DISTINCTIVE SIGNATURES OF THE SPIN- AND MOMENTUM-FORBIDDEN DARK EXCITON STATES UNDERLYING IN THE TEMPERATURE DEPENDENT PHOTO-LUMINESCENCE OF WSe2 MONOLAYERS

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Atomically thin transition-metal dichalcogenide monolayers (TMD-MLs) have recently drawn broad attention because of the extraordinary spin-, valley- and excitonic properties. [1,2] With the combined spin- and valley-degrees of freedom, an exciton in a TMD-ML under photo-excitation exhibits the complex fine structures, composed of the bright exciton (BX) states, and spin-forbidden (SF) and momentum-forbidden (MF) dark states (DXs) as well. Because of the optical invisibility, the latter two kinds of the dark states are in principle hardly observed and even distinguished in conventional spectroscopies though their impacts on the optical and dynamical properties of TMD-MLs have been well noticed. In this work, we present a theoretical and computational investigation of the excitonic band structures and the temperature-dependent photo-luminescence (TD-PL) spectra of WSe_2-MLs under thermalization by solving the density-functional-theory (DFT)-based Bethe-Salpeter equation (BSE) with the full consideration of both electron-hole direct and exchange Coulomb interactions.[3] As main results, we reveal the distinctive signatures of the SF- and MF-DX underlying in the TD-PL, featured by the quickly rising and slowly descending PL intensities with increasing the temperature at low T~80K and high T~260K, respectively. Further, the exciton dynamics in the W-based TMD-MLs subjected to the exciton-phonon interactions is also studied by using the Lindblad quantum master equation based on the multi-exciton-level model. The computational results agree well with the existing experimental data, [4] and account for the impact of the high lying MF-DX states on the optical properties of W-based TMD-MLs.

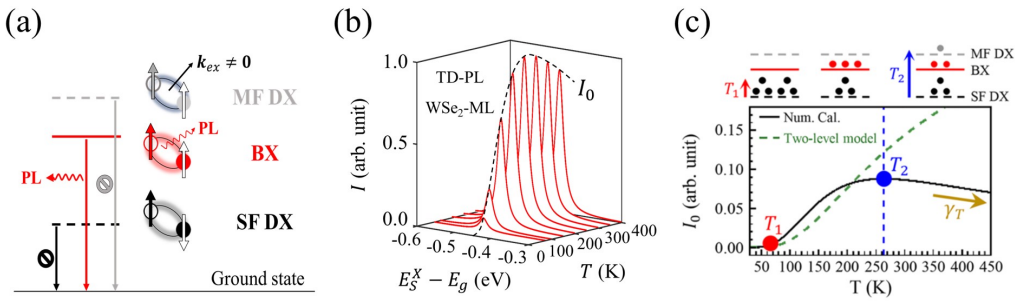

Fig. 1. (a) Schematics of the fine structure of the bright exciton (BX) states, and spin-forbidden (SF) and momentum-forbidden (MF) dark states (DX) of a WSe_2-ML. (b) The calculated temperature-dependent PL spectra and (c) the temperature-dependent PL intensities of a WSe_2-ML featured by the characteristic temperatures T_1 and T_2 as the signatures of the existing SF- and MF-DX, respectively. [3]

References

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