

# Resonant Electronic and Excitonic Hybridisation in Transition Metal Dichalcogenide Alloys Heterobilayers

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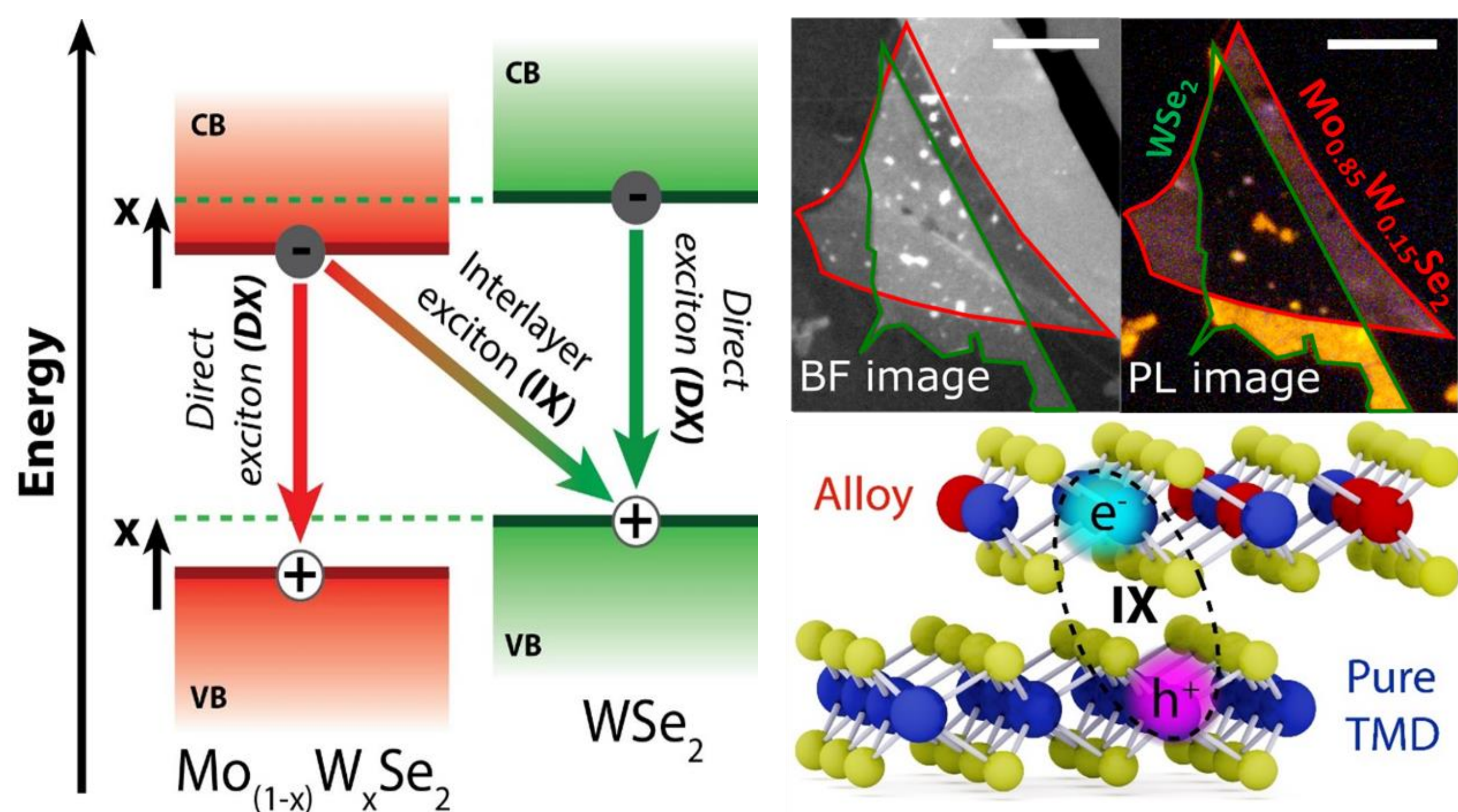
Dedicated to the memory of Alessandro Catanzaro who recently passed away

## 1 – Introduction

- Transition metal dichalcogenide (TMD) alloys are valid candidates to achieve fine tunability of the optical properties of Van der Waals (vdW) devices.[1]

## 2 - Van der Waals heterobilayer (HBL)

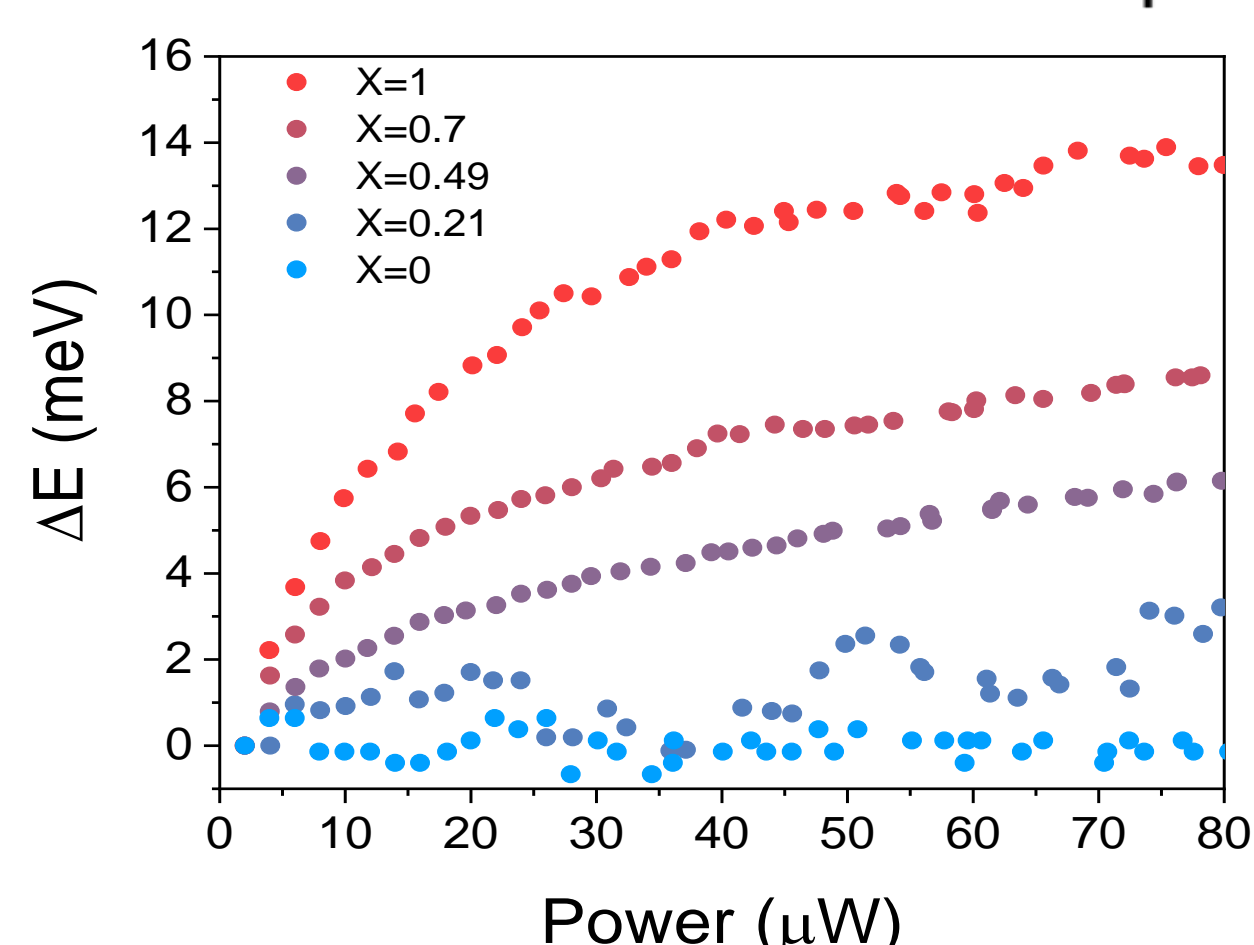
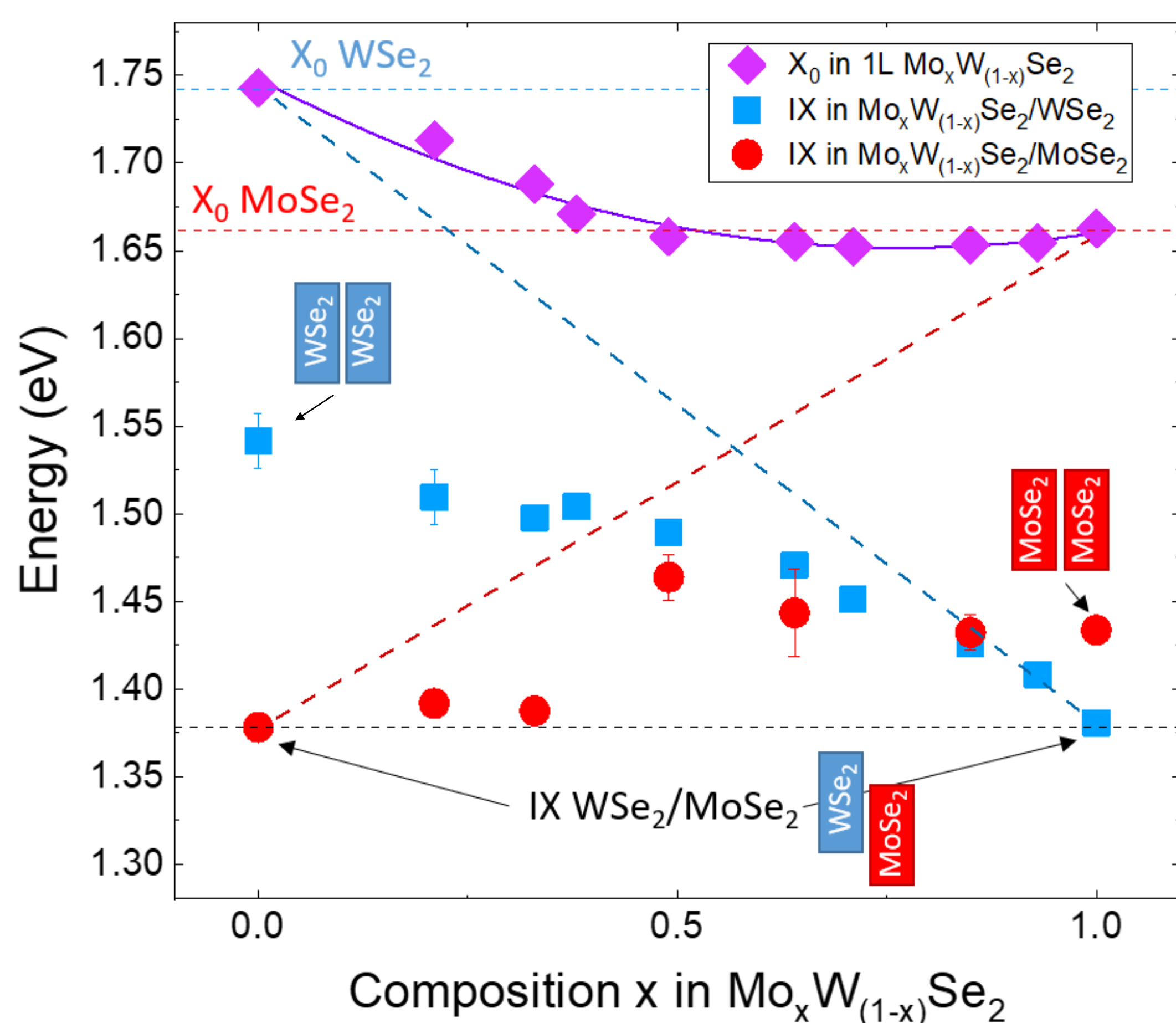
- TMD heterobilayers (HBLs) have a type II band alignment
- Ultrafast interlayer charge separation and interlayer excitons (IX)
- Intralayer excitons(DX) photoluminescence(PL) quenching.[2]



- IX emission energy is determined by the band offset; [2]
- $\text{Mo}_{1-x}\text{W}_x\text{Se}_2\backslash\text{WSe}_2$  and  $\text{Mo}_{1-x}\text{W}_x\text{Se}_2\backslash\text{MoSe}_2$  HBL IX peak control through alloy composition.

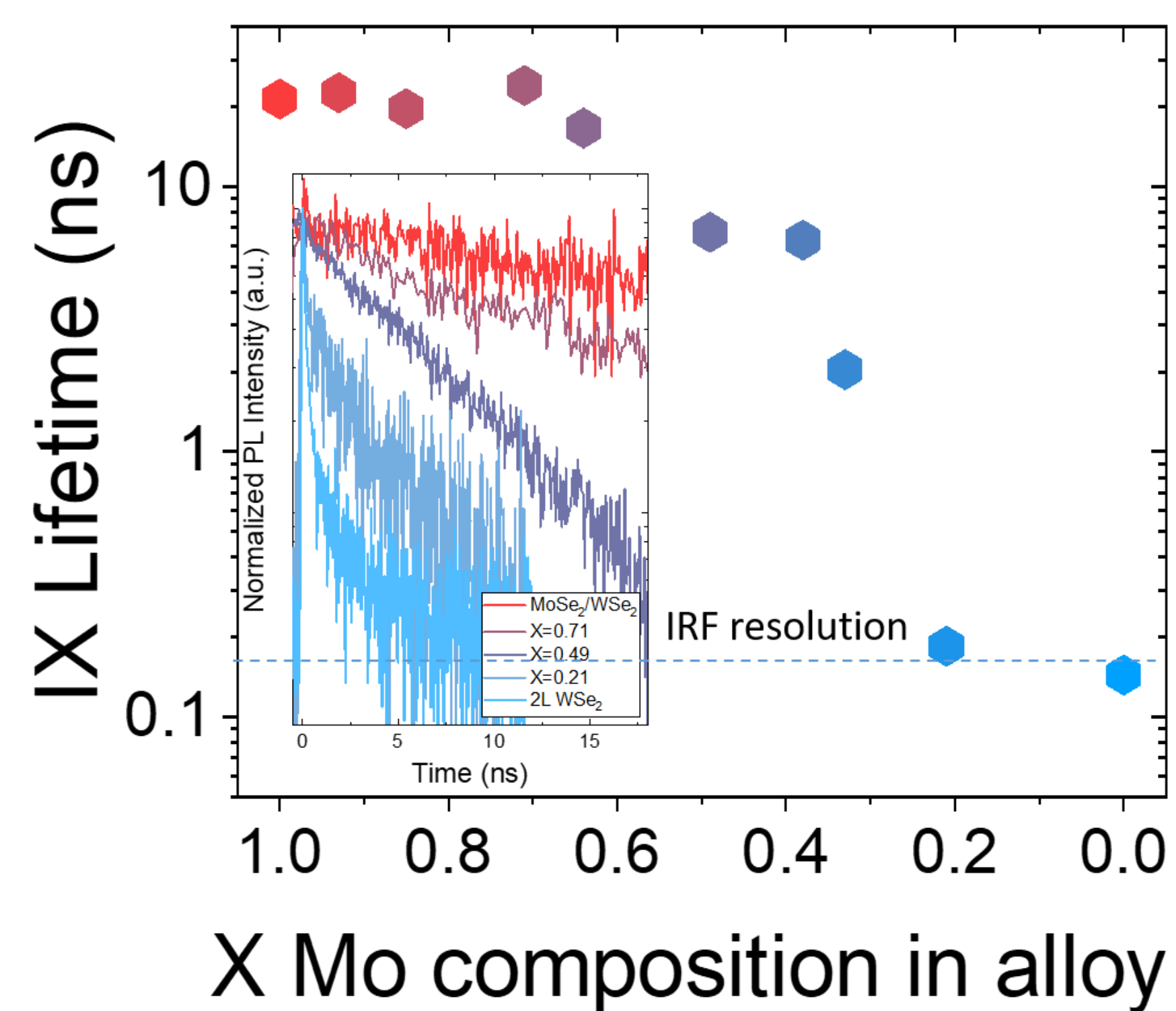
## 3 –Results

- For  $\text{Mo}_{1-x}\text{W}_x\text{Se}_2\backslash\text{WSe}_2$  HBLs IX emission energy blueshift from 1.38 to 1.52 eV as the alloy composition becomes more W-rich and the band offset is reduced. Same trend was confirmed in  $\text{Mo}_{1-x}\text{W}_x\text{Se}_2\backslash\text{MoSe}_2$  HBL.
- The IX PL peak deviate from the expected trend, pinning instead to the energy of the momentum indirect transition of bilayer configuration



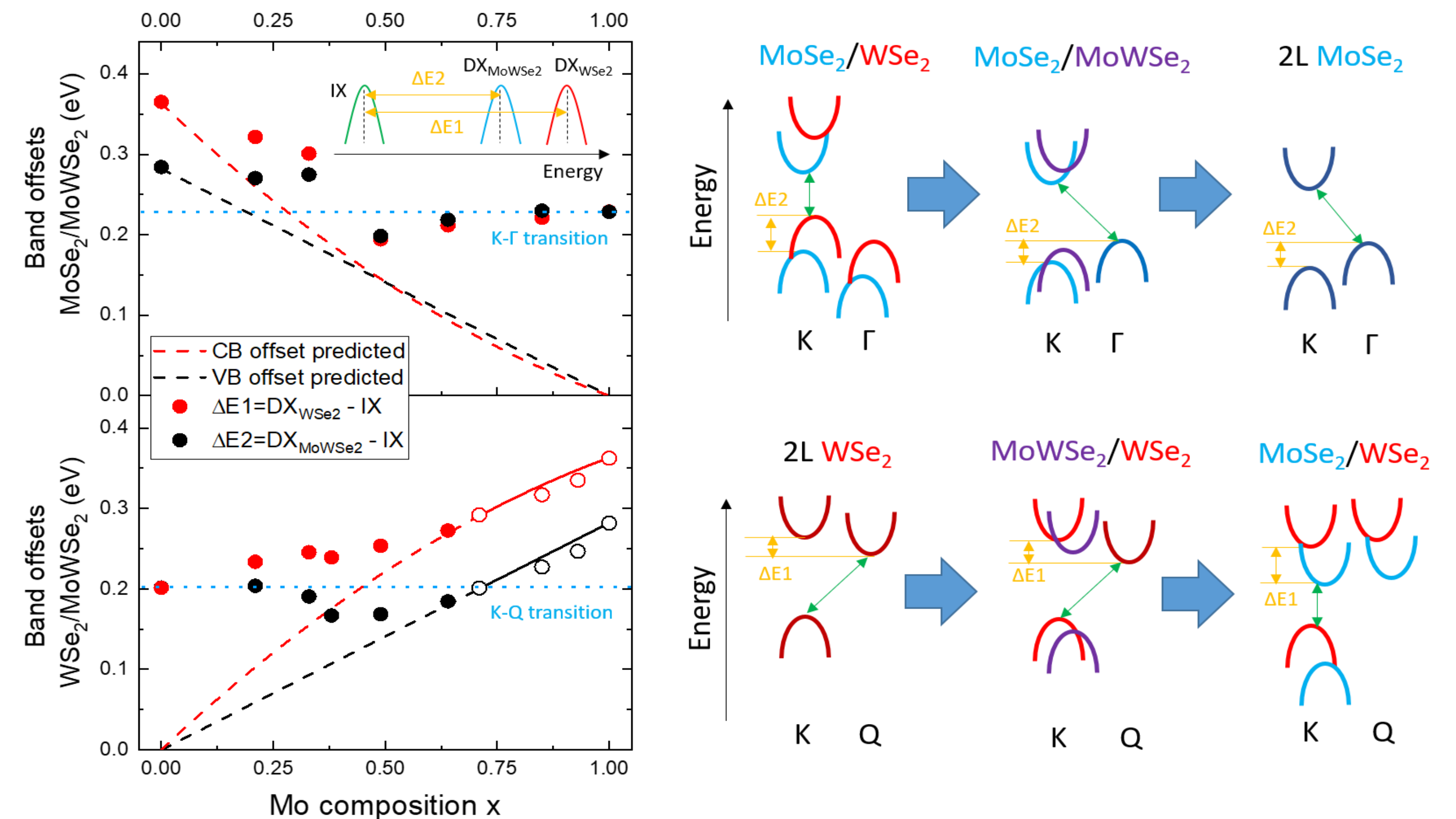
- Blueshift of the IX with increasing pump power due to the increase of IX density and IX-IX interactions.
- Blueshift reduces with increasing x, in  $\text{Mo}_{1-x}\text{W}_x\text{Se}_2\backslash\text{WSe}_2$  HBLs.

## 4 – Lifetime in $\text{Mo}_{1-x}\text{W}_x\text{Se}_2\backslash\text{WSe}_2$ HBLs



- Shortening of the lifetime from tens of nanoseconds (typical of IX) to few picoseconds (typical of momentum indirect transitions) with increasing x composition.

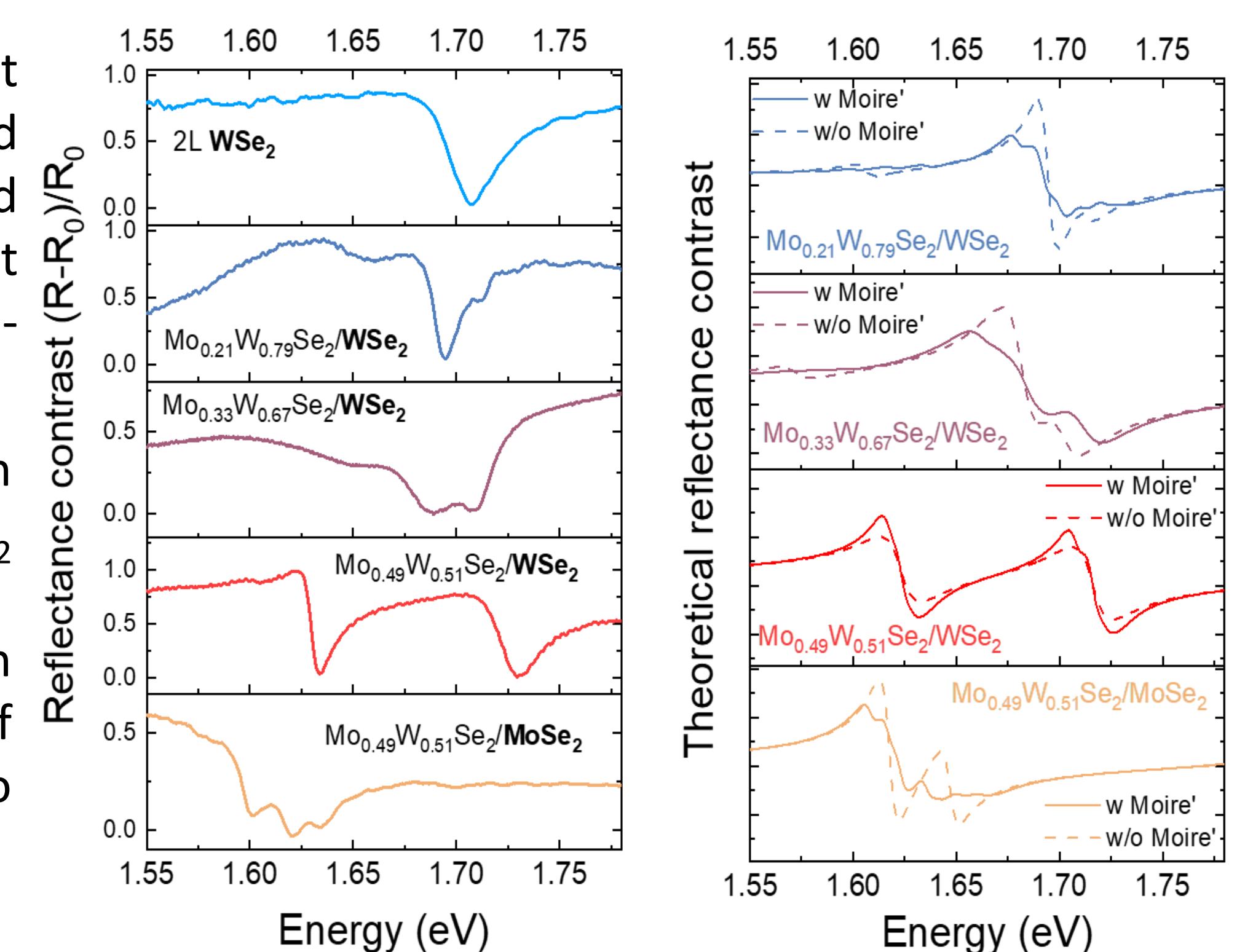
## 5 – Band Offset Consideration



- By considering measured DX and IX energies the band offsets deviate from those predicted for K-K transitions, approaching homobilayer (2L) configuration
- K-Q indirect transition for 2L WSe2 and  $\Gamma$ -K for 2L MoSe2 explain our results, in agreement with other studies in literature [3-5].

## 6 – Reflectance contrast and hybridization

- Reflectance contrast measurements revealed the rise of a third absorption peak at concentrations of quasi-degenerate energy states.
- Same trend observed in both  $\text{Mo}_{1-x}\text{W}_x\text{Se}_2\backslash\text{MoSe}_2$  and  $\text{Mo}_{1-x}\text{W}_x\text{Se}_2\backslash\text{WSe}_2$
- Attributed to exciton hybridization in presence of Moire' patterns, similar to [6].



## 7. Conclusions

- Tuning of the IX peak energy of more than 100 meV varying the composition of the alloy. IX peak pinning at the indirect exciton energy of the homobilayer pure TMD.
- Significant reduction of IX lifetime approaching homobilayer configuration
- Appearance of additional features in the absorption of nearly resonant HBLs.
- Reproducible pattern in both  $\text{Mo}_{1-x}\text{W}_x\text{Se}_2\backslash\text{MoSe}_2$  and  $\text{Mo}_{1-x}\text{W}_x\text{Se}_2\backslash\text{WSe}_2$ .

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## REFERENCES

[1] G. Wang *et al.*, *Nat. Commun.*, vol. 6, pp. 1–7, 2015.; [2] N. P. Wilson *et al.* *Nat. Nanotechnol.*, vol. 13, no. 11, pp. 1004–1015, 2018.; [3] Nguyen, P. V. *et al.*, *Nature*, 572(7768), 220–223, 2019.; [4] Zhang, Y. *et al.*, *Nature Nanotechnology*, 9(2), 111–115, 2014. [5] Wilson, N. R. *et al.*, *Science Advances*, 3(2), 1–8., 2017. [6] E. M. Alexeev *et al.*, *Nature*, vol. 567, no. 7746, pp. 81–86, 2019.