Full-Zone Valley Polarization Landscape of Finite-Momentum Excitons in Transition-Metal Dichalcogenide Monolayers

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Transition metal dichalcogenide monolayers (TMD-MLs) have attracted broad interest because of its spin-valley coupled characteristics in the electronic and excitonic structures [1,2]. The direct band gaps opened up at the K and K' corners of the first Brillouin zone (BZ) together with the valley-dependent optical selection rules make TMD-MLs a promising nanomaterials for the applications of valley-based photonics. However, it is known that the valley polarization of optically excited excitons are very likely to be degraded by the intrinsic electron-hole (e-h) exchange interaction which intermixes the interband transitions in distinct valleys. Moreover, the momentum-forbidden dark excitons (MFDXs) are also highly involved in the excitonic dynamics and various optical phenomena [3,4]. Therefore, a comprehensive theoretical understanding of the MFDXs in TMD-MLs over the extended momentum space is demanded.

In this study [5], we present a theoretical investigation of the full-zone landscape of finite-momentum dark excitons in WSe_2-MLs by solving the density-functional theory (DFT)-based Bethe-Salpeter equation (BSE) under the guidance of symmetry analysis. The studies reveal the comprehensive valley-depolarization landscape of finite-momentum exciton of WSe_2 monolayer. Dictated by the crystal symmetry, the valley pseudospin texture over the extended exciton-momentum $\boldsymbol{k}_{ex}$ space exhibits rich structures, featured by the inherently full valley polarizations in the excitonic K_{ex}, K'_{ex} and Q_{ex} valleys and also by the contrasted valley depolarization for the exciton states lying in the $\overline{\Gamma_{ex}M_{ex}}$ paths. Attractively, the superior valley polarizations of the intervalley dark excitons in WSe_2-MLs are shown almost fully transferrable to the optical polarization in the phonon-assisted photoluminescences because of the native suppression of exchange-induced depolarization in the second-order optical processes. The analysis of phonon-assisted photoluminescences accounts for the recently observed brightness, high degree of optical polarization, and long lifetime of the intervalley dark exciton states in tungsten-based TMD-MLs.

References

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