

MEASURING EXCITON POLARITON INTERACTIONS AND RELAXATION WITH MULTIDIMENSIONAL COHERENT SPECTROSCOPY

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Multidimensional coherent spectroscopy is based on transient four-wave mixing and has been used to reveal dynamics and interactions in a range of systems[1]. We have used this approach to reveal detail of the relaxation dynamics of exciton polaritons resonantly excited in the lower polariton branch of a GaAs-based quantum well in a cavity. Two-dimensional spectra, such as those shown below, map what is effectively the absorption energy (E_1) and emission energy (E_2), and in this case, these rephasing plots show homogeneous linewidths, even in the presence of inhomogeneous broadening.

After the initial excitation ($t_2=0$), the full range of energies of the lower polariton (LP) is excited, as demonstrated by the diagonal peak in the 2D spectrum at $t_2=0$. These polaritons rapidly (<500 fs) relax, however, they do not produce cross-peaks, (see plot below at 500fs) suggesting this process may not conserve wave-vector. Finally, at longer times, the LP peak on the diagonal becomes narrower as the excitation density decreases, and a distinct diagonal peak at the exciton energy is evident. In this figure, it is also clear that there is a cross-peak between the bottom of the polariton branch and the bare exciton, without any intermediate crosspeak. This suggests the relaxation from the bare excitons (with high k) to the bottom of the LP is rather direct and different to the relaxation within the LP band. Further experiments, analysis and modelling will help to understand the relaxation mechanisms and interactions in these exciton-polariton systems.

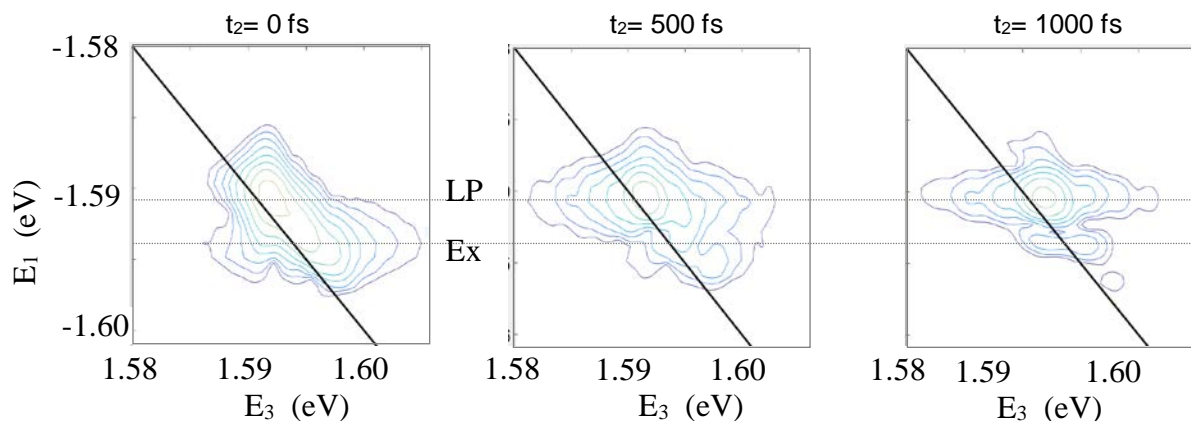


Fig. 1. Two-dimensional spectra from GaAs-based exciton-polaritons at population times $t_2= 0, 500$ fs, and 1000 fs. The E_1 (E_3) axis can effectively be considered the absorption (emission) energy, and the diagonal line marks the points where emission equals absorption energy. The dashed horizontal lines indicate the exciton energy (Ex) and the energy at the bottom of the lower polariton band (LP).

References

[1] Tollerud, J.O., and Davis, J.A. *Prog. Quant. Electr.* 2017, 55, 1-34.