

Optical properties of single-layer, double-layer, and bulk MoS₂

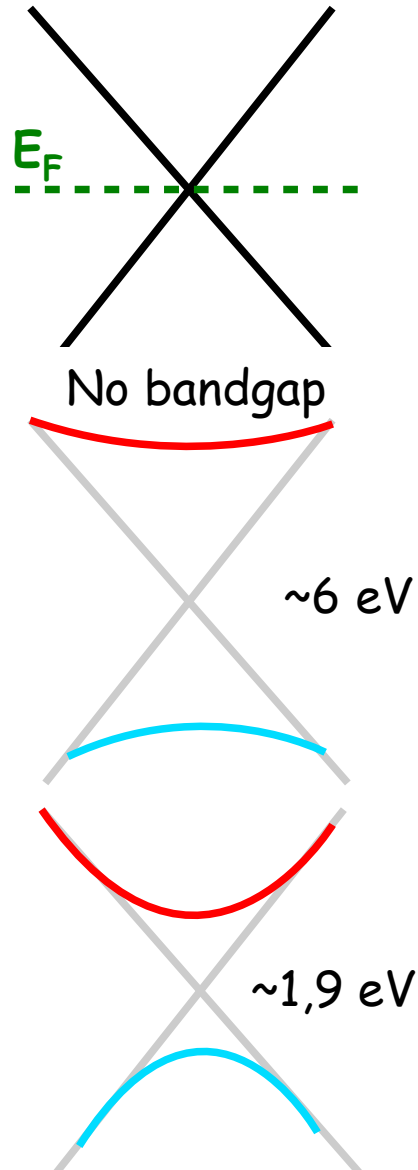
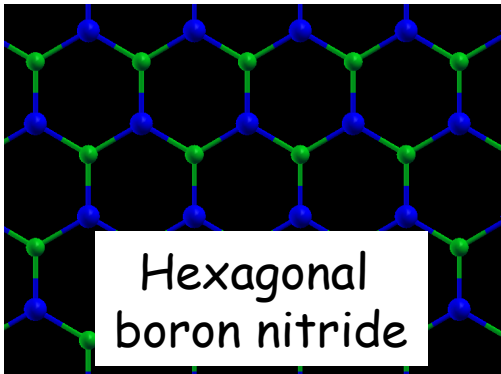
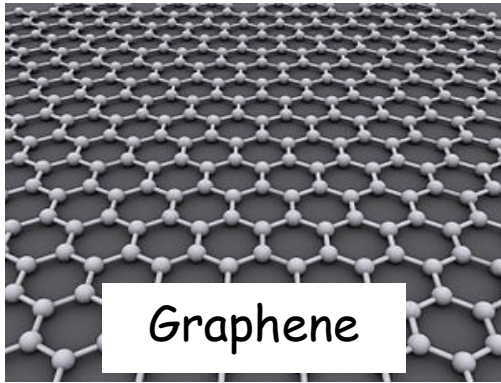
*Alejandro Molina-Sánchez, Ludger Wirtz,
Davide Sangalli, Andrea Marini, Kerstin
Hummer*



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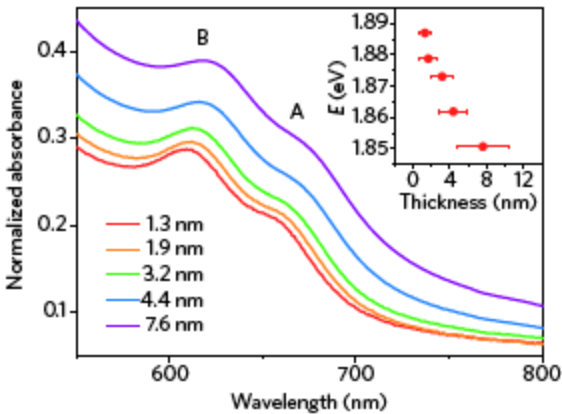
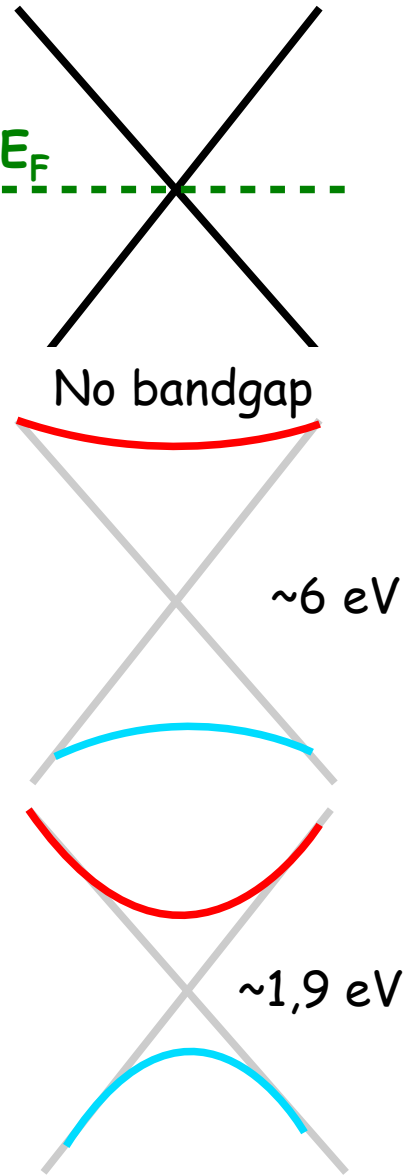
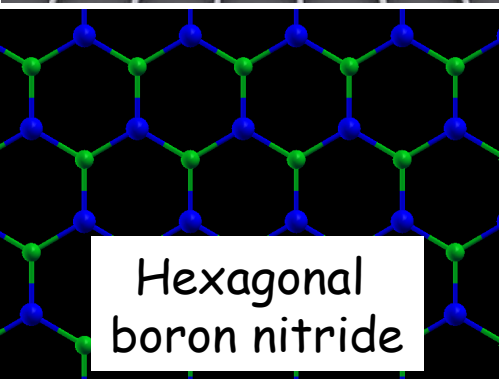
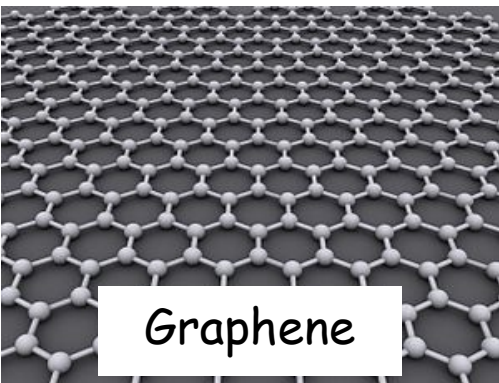
Single-layer semiconductors

From graphene to a new *family* of materials



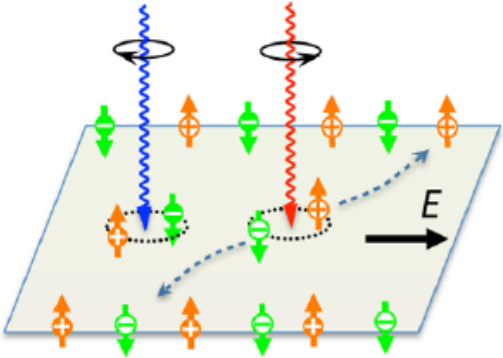
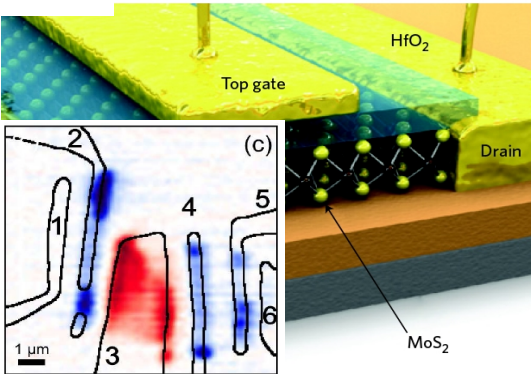
Single-layer semiconductors

From graphene to a new *family* of materials



The bandgap and the high mobility makes MoS₂ a suitable alternative to graphene or carbon nanotubes.

Appealing for transistors, tunable photo-thermoelectric effect, field-effect transistors ...



Spin-orbit coupling and class of symmetry lead to the spin and valley Hall effects of electrons and holes

Broad variety: MoSe₂, WS₂, MoS₂



See review Nature Nanotechnology 7, 699 (2012)

A. Castellanos, Nano Lett.,

Phys. Rev. Lett. 108, 196802 (2012)

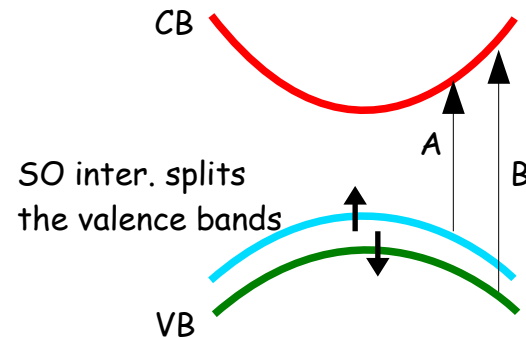
Transition metal dichalcogenides

Single-layer semiconductors

Crystal symmetry determines the electronic structure and optical properties

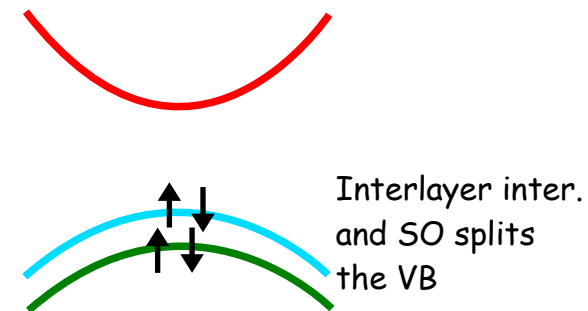
Single-layer (D_{3h})

No inversion symmetry



Double-layer and bulk (D_{6h})

Inversion symmetry

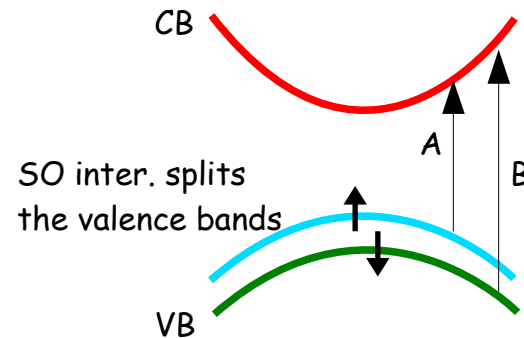


Transition metal dichalcogenides

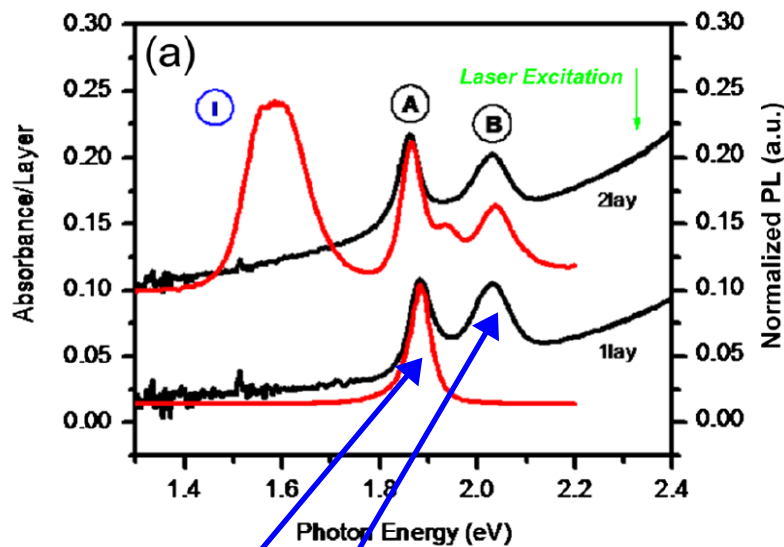
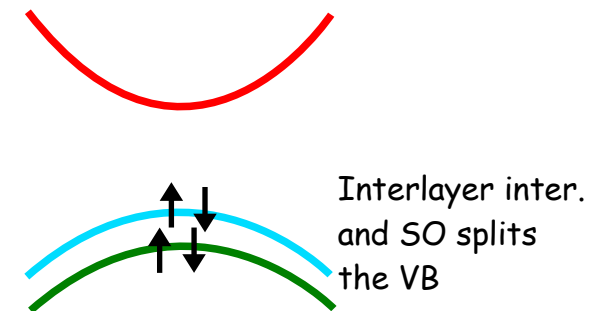
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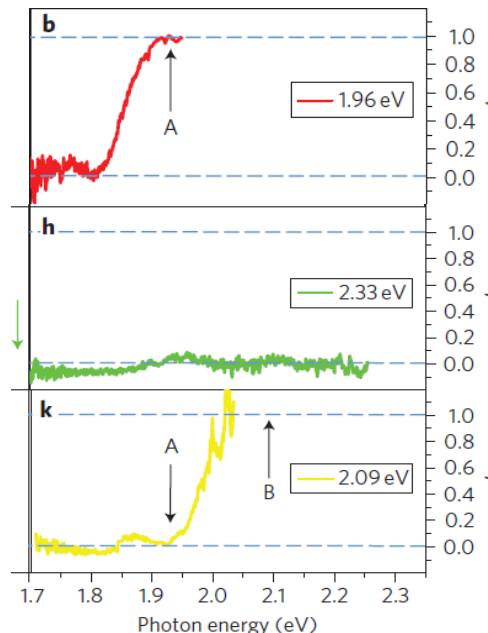
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VB splitting is exhibited in the PL and absorption by a two peaks structure



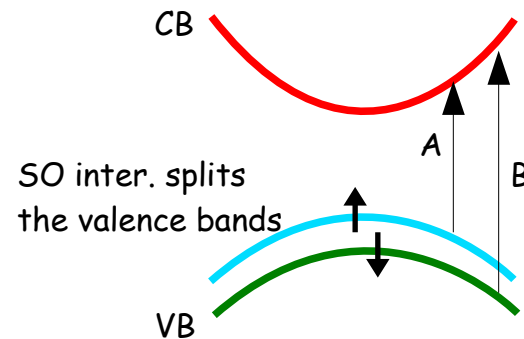
Control of the light polarization helicity by tuning the excitation energy (valley physics).

Transition metal dichalcogenides

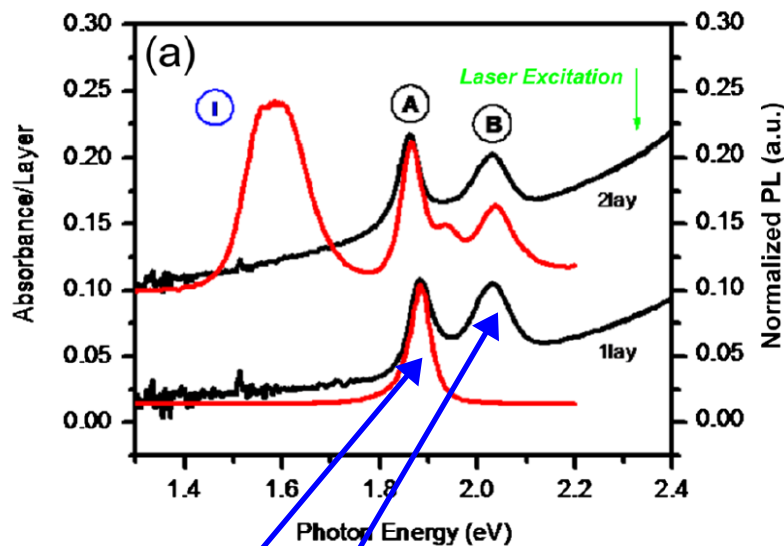
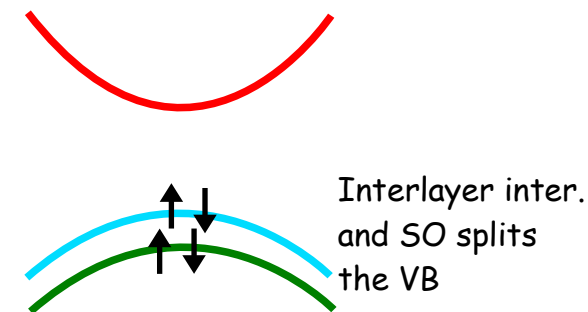
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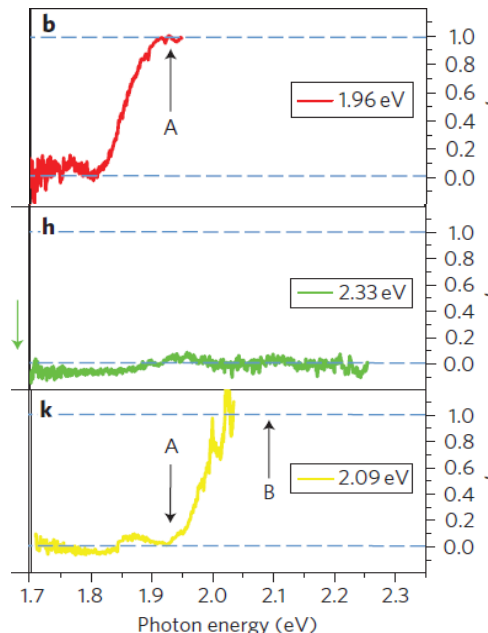
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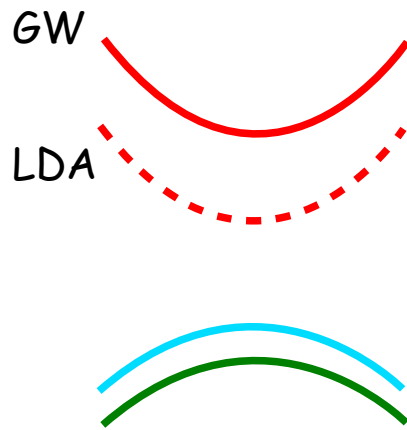
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Control of the light polarization helicity by tuning the excitation energy (valley physics).

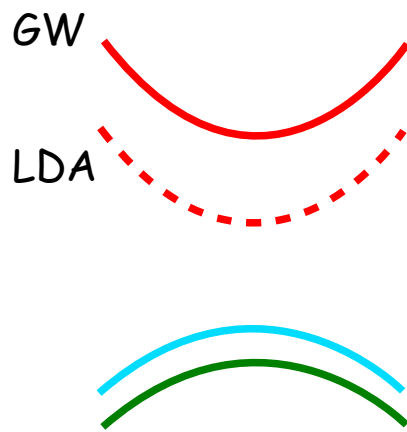
We have studied the electronic structure and the optical properties of MoS₂, and the dependence on the number of layers on the excitonic effects

MoS₂. Band Structure. LDA and GW method

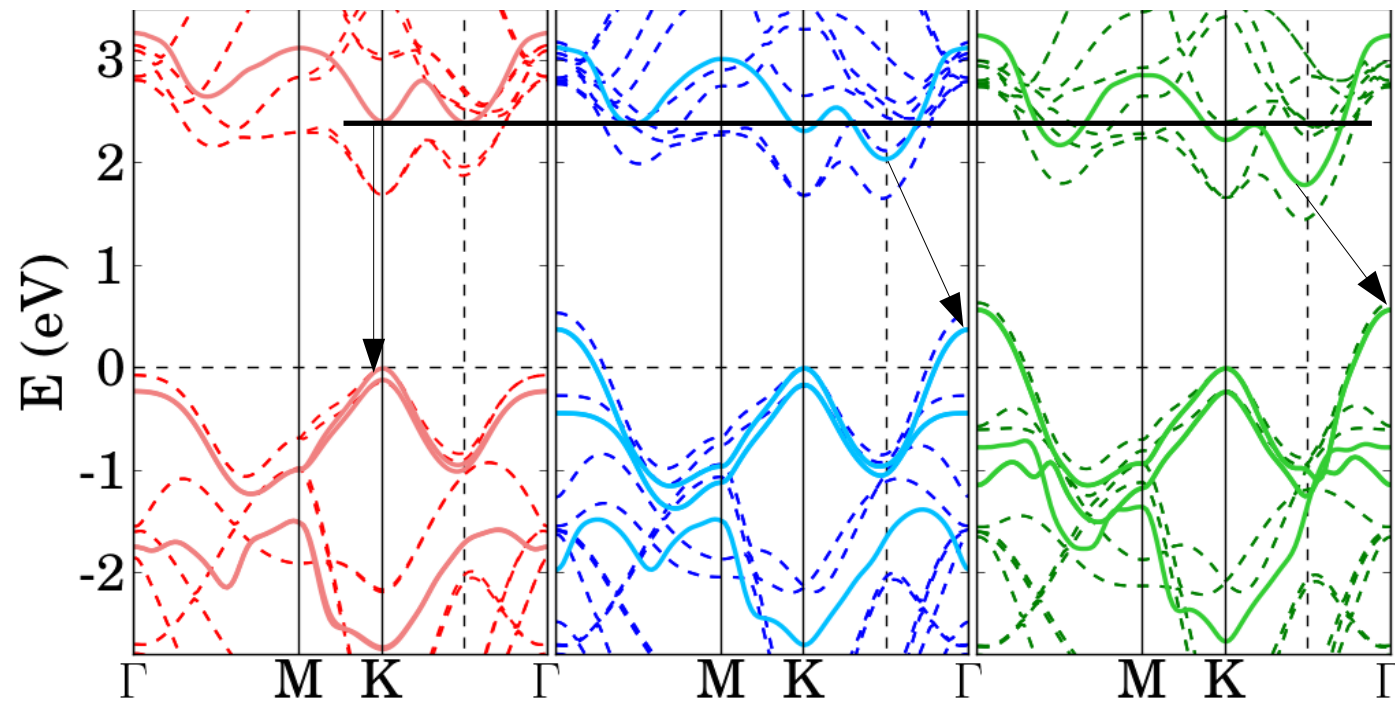


Correction of LDA bandgap underestimation by means of the GW method (spin-orbit interaction is included).

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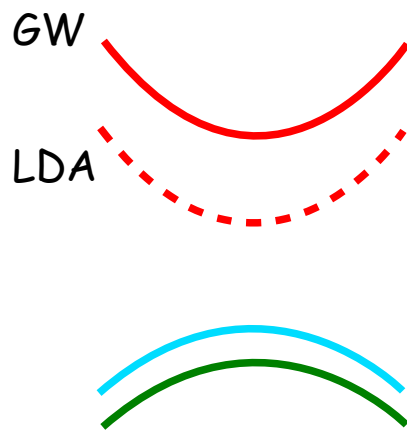
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Electronic correlation is very sensitive to variations in dielectric screening.

The addition of more layers changes the dielectric screening, the symmetry and the interlayer interaction.

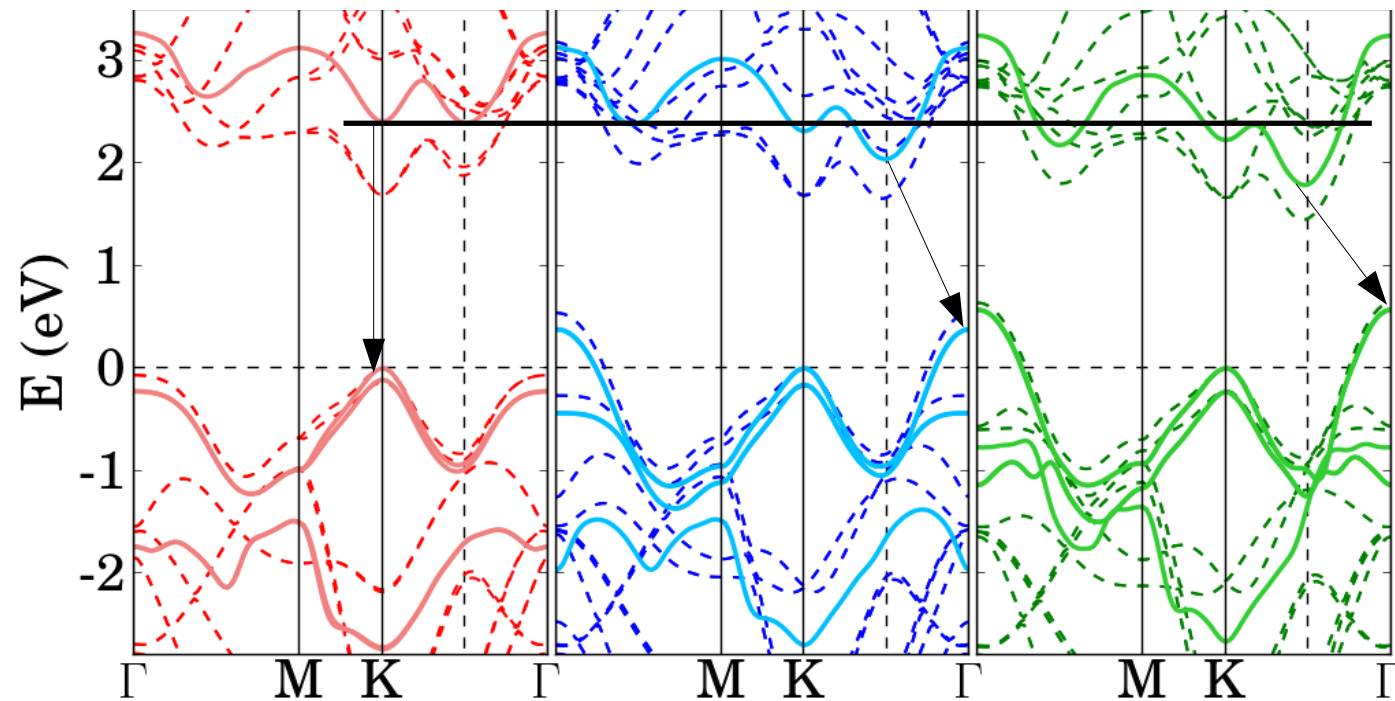
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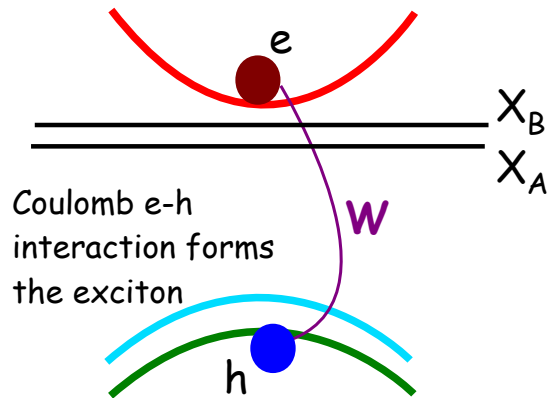
The GW correction is mainly a rigid shift of the conduction band. The correction is smaller when the number of layers increases (screening of the electron correlation).

Bandgap extremely sensitive to lattice optimization

The addition of more layers pushes up the valence band at Γ , making double-layer and bulk MoS₂ indirect SCs.

Excitonic effect on the optical properties...

MoS₂. Excitonic Effects. Bethe-Salpeter Equation



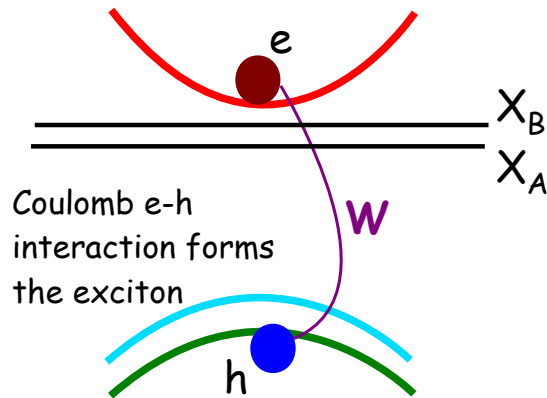
$$H^{exc}_{(n_1, n_2), (n_3, n_4)} = \boxed{(E_{n_2} - E_{n_1}) \delta_{(n_1, n_3)} \delta_{(n_2, n_4)}} \quad \text{Energy difference}$$

$$+ \boxed{i(f_{n_2} - f_{n_1}) \Xi_{(n_1, n_2), (n_3, n_4)}} \quad \text{Bethe-Salpeter Kernel}$$

$$\Xi_{\mathbf{K}_1 \mathbf{K}_2} = \boxed{-iV_{\mathbf{K}_1 \mathbf{K}_2}} + \boxed{iW_{\mathbf{K}_1 \mathbf{K}_2}} \quad \mathbf{K} \equiv (c, v, \mathbf{k})$$

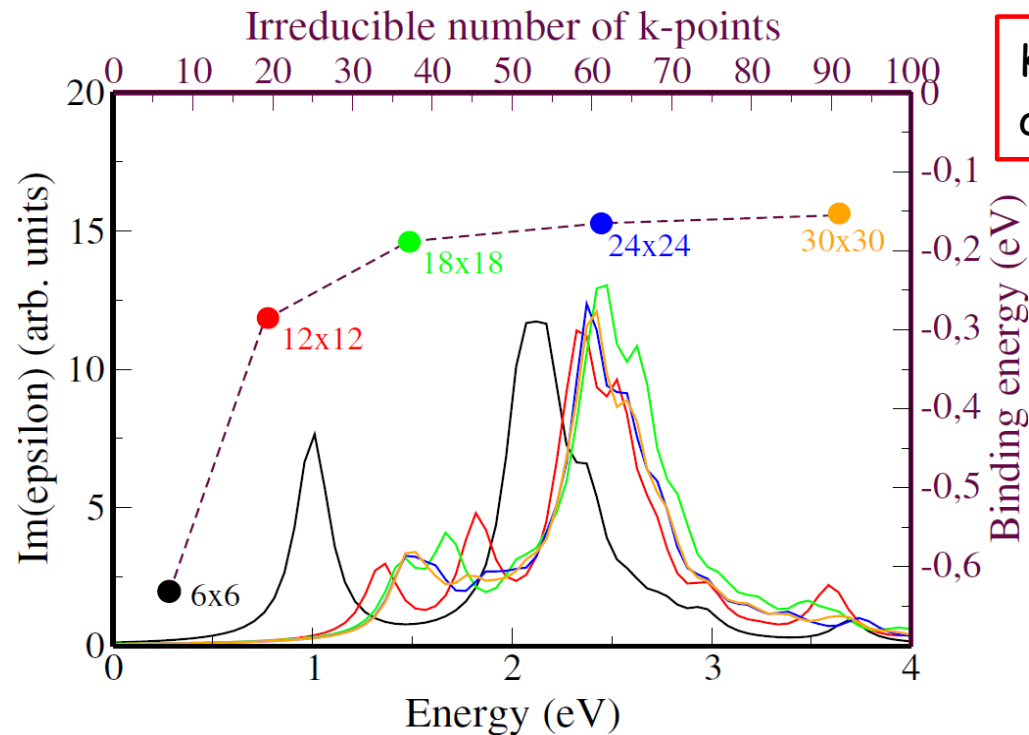
Unscreened short ranged exchange interaction Screened coulomb interaction

MoS₂. Excitonic Effects. Bethe-Salpeter Equation



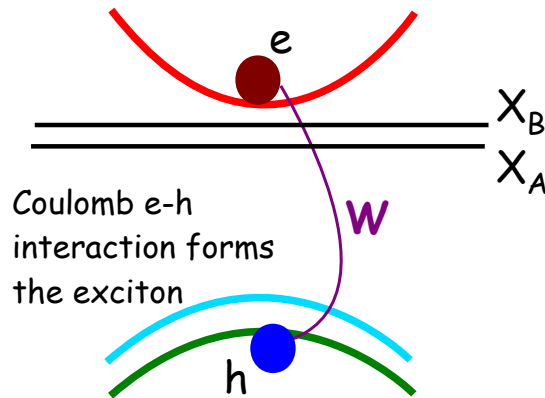
$$H_{(n_1, n_2), (n_3, n_4)}^{exc} = \underbrace{(E_{n_2} - E_{n_1}) \delta_{(n_1, n_3)} \delta_{(n_2, n_4)}}_{\text{Energy difference}} + \underbrace{i(f_{n_2} - f_{n_1}) \Xi_{(n_1, n_2), (n_3, n_4)}}_{\text{Bethe-Salpeter Kernel}}$$

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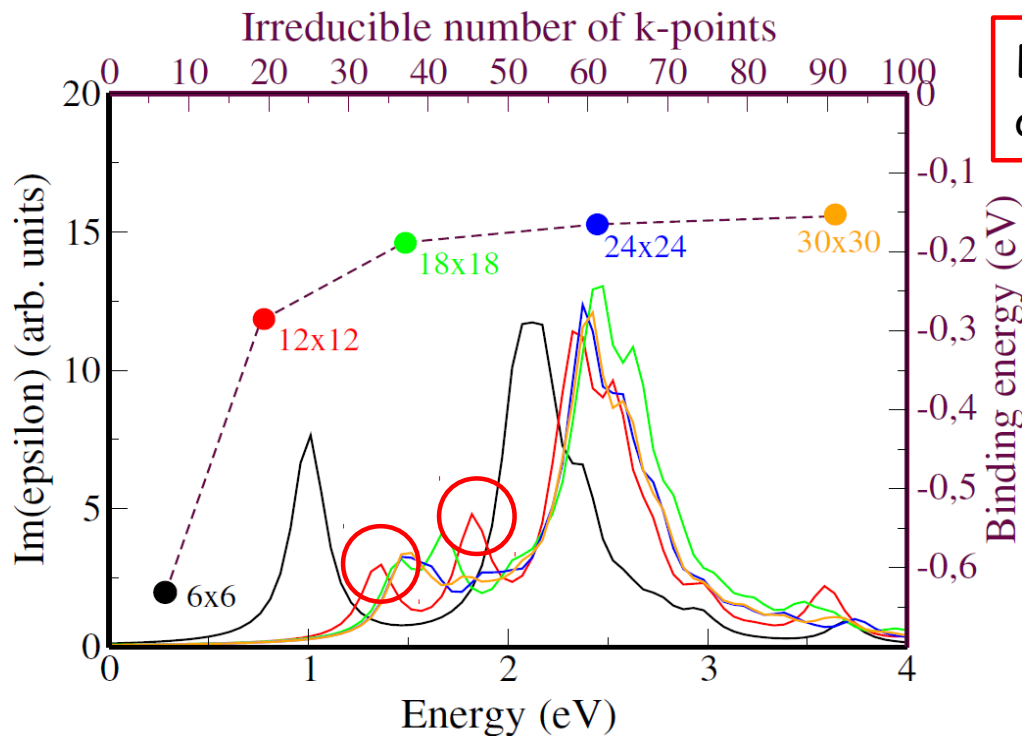
Key issue in reliable results: convergence in number of conduction and valence band states and k-points!

MoS₂. Excitonic Effects. Bethe-Salpeter Equation



$$H_{(n_1, n_2), (n_3, n_4)}^{exc} = \underbrace{(E_{n_2} - E_{n_1}) \delta_{(n_1, n_3)} \delta_{(n_2, n_4)}}_{\text{Energy difference}} + \underbrace{i(f_{n_2} - f_{n_1}) \Xi_{(n_1, n_2), (n_3, n_4)}}_{\text{Bethe-Salpeter Kernel}}$$

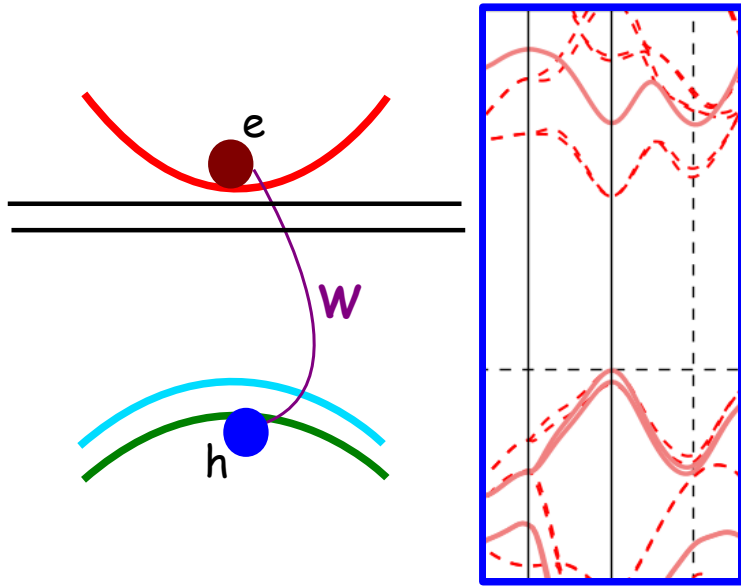
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Key issue in reliable results: convergence in number of conduction and valence band states and k-points!

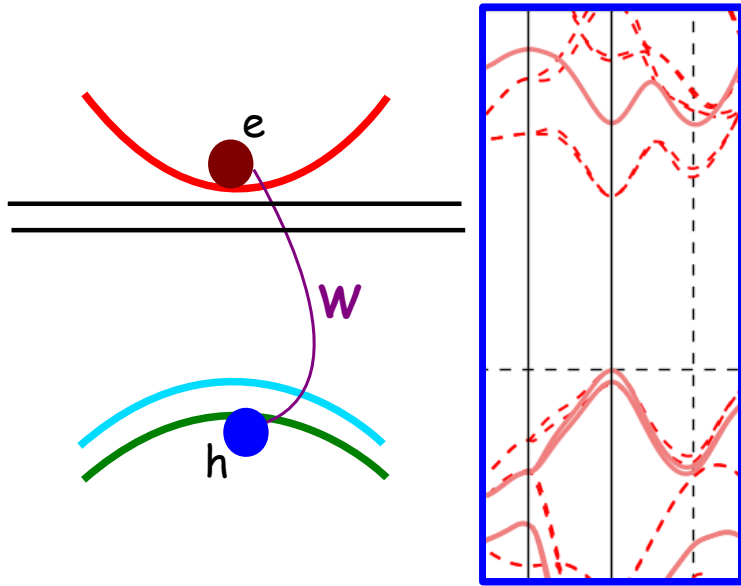
Lack of convergence in k-points normally overestimate the exciton binding energy. The k-sampling is directly related to the numbers of unit cells employed to map the exciton wave functions. Low k-sampling gives also artifacts in the optical absorption.

MoS₂. Excitonic Effects. Bethe-Salpeter Equation



The exciton binding energy decreases with the number of layers. Consequence of larger dielectric screening. This compensates partially the *GW* correction.

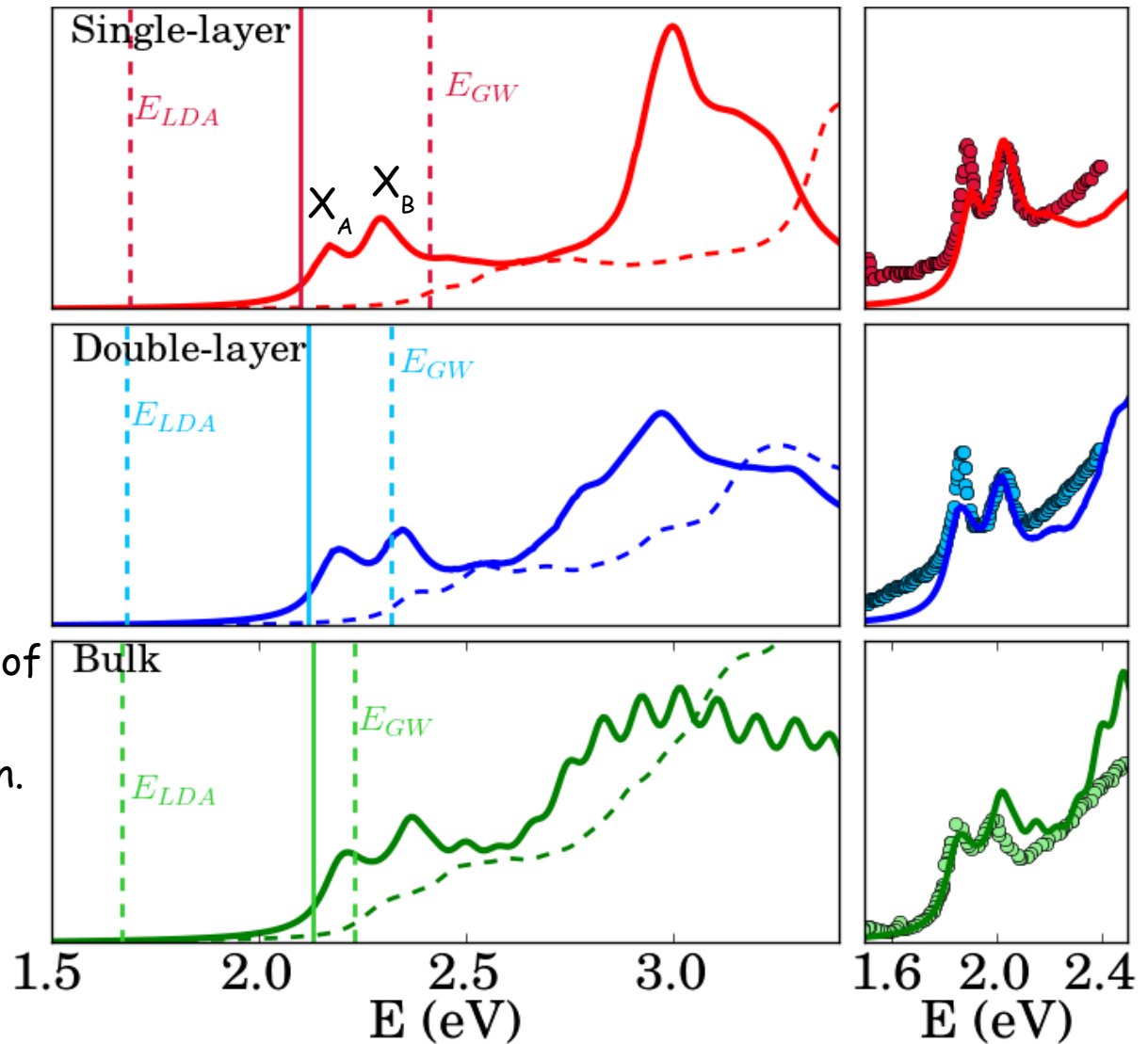
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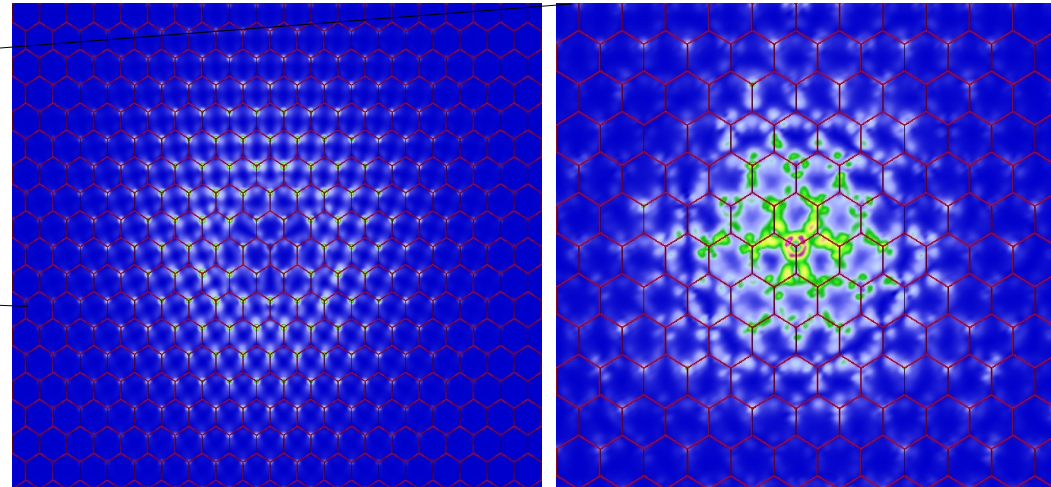
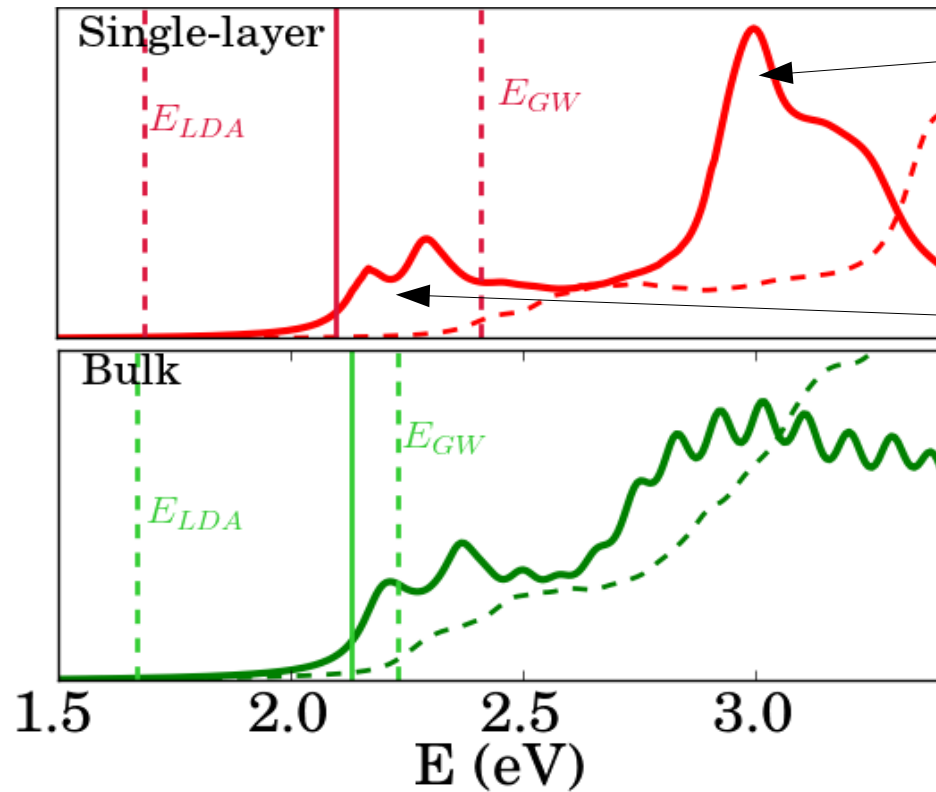
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For single-layers (around 3 eV) the absorption gains in efficiency (strongly bound exciton).

The theoretical spectra captures nicely the peaks separation for all the cases.

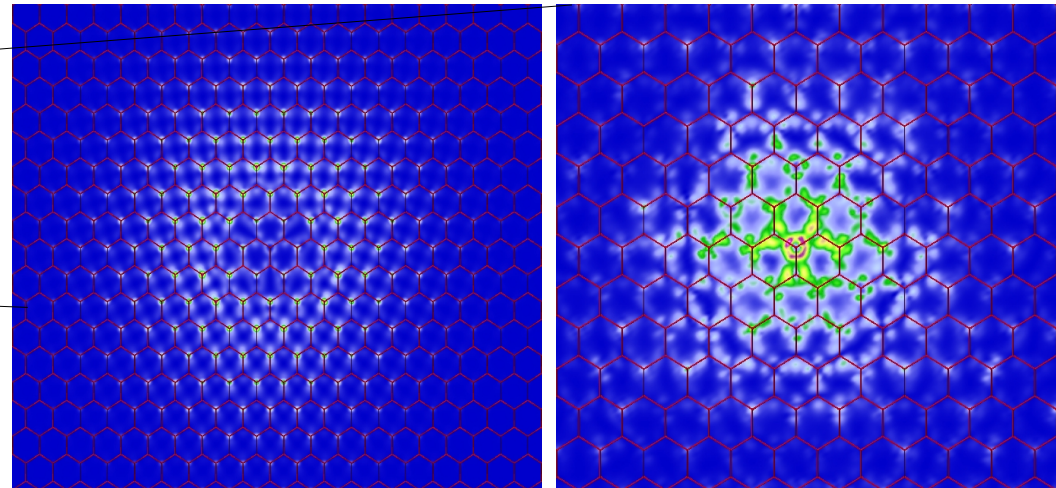
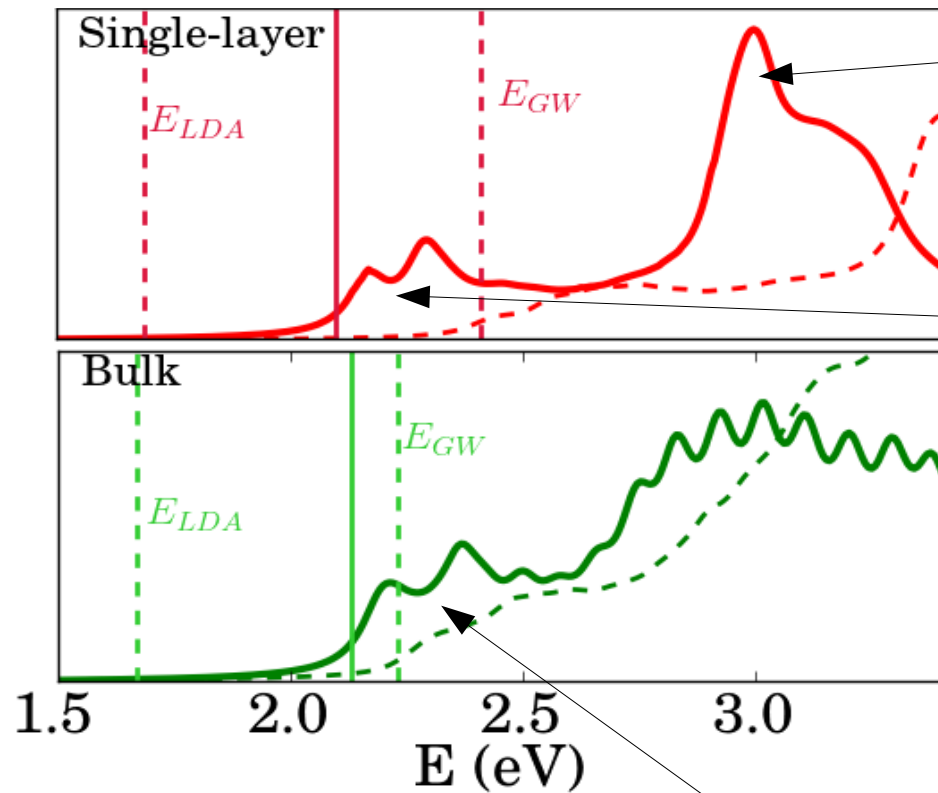


MoS₂. Excitonic Effects. Bethe-Salpeter Equation



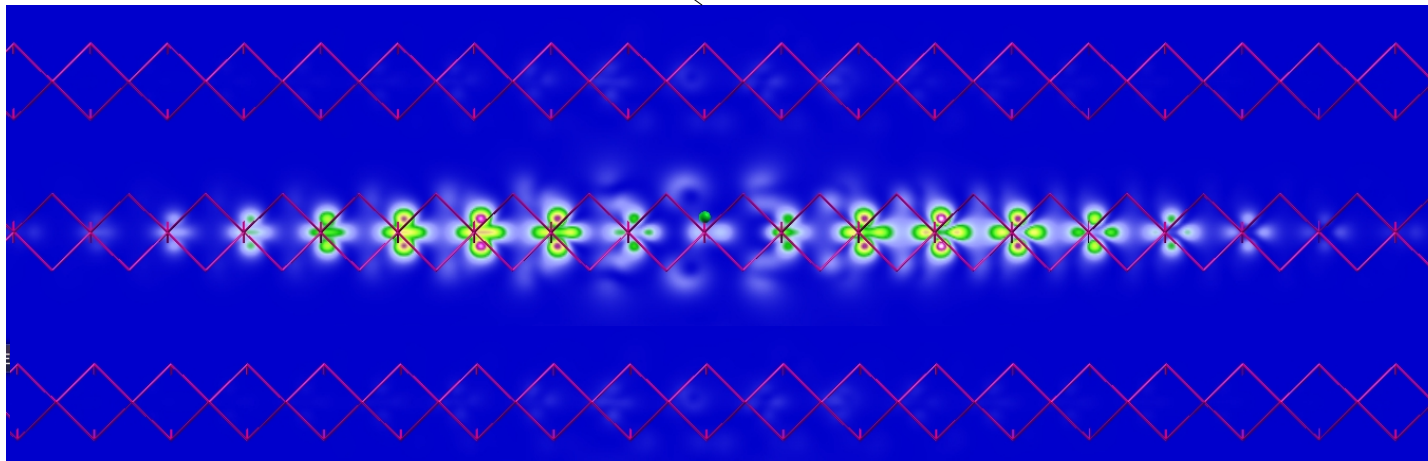
The intensity of the optical absorption is related with the localization of the excitons.

MoS₂. Excitonic Effects. Bethe-Salpeter Equation



The intensity of the optical absorption is related with the localization of the excitons.

For bulk, the exciton is confined in only one layer, due to the large interlayer distance.



We can appreciate the shape of the d-orbitals in the excitonic wavefunction

Conclusions and ongoing work

- The bandgap (direct or indirect) depends critically on the number of layers and lattice optimization.
- Importance of convergence for reliable results.
- Excitonic effects are stronger in environments with small dielectric constant (single-layers).
- Further studies will deal with strained layers and the influence on the optical response (tunability of the bandgap).

Acknowledgements

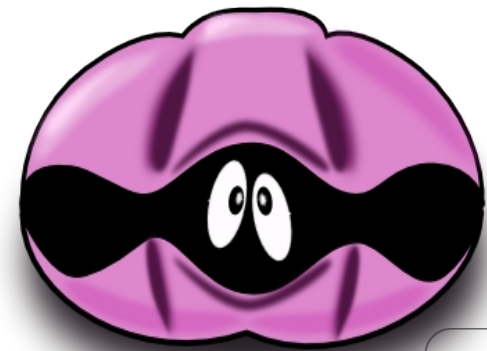


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Thank you for your attention!