

From pump–probe to six-wave mixing 2DES: Comparing coherence- and population-based detection

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Apart from the type of the detected signal, the main difference between coherence- and population-based coherent nonlinear spectroscopy is the number of tracked interactions with the electric field. In each order of nonlinearity, the population-based techniques feature one more interaction, bringing with itself an additional Liouville-space pathway observed in the response. In my talk, I will discuss the physical interpretation of this additional pathway and its consequences for the measured spectra. The considered effects include: isolation of excited-state dynamics, absence of excited-state absorption, estimation of electronic coupling, and sensitivity to exciton–exciton annihilation.