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Space- and Time-Resolved Exciton Dynamics

Quantum wavefunctions in general depend on space and time. Thus, if we want to understand nature, we should try to study spatial and temporal dynamics simultaneously. Experimental investigations are often hampered, however, by the small size of investigated systems as well as the ultrafast timescale of many time-dependent processes. Thus, special spectroscopic techniques are required, some of which will be introduced in this talk.

In particular, we develop and apply methods of coherent two-dimensional (2D) spectroscopy offering frequency resolution for both the excitation and the detection steps. It will be shown how this can be used, among other applications, to study the dynamics of excitons, i.e., the time evolution of excited states, in supramolecular systems. For quantitative results, we have derived a theorem proving that a unique “inversion” of experimental data is possible yielding the complete state-to-state population transfer kinetics under certain conditions. Extending the 2D method to higher nonlinear orders of light–matter interaction, we study exciton–exciton interactions directly, providing information on light harvesting and exciton diffusion in optoelectronic materials. We also implemented a 2D technique suitable for molecular beams in which environmental and interchromophore interactions can be switched off completely.

In heterogeneous and nanostructured systems exciton dynamics may vary strongly between different spatial domains. Thus we have developed variants of 2D spectroscopy with additional direct spatial resolution. In one approach, we employ time-resolved photoemission electron microscopy (TR-PEEM) to realize “2D nanoscopy” with sub-diffraction optical resolution. As an application, we demonstrate the existence of strongly (Anderson) localized photonic states that are responsible for perfect absorption in the long-wavelength region of nanotextured thin-film solar cells. We have also nanoengineered a system that displays long-range (over distances of twice the wavelength) coherent energy transfer and we observed unexpectedly long coherence times for molecular bilayers adsorbed on a metal surface.

