon-modulated exciton dynamics in halide perovskites.

[3] X.T. Nguyen et al, J. Phys. Chem. Lett. 10, 5414 (2019).

^[1] A. De Sio et al., Nature Commun. 7, 13742 (2016).

^[2] A. De Sio et al, Nature Nanotechnol. 16, 63-68 (2021).