INTERLAYER EXCITONS OF $MOSE_2$-WSE_2 HETERO-BILAYERS

L. Caussou[1], C. Lagoin[1], A. Reserbat Plantey[2], K. Watanabe[3], T. Taniguchi[3], V. Voliotis[1], and F. Dubin[1]

[1]Institut des NanoSciences de Paris, Sorbonne Université and CNRS, Paris, France
[2]Centre de recherche pour l'hétéroépitaxie et ses applications – CRHEA, Valbonne, France
[3]Research Center for Functional Materials, National Institute for Materials Science, Tsukuba, Japan

Monolayers of transition metal dichalcogenides (TMDs) exhibit remarkable optical properties. These rely on Coulomb bound electron-hole pairs realising model two-dimensional excitons. Monolayer excitons are characterised by strong optical absorption/emission, combined to binding energies possibly reaching a few 100 meV. Through the photoluminescence (PL) they radiate, excitons are then strikingly observable up to room temperature. In the low temperature limit, the exciton PL reaches the Fourier limit where its spectral width, of around 2 meV, is given by the very fast electron-hole recombination rate. The latter is then characterised by a time constant in the pico-second domain, which excludes that excitonic fluids are studied at thermodynamic equilibrium where quantum statistical correlations possibly emerge at sufficiently low temperatures ($\lesssim$ 10 K).

By interfacing two distinct TMD monolayers, thus realising a so-called hetero-bilayer, minimum energy states for electrons and holes possibly lie in a different layer. This geometry then favours the buildup of interlayer excitons made by opposite charge carriers confined in a distinct monolayer (Fig.1). The overlap between the carriers wave-functions is then greatly reduced so that interlayer excitons have radiative lifetimes extending beyond 100 ns. Furthermore, in hetero-bilayers excitons are characterised by a well oriented electric dipole, given by the separation between electrons and holes. They experience then repulsive dipolar interactions and explore at the same time a periodic two-dimensional confinement. Indeed, a triangular moiré lattice forms spontaneously due to both the mismatch between the monolayers lattice constants and the angle between their crytstalline axis. Remarkably both the period and the depth of the moiré potential are easily tuned by suitable hetero-bilayer engineering. Thus, interlayer excitons in moiré potentials provide a promising platform to explore the physics of the Bose-Hubbard Hamiltonian in the solid-state [1, 2, 3].

Here we study optically a $MoSe_2$-WSe_2 hetero-bilayer encapsulated in hBN (Fig.1). At low temperature ($\sim$5 K) interlayer excitons are directly evidenced by their low-energy PL emission, around 300 meV below that of intralayer excitons of individual $MoSe_2$ and WSe_2 monolayers. Importantly, we probe the role of the optical excitation, by comparing the situation where electrons and holes are non-resonantly injected, by exciting above the bandgap energies of $MoSe_2$ and WSe_2, to a resonant excitation of the $MoSe_2$ excitonic absorption. We observe that the latter excitation optimises both the PL intensity and its spectroscopic features. Thus, at low excitation powers, i.e. for very dilute excitonic fluids, we evidence that the PL spectrum is made of narrow-band emission lines, separated by a few 10 meV (right panel of Fig.1). Performing polarisation resolved excitation/detection spectroscopy, we observe that PL lines display the same degree of circular polarisation as the one of the excitation laser injecting electronic carriers. This behaviour reveals that interlayer excitons are delocalised, nevertheless we also resolve unpolarised PL lines marking the radiative recombination of localised excitons. We then discuss the role that the moiré potential can play in our experiments.

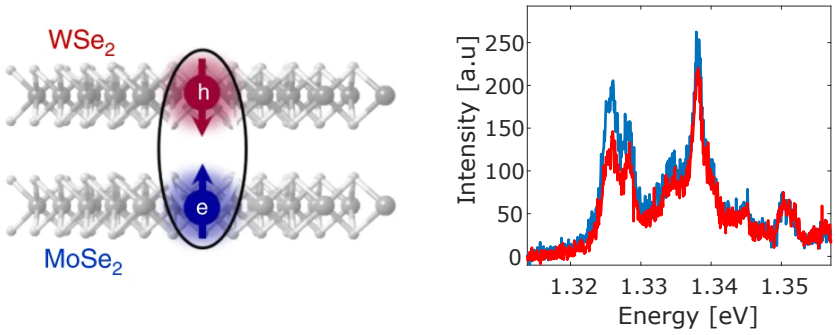

Figure 1: *(left)* We study a $MoSe_2$-WSe_2 hetero-bilayer hosting interlayer excitons resulting from the Coulomb attraction between electrons confined in a $MoSe_2$ monolayer and holes in the WSe_2 one. *(right)* Polarisation resolved PL spectrum of interlayer excitons at 5K. In these experiments electronic carriers are injected by a circularly polarised (σ^+) excitation at 1.62 eV, with an average power set to 50 nW. The PL is collected from the 1 μm wide region which is laser excited. The blue spectrum has the the same circular polarisation as the laser excitation, whereas the red one is detected along the orthogonal one.

References

[1] C. Lagoin and F. Dubin, Phys. Rev. B **103**, L041406 (2021)
[2] NP Wilson, W Yao, J Shan, X Xu, Nature **599**, 383-392 (2021)
[3] N. Goetting, F. Lohof, and C. Gies, arXiv:2201.10877 (2022)