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Exitonic relaxation and coupling in semiconductor nanostructures studied with optical 2D Fourier transform spectroscopy

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Many next-generation photovoltaic schemes are built on nanoscale confinement effects, but successful implementation of these schemes requires efficient harvesting of energy from the confined states, which depends on fast carrier extraction or exciton diffusion to reaction centers. Understanding how nanoscale environment influences exciton diffusion dynamics is critical to the long-term goal of being able to direct excitons to optimal sites. This electronic structure and dynamics can be captured by optical 2D-Fourier-transform spectroscopy (2DFTS), which tracks the phase of the nonlinear signal during two time delays of a multi-pulse excitation sequence. We used optical 2DFTS to study the coherent response of an ensemble of interfacial "natural" GaAs quantum dots (QD), found within the monolayer fluctuations of a quantum well (QW). The QD and the QW are excited simultaneously and homogenous and inhomogeneous linewidths of both are measured. The absence of a phonon-activation peak in the 2D spectra reveals that elastic exciton-phonon scattering is the primary dephasing mechanism. Upon variation of the population time delay and lattice temperature, 2D spectra clearly reveal a coupling from the QW states to the lower energy QD mediated by incoherent phonon interactions.