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Abstract

Monolayer transition-metal dichalcogenides (TMDCs) show a wealth of exciton physics. Here, we present the existence of a new excitonic species, the high-lying exciton (HX), in single-layer WSe₂ with almost twice the band-edge A-exciton energy and with a linewidth as narrow as that of band-edge excitons (Figure 1) [1]. The HX is populated through momentum-selective optical excitation in the K-valleys, and is identified experimentally in upconverted photoluminescence (UPL) and theoretically in *ab initio* GW-BSE calculations. The coincidence of such high-lying excitonic species at around twice the energy of bandedge excitons enables the excitonic quantum-interference phenomenon revealed in optical second-harmonic generation (SHG) [2]. High-lying excitons in bilayer WSe2 can be largely tuned by twisting [3], which gives control over the excitonic quantum interference and the corresponding optical nonlinearity.

References

- [1] K. -Q. Lin et al., Nat. Commun. DOI: 10.1038/s41467-021-25499-2 (2021).
- [2] K. -Q. Lin, S. Bange, & J. M. Lupton, Nat. Phys. 15, 242-246 (2019).
- [3] K. -Q. Lin et al., Nat. Commun. 12, 1553 (2021).

Figures

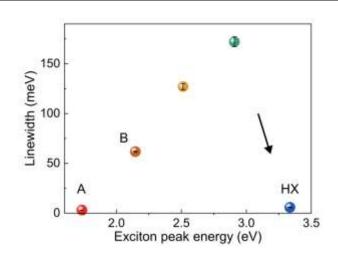


Figure 1: Energies and linewidths (full width at half maximum) of the prominent excitonic states seen in the PL of hBN encapsulated monolayer WSe₂. The PL linewidth increases with transition energy until the HX breaks the trend.