Center for Nanostructured Graphene

Technical University of Denmark

DTU

Excitons in van der Waals Heterostructures

PhD Student

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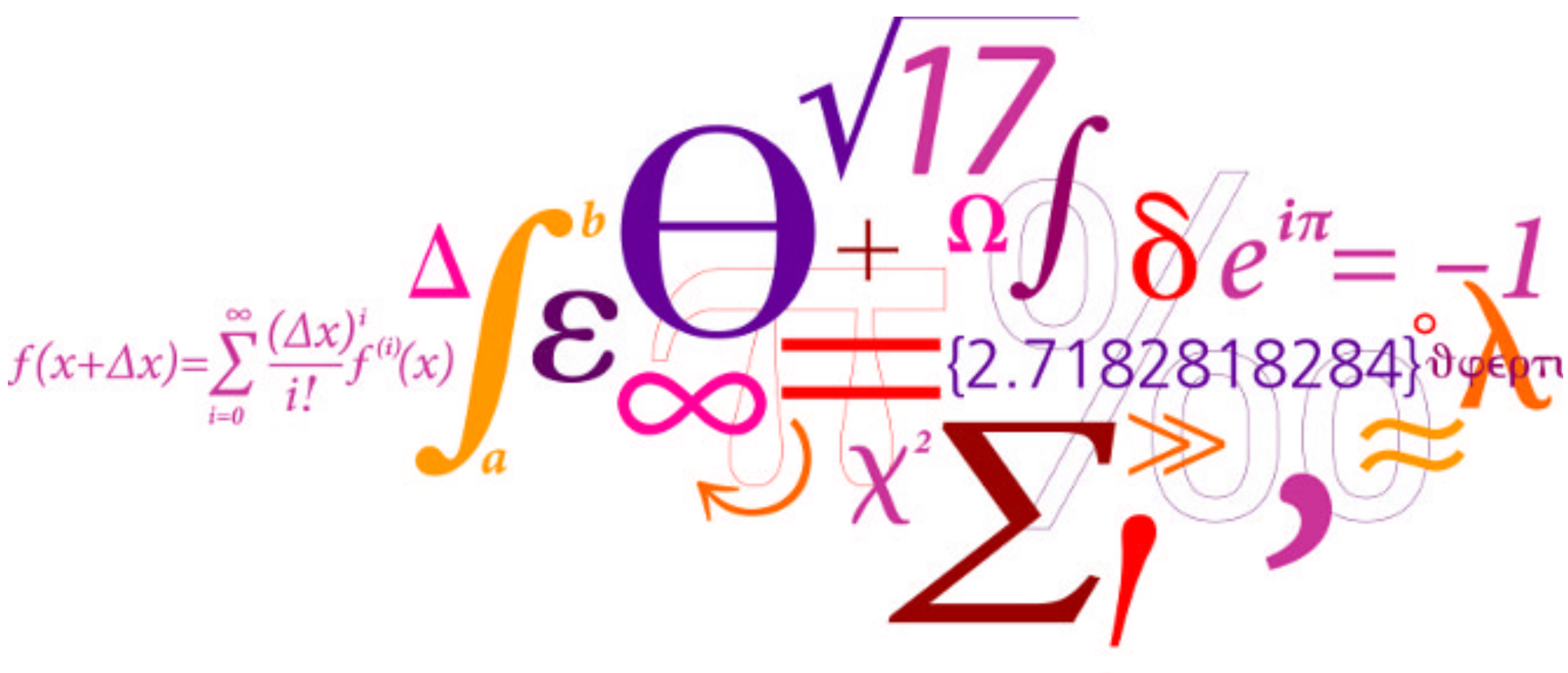
Graphene Canada 2015

Montreal

Oct. 16th 2015

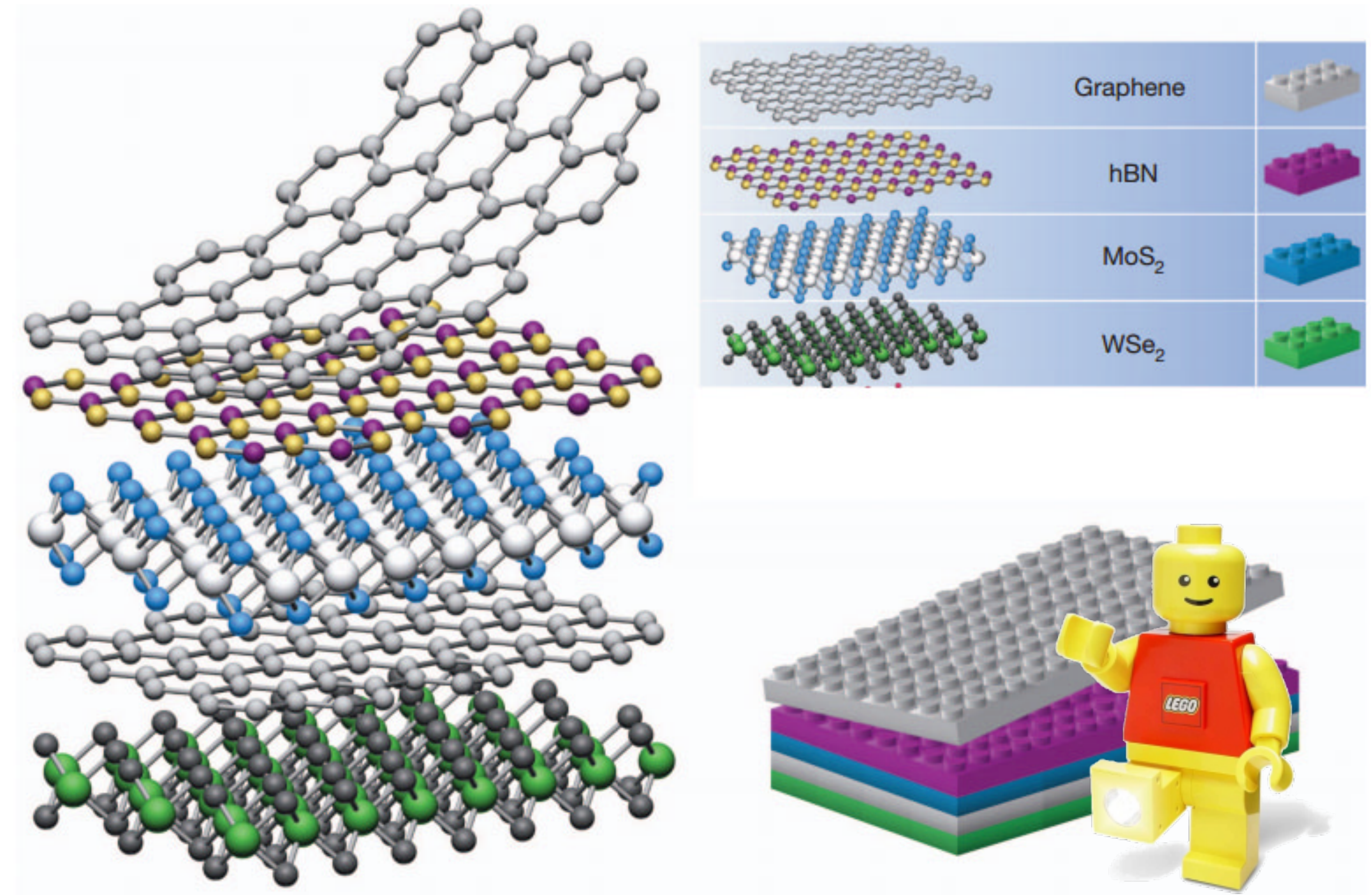


Why van der Waals Heterostructures?

Extraordinary Properties

- Bandgaps in the visible range;
- High optical absorbtion coefficient;
- Mechanical resistance and Flexibility:
- Atomically well defined interfaces.
- Transparency.

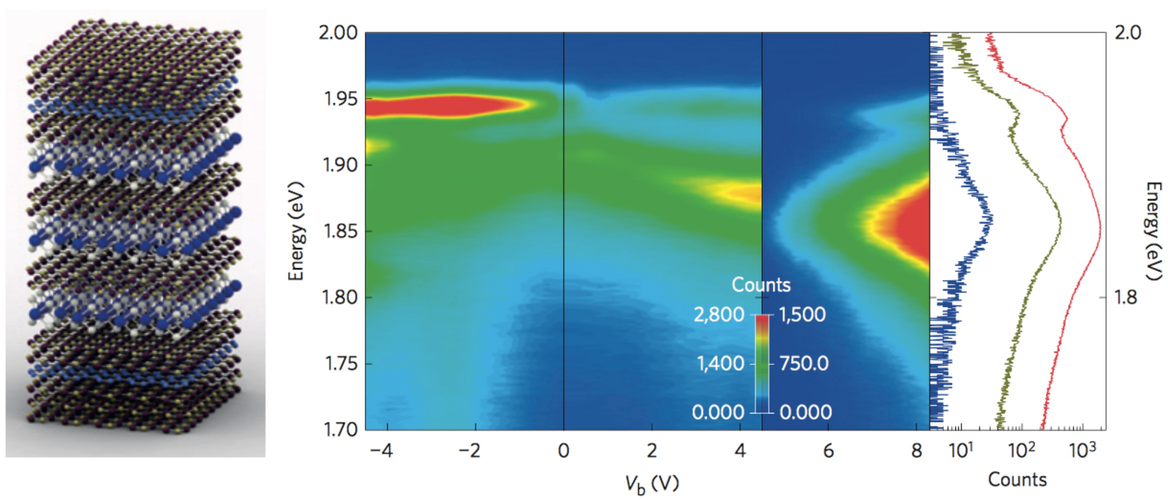
Most of these are inherited from the isolated 2D layers!



vdWHs are a playground for material scientists: their properties can be tuned just by stacking 2D layers together

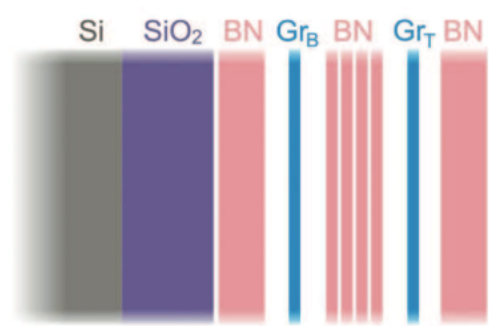
Several (opto)electronic Applications

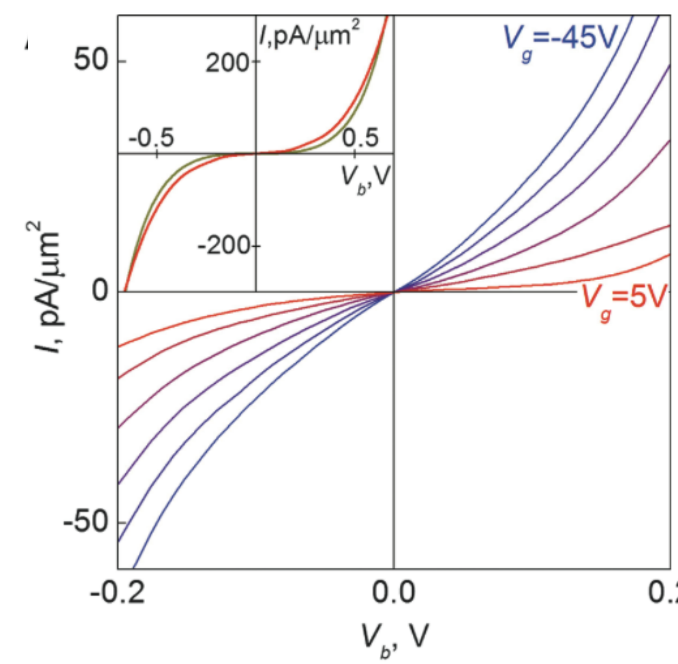
Light Emitting Diode



F. Withers et al, Nature Mater. **14**, 301–306 (2015)

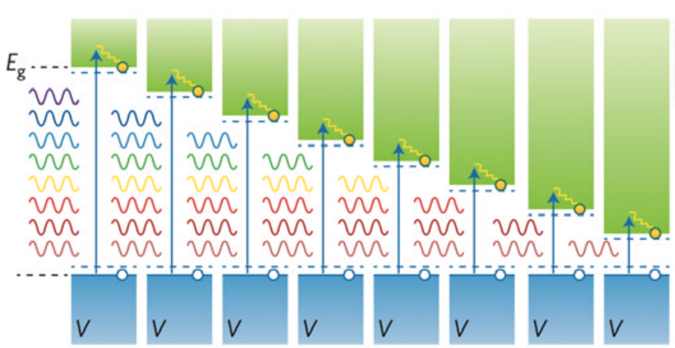
Field effect transistor

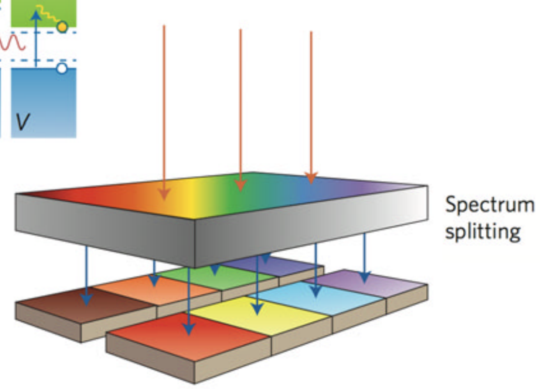




L. Britnell et al, Science **355**, 947 (2012)

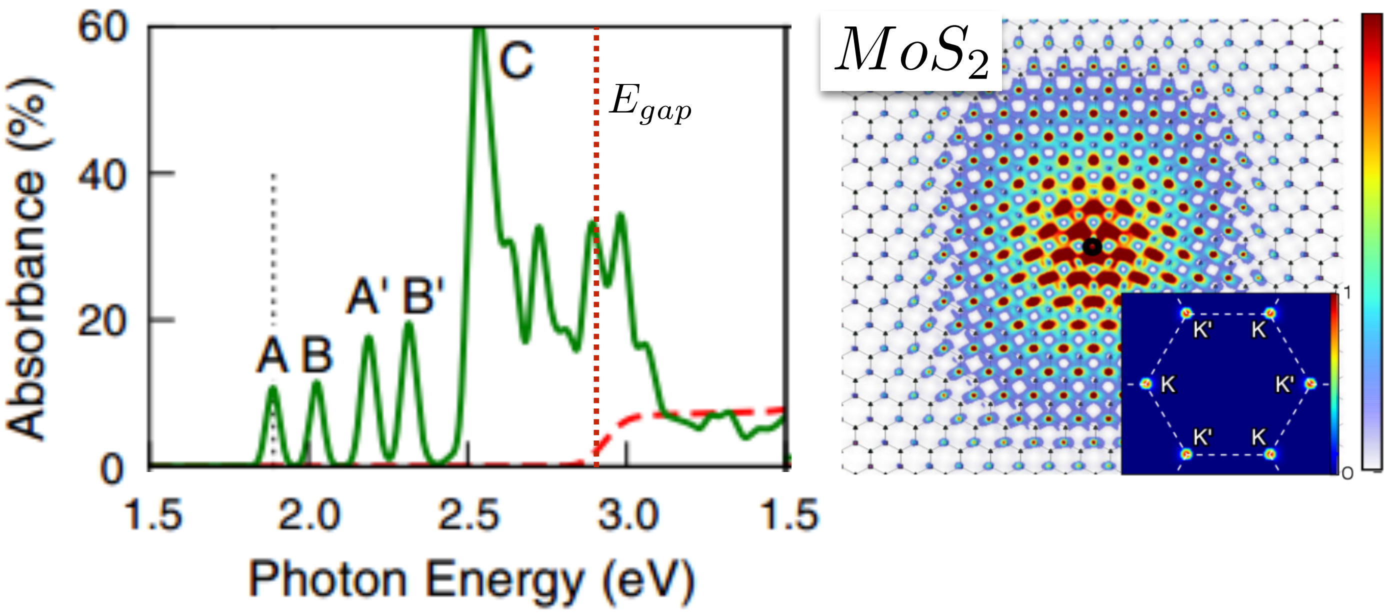
Multi-junction solar cell





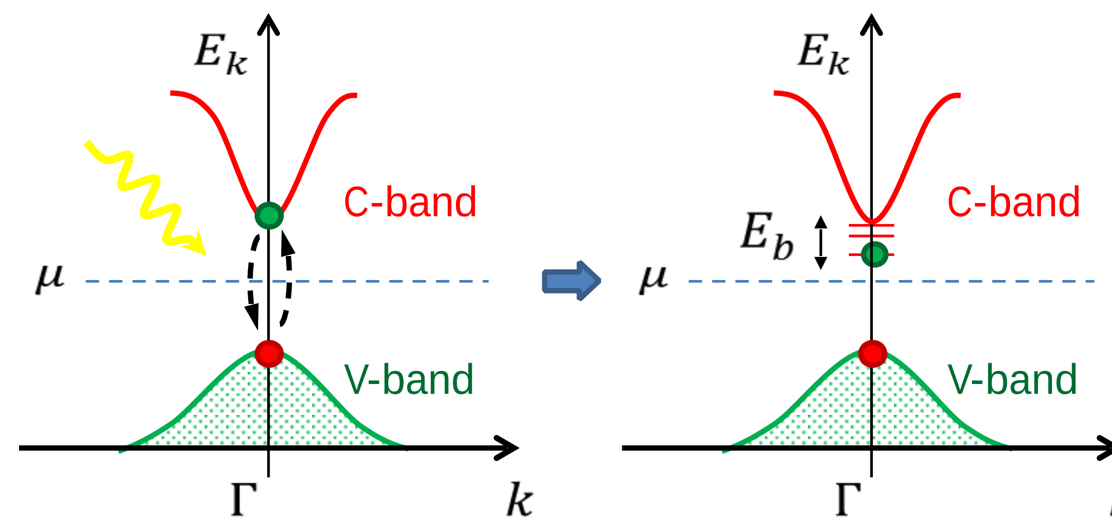
A. Polman et al, Nature Mater. **11**, 174–177 (2012)

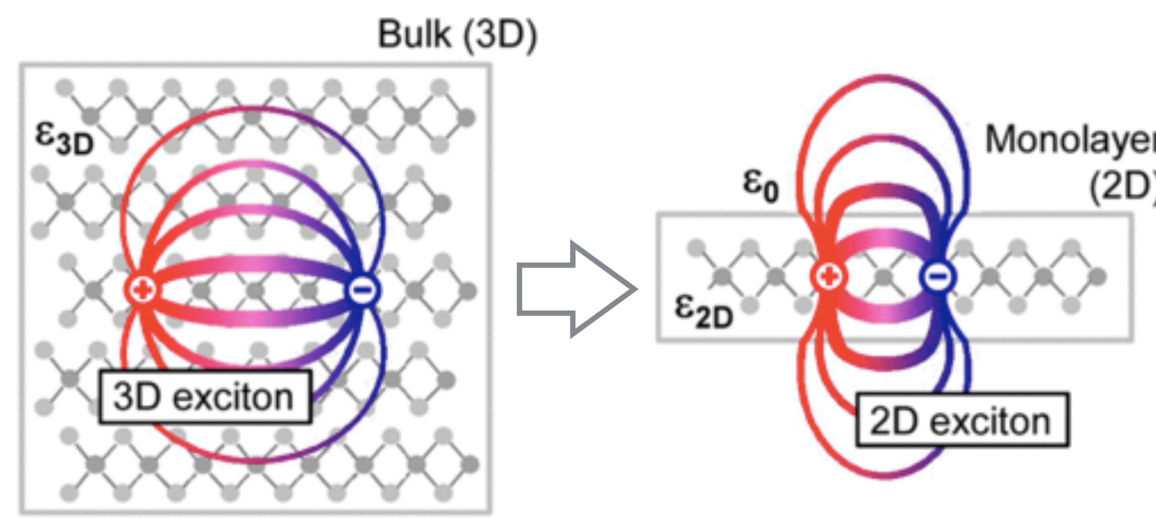
Excitons are strongly bound in 2D



D.Y. Qiu et al, PRL **111**, 216805 (2013)

Absorption below the gap!
What is it going on?





Alexey Chernikov et al, PRL **113**, 076802 (2014)

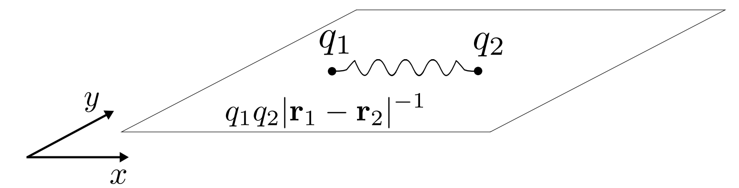
The reduced screening in 2D results in enhanced e-h interaction!

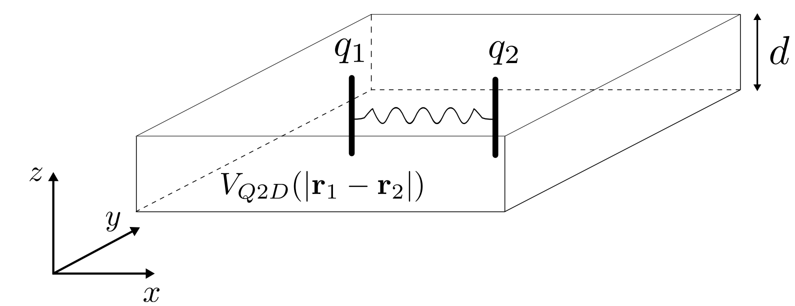
Screened Electron-Hole Interaction

The electron-hole interaction is screened by all the other electrons in the material

Strict 2D picture:

$$W_{2D}(\mathbf{q}_{\|}) = -\frac{2\pi}{|\mathbf{q}_{\|}|}\epsilon^{-1}_{2D}(\mathbf{q}_{\|}) \;, \qquad \epsilon_{2D}(\mathbf{q}_{\|}) = 1 + 2\pi\alpha|\mathbf{q}_{\|}|$$





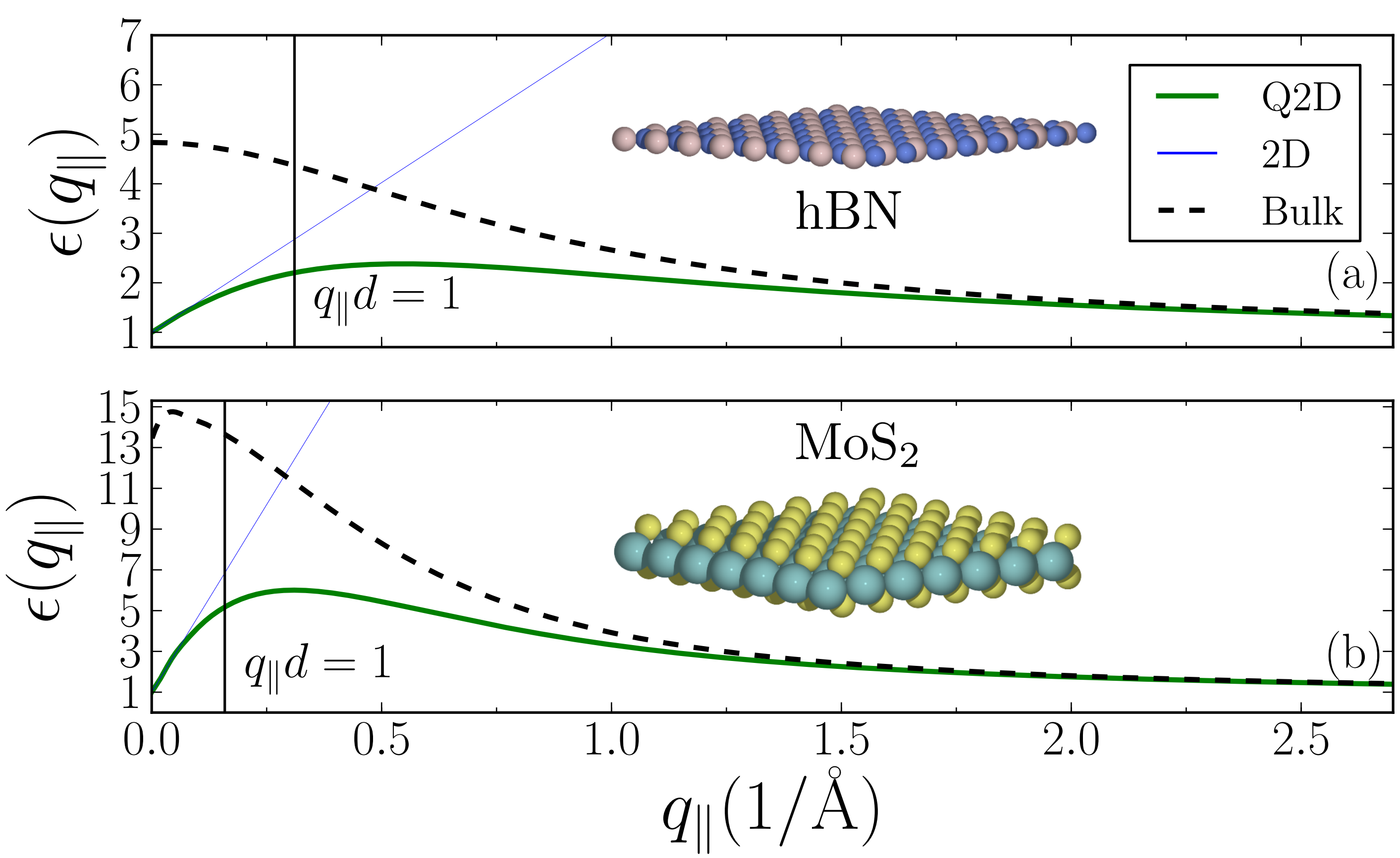
Q2D picture:

$$W_{Q2D}(\mathbf{q}_{\|}) = V_{Q2D}(\mathbf{q}_{\|})\epsilon^{-1}_{Q2D}(\mathbf{q}_{\|}) \;,$$

$$V_{Q2D}(\mathbf{q}_{\|}) = -\frac{4\pi}{d|\mathbf{q}_{\|}|^2}\left[1 - \frac{2}{|\mathbf{q}_{\|}|d}e^{-|\mathbf{q}_{\|}|d/2}\sinh\left(\frac{|\mathbf{q}_{\|}|d}{2}\right)\right]$$

$$= \begin{cases} -\frac{2\pi}{|\mathbf{q}_{\|}|} & q_{\|}d \ll 1 \\ -\frac{4\pi}{|\mathbf{q}_{\|}|^2} & q_{\|}d \gg 1 \end{cases}$$

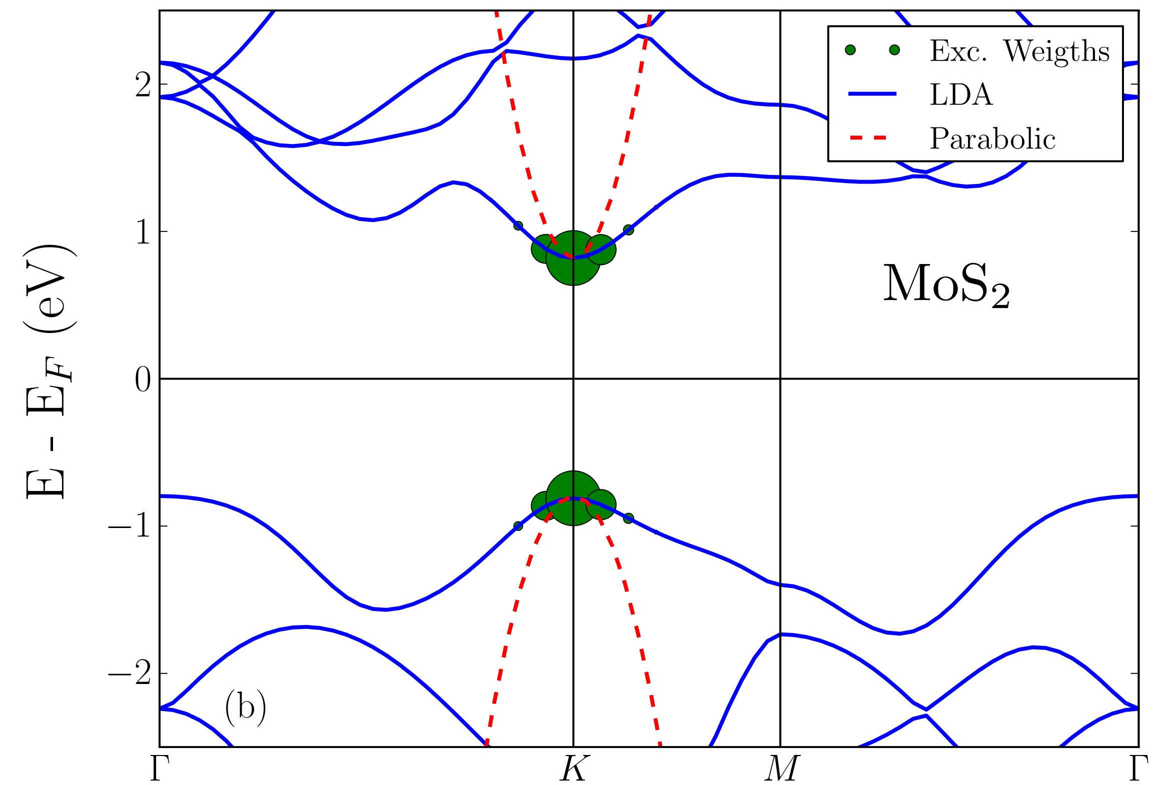
$$\epsilon^{-1}_{Q2D}(\mathbf{q}_{\|}) = \frac{2}{d}\sum_{G_{\perp}} e^{iG_{\perp}z_0}\frac{\sin(G_{\perp}d/2)}{G_{\perp}}\epsilon^{-1}_{G_{\perp}\,0}(\mathbf{q}_{\|})$$



Binding Energies for Isolated Layers

Exciton binding energies using the Mott-Wannier Hamiltonian:

$$\left[-\frac{\nabla^2_{2D}}{2\mu_{ex}} + W(\mathbf{r}_\parallel)\right] F(\mathbf{r}_\parallel) = E_b F(\mathbf{r}_\parallel)$$



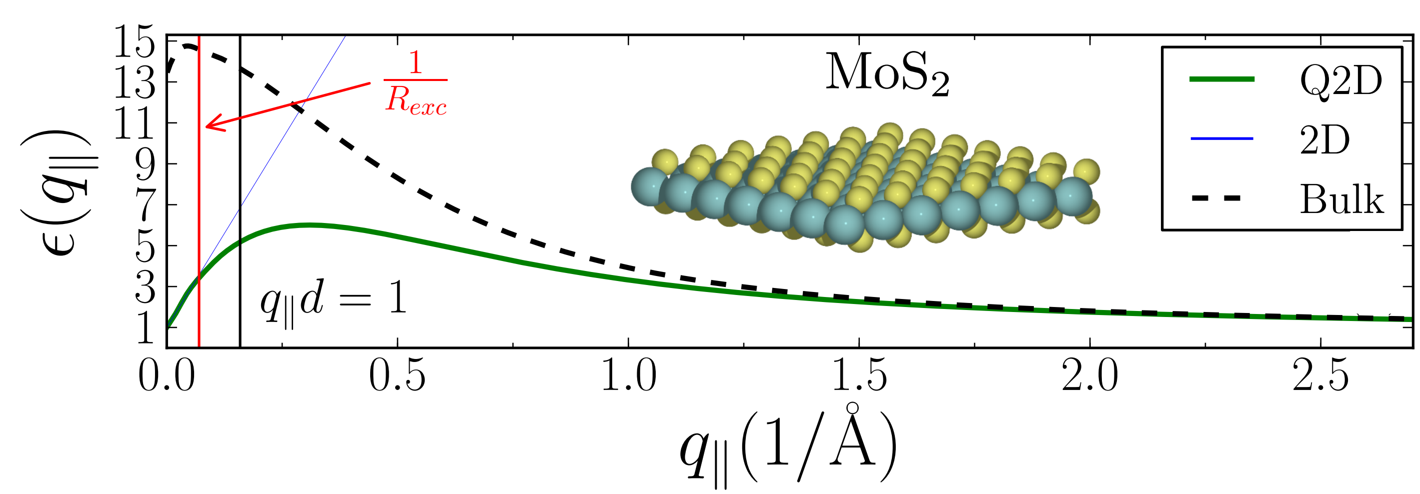
2D and Q2D model agree very well, why?

	$\mathbf{E_b^{BSE}}$(eV)	$\mathbf{E_b^{Q2D}}$(eV)	$\mathbf{E_b^{2D}}$(eV)
hBN	2.05	2.35	2.34
MoS2	0.43	0.61	0.60

The exciton is localised in q-space:

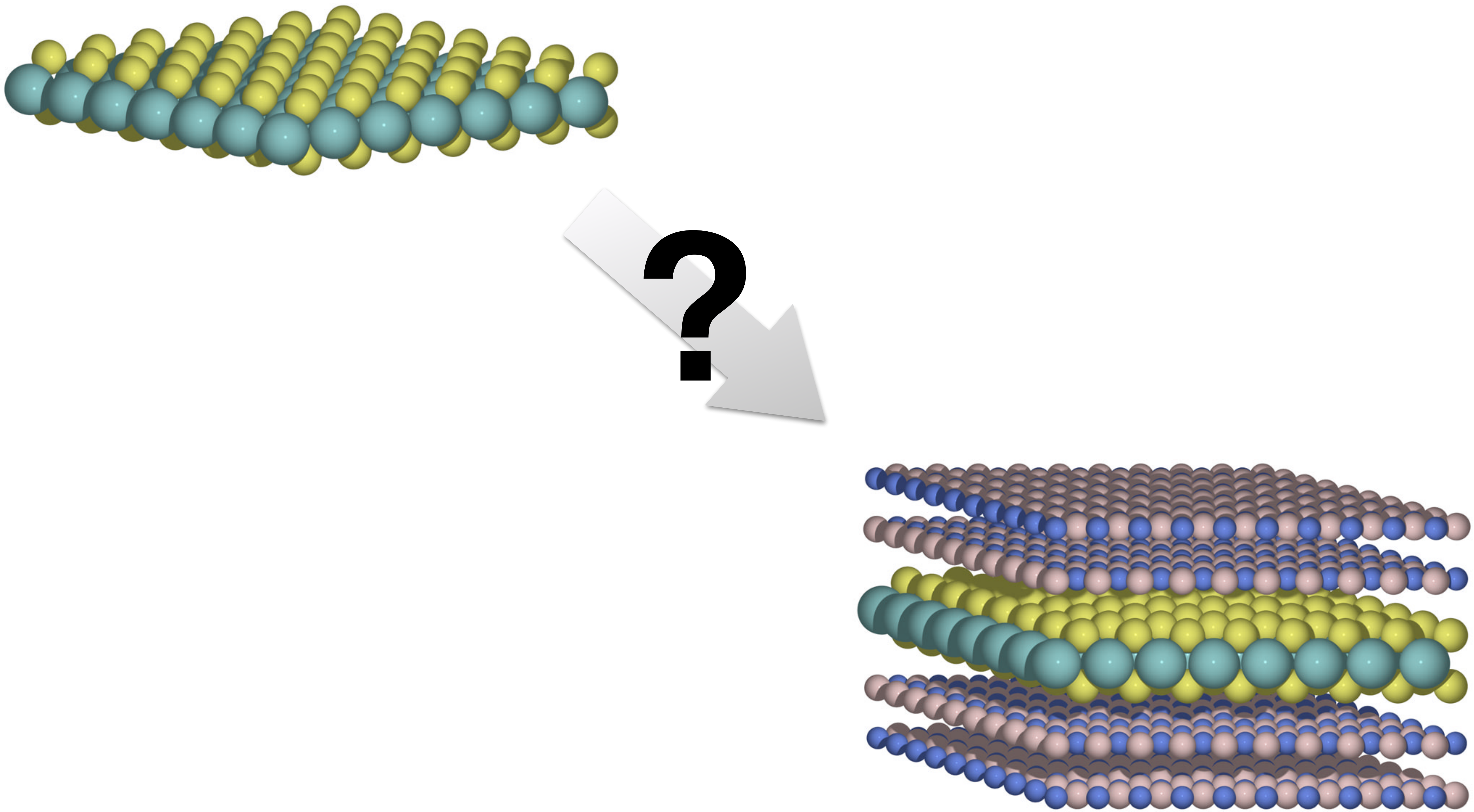
$$\frac{1}{R_{exc}} < \frac{1}{d}$$

regime for which 2D and Q2D electron-hole interaction are essentially equivalent

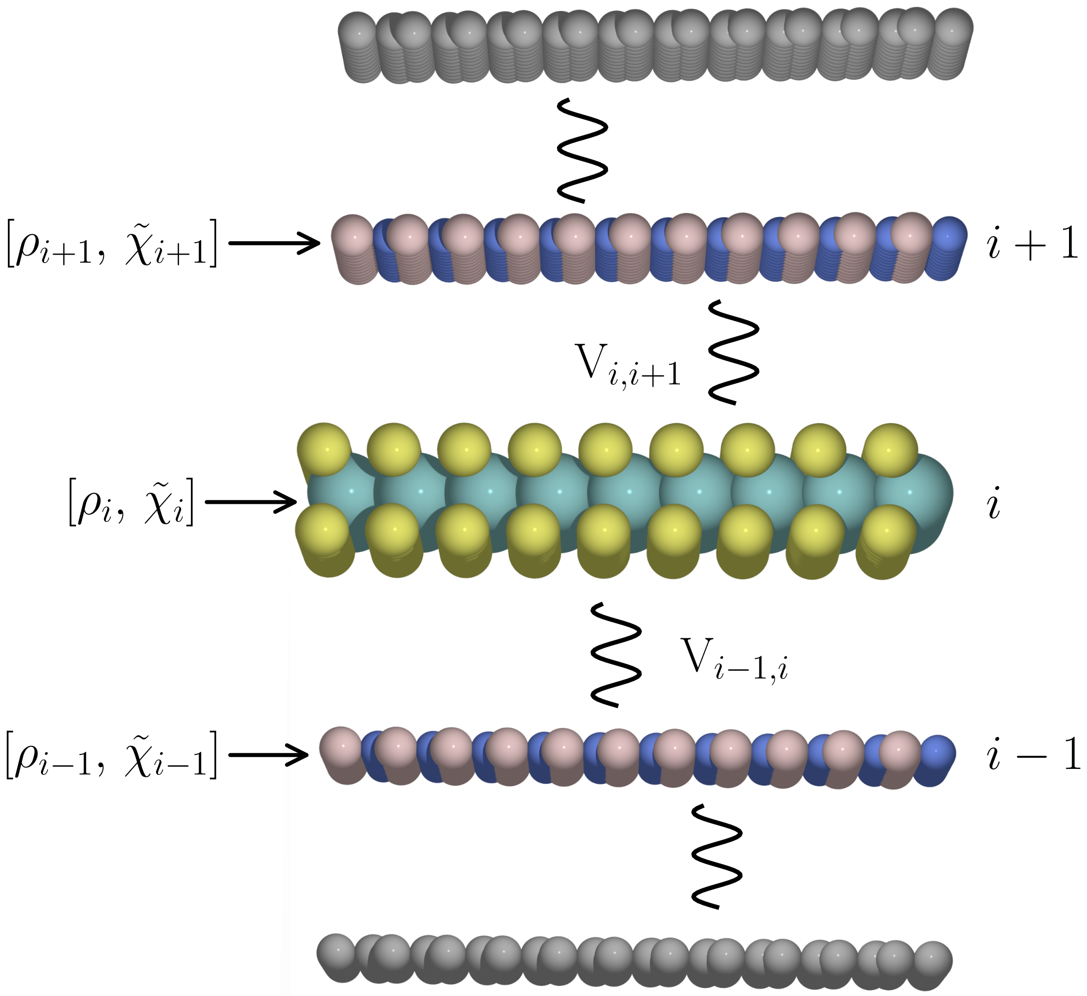


S. Latini, T. Olsen and K.S. Thygesen, submitted arXiv:1509.07972

From 2D to van der Waals Heterostructures



Quantum Electrostatic Heterostructure (QEH) Model



vdHs are unfeasible for traditional ab-initio calculations:

- incommensurable structures;
- too many layers.

... but a **LEGO bricks** picture can be employed!

Multiscale approach (QEH), which combines:

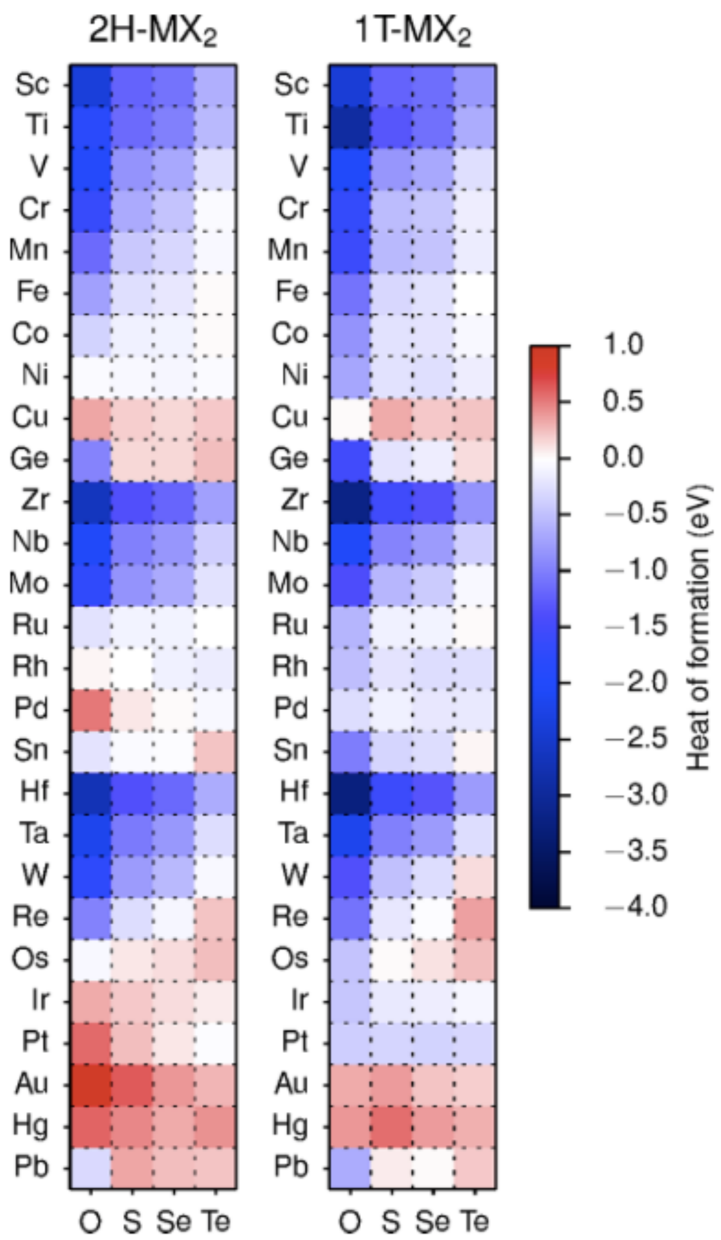
- quantum accuracy at the single layer level (dielectric building blocks);
- electrostatic coupling of the layers.

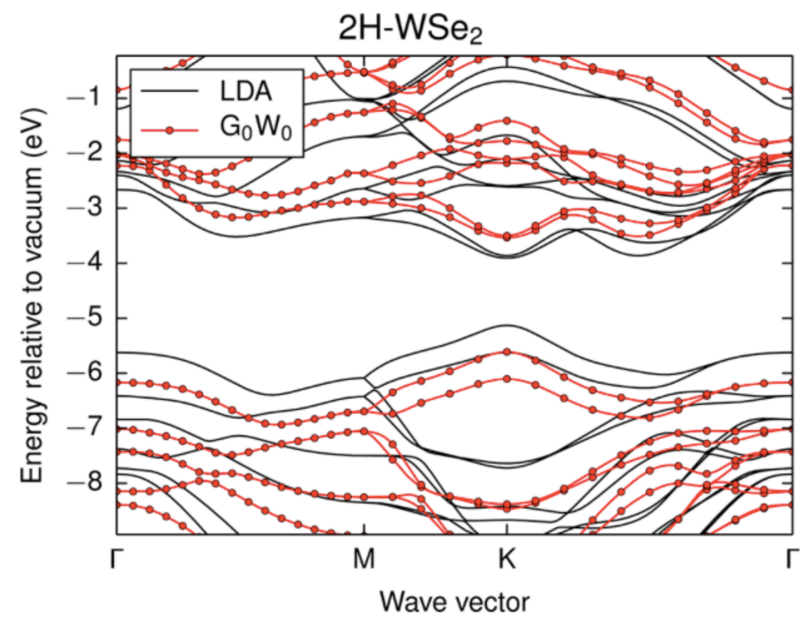
K. Andersen, S. Latini and K.S. Thygesen, Nanoletter **15**, 4616 (2015)

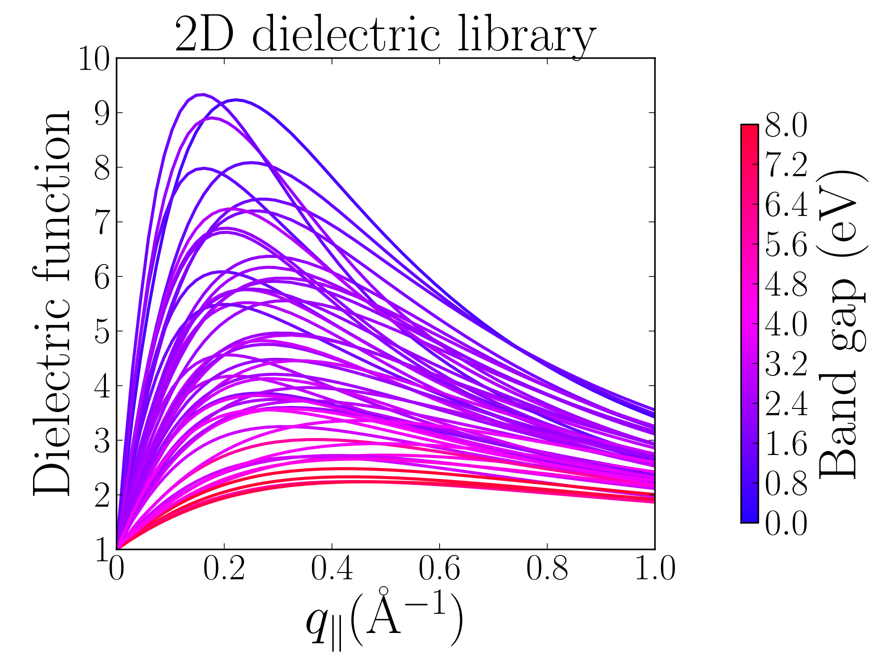
Two-dimensional Materials Database

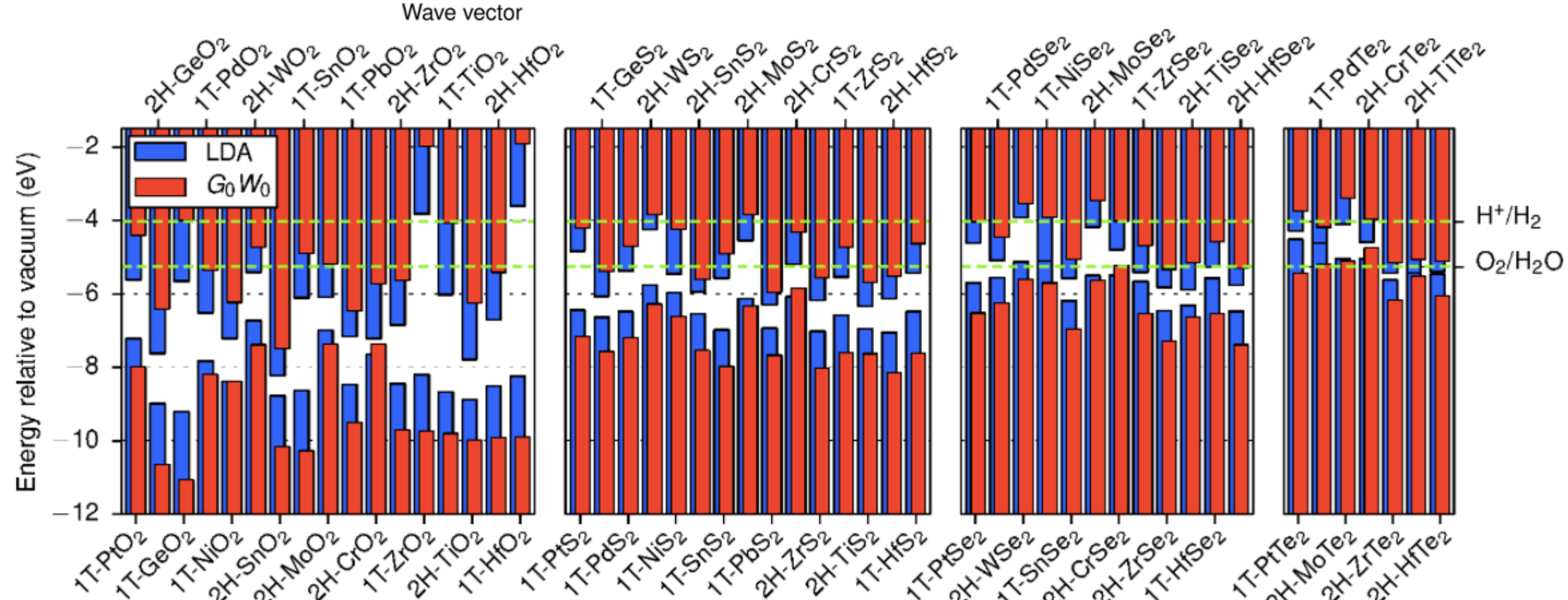
Dielectric building blocks for more than 50 materials and QEH software

https://cmr.fysik.dtu.dk







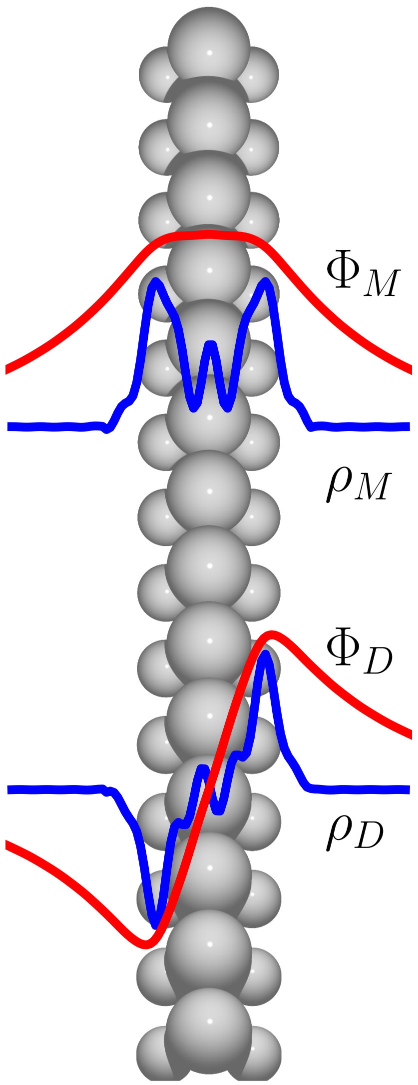


F. Rasmussen and K.S. Thygesen, J. Phys. Chem. C **119**,13169–13183 (2015)

QEH: Formalism

The QEH model gives us access to the dielectric properties of the full heterostructure:

$$\epsilon^{-1}_{i\alpha,j\beta}(\mathbf{q}_{\|},\omega) = \delta_{i\alpha,j\beta} + \sum_{k\gamma} V_{i\alpha,k\gamma}(\mathbf{q}_{\|})\chi_{k\gamma,j\beta}(\mathbf{q}_{\|},\omega)$$



where

$$\chi_{i\alpha,j\beta} = \tilde{\chi}_{i\alpha}\delta_{i\alpha,j\beta} + \tilde{\chi}_{i\alpha}\sum_{k\neq i,\gamma} V_{i\alpha,k\gamma}\,\chi_{k\gamma,j\beta}$$

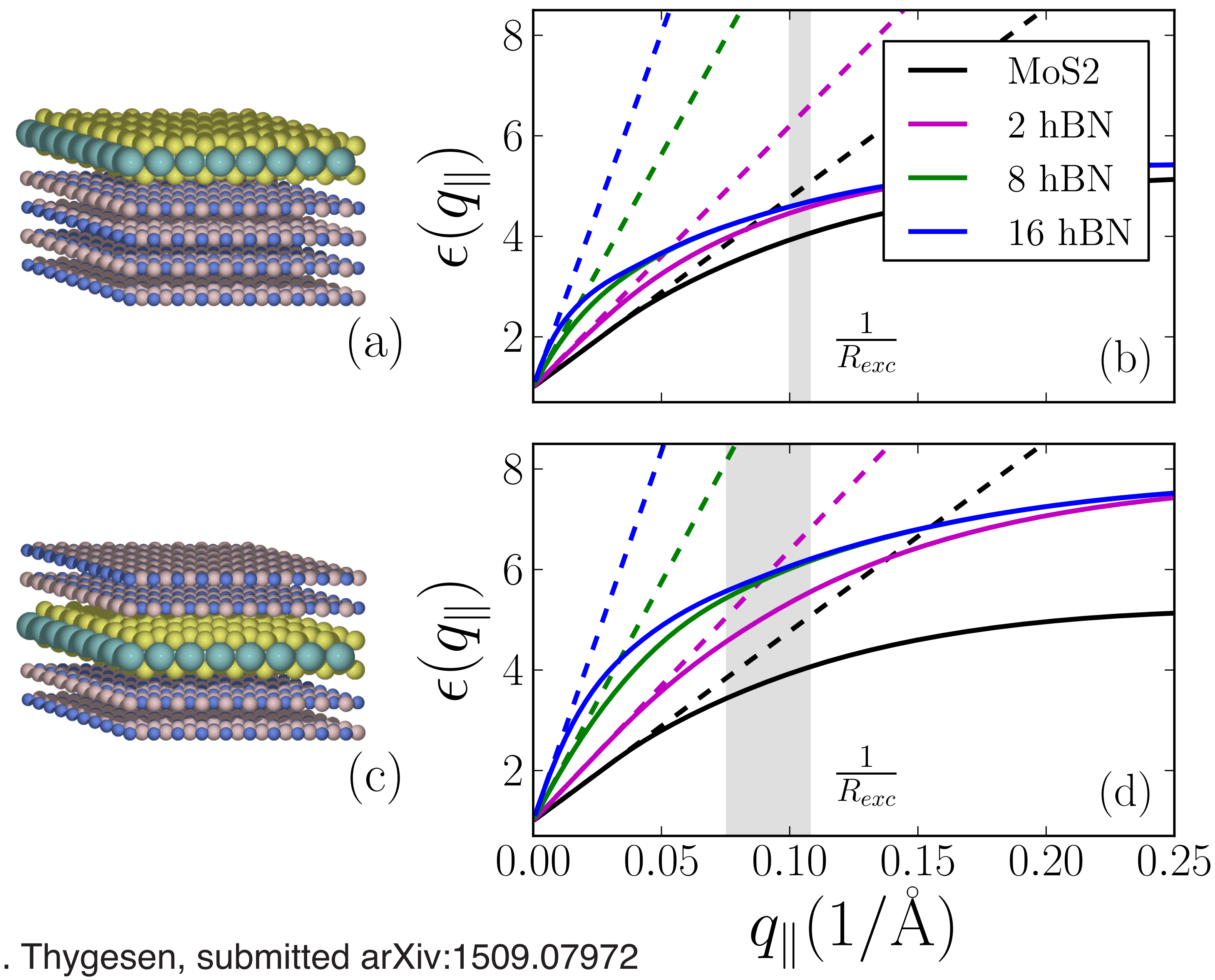
$$V_{i\alpha,k\gamma}(\mathbf{q}_{\|}) = \int \rho_{i\alpha}(z,\mathbf{q}_{\|})\Phi_{k\gamma}(z,\mathbf{q}_{\|})dz$$

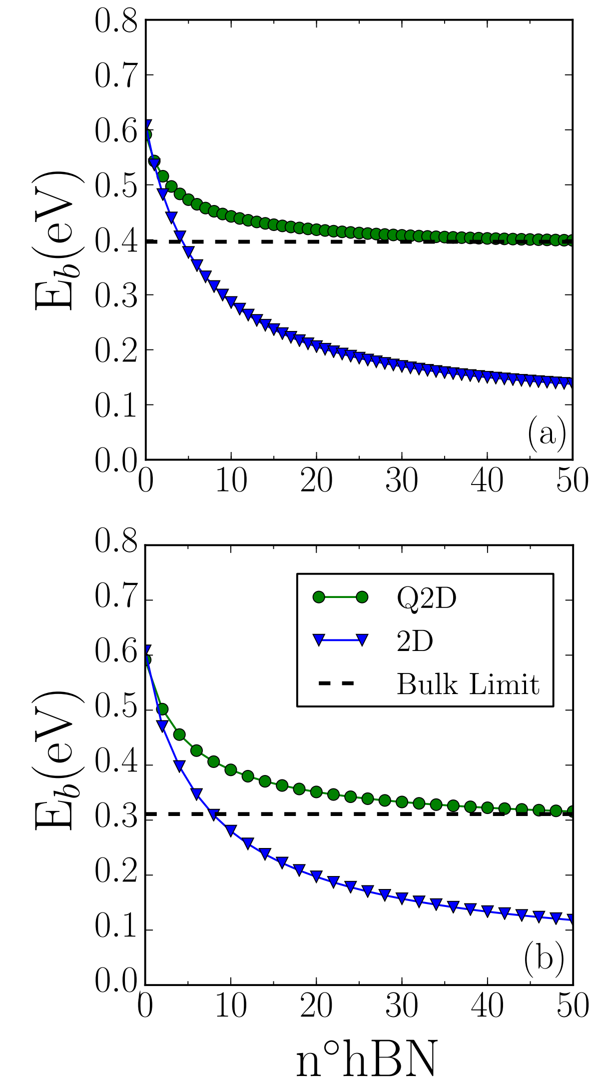
… and to the screened electron-hole potential:

$$W(q_{\|}) = \underline{\rho}^{\mathsf{T}}_{e}(q_{\|})\ \underline{\underline{\epsilon}}^{-1}(q_{\|})\ \underline{\phi}_{h}(q_{\|})$$

Breakdown of the Linear Screening

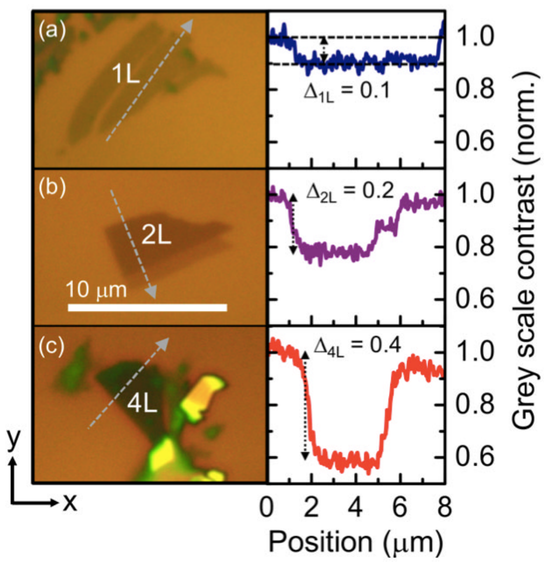
- Screening from neighboring layers reduces Eb and increases R_{exc};
- Dielectric function far from being linear for $q_{\parallel} < \frac{1}{R_{exc}}$;
- Non-linearity increases with the thickness of the structure.

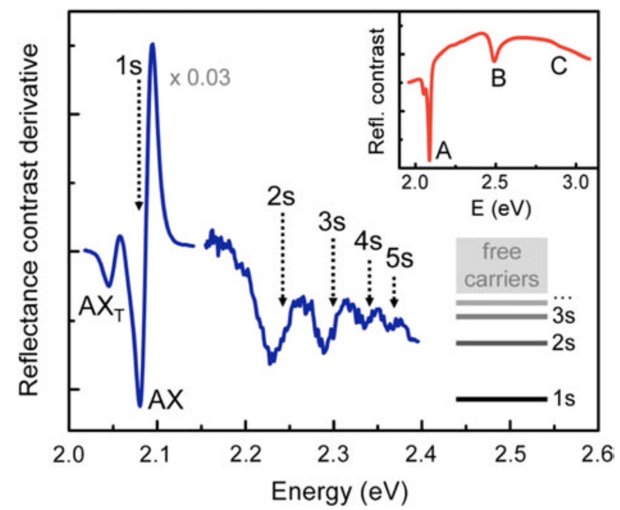




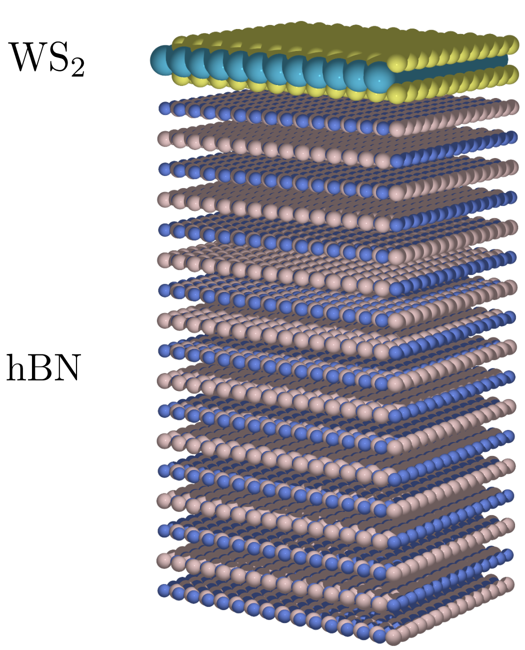
S. Latini, T. Olsen and K.S. Thygesen, submitted arXiv:1509.07972

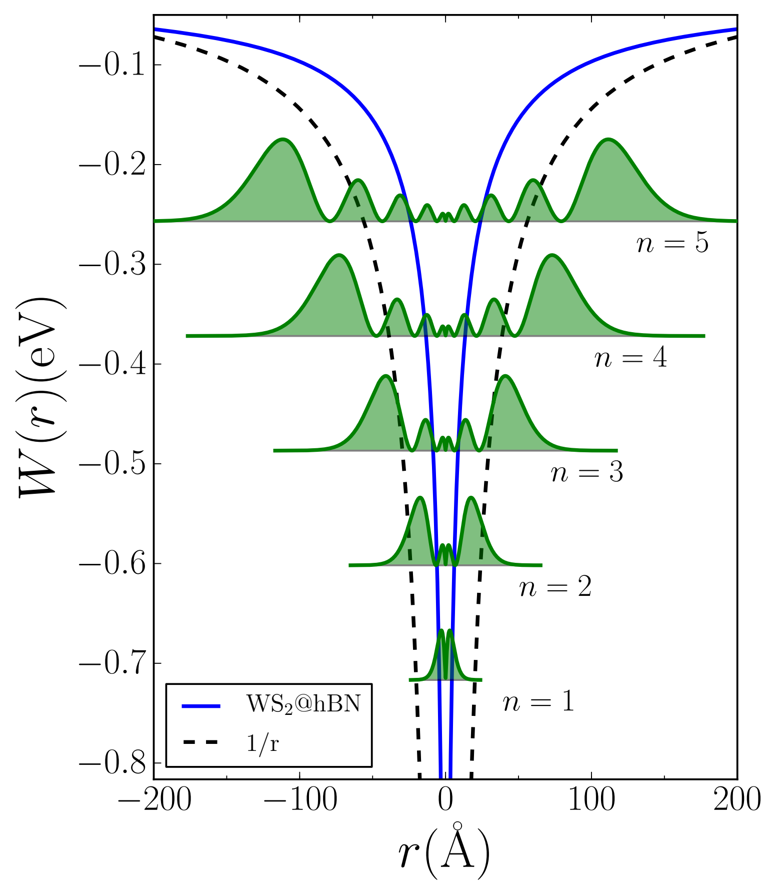
Non-Hydrogenic Rydberg Series in WS2

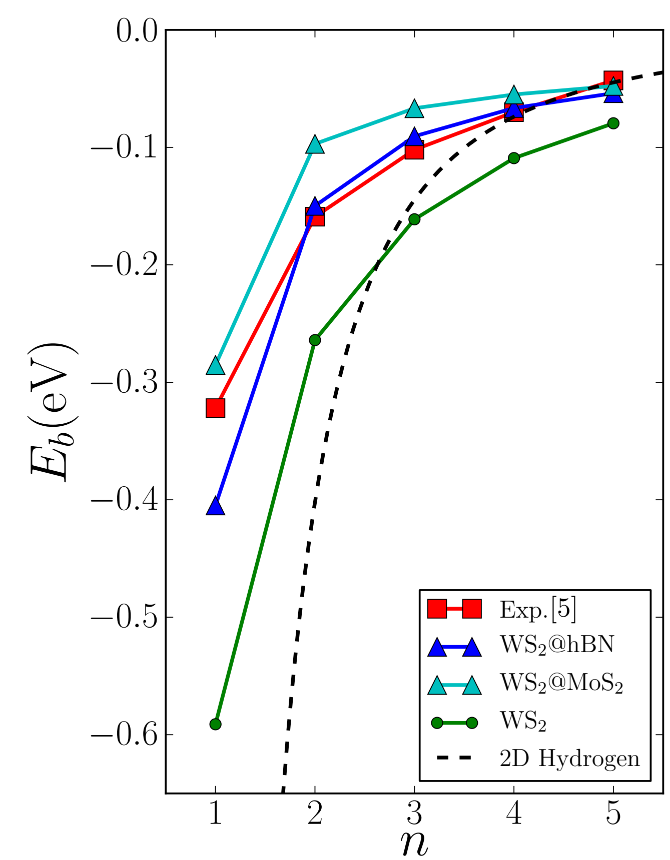




Alexey Chernikov et al, PRL **113**, 076802 (2014)





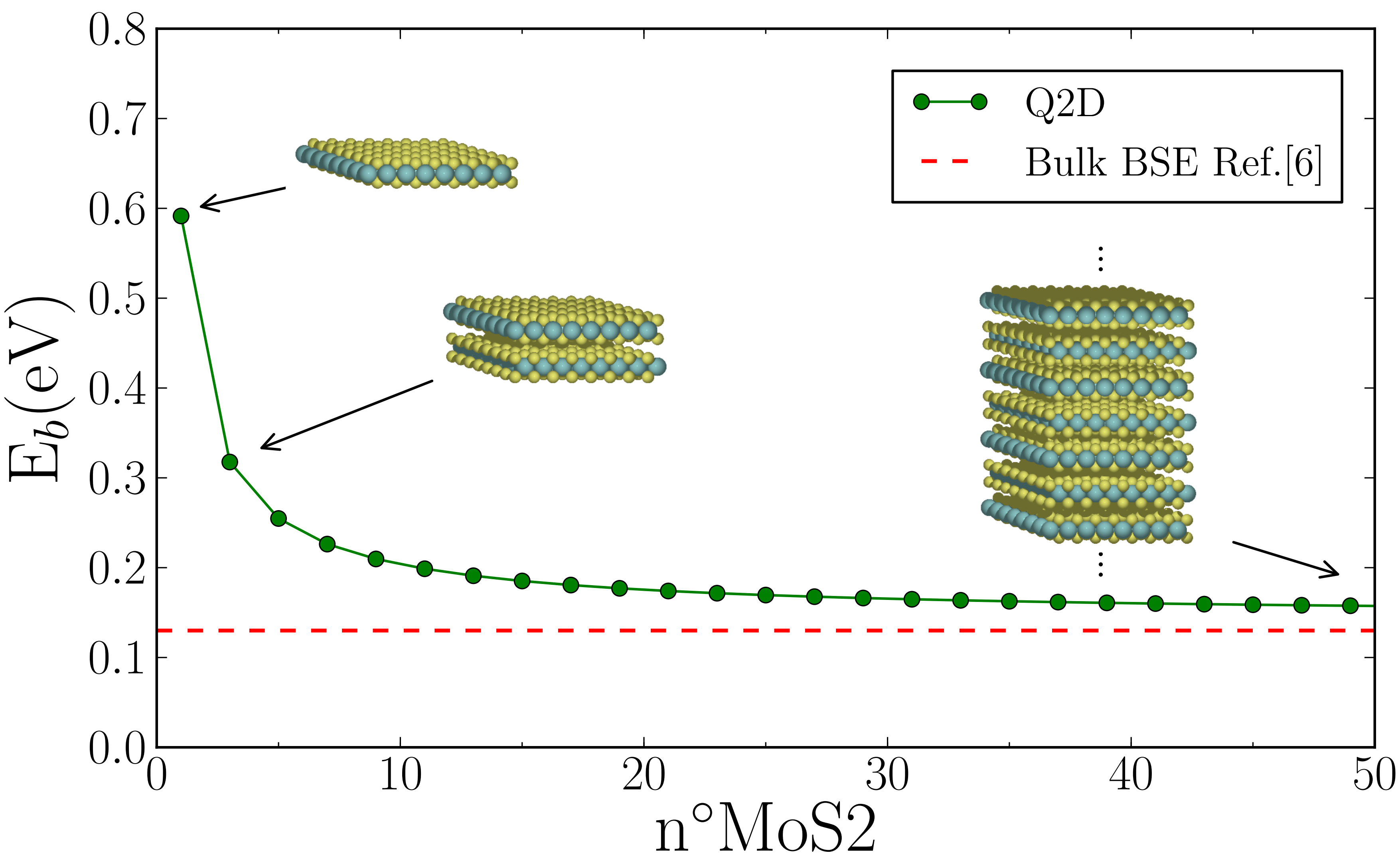


K. Andersen, S. Latini and K.S. Thygesen, Nanoletter **15**, 4616 (2015)

From 2D to 3D Excitons in MoS2

Mott-Wannier Hamiltonian for 3D layered structures:

$$\left[-\frac{\nabla_{\parallel}^2}{2\mu_{\parallel}^{ex}} - \frac{\nabla_{\perp}^2}{2\mu_{\perp}^{ex}} + W(\mathbf{r})\right] F(\mathbf{r}) = E_b F(\mathbf{r})$$



- $\mu_{\parallel}^{ex} \ll \mu_{\perp}^{ex}$: excitons are Q2D even in bulk;
- The variation in Eb is mainly due to the interlayer screening!

S. Latini, T. Olsen and K.S. Thygesen, submitted arXiv:1509.07972

Summing up

- We proposed multi-scale method that seamlessly connects exciting effects in the isolated layer limit to the multi-layer case;
- With the QEH model we can address scientific questions beyond the capability of the state of the art techniques;
- We provide a clear connection between 2D and 3D dielectric and exciting properties;
- We find good agreement with experiments

A new era for predicting and designing dielectric and excitonic properties of vdWHs

Acknowledgements

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Center for Nanostructured Graphene

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Thanks for the Attention!!!