Large scale calculations of electronic structure of 2D Crystals

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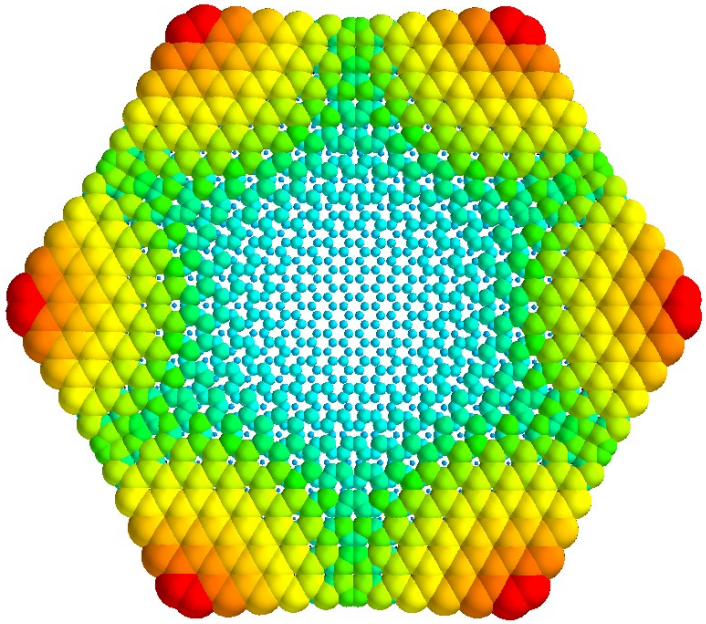
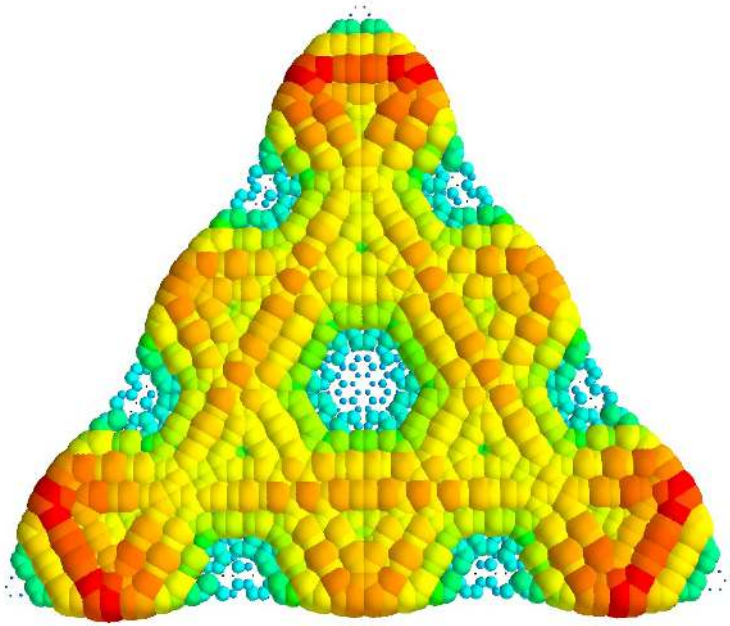
International Iberian Nanotechnology Laboratory, Portugal





NanoPT, 17th February 2016

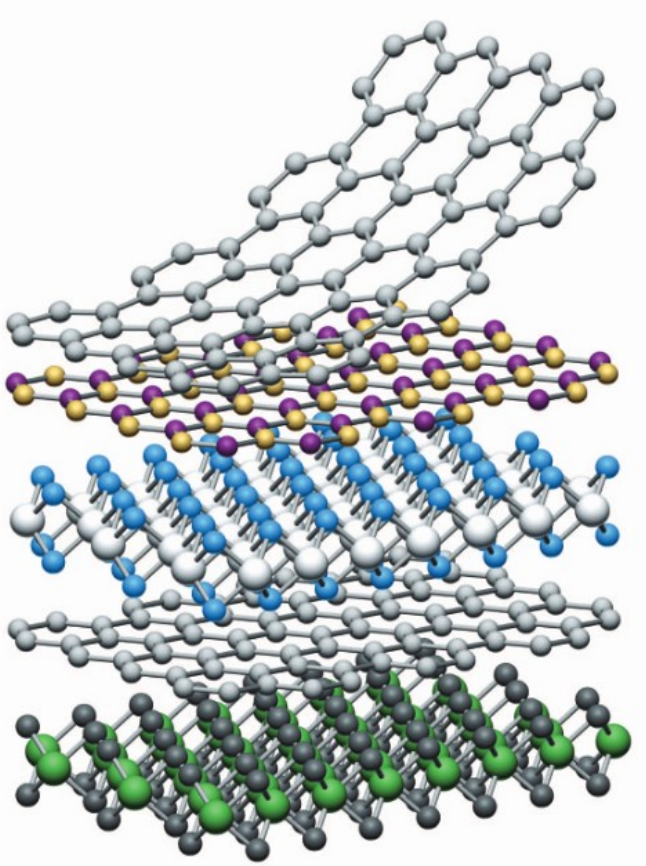
Why we care about electronic properties of materials

Health imaging techniques

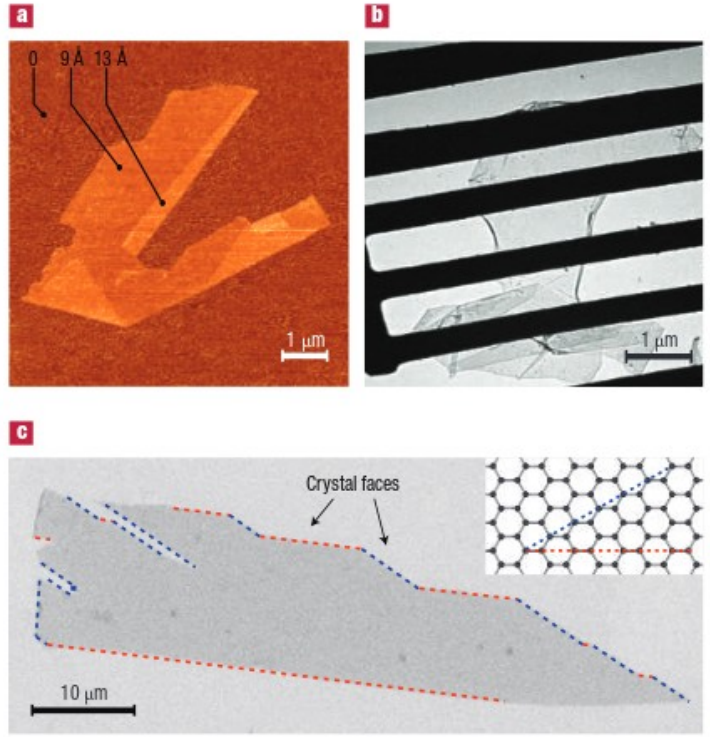
Personal computing

Space exploration

The rise of 2d materials



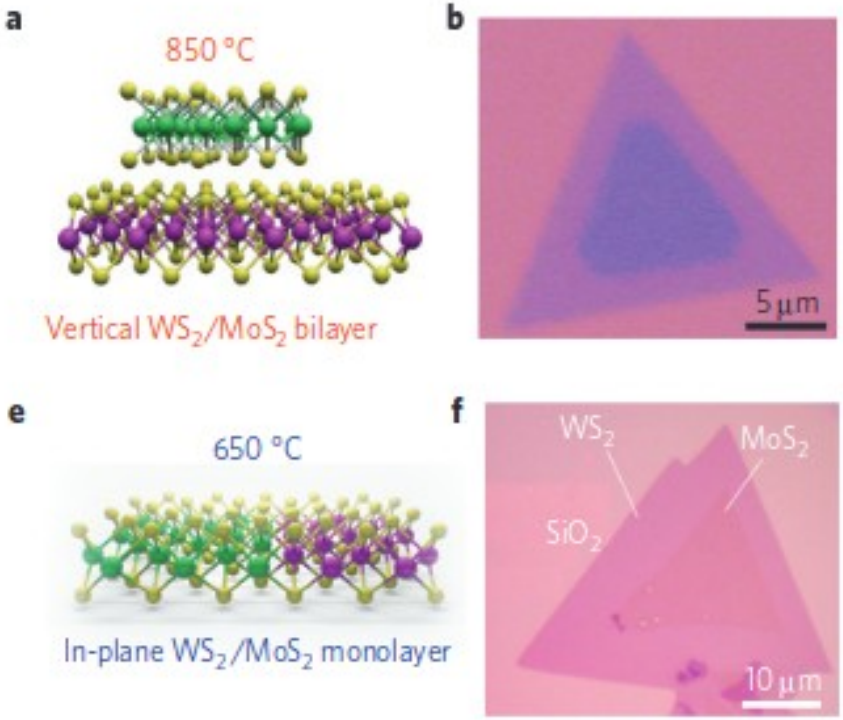
Nature **499**, 419–425 (25 July 2013)



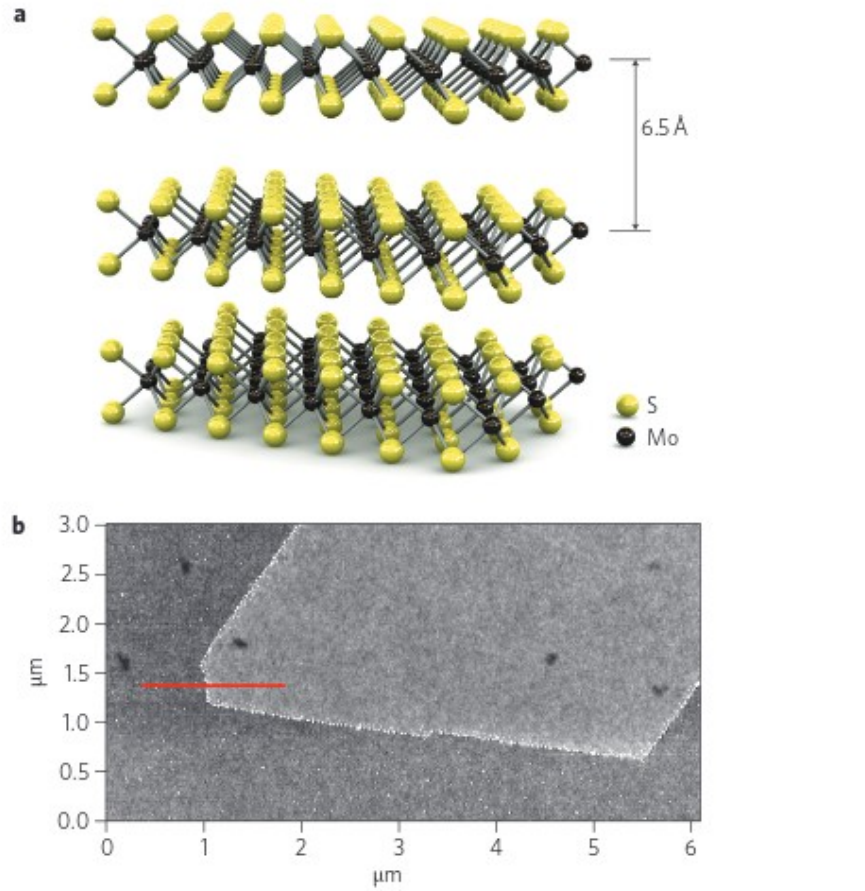
Nature Materials **6**, 183 - 191 (2007)

New materials with new properties

New materials: Molybdenite

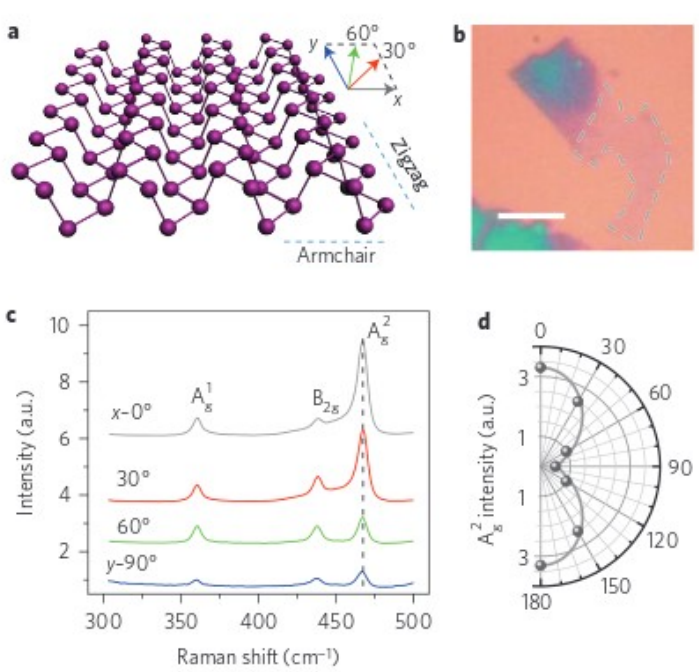


Nature Materials **13**, 1135–1142 (2014)

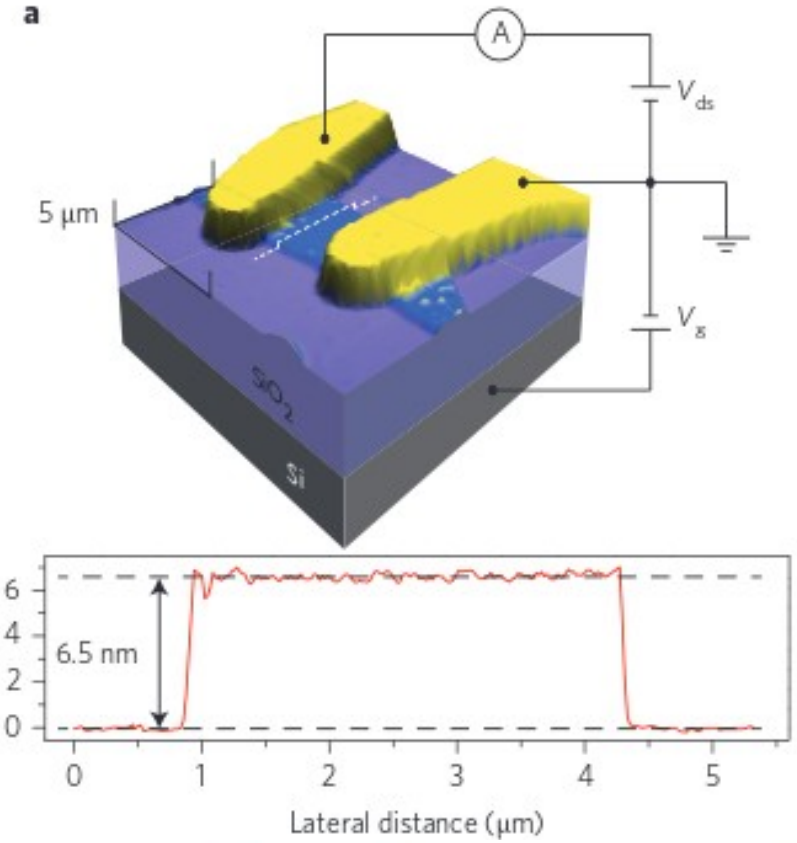


Nature Nanotechnology **6**, 147–150 (2011)

New materials: black phosphorus



Nature Nanotechnology **10**, 517–521 (2015)



Nature Nanotechnology **9**, 372–377 (2014)

New materials with unknown electronic properties

How to model electronic properties in nanoscale materials?

$$i\hbar\frac{\partial}{\partial t}\Psi(\mathbf{r},\,t) = -\frac{\hbar^2}{2m}\nabla^2\Psi(\mathbf{r},\,t) + V(\mathbf{r})\Psi(\mathbf{r},\,t).$$

Goal: solve the Schrodinger equation for the material

Three approaches to calculate electronic properties

k.p analytical Hamiltonian

Elegant and clear

Only simple electronic structures

Density functional theory

General, useful for any system

Only small systems

Tight binding modeling

Large scale systems

Needs reliable external input

What if no approach works?

The problem:

Quantum Hall effect in an arbitrary material

Different approaches fail

- Analytical model
  - **No clue of the analytical Hamiltonian**
- Tight binding
  - **No clue of parameters**
- Density functional theory
  - **Way beyond current computational power**

What do we do when the three of them fail?

Get the best of two worlds

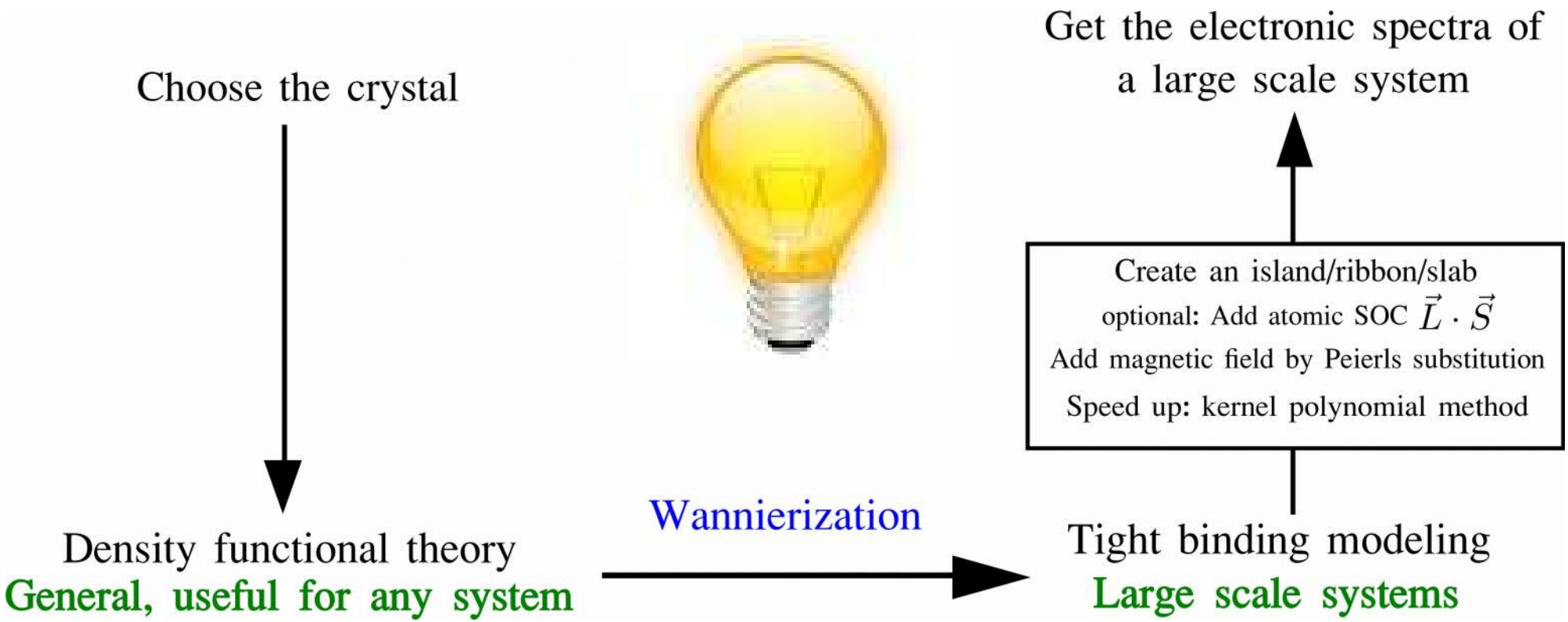
k.p analytical Hamiltonian
Elegant and clear
Only simple electronic structures

Density functional theory
General, useful for any system
Only small systems

Tight binding modeling
Large scale systems
Needs reliable external input

Wannierization

The solution: start with DFT, end up in tight binding



Wannier90 open source package

Comput. Phys. Commun. 178, 685 (2008)

A method for large scale systems

- Calculation of the spectrum for a tight binding can be expensive for very large scale

Time scales as (atoms)^3

1000 atoms in 1 second  100000 atoms in 10 days

Using Kernel polynomial method

No need of diagonalization, expansion in Chebyshev polynomials

$$D_i(E) = \frac{1}{\pi\sqrt{1-E^2}}\left[\mu_0 + 2\sum_n \mu_n T_n(E)\right]$$

$$T_0(x) = 1 \qquad T_1(x) = x$$

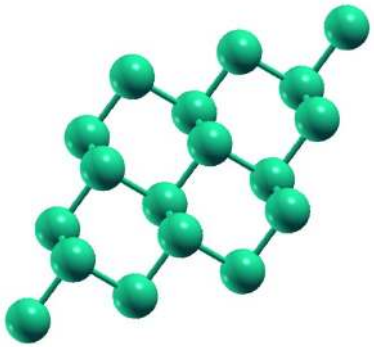
$$T_{m+1}(x) = 2xT_m(x) - T_{m-1}(x)$$

$$\mu_n = \sum_k \int_{-1}^{1} |\langle i|k\rangle|^2 \delta(E_k - E) T_n(E) = \sum_k |\langle i|k\rangle|^2 T_n(E_k) = \langle i|T_n(H)|i\rangle$$

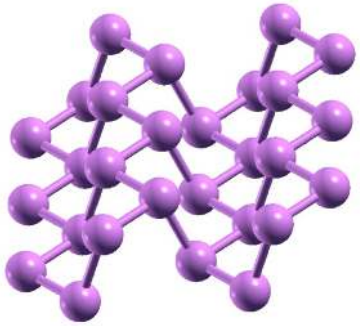
Rev. Mod. Phys. 78, 275 (2006)

Applying the general technique to new 2D materials

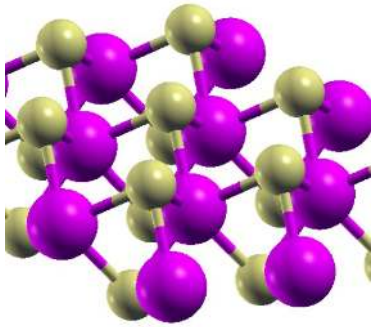
Topological insulator
Bi (111)



Novel 2D semiconductor
black phosporus

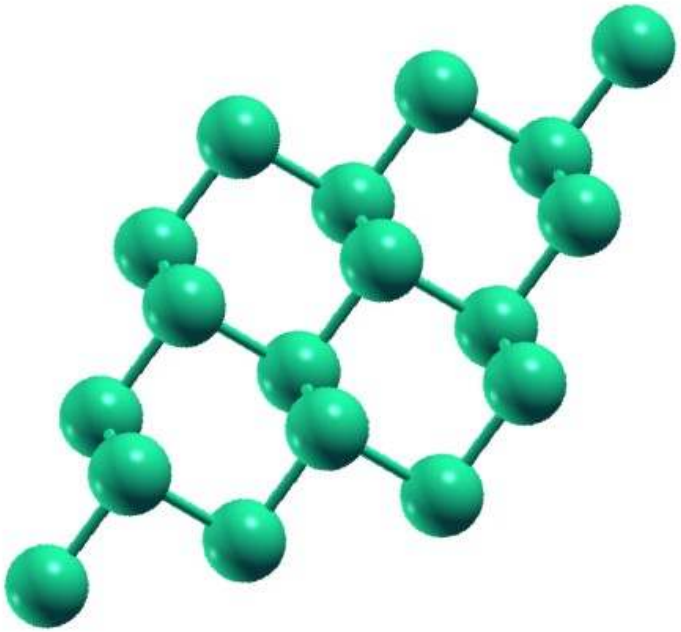


Gapped Dirac material
MoS_2

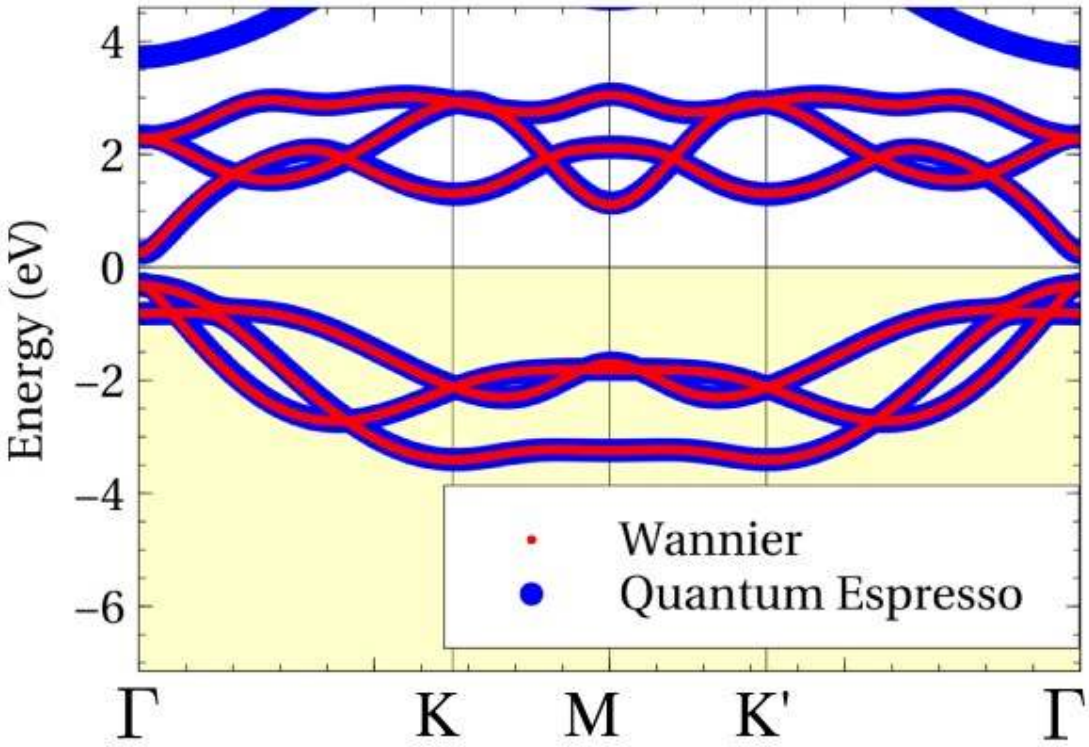


Bismuth 111

Buckled honeycomb structure



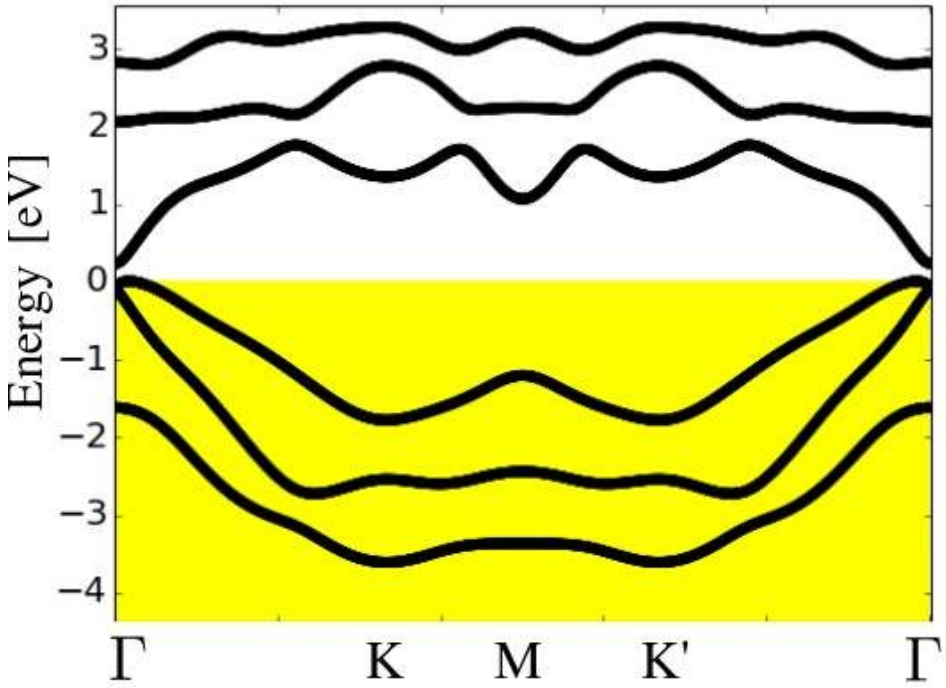
Band structure without SOC



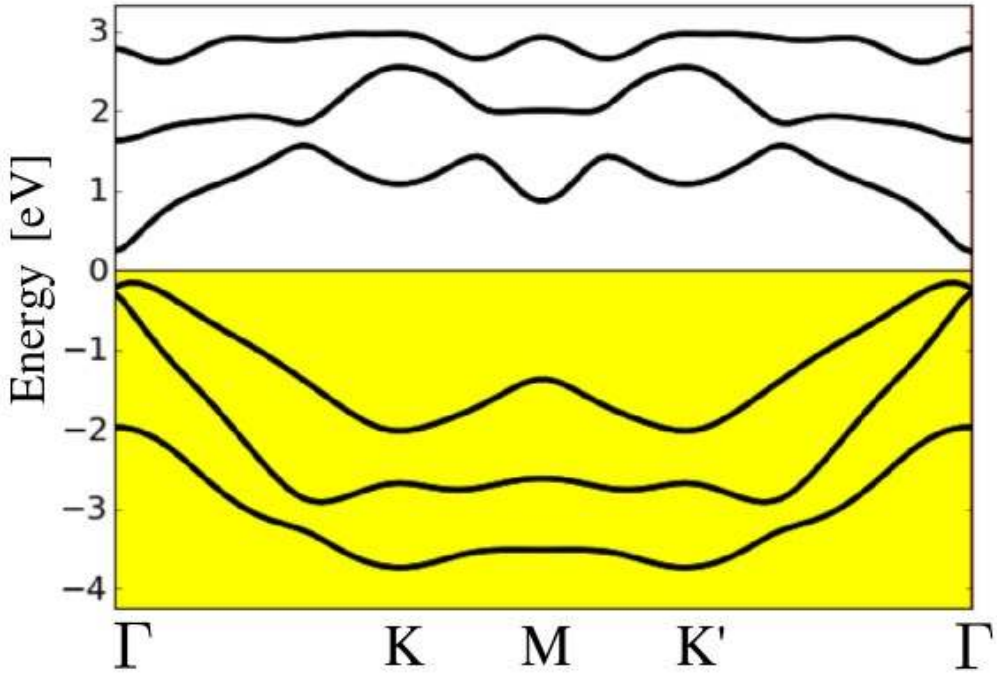
Wannier band structure with p-like states matches DFT

Bismuth 111 with spin-orbit coupling

Wannier + atomic SOC



All electron DFT with SOC

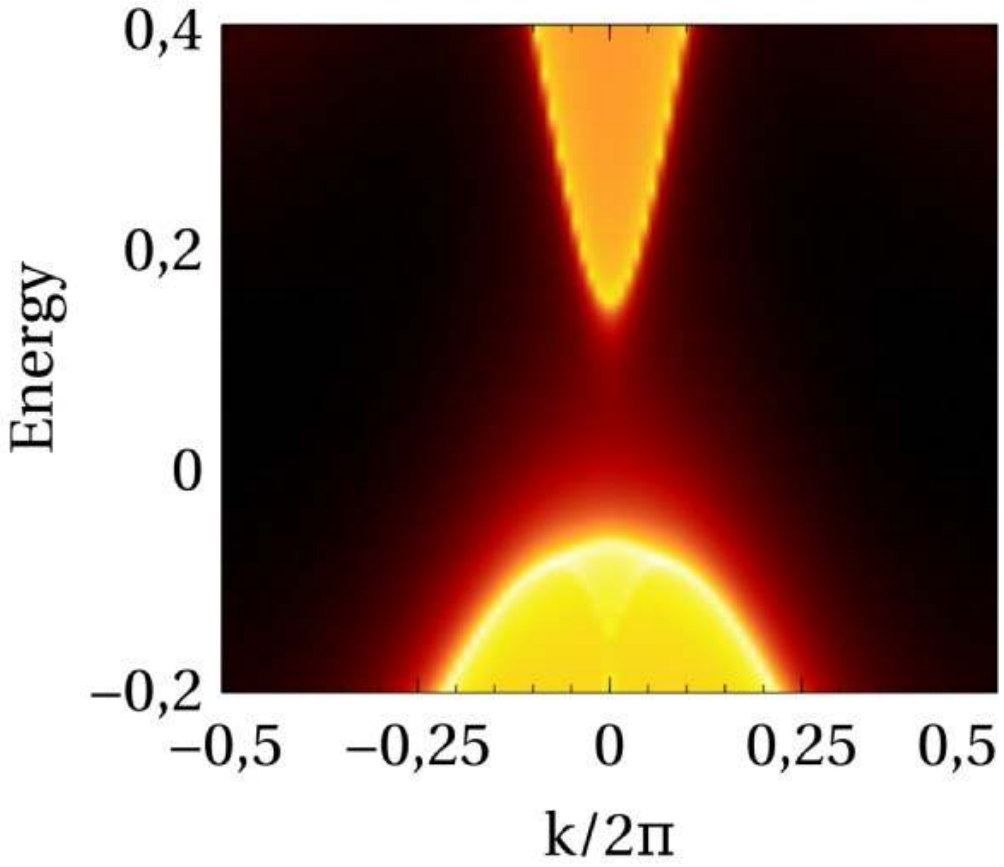


Wannier + atomic SOC agrees with fully relativistic DFT

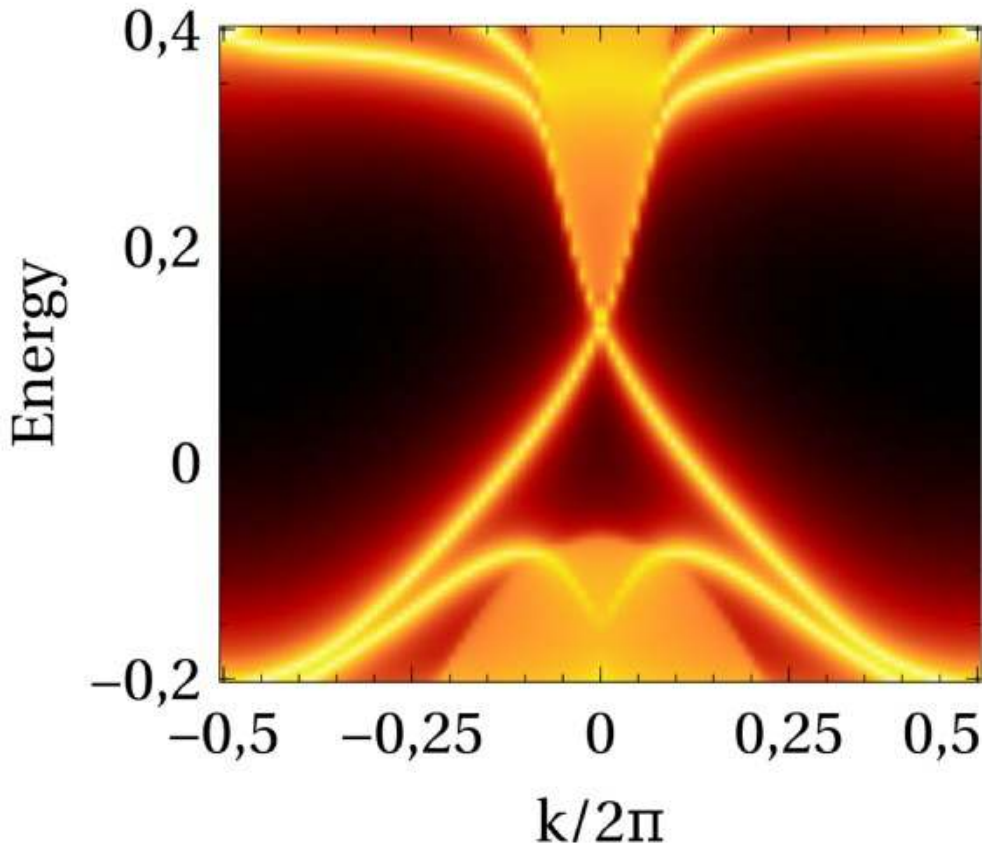
k-resolved spectral DOS Bi(111)

Gapless surface states and gapped bulk spectrum

Bulk k-resolved DOS



Surface k-resolved DOS

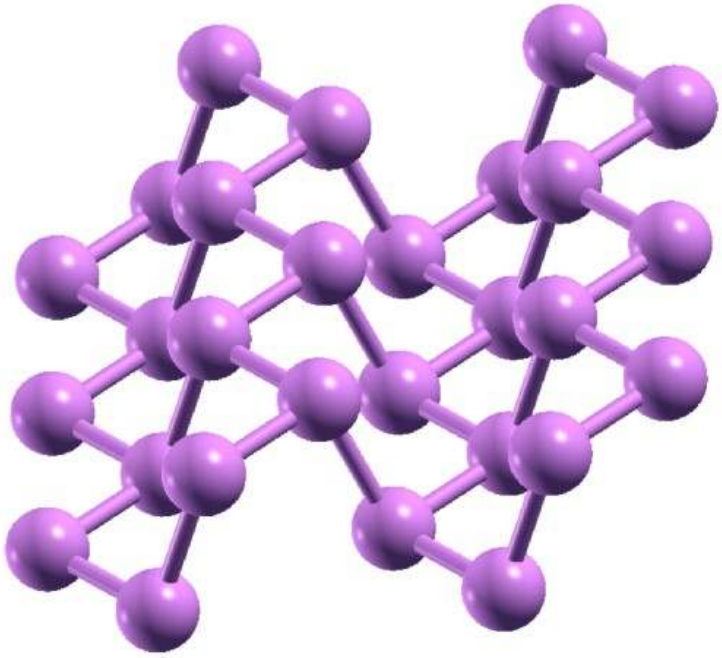


Non trivial topology

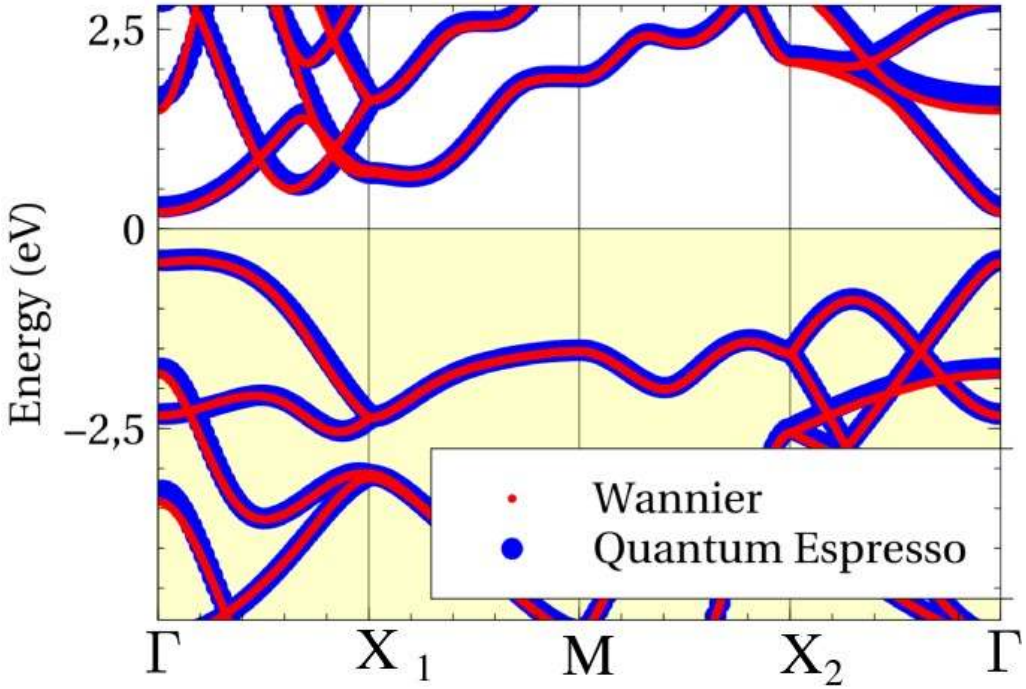
Phys. Rev. Lett. 107, 166801 (2011)

Novel semiconductor: black phosphorus

Distorted hexagonal lattice

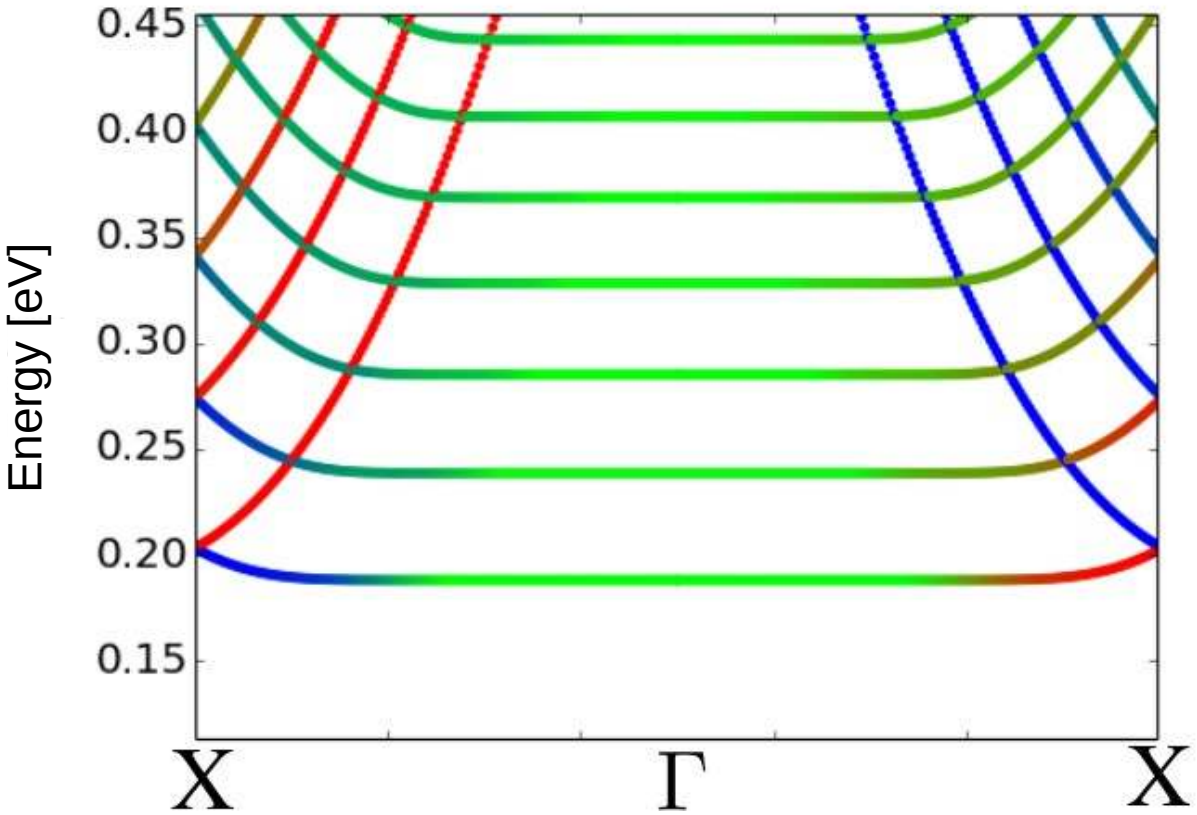


Bulk band-structure



Band structure is well captured with Wannier p-like states

Quantum Hall effect in black phosphorus

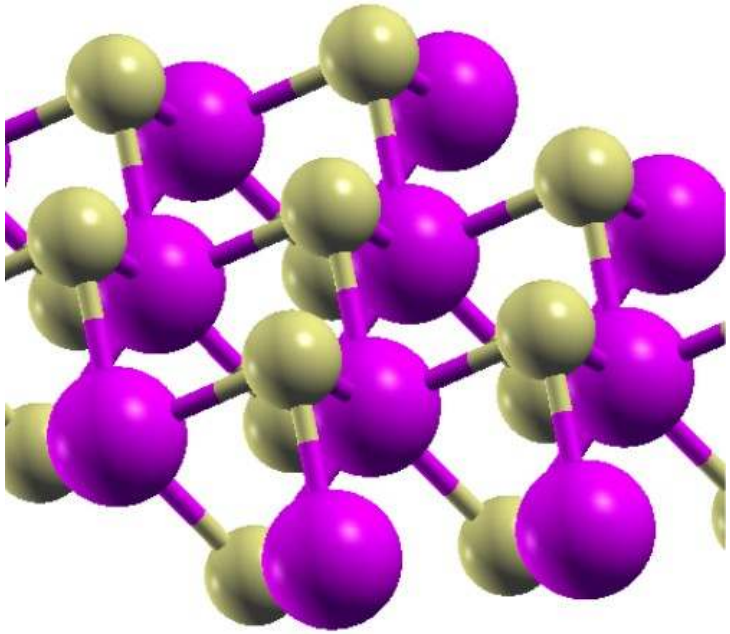


Landau spectrum in black phosphorus

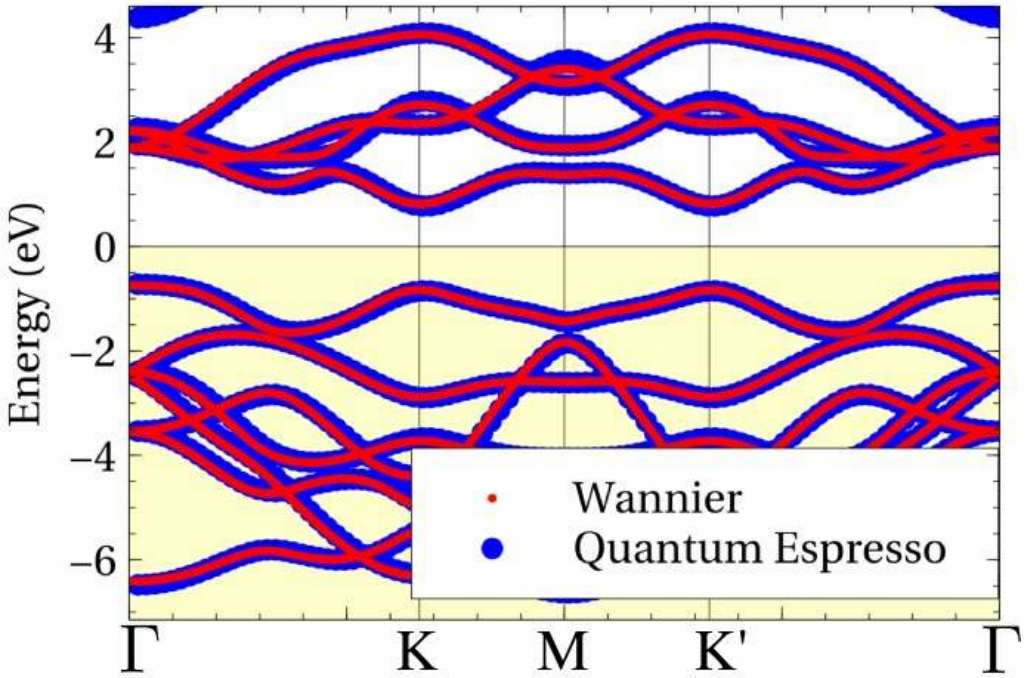
Phys. Rev. B 92, 075437 (2015)
arXiv:1504.07155 (2015)

Gapped Dirac material: MoS_2

Hexagonal-like structure



Bulk band structure

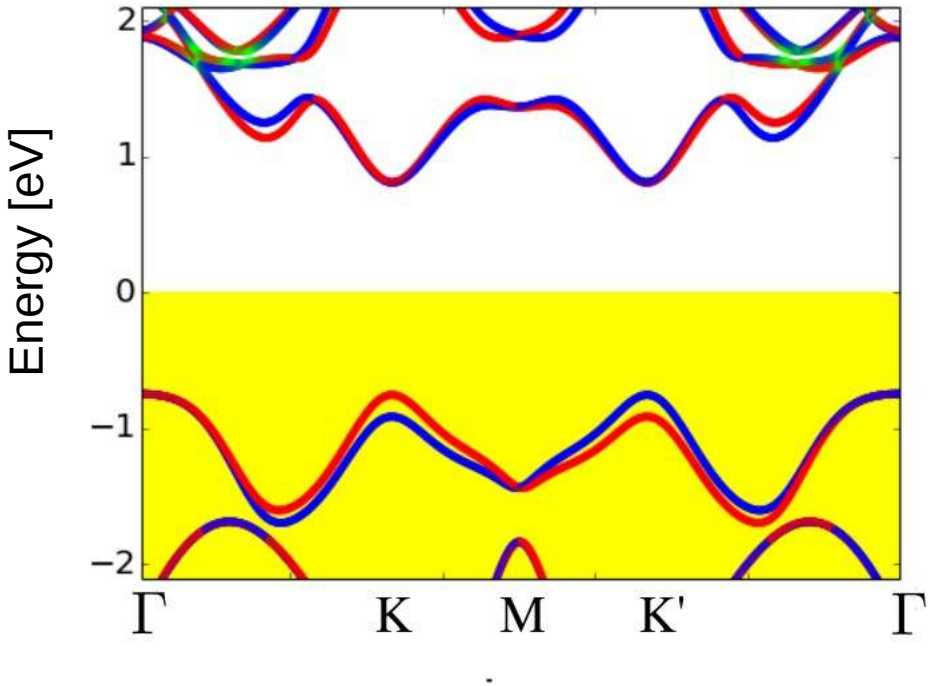


Band structure well captured with Mo d-like and S p-like Wannier states

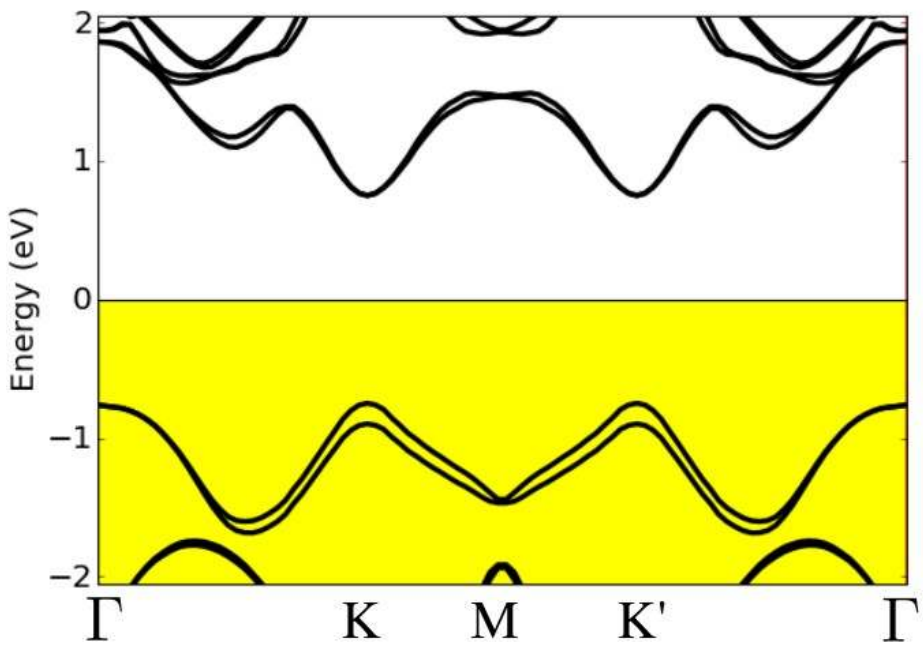
Relativistic effects in in MoS_2

Spin orbit coupling can be included in the Wannier Hamiltonian

Wannier + atomic SOC



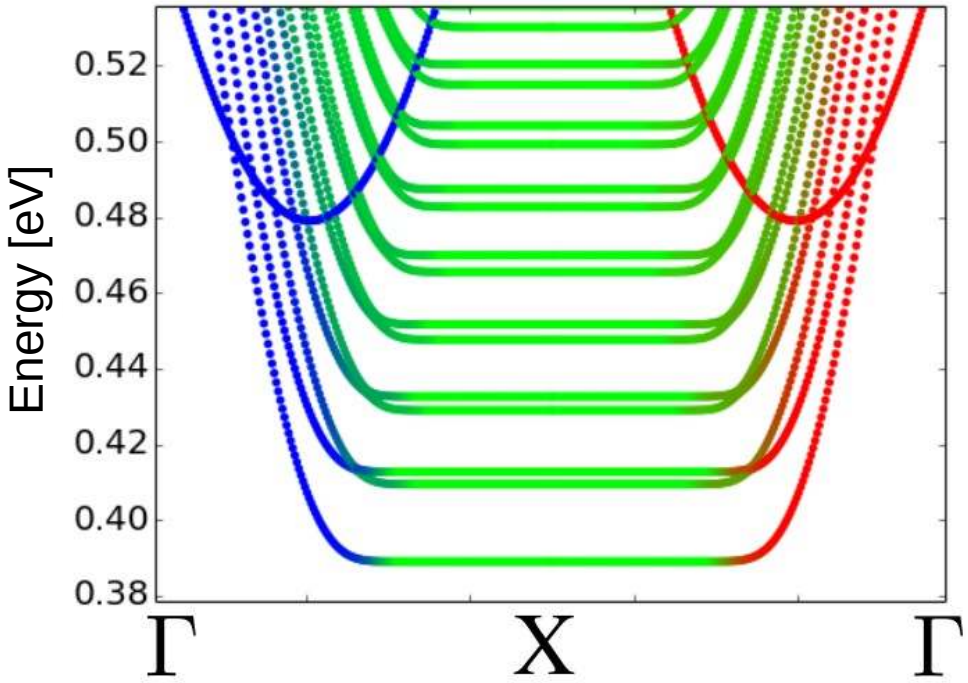
All-electron DFT with SOC



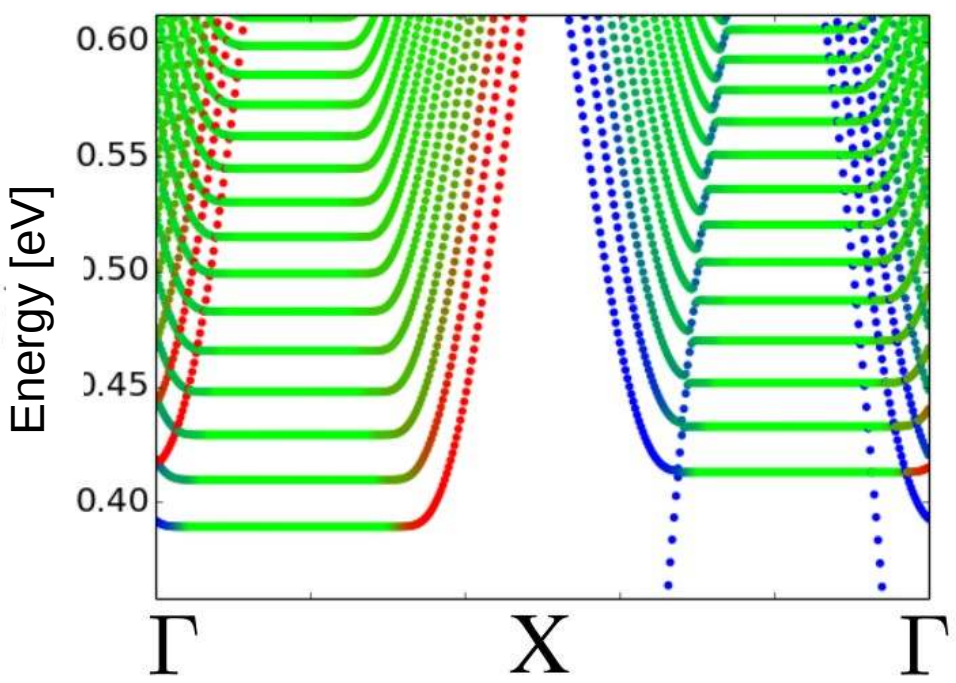
Wannier + atomic SOC captures valley dependent spin splitting

Quantum Hall in MoS_2

Armchair edge



Zigzag edge

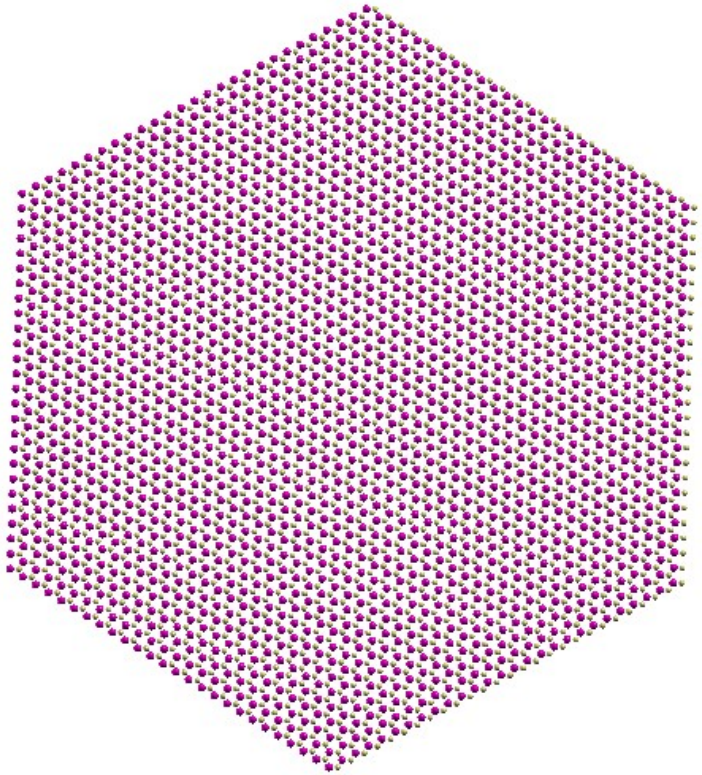


Anomalous Landau level spectra, valley dependent splitting

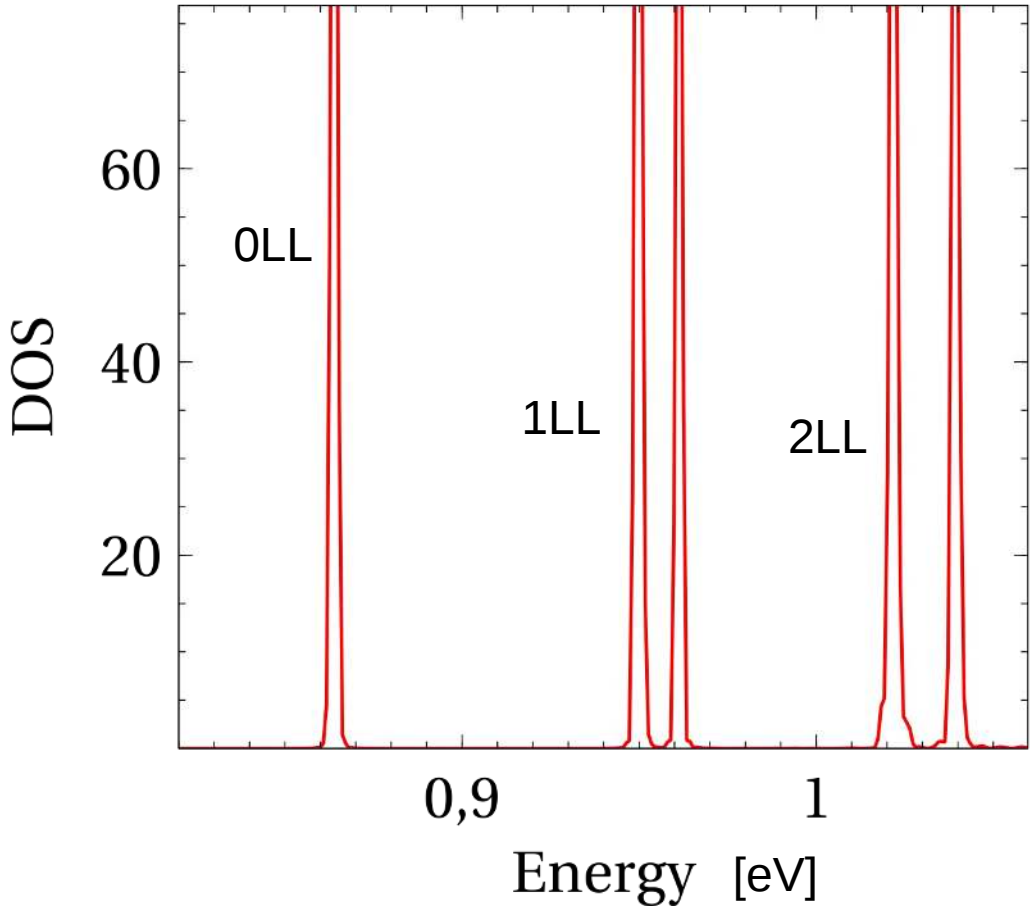
Phys. Rev. B 90, 045427 (2014)
Phys. Rev. B 91, 075433 (2015)

Quantum Hall in MoS_2 flake

Hexagonal MoS2 flake



DOS in the center of the flake



KPM allows to calculate electronic states in a big quantum Hall MoS2 flake

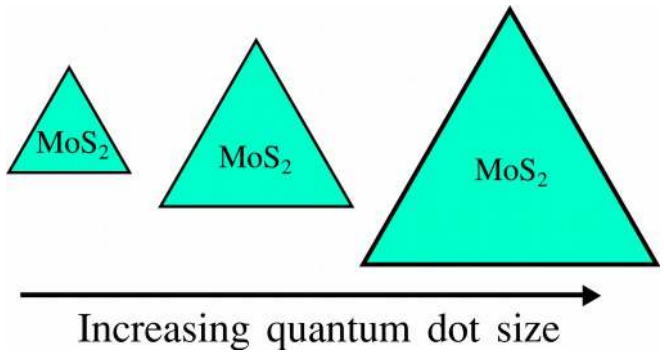
Pros and cons of the whole method

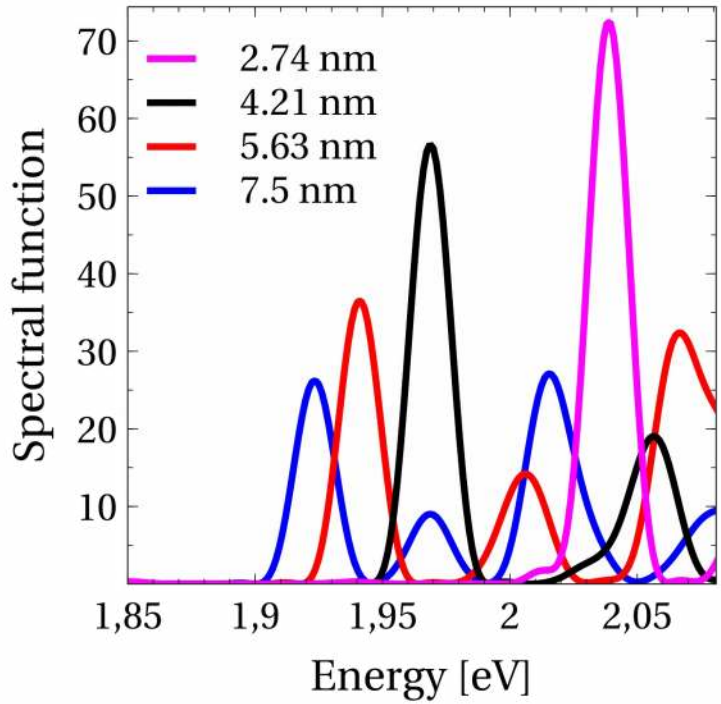
- **Pros**
  - Any material (single particle picture)
  - Large scale systems: ribbons, islands, slabs...
  - Allows to control SOC, magnetic field, defects, interfaces...
- **Cons**
  - Ignores structural/surface/electronic reconstruction
  - Inherits all the problems of DFT

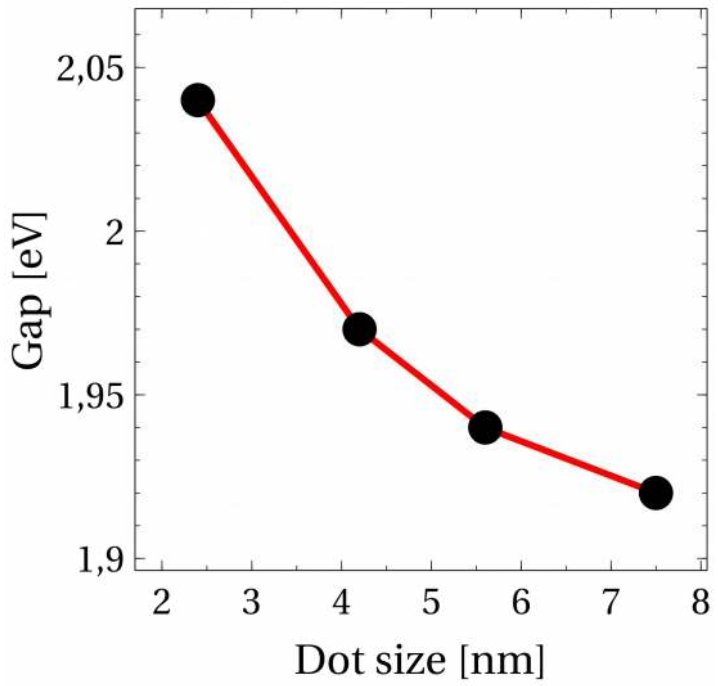
What else could you calculate?

- Dependence of gap with quantum dot size
- Transport between heterostructures
- Field effect modeling
- Magnetoresistance effects
- Doping dependent transport
- Thermoelectric effects
- Optical absorption
- Topological electronic states
- …..

Control of gap by quantum confinement in MoS_2







Control of gap by confinement in MoS_2

Summary

- Wannierization allows to study electronic properties in large scale systems, starting with DFT, for nearly any material

Thank you!!

We acknowledge financial support from

Spinograph
Spintronics in Graphene