EXCITONIC EFFECTS IN THE OPTICAL PROPERTIES OF SINGLE-LAYER, BI-LAYER AND BULK MoS_2

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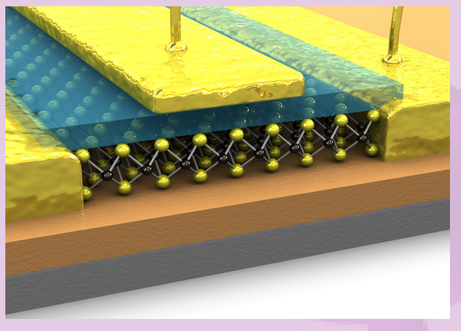
MOTIVATION



Single-layer MoS_2 is a promising semiconductor in the field of transistors and optoelectronic devices [1].
Bulk MoS_2 structure is a hexagonal lattice with layers attached by van der Waals forces.

The most remarkable optical properties of MoS_2 are [2,3]:

1) The bandgap changes from direct to indirect when it passes from single-layer MoS_2 to multi-layer MoS_2.
2) Photoluminescence is much more efficient in single-layer than in multi-layers.
3) However, optical absorption spectra are similar for any structure.

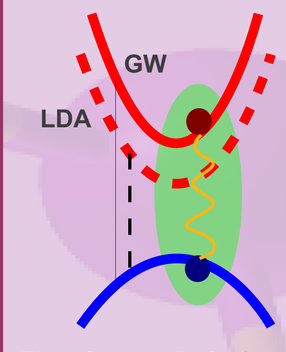
We analyze theoretically the importance of the electron-electron interaction and the excitonic effects in the optical properties, keeping in mind the influence of the dielectric screening in the Coulomb interaction.

THEORETICAL METHODS

We apply state-of-the-art ab initio methods to study the electronic structure and the optical absorption of MoS2 [4,5].

1) LDA band structure and correction of the bandgap with the GW method.
2) Electron-hole Coulomb interaction »»»»»» ***excitons!***

1) GW method: One-particle Green's function formulation



The electron-hole Coulomb interaction renormalizes the bandgap

Screened Potential $W(r,r',\omega)$

Quasiparticle = electron + screening cloud

Self-energy $\Sigma=GW$

$$H_0(\mathbf{r})\Psi_i(\mathbf{r})+\int\Sigma(\mathbf{r},\mathbf{r}',\epsilon_i)\Psi_i(\mathbf{r}')d^3\mathbf{r}' = \epsilon_i\Psi_i(\mathbf{r})$$

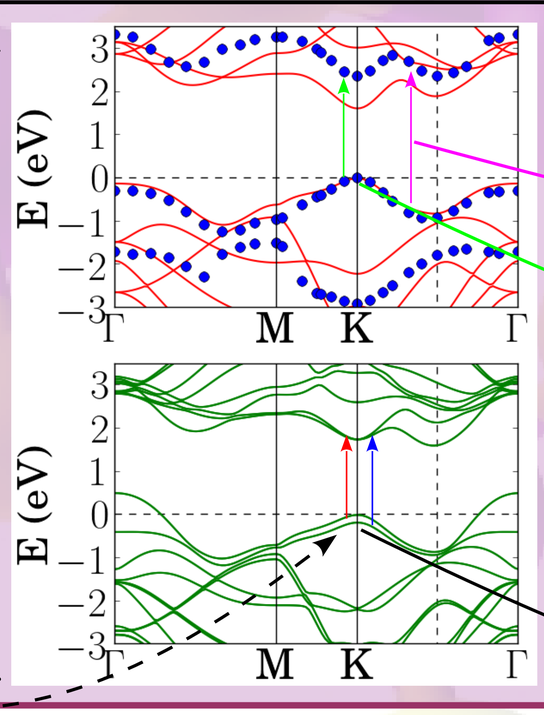
2) The excitonic effects are described with the Bethe-Salpeter equation

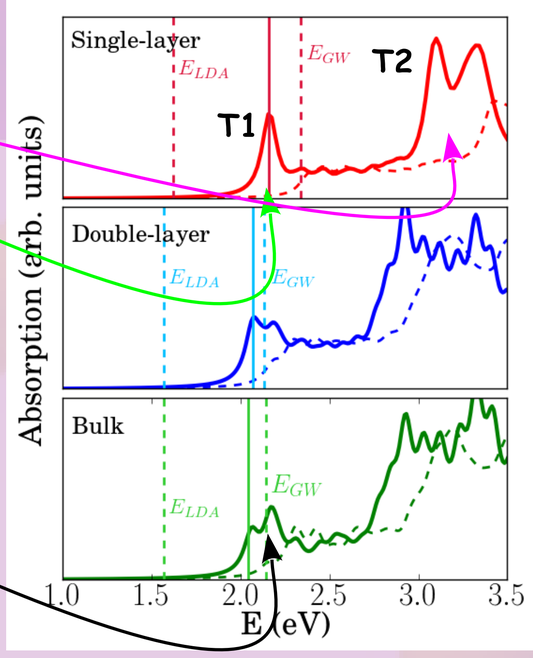
BAND STRUCTURE AND OPTICAL ABSORPTION

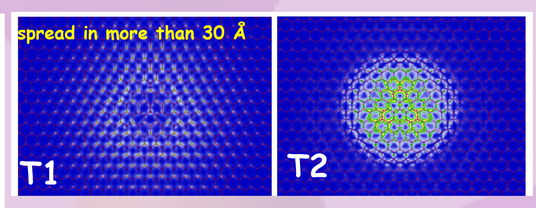
The GW correction is much larger in single-layer (SL) than bulk.
Semi-core *d*-orbitals or **Mo** are included.
Direct bandgap in LDA and GW calculation only for SL.

K-point	SL	DL	BULK
GW-corr. (eV)	0.72	0.56	0.56

The electron-electron correlation is attenuated in SL in comparison with bulk.
A more detailed description of SL needs of spin-orbit interaction.
In DL and BULK, the **splitting** at K at the valence band (VB) is only due to the interlayer interaction (inversion symmetry).

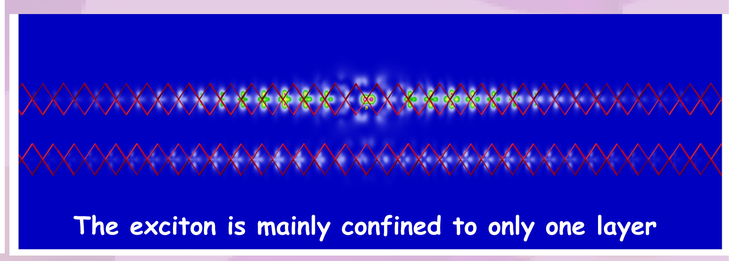




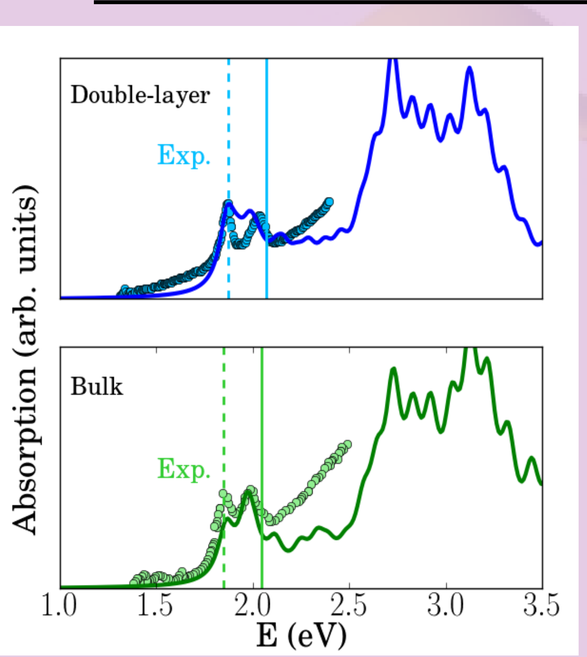


We find a remarkable second peak, at 3.0 eV, with the exciton confined in a few atoms.

The electron-hole interaction cancels partially the up-shift of the GW-correction in all the cases but more evidently in SL. The two-peaks in DL and BULK are due to interlayer interaction (absence in SL). All spectra are very similar (**Q1**).



COMPARISON WITH EXPERIMENTS



In the case of DL and BULK, the interlayer interaction produces the splitting and the inversion symmetry prevents of spin-orbit splitting. Therefore, we can compare the theoretical spectra with the experimental data of optical absorption for DL and BULK (we have corrected 0.2 eV our spectra).

The DL and BULK spectra are almost identical, theoretically and experimentally (**Q2**).

CONCLUDING REMARKS

Q1: The first peaks comes from the energy transitions at **K**, where SL, DL and BULK electronic structures are very similar. The main difference is the direct band gap of SL (as shown in GW calculations), that gives a more efficient photoluminescence.

Q2: The exciton is essentially confined in one-layer. In the case of DL and BULK, the stacking of more layers will influence in the dielectric screening but won't substantially modify the wave functions.

The electron-electron correllation and the electron-hole interaction are much larger in SL than in DL and bulk. There is an effect of cancellation observed in the optical spectra..

REFERENCES

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[4] ABINIT http://www.abinit.org/
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[7] A. Molina-Sánchez and L. Wirtz. *To be submitted.*