Controlling interlayer excitons in MoS₂ layers grown by chemical vapor deposition

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Chemical vapor deposition (CVD) allows growing wafer scale thin films of transition metal dichalcogenides (TMDs). In this work, we show the improved optical quality of MoS₂ monolayers (ML) with a reduced transition linewidth down to 5 meV at low temperature (T=4 K) by reducing dielectric disorder [1]. These results for the free excitons in emission and absorption are comparable to the best ML samples obtained by mechanical exfoliation of bulk material.

Combining CVD grown MoS₂ monolayers to form multilayers allows to access new functionalities. We examine the correlation between the stacking order and the interlayer coupling of valence states in MoS₂ as-grown CVD bilayers and artificially stacked bilayers from CVD monolayers [2, 3]. We show that hole delocalization over the bilayer in 2H stacking is allowed and results in strong interlayer exciton (IE) absorption and also in a larger A-B exciton separation as compared to 3R bilayers, where both holes and electrons are confined to the individual layers. Comparing white light reflectivity spectra for 2H and 3R stacking allows extracting an interlayer coupling energy of about $t_{\perp} = 50$ meV. Obtaining very similar results for as-grown and artificially stacked bilayers is promising for assembling large area van der Waals structures with CVD material, using inter layer exciton formation and A-B exciton separation as indicators for interlayer coupling [3].

References

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Figure 2: (a) Schematic of 3R stacked bilayer with intralayer excitons (top) compared to 2H stacked bilayer. (b) Reflectivity spectra of as-grown 2H-bilayer (blue) and as-grown 3R-bilayer (red). A, B and interlayer exciton are marked. (c) Energy difference between B and A exciton for the as-grown (orange), as well as artificially-assembled (black) 2H and 3R MoS₂ homobilayers.

Graphene2020