Layered transition metal dichalcogenides (TMDCs) host a variety of strongly bound exciton complexes that control the optical properties in these materials. Apart from spin and valley, layer index provides an additional degree of freedom in a few-layer thick film. Using the solution of Bethe-Salpeter equation, we show the existence of two near-degenerate $A_{1s}$ excitons in a bilayer TMDC, out of which one (intra-layer exciton) is an order of magnitude higher in terms of radiative decay rate (and hence luminescence) compared to the other (inter-layer exciton), as explained in Figure 1.

In a more general way, we show that in a few-layer TMDC film, the wavefunctions of the conduction and valence band edge states contributing to the $K$ ($K'$) valley are spatially confined in the alternate layers giving rise to (quasi-)intra-layer bright exciton and inter-layer dark excitons. Depending on the spin and valley configuration, the bright exciton state is further found to be a coherent superposition of two layer-induced states, one (E-type) distributed in the even layers and the other (O-type) in the odd layers (Figure 2). Such layer index ($l_z$) is coupled to the spin ($s_z$) and valley ($t_z$) indices by the rule $l_zs_zt_z = +1$. The intra-layer nature of the bright exciton manifests as a relatively weak dependence of the exciton binding energy on the thickness of the few-layer film (Figure 3), and the binding energy is maintained up to 50 meV in the bulk limit – which is an order of magnitude higher than conventional semiconductors.

References