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Brightening the Dark Excitons in 2D Systems

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Abstract

Transition metal dichalcogenides have attracted a lot of attention in recent years due to their unique indirect to direct band gap transition from bulk to monolayer thickness. Combination of lattice inversion asymmetry and the presence of strong spin-orbit coupling in the valence band and weak spin-splitting in the conduction band results in the lowest lying exciton in WX2 (X = S, Se) being spin forbidden and optically dark. The dark nature of this exciton makes it particularly challenging to probe at room temperature using conventional photoluminescence setups [1-3]. Because of their long lifetimes, however, dark excitons are highly attractive for quantum optics and optoelectronic applications.

In this contribution, we demonstrate a novel approach of probing the radiative decay of dark exciton related emissions in WSe_2 monolayers studied by conventional and tip-enhanced photoluminescence (TEPL) and electric field dependent TEPL at room temperature. Monolayer WSe_2 flakes were sandwiched by noble metal (Au or Ag) substrates and polydimethylsiloxane (PDMS) nano-patches providing a strong local out-of-plane dipole moment with respect to the two-dimensional plane. This strong dipole moment not only enhances the dark excitons in WSe_2, it also produces bound excitons due to extrinsic charge defects visible at room temperature. The spatial distributions of these dark exciton related emissions were studied by TEPL with a spatial resolution < 10 nm confirming the confinement of these excitons within the PDMS nano-patches. Moreover, we also observed a direct correlation between the relative defect density and the radiative emission of dark excitons as revealed by tip-enhanced Raman scattering (TERS). Finally, by removing the nano-patches from the top of the flakes we are able to recover the optically bright excitons in the WSe_2 monolayer.

References

[1] Zhou et al., Nature Nanotechnology, 2017, 12, 856

[2] Zhang et al., Nature Nanotechnology, 2017, 12, 883

[3] Park et al., Nature Nanotechnology, 2018, 13, 59

Figures

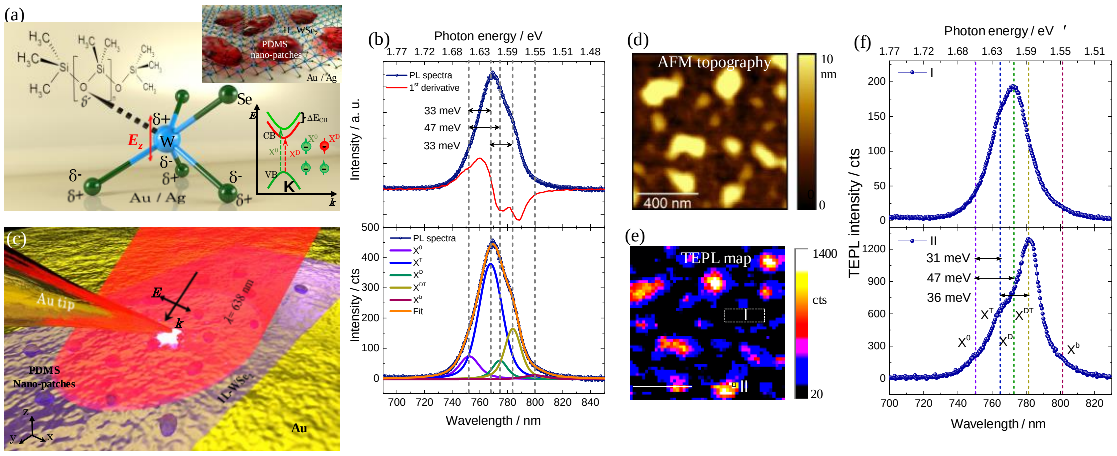

Fig. (a) Schematic of the sample structure, formation of out-of-plane electrostatic dipole, and the electronic band structure illustrating the dark exciton formation. (b) Micro-PL probing of dark exciton in WSe_2 on an Au substrate. (c) Schematic of TEPL experiments in side-illumination geometry. (d) AFM topography and (e) corresponding TEPL map of an area of interest on a WSe_2/Au sample. (f) Two representative TEPL spectra of WSe_2 as indicated in fig. (e)