Isolating specific pathways in double quantum 2D spectroscopy

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By combining spectral amplitude shaping and select pulse delays it is possible to filter out unwanted signal contributions in double quantum 2D spectra, analogous to techniques in multidimensional NMR. In coupled semiconductor quantum wells this allows removal of broad free-carrier contributions and isolation of the mixed 2-exciton states.

Techniques in multidimensional NMR have developed over decades to the point where modern pulse sequences can contain hundreds of pulses, all carefully controlled. The main purpose of these pulse sequences is to filter out specific signal pathways and infer molecular structure. Here we extend these ideas to electronic multidimensional spectroscopy.

Specifically, we explore the dynamics in semiconductor double quantum wells using a sensitive pulse-shaper based multidimensional spectroscopy apparatus [1]. In rephasing experiments we identify coherent interactions between QW excitons, nominally dark excitons and barrier excitons. These coherent interactions also lead to cross-peaks in the 2Q spectrum representing mixed 2-exciton states. The regions below and above the diagonal corresponding to the mixed 2-exciton state involving a barrier exciton and a quantum well exciton (either wide-well or narrow well) are shown In Fig1 (a) and (d). In both cases there is a broad continuum masking the peaks and below the diagonal (a) there are no clear cross-peaks.

Two pathway selective schemes are applied, such that one pulse is resonant with the QW excitons and one with the barrier exciton, with a 200fs delay between them, as indicated in Fig 2 (h,i), while the delay of the third, broadband pulse is scanned. In both pathway selective orderings the 2-exciton state being probed is nominally the same, yet they produce vastly different results. For PS2, the above diagonal peaks are isolated, while for PS1 the below diagonal peak is more clearly isolated. The differences arise because the delay between the first two pulses effectively filters out pathways involving free-carriers which rapidly dephase.

Filtering the signal by applying a time window on the experiment in combination with the spectral amplitude shaping allows selectivity and an ability to quantitatively assess different contributions to the overall signal.

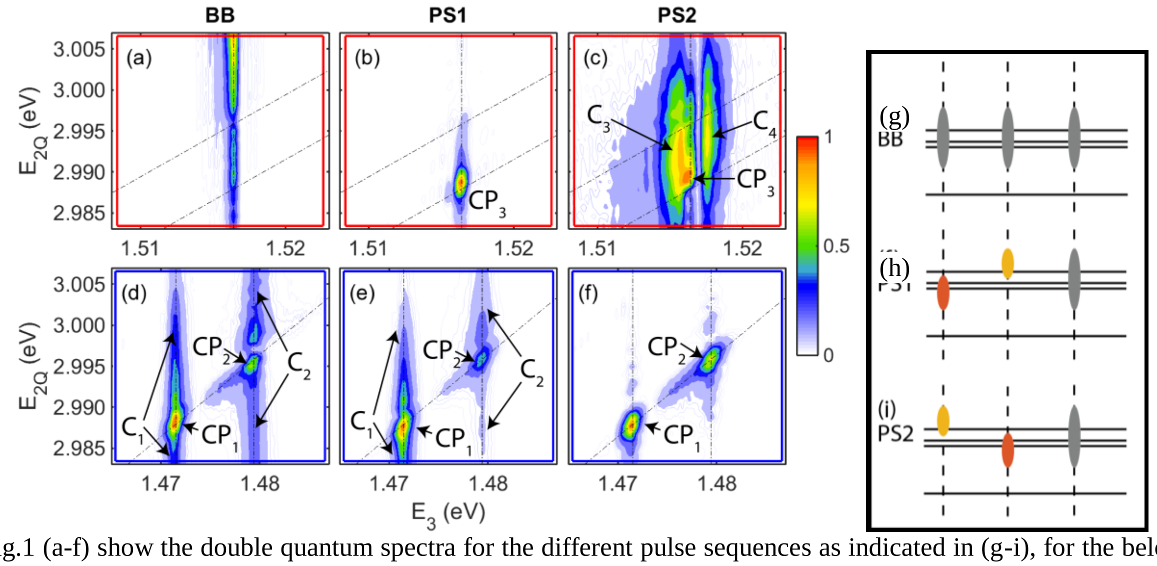

Fig.1 (a-f) show the double quantum spectra for the different pulse sequences as indicated in (g-i), for the below (a-c) and above (d-f) diagonal regions involving the mixed 2-exciton states.

[1] J. Tollerud *et al*., Optics Express **22,** 6719 (2014).