Probing Exciton Dynamics in 2D $MoSe_2$ with Multidimensional Coherent Spectroscopy

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Two-dimensional transition metal dichalcogenides (TMDs) are a family of atomically thin semiconductors with tunable optical and electronic responses, offering a wide range of applications in optics and optoelectronics. Monolayer TMDs such as $MoSe_2$ exhibit a strong Coulomb interaction, which leads to tightly bound excitons with binding energies of several hundred meV [1]. Multidimensional coherent spectroscopy (MDCS) is a powerful optical technique for probing many-body interactions in semiconductors. Unlike conventional one-dimensional linear and nonlinear spectroscopic methods, MDCS is able to overcome the inhomogeneity in the sample and reveal the intrinsic linewidth of excitons, which is associated with the lifetime of the excited state as well as the optical coherence time related to the coherent superposition between the ground and excited states.

In this work, we probed the exciton dynamics and coupling among them in several hBN-encapsulated $MoSe_2$ structures including monolayer and homobilayers with various stackings. Two-dimensional coherent spectroscopy was applied in conjecture with a home-built ultrafast tunable optical parametric oscillator (OPO) in the near-infrared regime to study these samples. The optical coherence times were measured from liquid helium to room temperature. While different types of MDCS spectra are possible by varying different delays between the pulses, our work focused on one-quantum rephasing spectra from which homogeneous linewidths were extracted.

References

[1] Ye, Z., Cao, T., O'Brien, K. *et al.* Probing excitonic dark states in single-layer tungsten disulphide. *Nature* **513**, 214–218 (2014).