Diffusion of quantum well excitons measured with nonlinear spectroscopy

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Optical spectra of semiconductors display narrow resonances below the absorption edge, which are attributed to the excitons, i.e. pairs of electrons and holes, bound by the Coulomb interaction. We here perform coherent nonlinear spectroscopy of excitons in epitaxially grown CdTe-based quantum wells. By performing heterodyne four-wave mixing (FWM) microscopy, we find that the exciton absorption is dominated by homogeneous broadening on areas exceeding a few hundred microns squared. The photon-echo sequence of the FWM reveals no traces of the inhomogeneous broadening, indicating that excitons experience no localization due to the microscopic potential fluctuations. Under such conditions, the excitons with a large in-plane momentum of their center of mass (out of the light cone), can freely roam across the quantum well. After reaching mesoscopic distances, within $10\,\mu$m range, they can relax back to the radiative cone and recombine radiatively. In these novel experiments, we take advantage of the microscopic configuration of the FWM to probe the diffusion of the exciton density and coherence. This is achieved by measuring FWM as function of the spatial separation between pumps, inducing density or coherence of the excitons, respectively, and the probe converting them into the FWM. An extra care is taken to distinguish between FWM originating from diffusive excitons, i.e. those traveling between the pump and the probe, and the FWM signal created directly by the scattered light impinging the probe. This proof-of-principle experiment demonstrates that heterodyne FWM can be employed to infer propagative effects of excitons in semiconductor nanostructures.

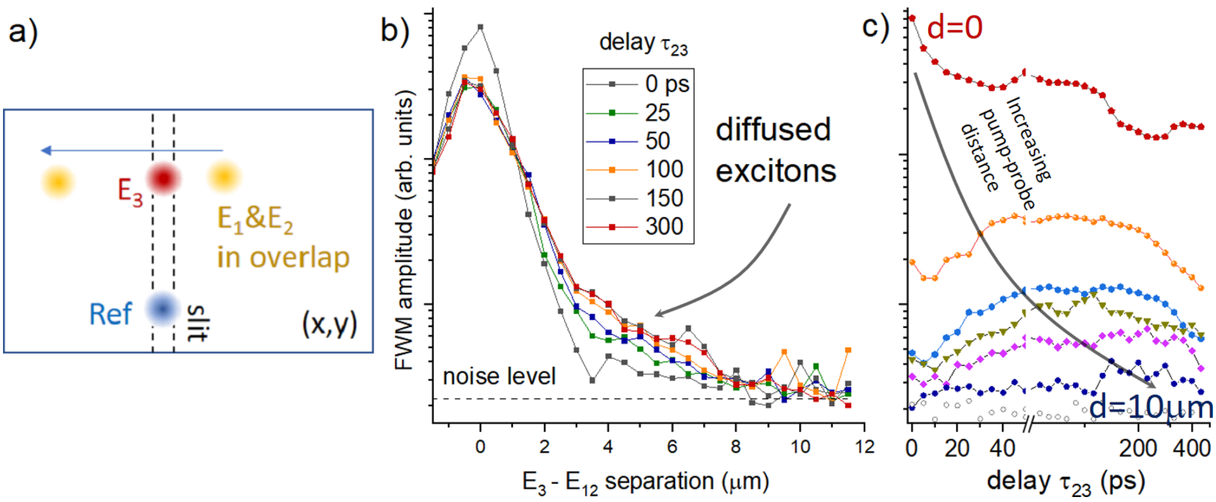

Figure 1: a) A geometry of the spatially-resolved FWM experiment, FWM generated at the probe position E_3, is detected via heterodyne spectral interference with the reference Ref. b) build up of the exiton diffusion with increasing the delay τ_{23}, c) signatures of the diffusive excitons in the delay dependence: with increasing the pump-probe the maximum of the FWM shifts to larger delays τ_{23}