Fluorescence-Detected 2D Spectroscopy of Confined Materials

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Fluorescence detection avoids unwanted solvent contributions and scattering artefacts in 2D spectroscopy. Using femtosecond pulse shaping and phase cycling, we retrieve and separate various nonlinear signals. Exemplarily, we compare fourth-order and sixth-order spectra for a molecular dimer with coherently detected spectra. In combination with optical microscopy, we investigate microstructured samples and obtain 2D spectra of a $MoSe_2$ monolayer. Analysis of long-term beating allows us to retrieve the exciton–phonon coupling strength at room temperature, a quantity previously unknown for 2D materials. Further, we show that placing an inorganic 2D semiconductor in a microcavity leads to mixed exciton–phonon–photon states. We also apply the technique to 1D and 0D materials.