Prize Talk PRV III Thu 13:15 HSZ 01

High-lying excitons and excitonic quantum interference in 2D semiconductors — •Kai-Qiang Lin — Department of Physics, University of Regensburg, Regensburg, Germany — College of Chemistry and Chemical Engineering, Xiamen University, Xiamen, China — Laureate of the Walter-Schottky-Prize 2023

Two dimensional semiconductors such as transition-metal dichalcogenide (TMDC) monolayers show a wealth of exciton physics. We present the existence of a novel excitonic species, the high-lying exciton (HX), in TMDC monolayers with almost twice the energy of the band-edge A-exciton but with a linewidth as narrow as that of band-edge excitons. The HX is populated through momentum-selective optical excitation in the K-valleys, and is identified experimentally in up-converted photoluminescence and theoretically in ab initio GW-BSE calculations. These calculations show that the HX is comprised of electrons of negative effective mass. The coincidence of such high-lying excitonic species at around twice the energy of band-edge excitons gives rise to a well-defined excitonic three-level system, which enables quantum-interference phenomenon revealed in optical second-harmonic generation. We show that the temporal dynamics in such a three-level system can be probed through time-resolved sum-frequency generation and four-wave mixing. The HXs can also be tuned over a wide range by twisting and Stark effect in bilayer WSe2, which gives control over the excitonic quantum interference and the corresponding optical nonlinearities. Finally, we show how an electrical gate can be used to tune excitonic quantum interference in a monolayer TMDC transistor device by forming trions.