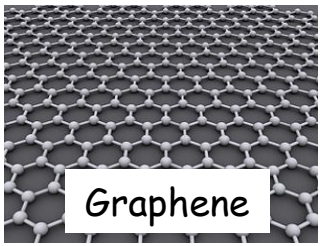
Optical and vibrational properties of layered MoS2

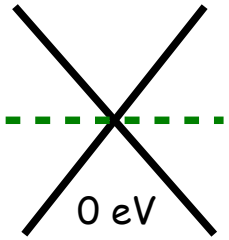
Alejandro Molina-Sánchez and Ludger Wirtz

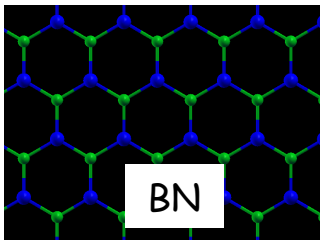
Theory, Modelling and Computational Methods for Semiconductors
Manchester, January 2014

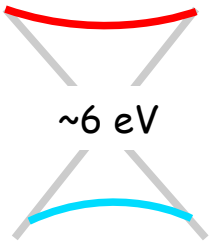
Layered materials. Semiconductor MoS_2

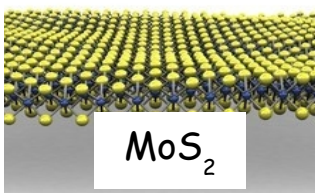
Two-dimensional materials. Search of new functionalities.

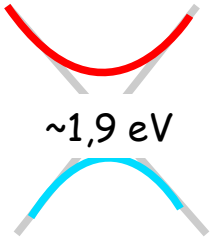


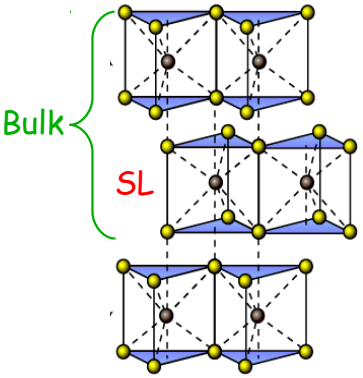












See review Nature Nanotechnology 7, Nano Lett. 13, 358 (21013) .
Incoming Review in Surface Science Reports

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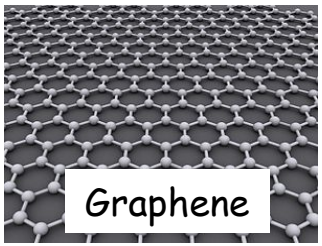
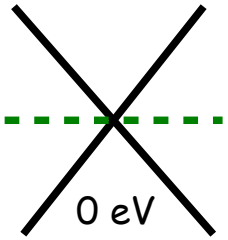
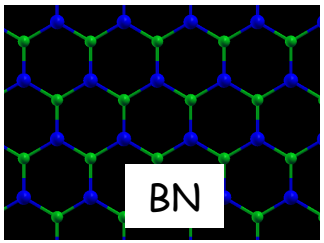
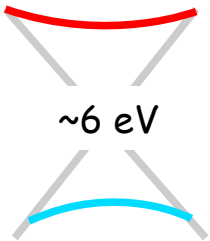
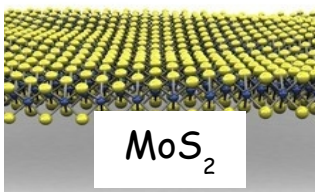
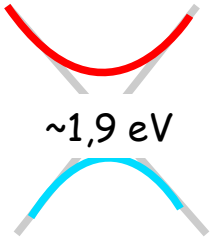


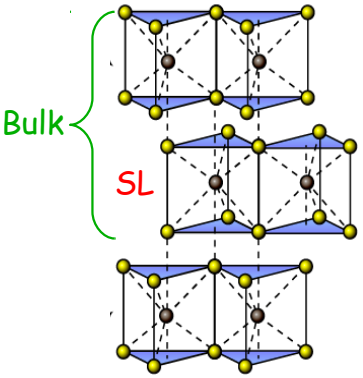




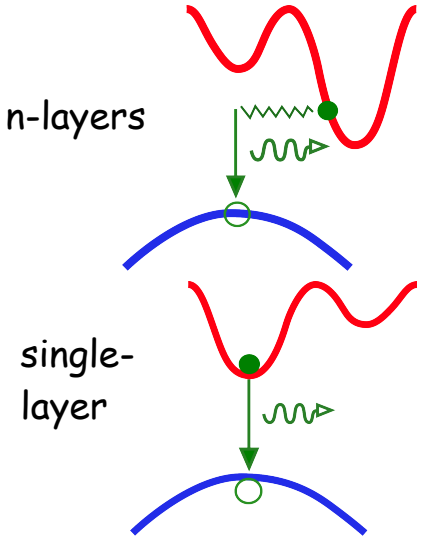








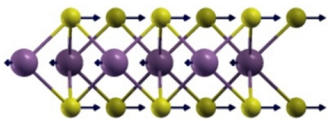
Electronic and optical properties



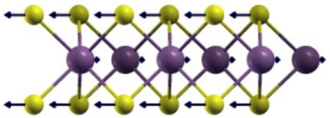
Direct/indirect bandgap depending on <u>number of layers</u> and <u>strain</u>

Higher efficiency of photoluminescence in single-layers

Vibrational properties



Phonon modes are very sensitive to layer thickness.



Counter-intuitive trend of the in-plane mode.

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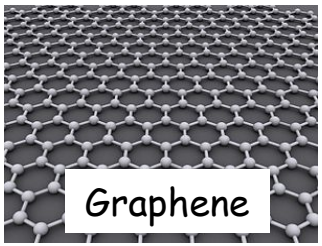

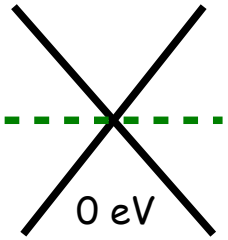

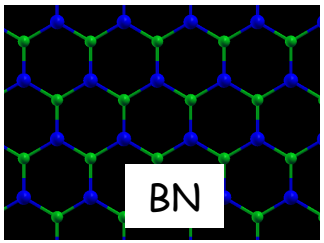

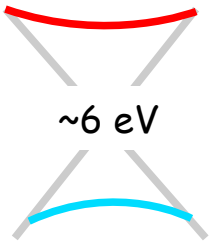

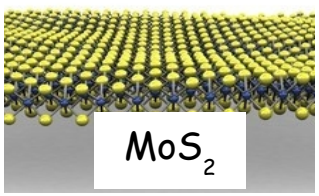

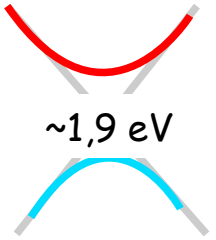

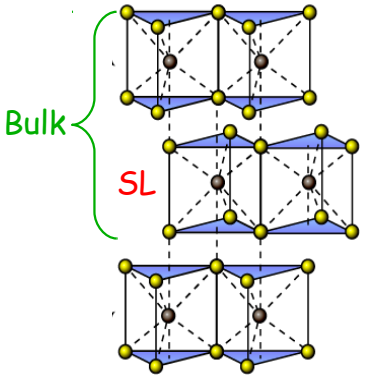

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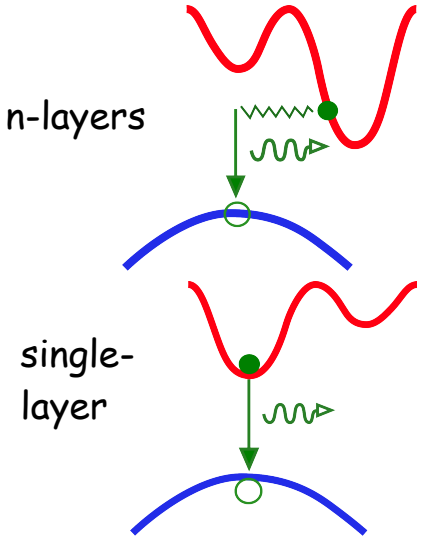

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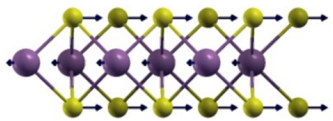

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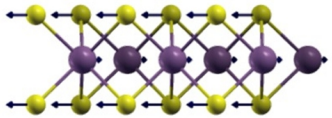

Counter-intuitive trend of the in-plane mode.

Ab-inito approach for the study of band structure, optical spectra, phonons

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See review Nature Nanotechnology 7, Nano Lett. 13, 358 (21013) .
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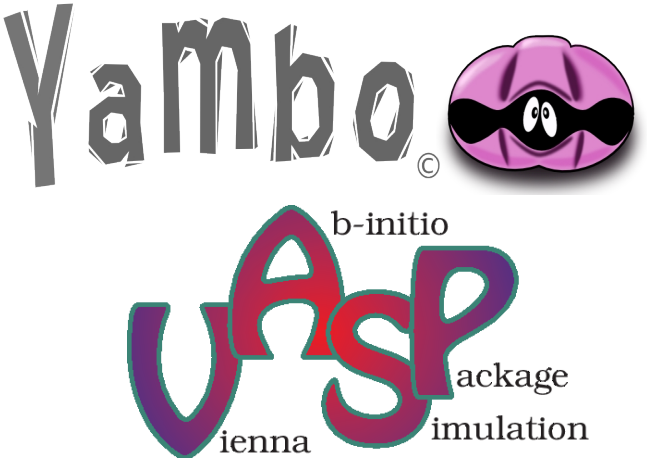
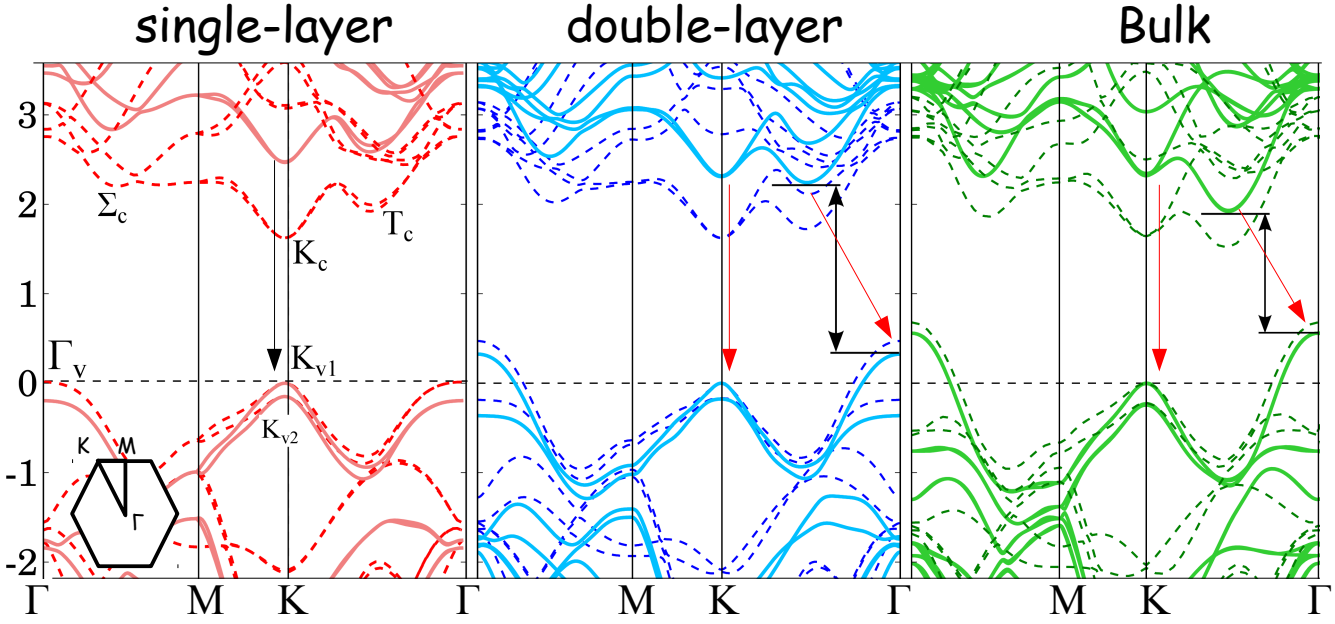
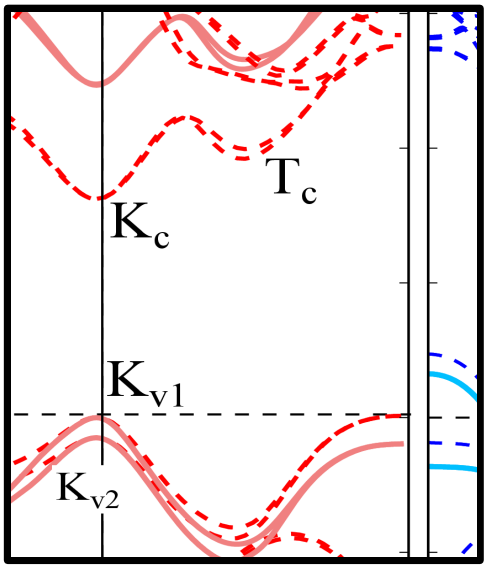
Electronic structure

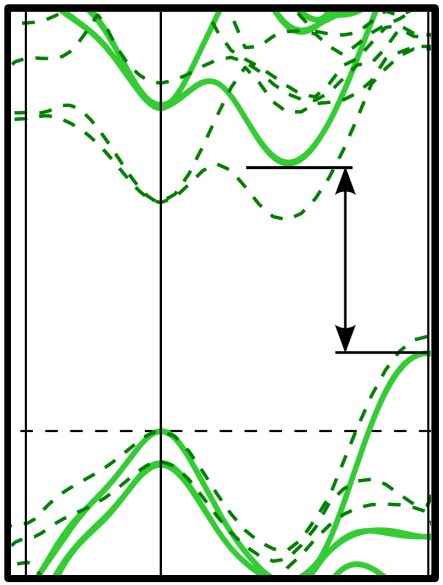
Beyond LDA. scGW-method
Spinor fully described
Semi-core d-orbitals.

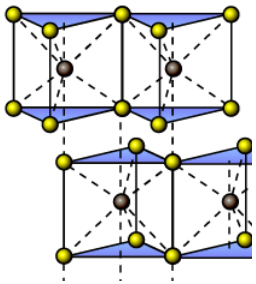




Conduction band is very sensitive to the crystal structure and to the GW "flavour"







Interaction between layers

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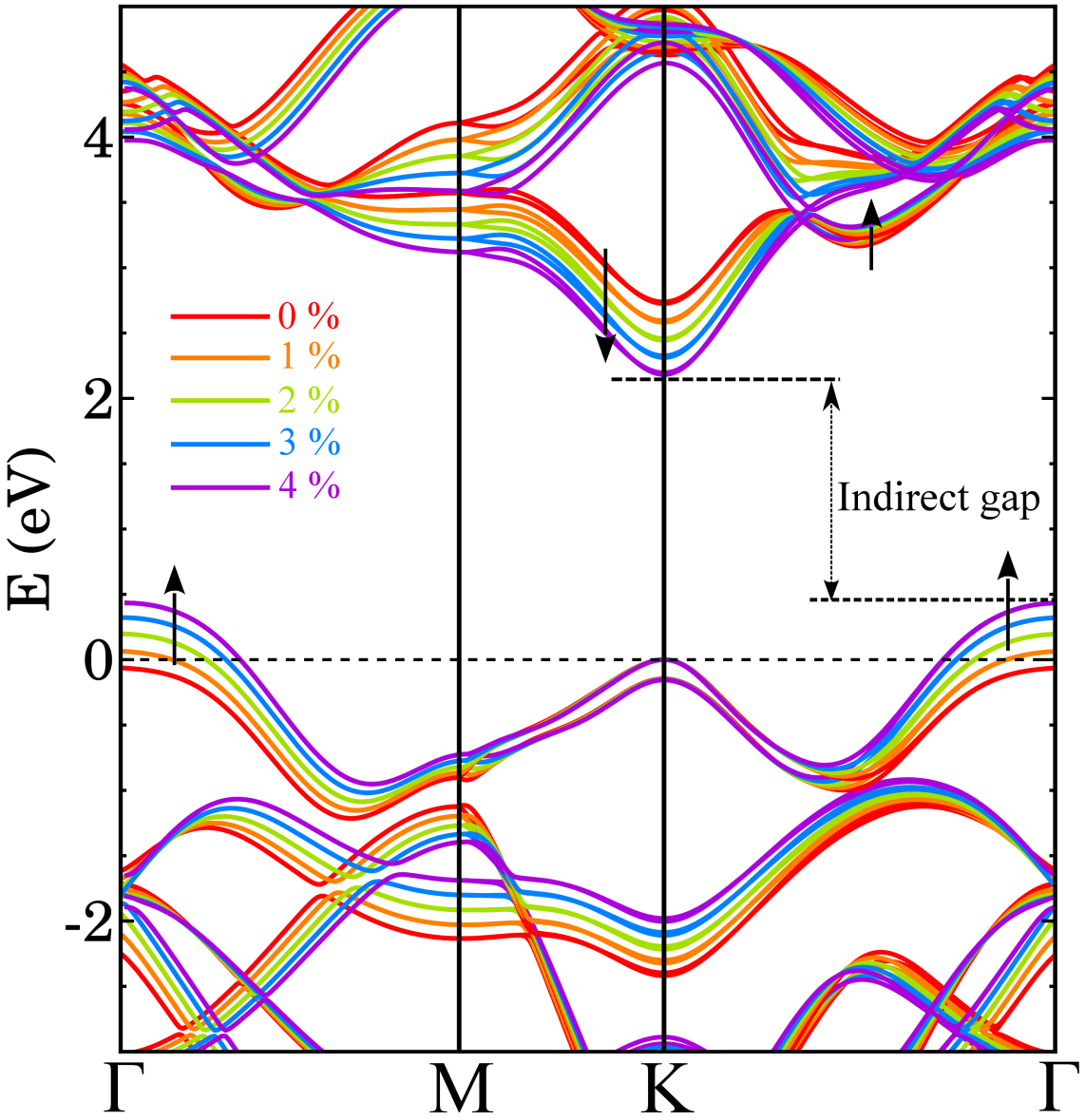
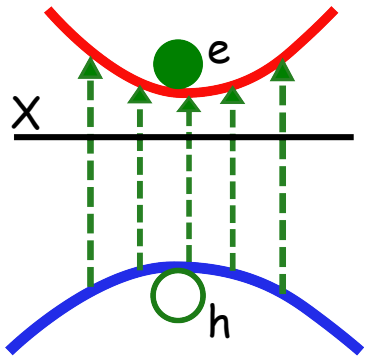
Electronic structure. Strained single-layer.



Strain changes the bandgap from direct to indirect

Optical properties. Excitonic effects

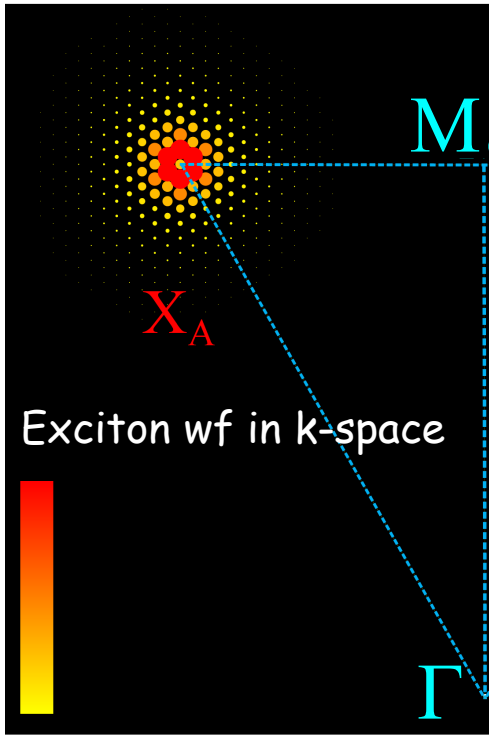


e-h pairs interact and form the exciton

$$H^{X}_{(n_1,n_2),(n_3,n_4)} = \left(E_{n_2} - E_{n_1}\right)\delta_{n_1,n_3}\delta_{n_2,n_4} + i\left(f_{n_2} - f_{n_1}\right)\Xi_{(n_1,n_2),(n_3,n_4)}$$

Energy difference — Bethe-Salpeter Kernel

$$\Psi(\mathbf{r}_e,\mathbf{r}_h) = \sum_{i,j} A_{i,j,\mathbf{k}}\phi_i(\mathbf{r}_e)\phi_j(\mathbf{r}_h)$$

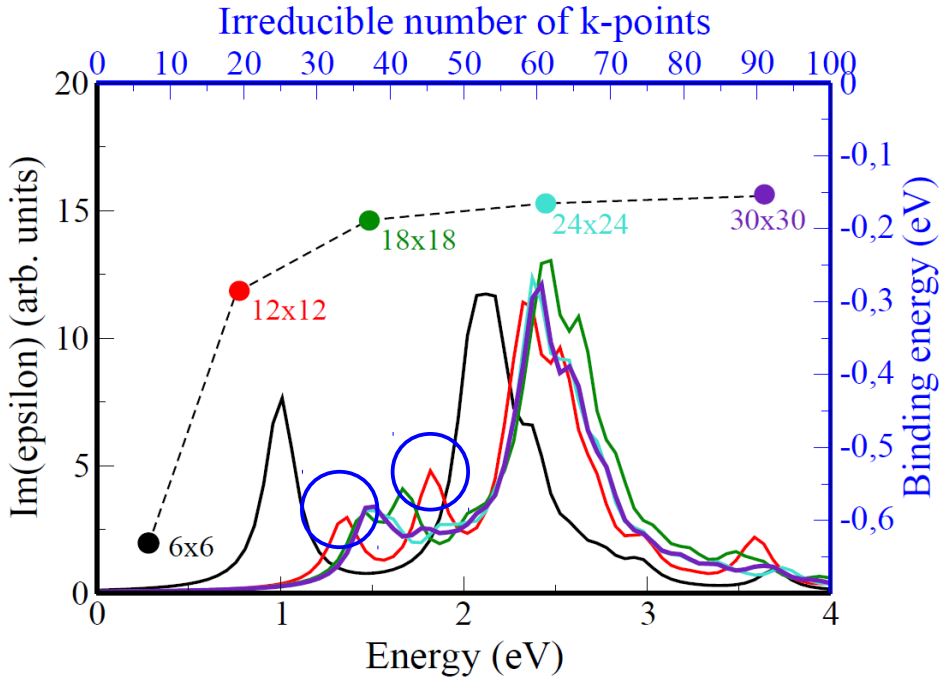


Exciton wf in k-space

Key parameters for reliable results

- Number of bands

- k-sampling of the Brillouin zone



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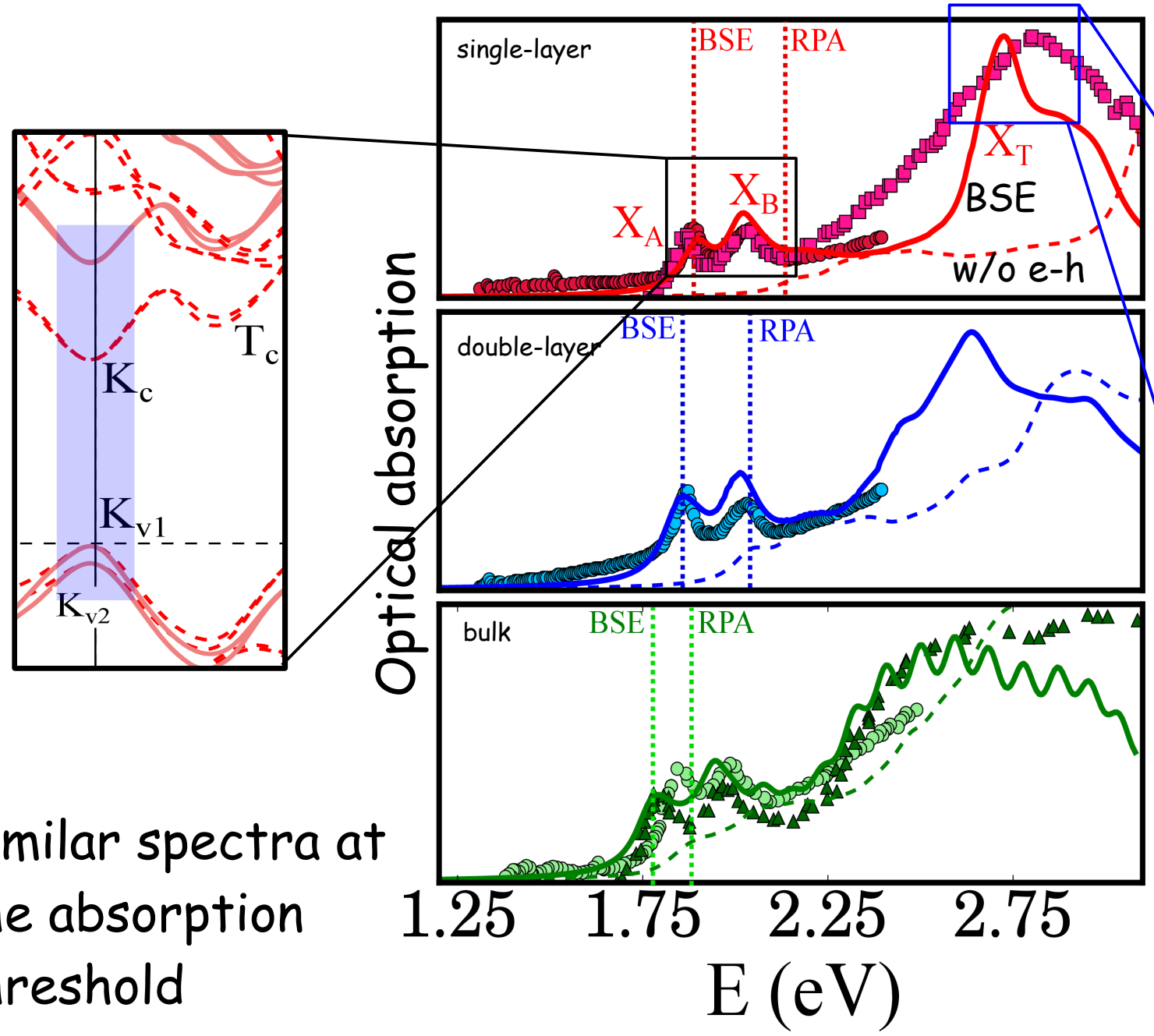
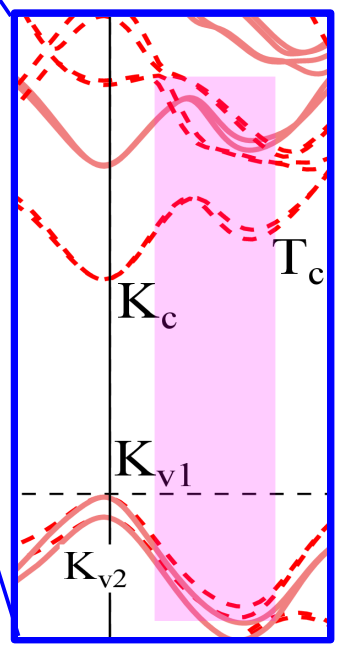


Sharp peak comes from a very localized exciton. High DOS

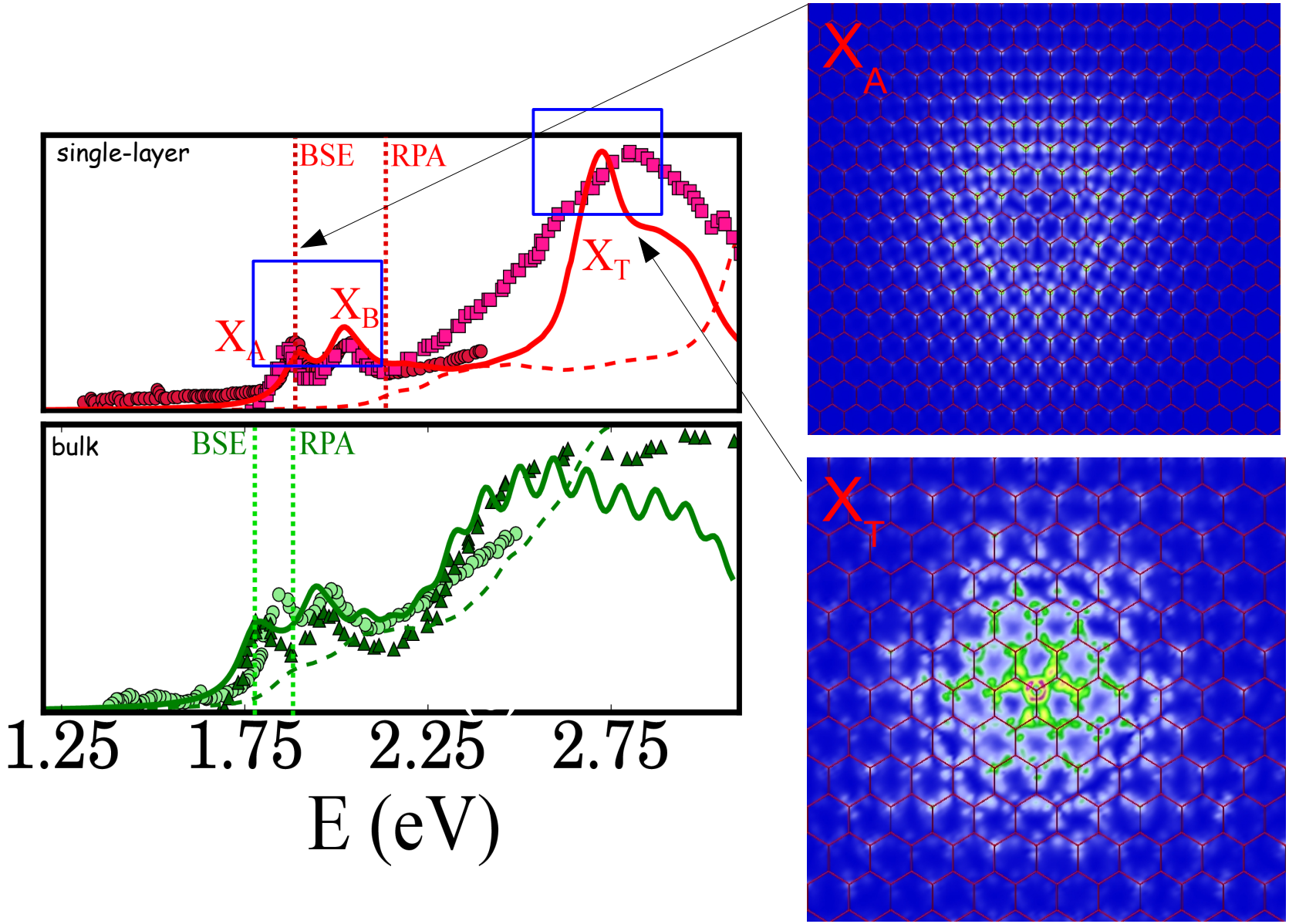


Similar spectra at the absorption threshold

Experimental confirmation PRB 87, 201401(R) 2013

Optical properties. Excitonic effects

Exciton wavefunctions (real-space)



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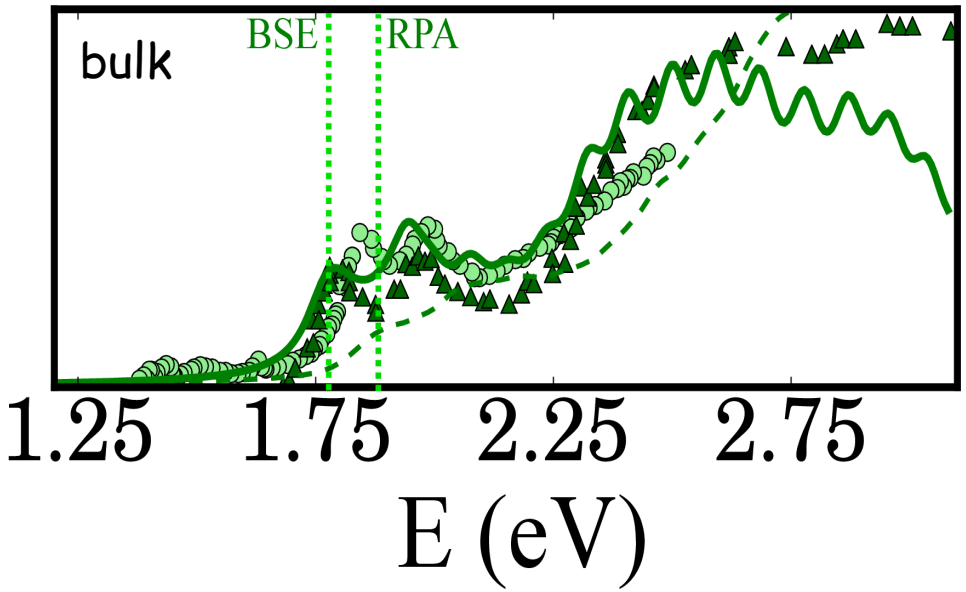
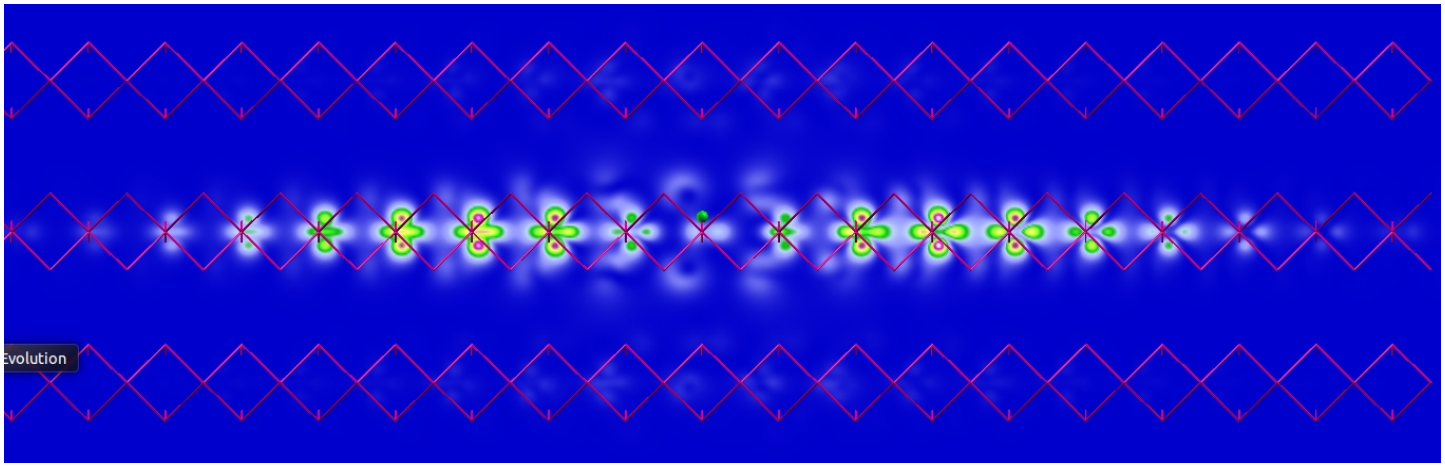
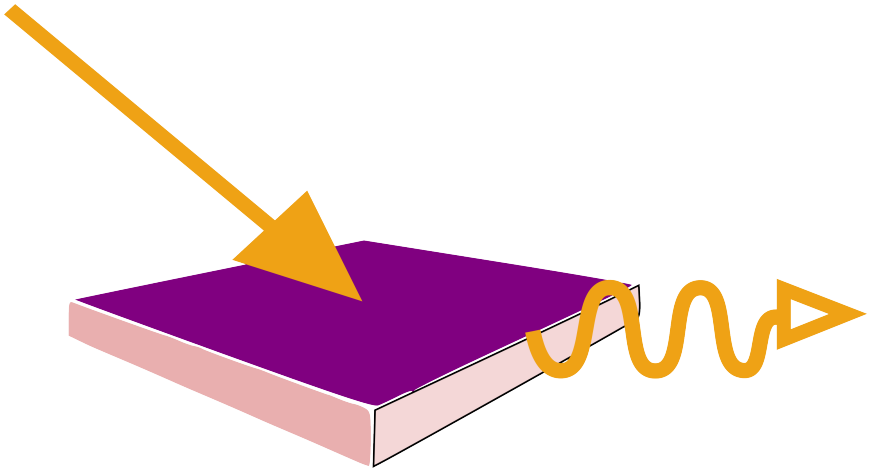


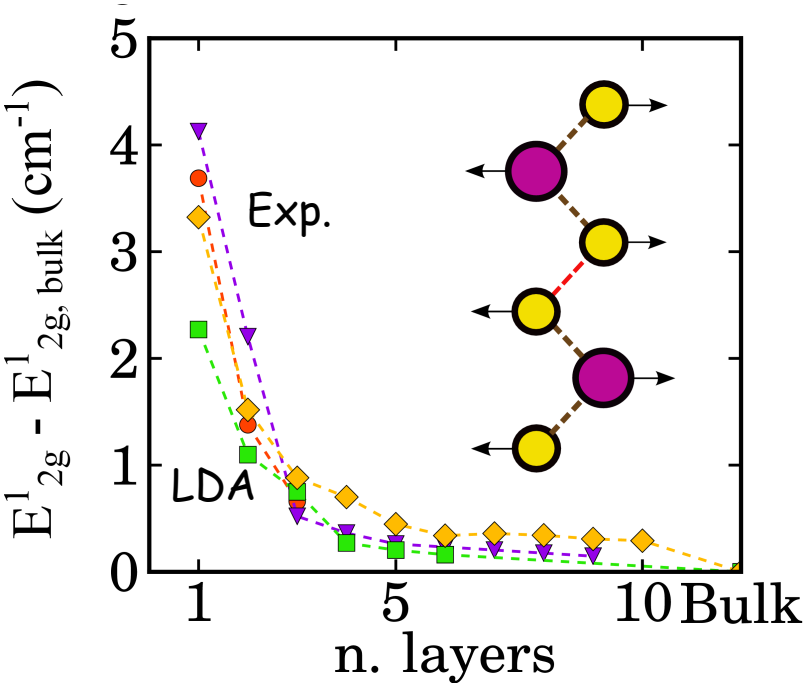


Exciton is confined in one layer

Large interlayer distance

Vibrational properties.



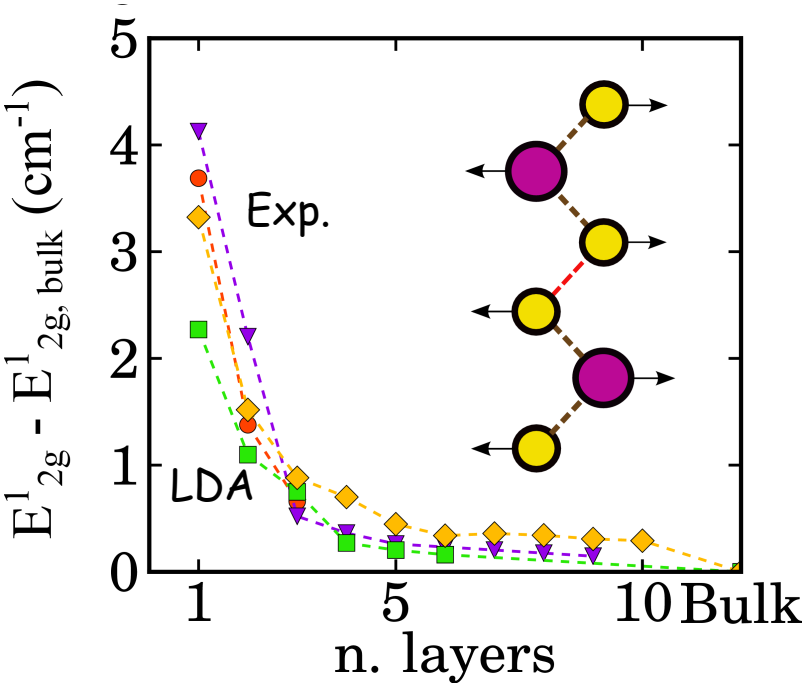


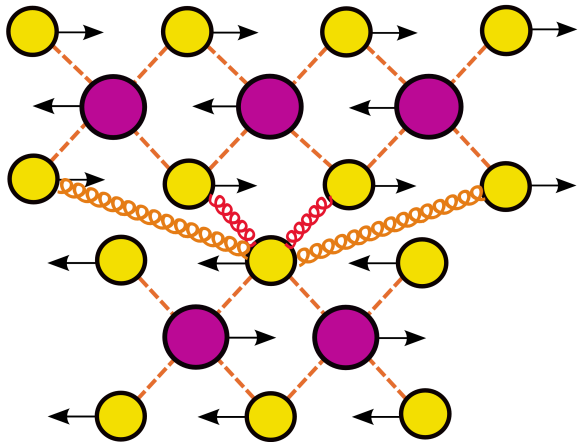
Raman spectroscopy - structural properties

Anomalous behaviour of the phonon mode E_{2g}

Increasing number of layers >> frequency decreases

Vibrational properties.



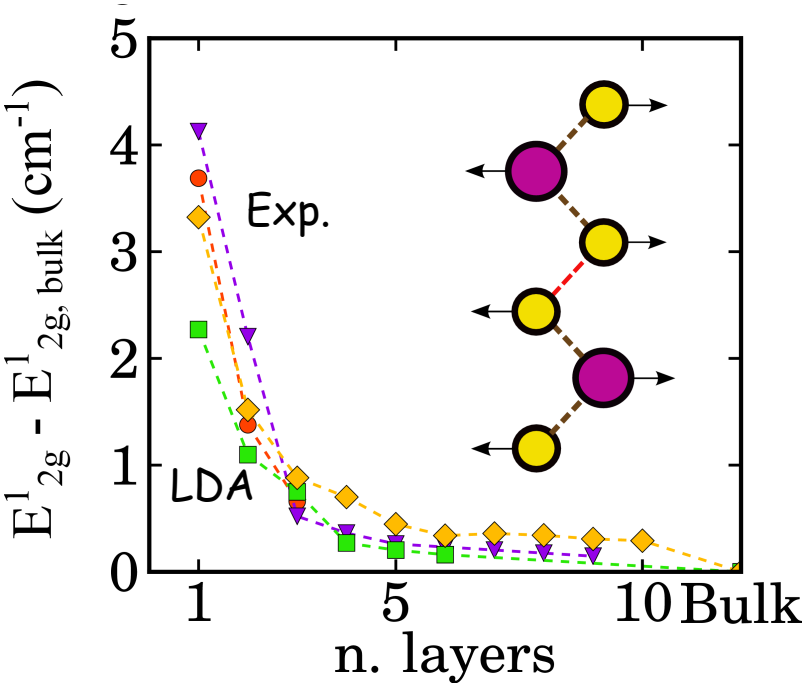


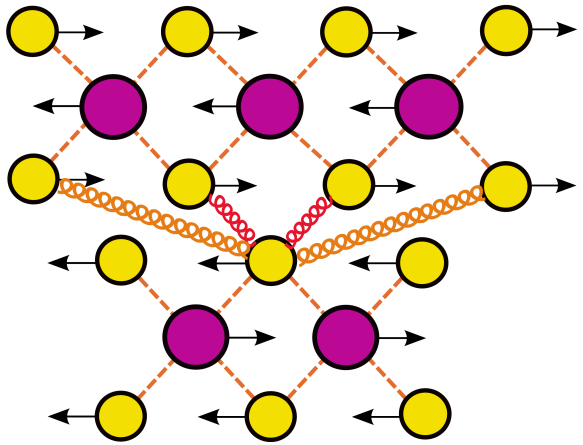
Interlayer interaction?

Surface effects, changes in lattice parameter?

Dielectric screening?

Vibrational properties.





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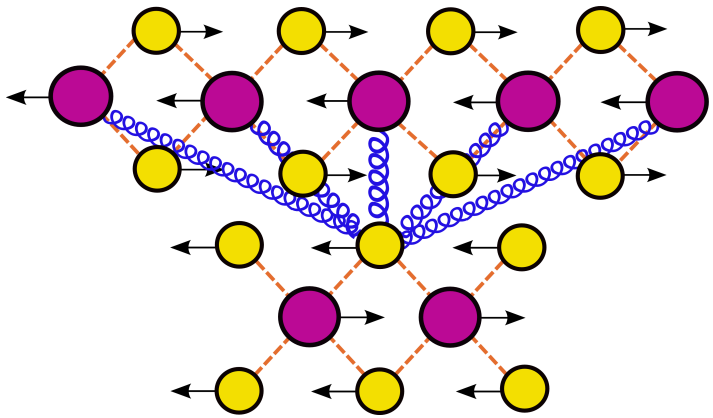
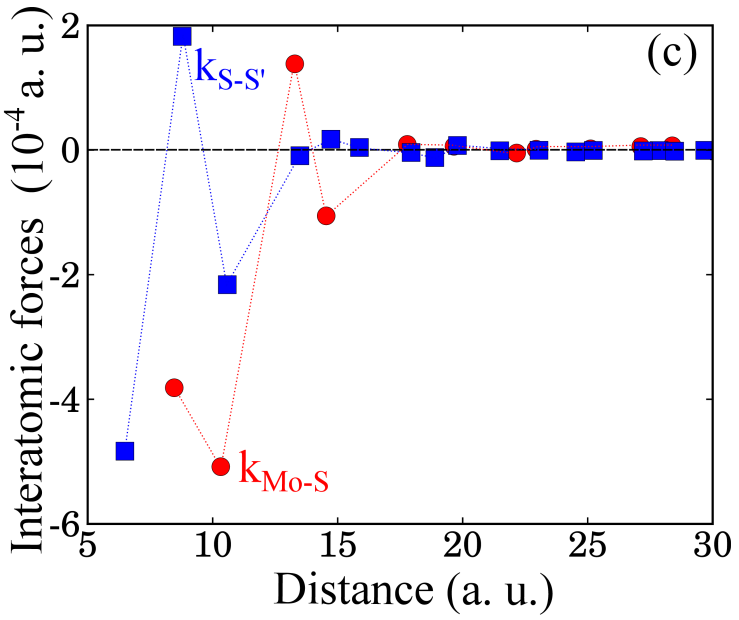
Interlayer interaction... but considering farer neighbours, Mo-S interaction



Further examination of the force constant



Conclusions

- Layered materials, and in particular MoS_2, show interesting physical properties, very sensitive to the number of layers.
- External strain can tune the bandgap and its character in MoS_2 single-layer.
- Optical properties and excitons: Very localized exciton at high energy (confirmed experimentally).
- The anomalous trend of the in-plane phonon mode is due to the interaction between sulphur and molybdenum atoms from different layers.

Acknowledgements

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Physics and Material Sciences Research Unit (PHYMS)

Davide Sangalli and Andrea Marini, Istituto di Struttura della Materia (ISM), Consiglio Nazionale delle Ricerche (CNR), Rome, Italy.

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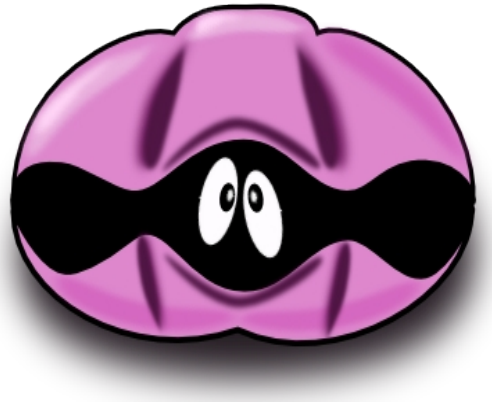

Kerstin Hummer, University of Vienna.

Thank you for your attention!