Resonant electronic and excitonic hybridisation in transition metal dichalcogenide alloys heterobilayers

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Van der Waals heterobilayers of vertically stacked atomically thin transition metal dichalcogenides (TMDs) can exhibit interlayer excitons (IX), formed by electrons and holes spatially located in the adjacent materials. IXs possess long radiative lifetimes and an electric dipole oriented perpendicularly to the plane of the TMDs, which result in enhanced non-linear exciton-exciton interactions.[1] Furthermore, the twist angle and lattice constant mismatch between the two materials of a bilayer can lead to a Moire' superlattice that strongly modifies the excitonic landscape of the structure.[2]
In our work, we adopt several $Mo_xW_{(1-x)}Se_2$ alloys stacked to WSe_2 and $MoSe_2$ monolayers to continuously tune the heterobilayer properties through modifications of the bandgap and band offset. Spanning the whole range of alloys composition, the devices clearly showed an IX photoluminescence signal that can be tuned in energy up to 100 meV at T<10K (Fig.1a). By reaching an alloy composition close to the pure TMDs, IXs energies bend towards the momentum- indirect transition energy of the homo-bilayers, changing from space- to momentum-indirect. This is confirmed by a 2-orders of magnitude decrease of the IX lifetimes. Moreover, when the electron bands are close to resonance and the layers crystals axis misalignment is small, multiple transitions appear in reflectance contrast (Fig.1b), due to the effect of excitons hybridization and Moire' patterns as also confirmed by the theory.

References

[1] Mak, K. F.; Shan, J.; *Nat. Nanotechnol.* **2018**, *13* (11), 974–976.
[2] Tartakovskii, A.; Nature Reviews Physics **2020**, 2(1), pp.8-9.

Figures

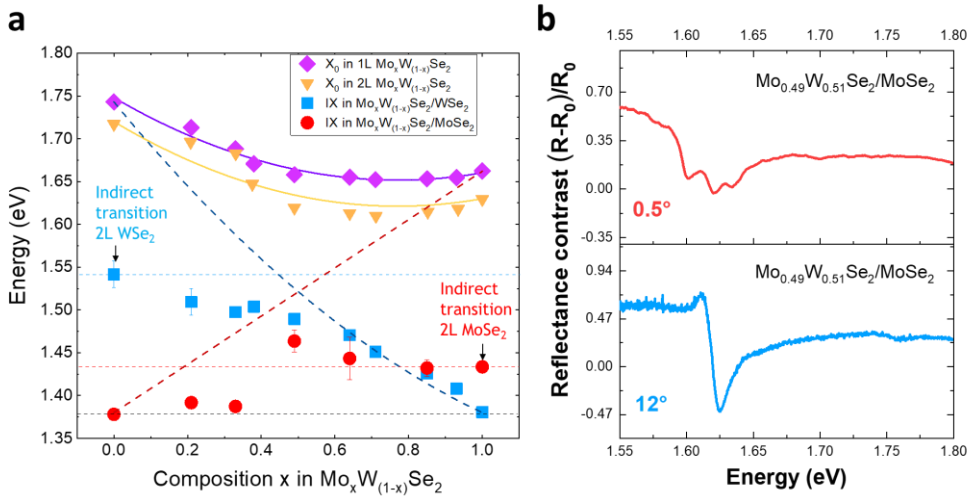

Figure 1: (a) IX energy for $Mo_xW_{(1-x)}Se_2/MoSe_2$ and $Mo_xW_{(1-x)}Se_2/WSe_2$ heterobilayers as a function of the Mo composition in the alloy. Dashed thick lines are the expected IX energy trend not considering hybridization. **(b)** Reflectance contrast spectra of an aligned and misaligned $Mo_{0.49}W_{0.51}Se_2/MoSe_2$.