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Multidimensional coherent spectroscopy of semiconductor excitons, biexcitons and polaritons (I)

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Three-pulse multidimensional coherent spectroscopy allows for isolation of zero-, one- and two-quantum pathways for separate and coupled, as well as their homogeneous and inhomogeneous linewidths [1]. Excitons and biexcitons are examined in semiconductor quantum wells to determine the lineshapes and quantum pathways associated with the optical excitation. These result are compared and contrasted with those from a semiconductor microcavity where the excitons form polaritonic modes due to normal-mode splitting [2]. One-quantum rephrasing spectroscopy maps the detuning dependence of the exciton-polariton branches. Increasing detuning moves all features to higher energy and the expected anti-crossing is observed. An isolated biexciton is seen only at negative detuning, separated by a binding energy of approximately 2 meV. For positive detuning, the spectral weight of the off-diagonal features swap, as the lower polariton branch and biexciton come into resonance. This indicates that the off-diagonal features are sensitive to the interactions including two-quantum contributions and that a situation similar to a Feshbach resonance exists. Polarization dependence of two-quantum contributions show spin sensitive two-polariton and new biexciton correlations. The latter likely influence the Feshbach resonance between biexcitons and two-polariton states. The two-quantum signatures also demonstrate that biexcitons perturb the light-matter coupling in the microcavity to reduce the mixed two-polariton contributions. Detuning dependence of zero-quantum contributions show Raman-like coherences that are enhanced near zero detuning. Asymmetry of the Raman coherences are indicative of many-body interactions, which also grow stronger as the light-matter interactions are enhanced near zero detuning.
[1] Bristow et al, Rev Sci Instrum 80, 073108–8 (2009).
[2] Wilmer et al, Phys. Rev. B 91, 201304(R) (2015).

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