Propagative coherence and density of quantum well excitons measured with nonlinear spectroscopy

M. Raczyński [1], A. Dydniański [1], K. Połczyńska[1], G. Szwed[1], J.-W. Jung[2], G. Nogues[2], W. Langbein[4], W. Pacuski[1], P. Kossacki[1], J. Kasprzak [1,2,3]*

[1]Faculty of Physics, University of Warsaw, ul. Pasteura 5, 02-093 Warszawa, Poland
[2]Univ. Grenoble Alpes, CNRS, Grenoble INP, Institut Néel, 38000 Grenoble, France
[3]Japanese-French Laboratory for Semiconductor Physics and Technology (J-FAST), CNRS, Université Grenoble Alpes, Grenoble INP, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8573, Japan
[4]School of Physics and Astronomy, Cardiff University, The Parade Cardiff, UK

The optical spectra of semiconductors display narrow resonances below the absorption edge, which are attributed to excitons, i.e. pairs of electrons and holes bound by the Coulomb interaction. Here, we perform coherent nonlinear spectroscopy of excitons in epitaxially grown CdTe-based quantum wells. By performing heterodyne four-wave mixing (FWM) microscopy [1], we find that exciton absorption is dominated by homogeneous broadening in areas exceeding a few hundred square microns. The FWM photon-echo sequence reveals virtually no traces of inhomogeneous broadening, indicating that excitons experience weak localisation due to the microscopic potential fluctuations. Under such conditions, excitons generated by a resonant excitation can acquire a large momentum via scattering with acoustic phonons, permitting them to propagate in-plane of the quantum well. After reaching mesoscopic distances within the 10-micron range, they eventually relax back into the radiative cone and recombine. In these novel experiments, we take advantage of the microscopic configuration of the FWM to investigate the propagation of exciton density and coherence. This is achieved by introducing spatial separation between the pumps, inducing excitonic coherence or density, respectively, and the probe that converts them into FWM, as presented in Figure 1. Extra care is taken to distinguish between FWM originating from propagative excitons (i.e. those travelling between the pumps and the probe) and the FWM signal created directly by scattered light impinging onto the probe. This proof-of-principle experiment demonstrates that heterodyne FWM can be used to infer the propagative effects of excitons in semiconductor nanostructures, paving the way towards non-local, coherent optical control of solid-state qubits in photonic circuits.

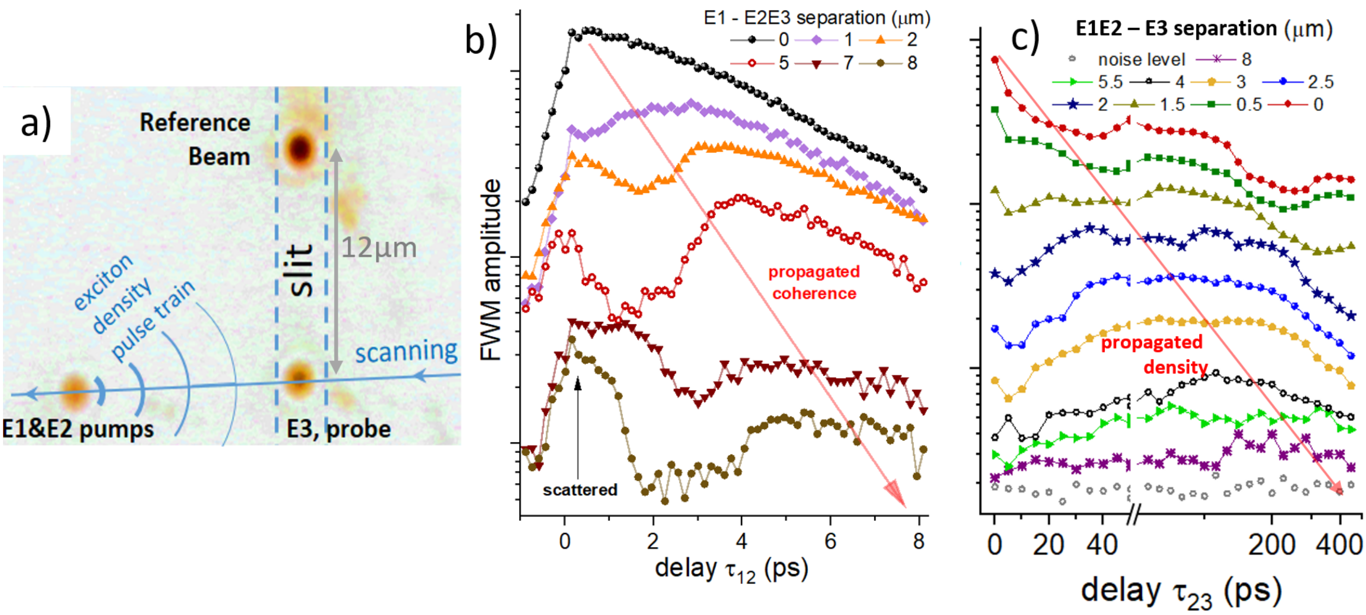

Figure 1: a) A geometry of the spatially-resolved FWM experiment: FWM generated at the probe position E3, is detected via heterodyne spectral interference with the reference. b) Signatures of the propagative exciton coherence in "time-of-flight" configuration: with increasing the pump-probe separation the FWM distribution shifts toward longer delays τ_{12}. c) Signatures of exciton diffusion: the maximum of the FWM distribution shifts towards longer delays τ_{23}.

References

[1] D. Groll, T. Hahn, P. Machnikowski, T. Kuhn, J. Kasprzak, and D. Wigger *"Fundamentals of heterodyne wave mixing spectroscopy: a tutorial"*, arXiv:2503.19750v1 in review (2025).

*E-mail: jacek.kasprzak@cnrs.fr