SFB1242

SFB 1242 Nichtgleichgewichtsdynamik kondensierter Materie in der Zeitdomäne

UNIVERSITÄT DUISBURG ESSEN

Open-Minded

19.10.2021 / 10 Uhr c.t., Raum MG 272
Campus Duisburg

Coherent ultrafast charge and energy transfer processes in nanostructures

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The function of essentially all present and future quantum devices, from quantum computers over quantum sensors to photocatalytic systems or solar cells, relies on the motion of charges and spins on ultrafast time and exceedingly short length scales. Usually, these dynamics are governed by such a complex interplay between electronic and nuclear motion that their understanding is quite limited and we rely on particle-like transport models for describing the dynamics and function of those systems.

In my talk, I will introduce and discuss several systems in which this classical, particle-like transport regime breaks down and the wave-like coherent transport of energy and charge becomes dominant, even in disordered nanostructures and at room temperature. I will try to demonstrate this for different transport processes and in quite diverse systems, ranging from artificial light-harvesting molecules to technologically-relevant materials such as halide perovskite crystals. Specifically, I will discuss coherent intramolecular charge transport processes in donor/acceptor molecules (1) and organic thin films (2), and coherent intermolecular charge transfer in P3HT/PCBM solar cell layers (3). More recently, we have found strong evidence for the role of intermolecular conical intersections (CoIns) in the energy transport in aggregated donor/acceptor thin films (4). Using an advanced spectroscopic method, two-dimensional electronic spectroscopy (2DES), we have followed the coherent wavepacket motion across such a CoIn (4). Finally, I will comment on what 2DES can tell us about coherent couplings between excitons and surface plasmon polaritons in artificially designed nanostructures. Making use of recent developments in laser technology, we improve the time resolution and sensitivity in 2DES and resolve coherent energy exchange processes (strong coupling) and two-particle excitations in such hybrid systems for the first time. The experiments illustrate a conceptually new approach for understanding how nanostructures “talk to each other”.

Our studies of charge transport in organic materials give unexpected evidence for long-lived vibronic quantum coherence and outline strategies for molding the flow of charge in nanostructures by tailoring and controlling their coherent coupling to vibrational modes of the materials. These advances became possible by probing the optical properties of nanostructures with a time resolution of few femtoseconds only, faster than any of the functionally relevant vibrational modes of the material, and by comparing the experimental results to advanced theoretical modelling.

[1] C. A. Rozzi et al., Nat. Commun. **4**, 1602 (2013).
[2] A. De Sio et al., Nat Commun **7**, 8 (2016).
[3] S. M. Falke et al., Science **344**, 1001 (2014).
[4] A. De Sio et al., Nature Nano **16**, 63 (2021).
[5] J. H. Zhong et al., Nature Commun. **11**, 1 (2020)

Für diese Zeit steht eine Kinderbetreuung nach vorheriger Anmeldung zur Verfügung.

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