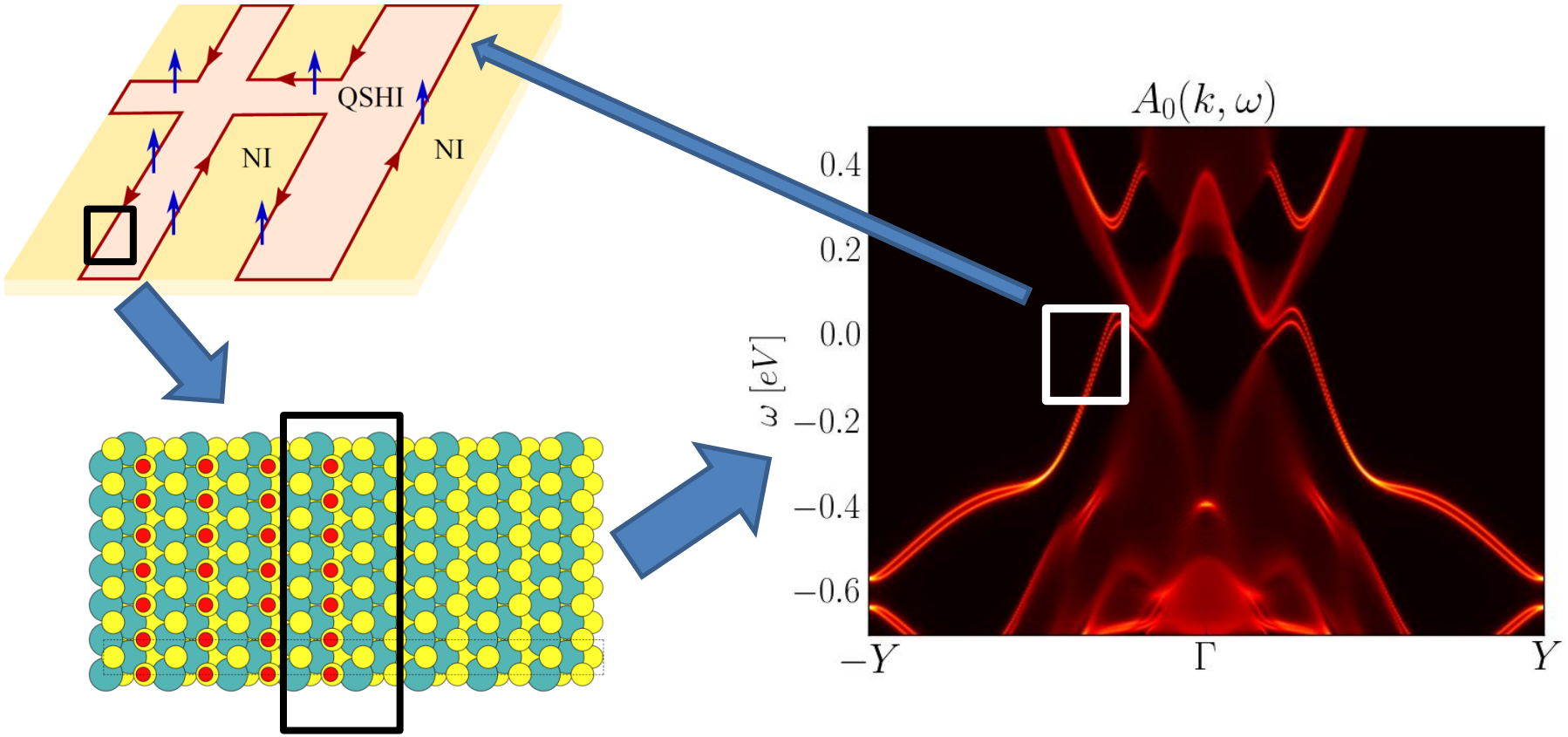
# Designing topological in-plane heterostructures from first principles: 1T'-$MoS_2$



# Outline

- Spin-orbit coupling in GPAW
- Two-dimensional topological insulators
- Topological in-plane heterostructure with 1T'-$MoS_2$
- Metallic boundary states

# Standard derivation

The Dirac equation is

$$\left[\beta mc^2 + c\boldsymbol{\alpha}\cdot\boldsymbol{p} + V(x)\right]\psi(x) = E\psi(x)$$

where **α** and β are 4x4 matrices, which can be written in terms of the Pauli matrices.

Writing

$$\psi(x) = \begin{bmatrix}\xi(x)\\ \eta(x)\end{bmatrix}$$

eliminating η and expanding to second order in $(E - V)/mc^2$ yields

Kinetic correction  Darwin  Spin-orbit

$$\left[\frac{p^2}{2m} + V(x) - \frac{p^4}{8m^3c^2} - \frac{i\hbar\boldsymbol{p}\cdot\boldsymbol{\nabla}V}{4m^2c^2} + \frac{\hbar\boldsymbol{\sigma}\cdot\boldsymbol{p}\times\boldsymbol{\nabla}V}{4m^2c^2}\right]\xi(x) = E_s\xi(x)$$

spherical symmetry

$$-\frac{\boldsymbol{S}\cdot\boldsymbol{L}}{2m^2c^2}\frac{1}{r}\frac{dV}{dr}$$

where ξ is a two-component spinor

# Implementation of SO in GPAW

Since the spin-orbit coupling involves the gradient of the potential, the dominant contribution is near the nucleii

We assume that all SO is captured inside the PAW augmentation sphere where

$$|\psi_{n\sigma}\rangle = \sum_{ai} \langle \widetilde{p_i^a} \, |\tilde{\psi}_n\rangle |\phi_i^a \sigma\rangle$$

The full SO Kohn-Sham Hamiltonian can then be set up in a basis of scalar-relativistic states

$$\mathrm{H}_{\mathrm{n_1 n_2 \sigma_1 \sigma_2}} = \delta_{\sigma_1\sigma_2}\delta_{n_1 n_2}\varepsilon_{n_1\sigma_1} + \langle \psi_{n_1\sigma_1} | H^{SO} | \psi_{n_2\sigma_2} \rangle$$

$$\langle \psi_{n_1\sigma_1} | H^{SO} | \psi_{n_2\sigma_2} \rangle = \sum_{a i_1 i_2} \langle \tilde{\psi}_{n_1} | \tilde{p}_{i_1}^a \rangle \, \langle \phi_{i_1}^a \sigma_1 | H^{SO} | \phi_{i_2}^a \sigma_2 \rangle \langle \tilde{p}_{i_2}^a | \tilde{\psi}_{n_2} \rangle$$

# Implementation of SO in GPAW

For spherically symmetric potentials the atomic orbitals factorizes into a radial part and spherical harmonic

$$\phi_i^a(\boldsymbol{r}) = f_i(r)Y_i(\phi,\theta)$$

We thus have to evaluate

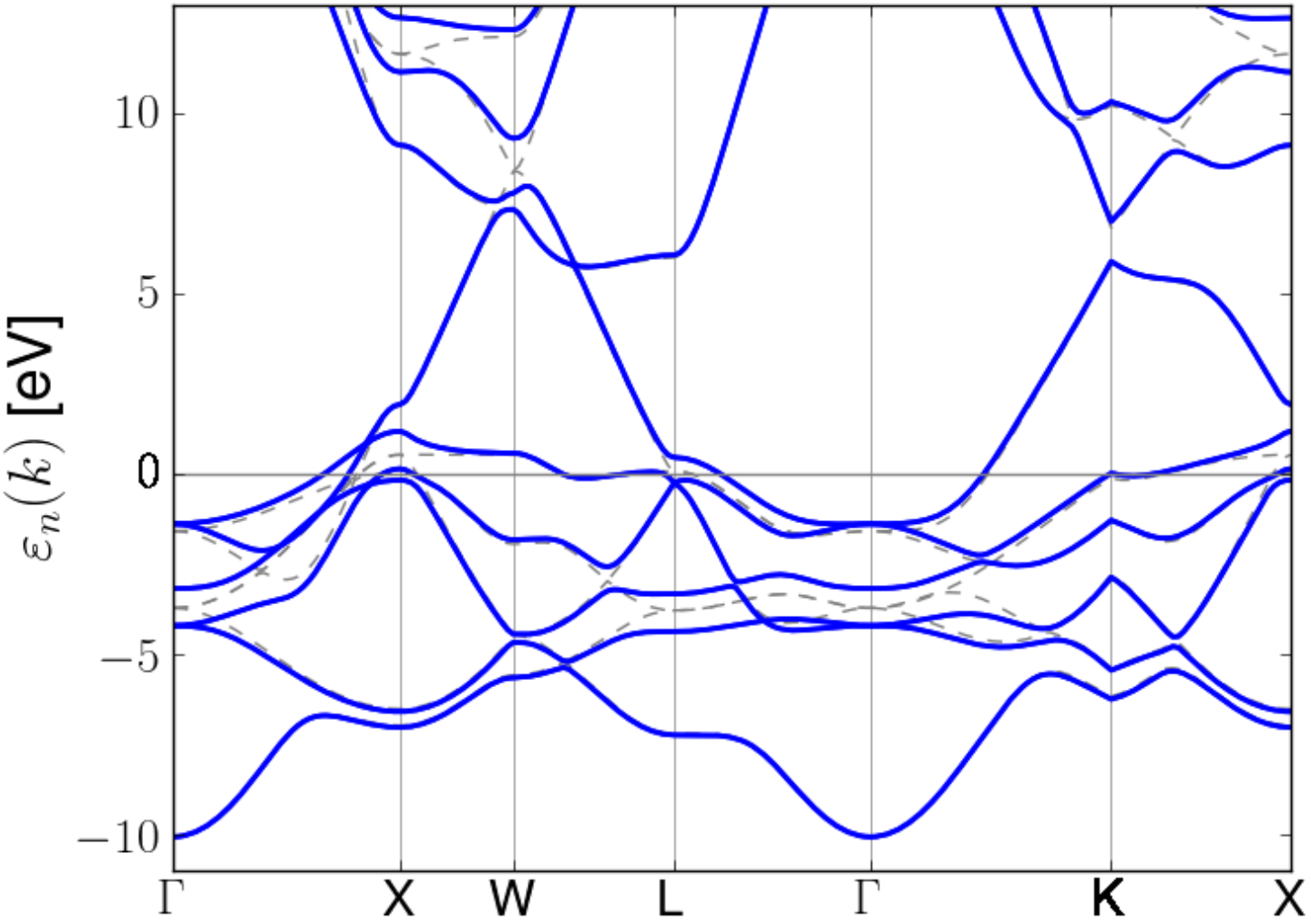
$$\langle \phi_{i_1}^a \sigma_1 | H^{SO} | \phi_{i_2}^a \sigma_2 \rangle = \langle f_{i_1}^a Y_{i_1}^a \sigma_1 | H^{SO} | f_{i_2}^a Y_{i_2}^a \sigma_2 \rangle$$

$$= -\frac{1}{2m^2c^2} \langle Y_{i_1}^a \sigma_1 | \boldsymbol{S}\cdot\boldsymbol{L} | Y_{i_2}^a \sigma_2 \rangle \langle f_{i_1}^a | \frac{1}{r}\frac{dV}{dr} | f_{i_2}^a \rangle$$

The radial part is calculated numerically on a non-uniform grid. The angular matrix elements have been tabulated. For example

$$\langle p_i | L_x | p_j \rangle = \begin{bmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \qquad i,j \in \{y,z,x\}$$

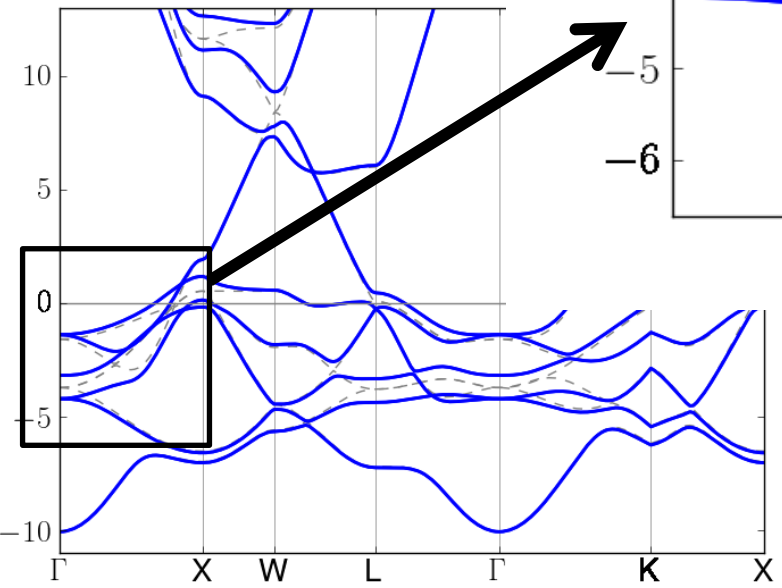
# fcc Pt

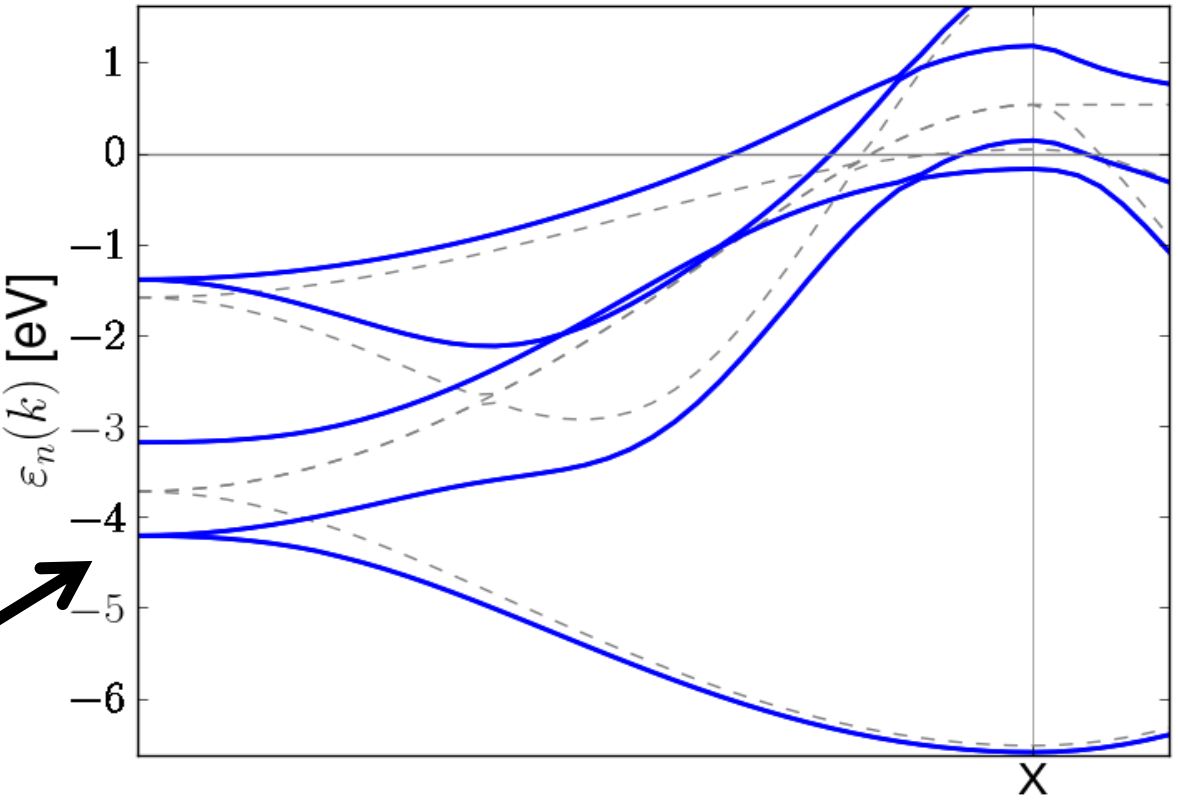



Spin degeneracy is protected by inversion symmetry

# fcc Pt

Spin-orbit corrections of up to 1.5 eV

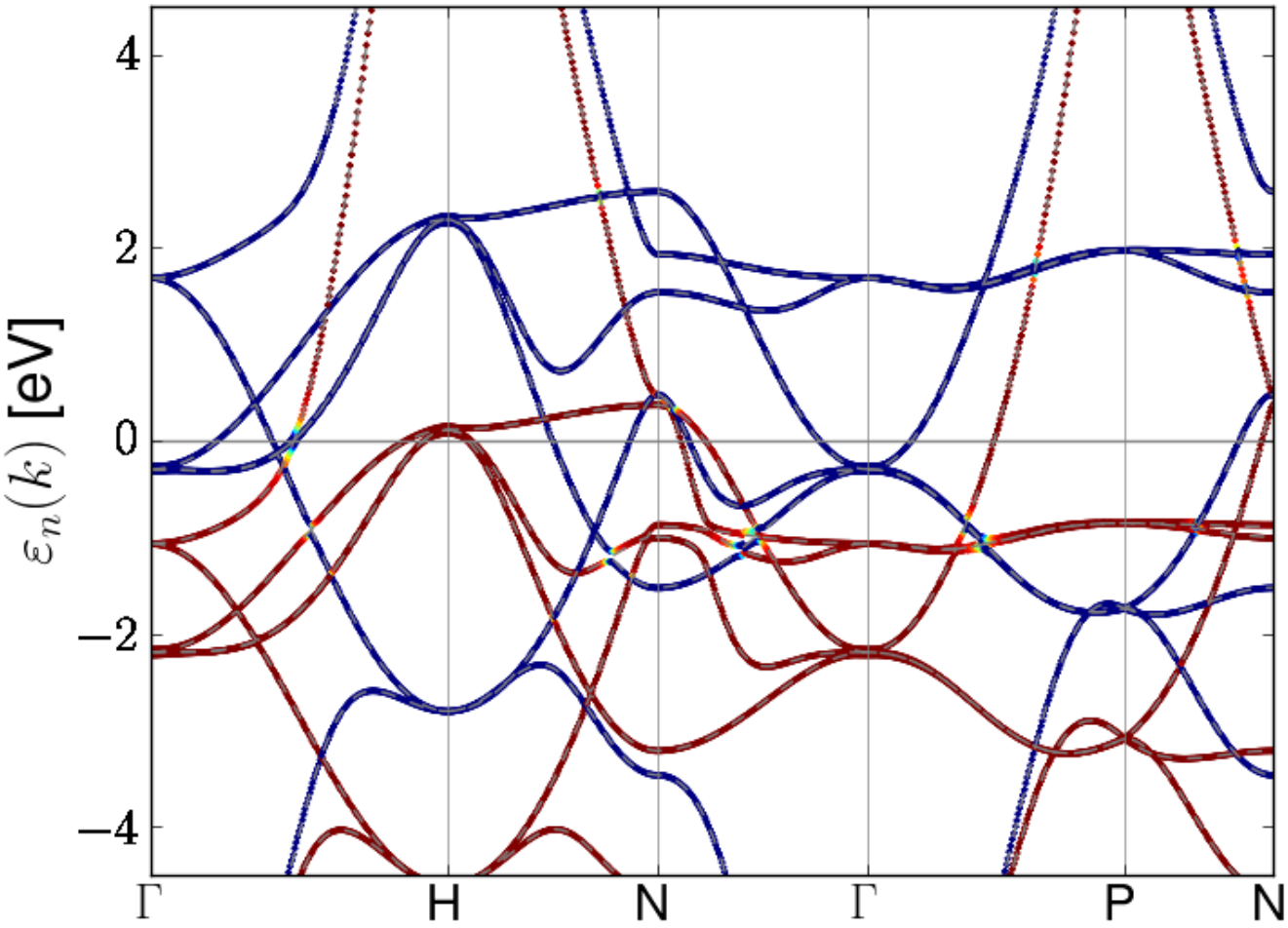



The double degenerate *d* states carry a two-dimensional irreducible representation of $C_{4v}$

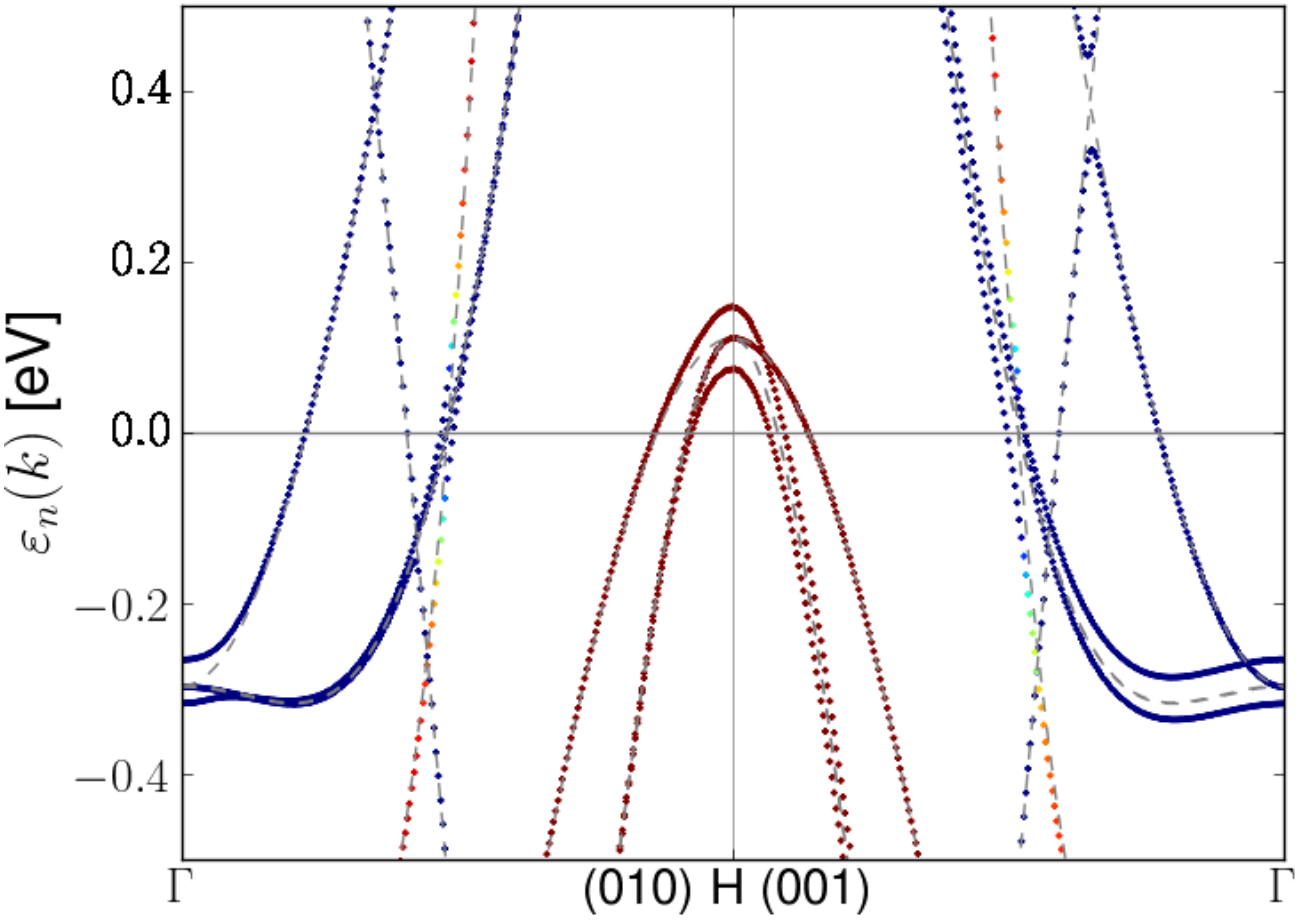
Symmetry is reduced to $C_4$ by spin-orbit coupling

# bcc Fe




Color scale denotes spin character: $\mathrm{S_n(k)} = \langle \psi_{kn} | \sigma_z | \psi_{kn} \rangle$

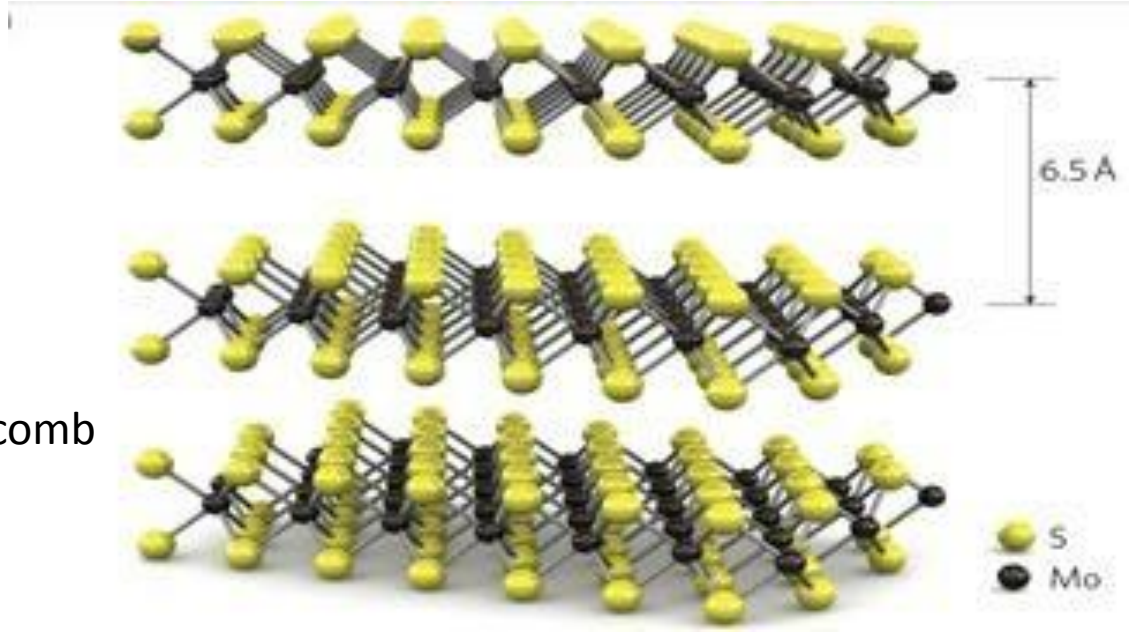
# bcc Fe



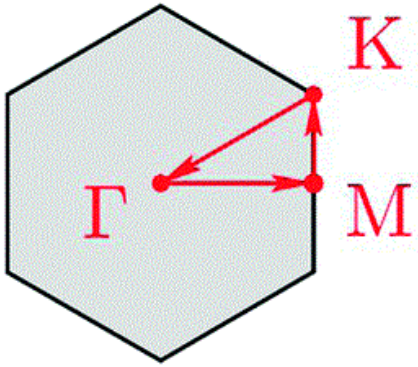

The band structure depends on the direction relative to the spin projection

# $MoS_2$

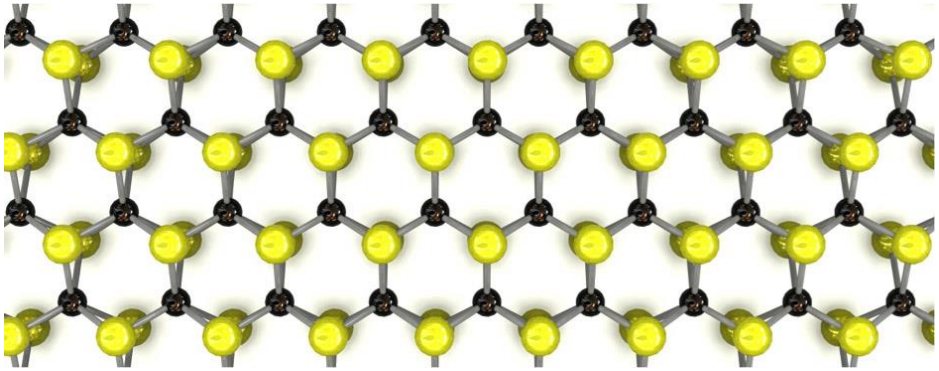
Bulk $MoS_2$ is composed of stacked two-dimensional sheets bound by van der Waals interactions




The indvidual sheets have a honeycomb lattice similar to graphene or hBN

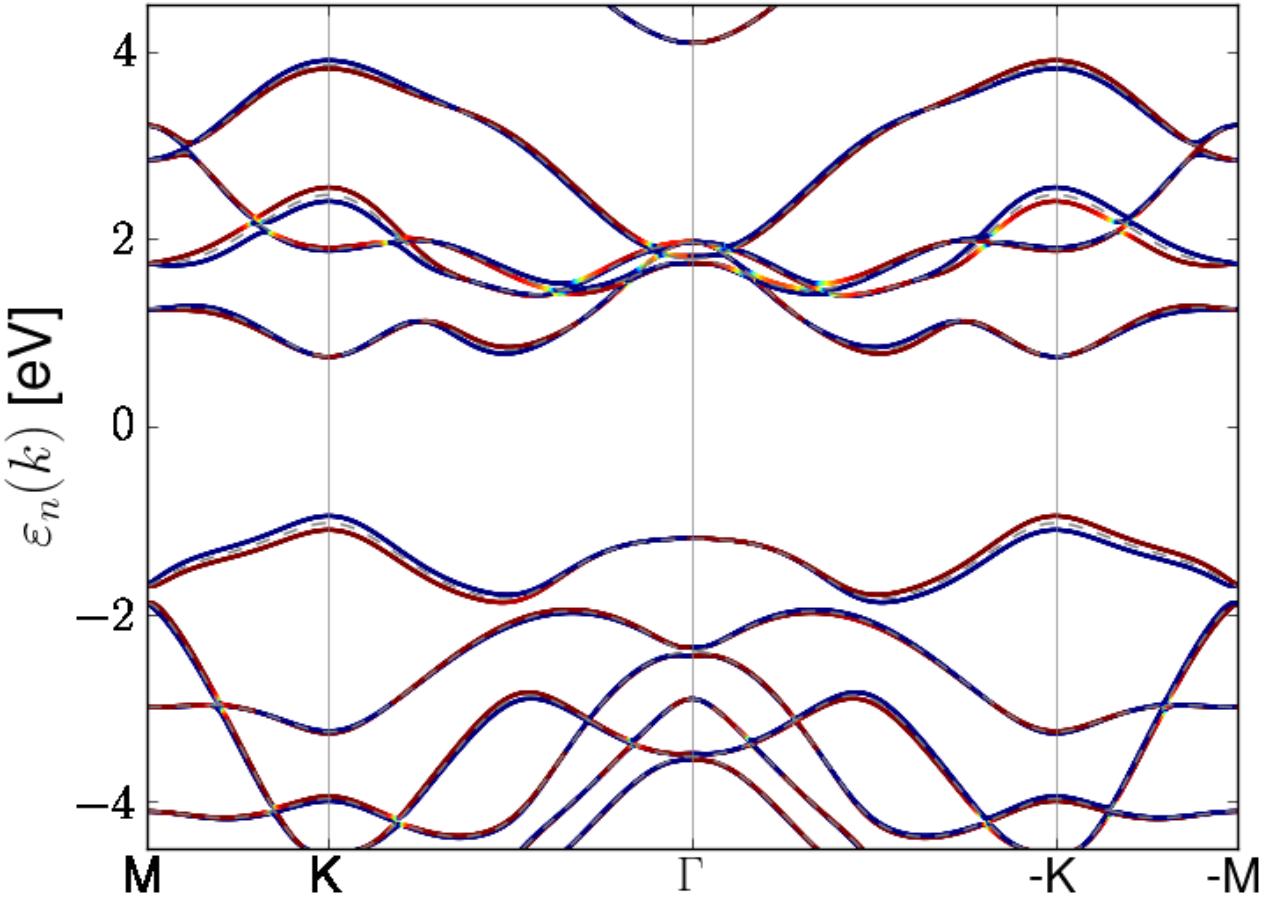


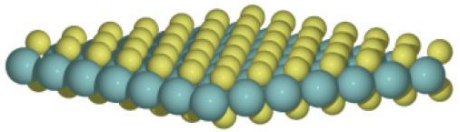

Brillouin zone

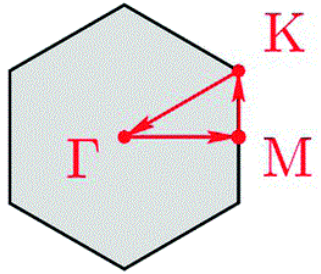


# Monolayer $MoS_2$

Lack of inversion symmetry



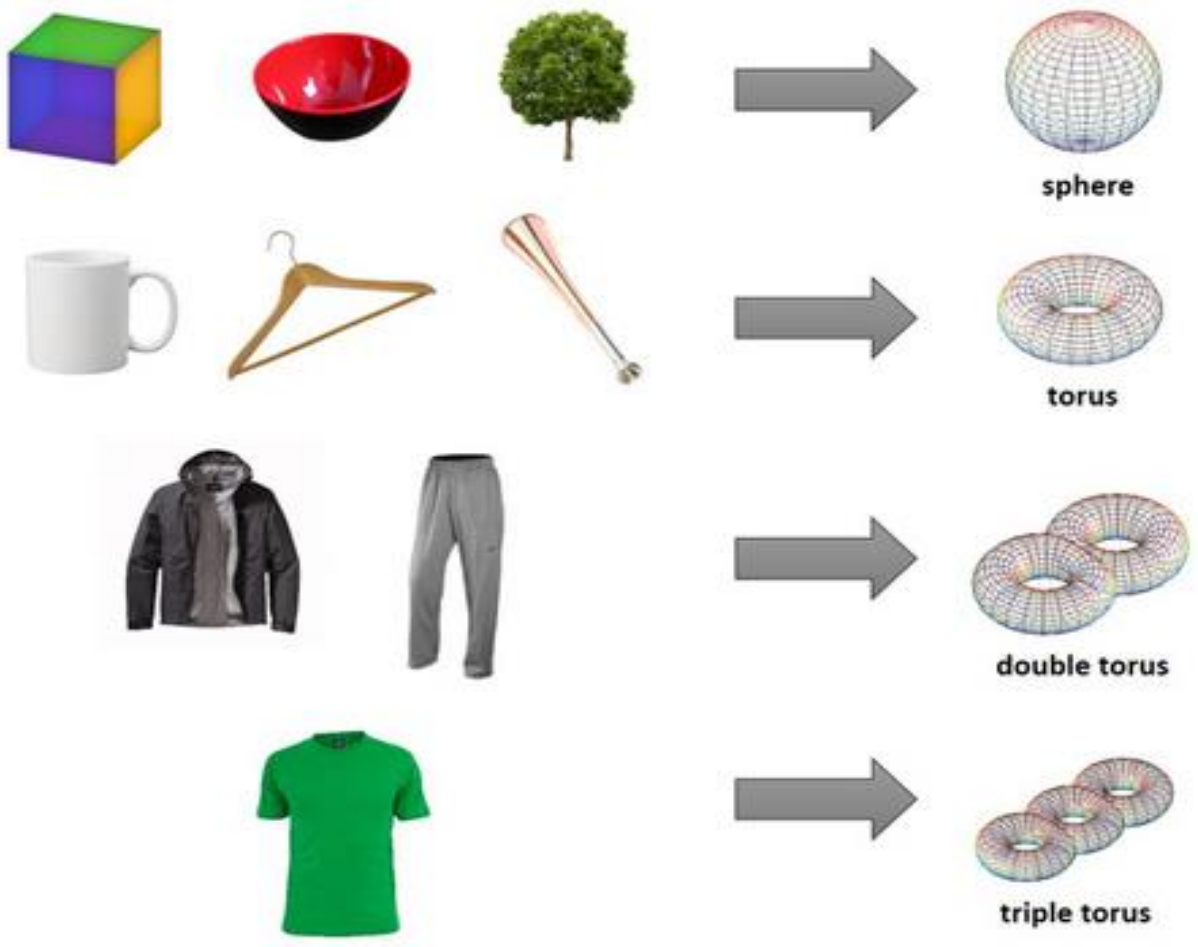






= 0.149 eV

Color scale denotes spin character: $S_n(k) = \langle \psi_{kn} | \sigma_z | \psi_{kn} \rangle$

# Topology

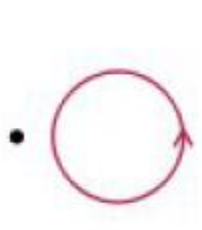



Number of holes in two-dimensional manifolds

# Topology

-2

-1



0

1

2

3

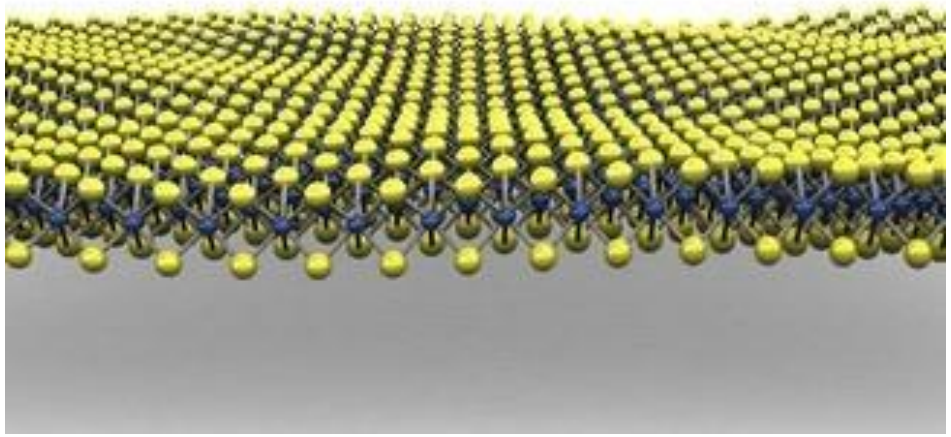
Winding number around ”stick”

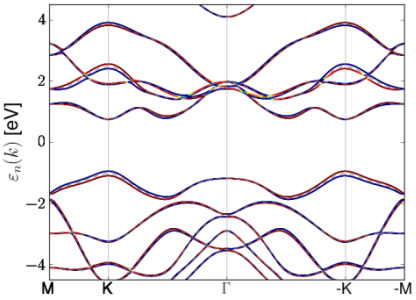
Quantum Hall conductivity: $\sigma_H = n\frac{e^2}{h}$

Winding number of occupied Bloch states around Brillouin zone edge

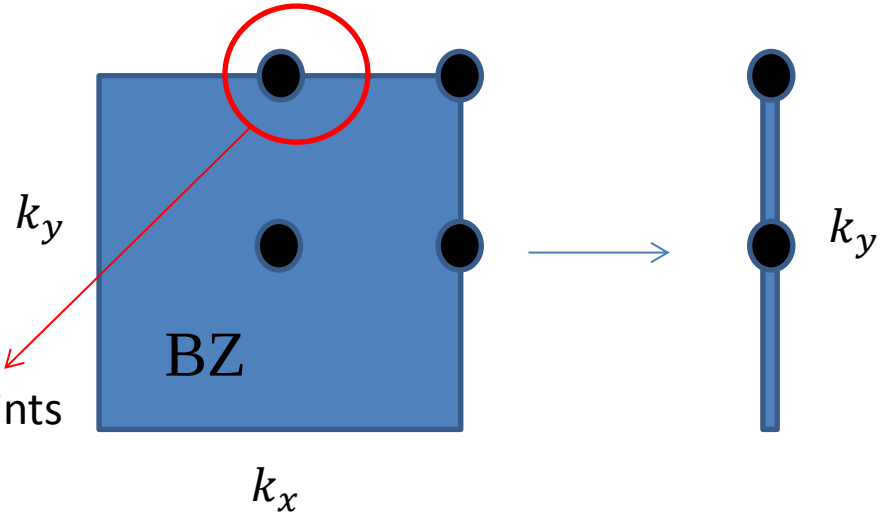
Thouless et al. *Phys. Rev. Lett.* **49**, 405

# Edges of two-dimensional insulator



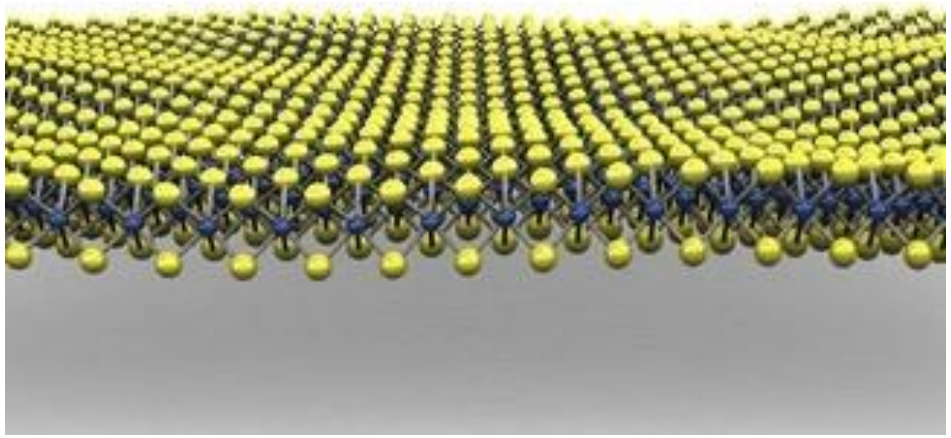


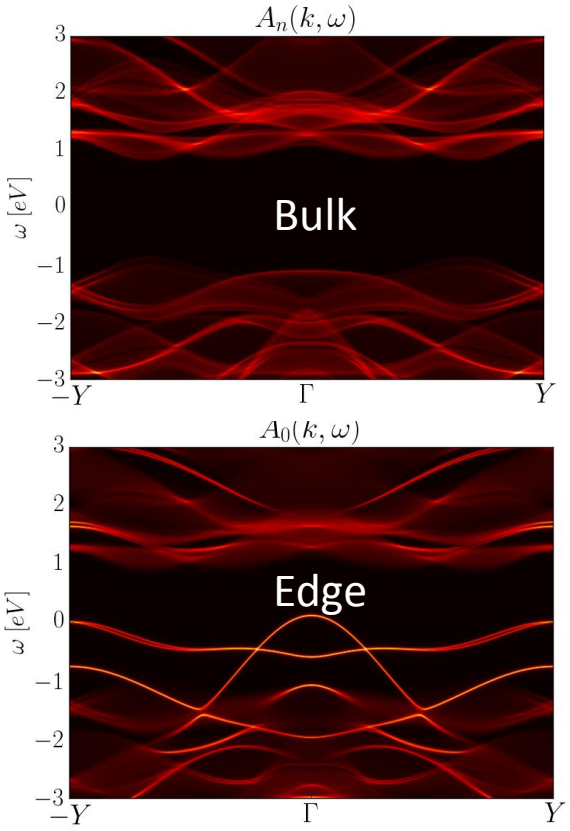

Bulk 2H-$MoS_2$



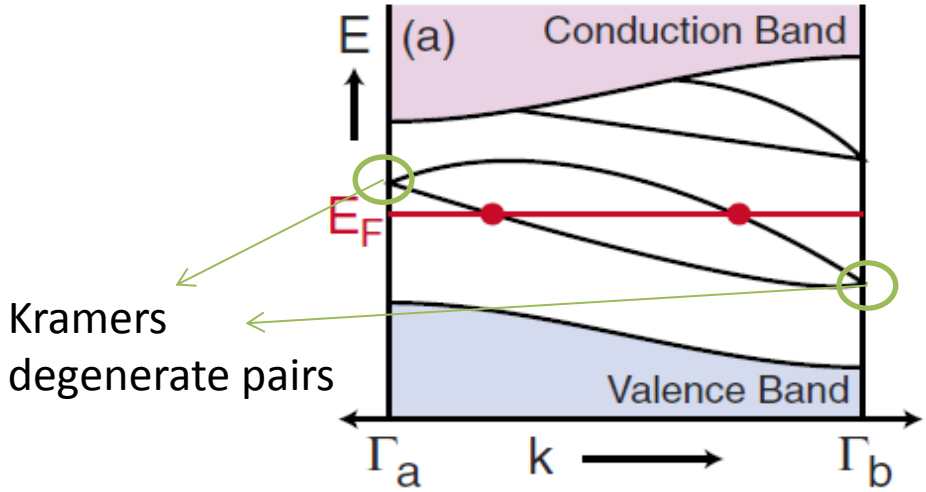

Time-reversal invariant points

# Edges of two-dimensional insulator

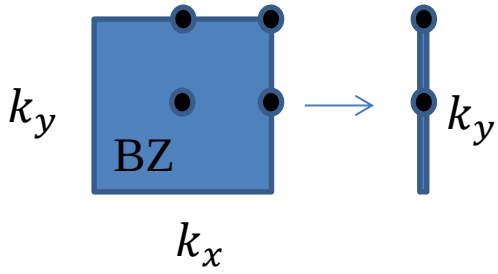




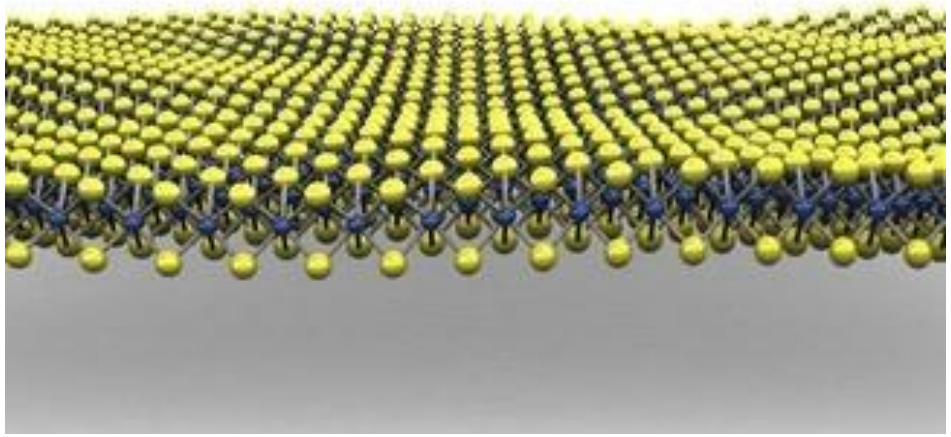

Two things may happen at the edge:

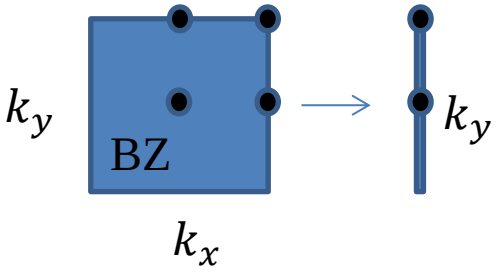



Kramers degenerate pairs

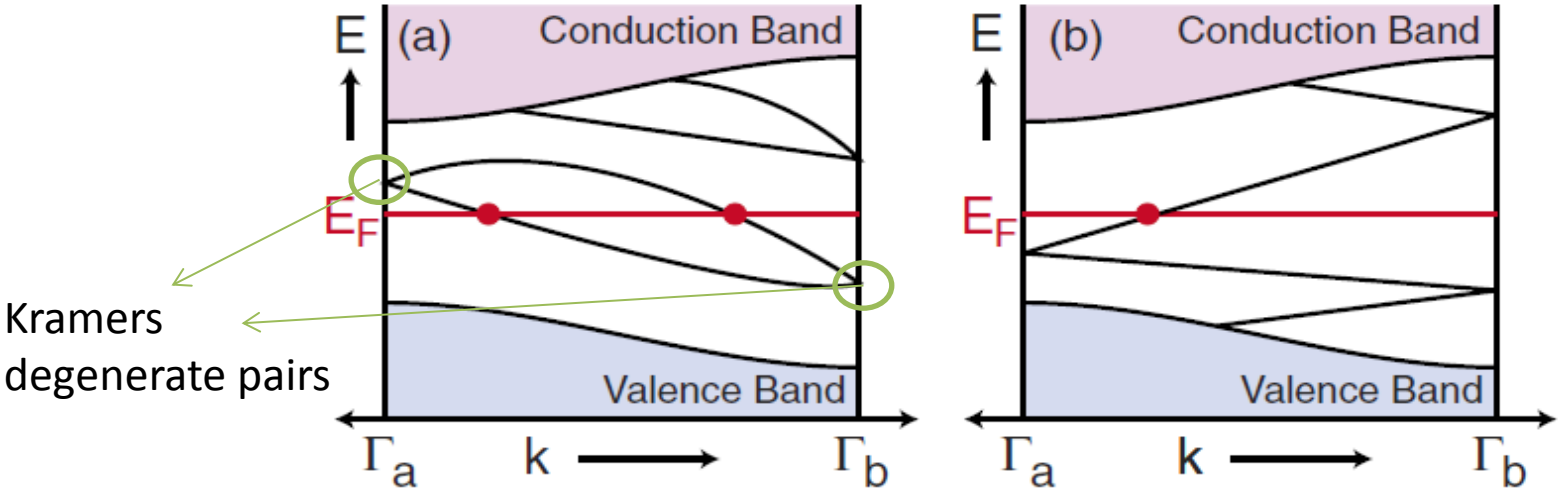


# Edges of two-dimensional insulator



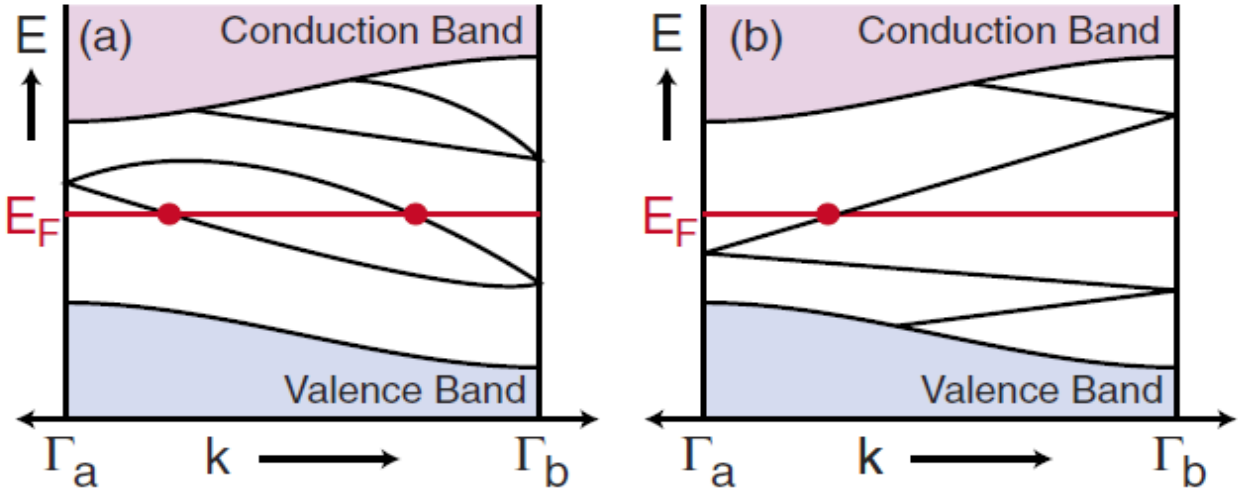



Or:



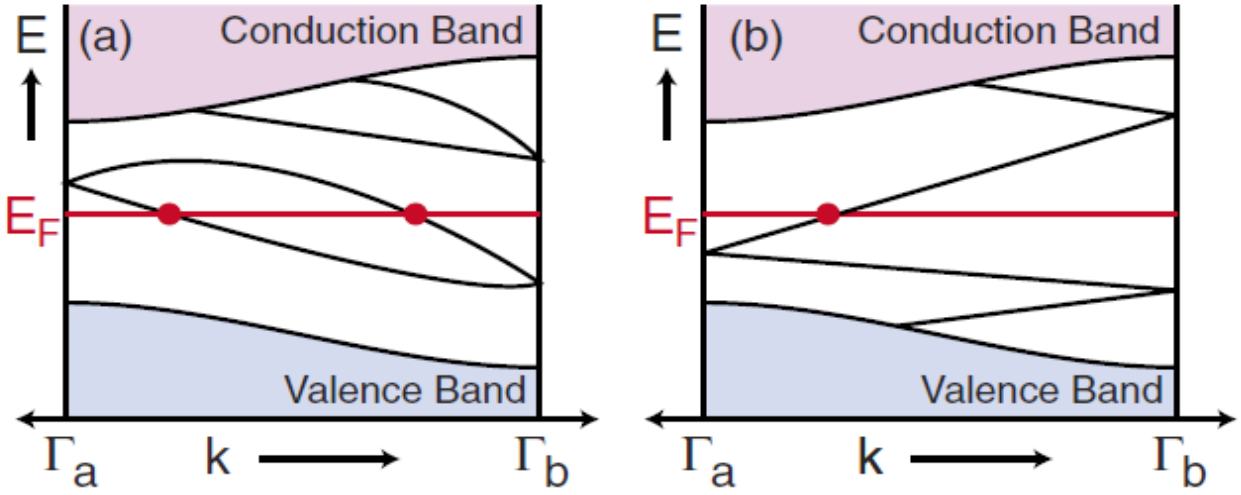

Kramers degenerate pairs

# Edges of two-dimensional insulator




The distinction between odd or even number of crossings at edges is a property of the bulk material and comprises a topological $Z_2$ classification of 2D insulators

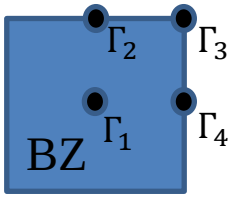
# Edges of two-dimensional insulator




The distinction between odd or even number of crossings at edges is a property of the bulk material and comprises a topological $Z_2$ classification of 2D insulators

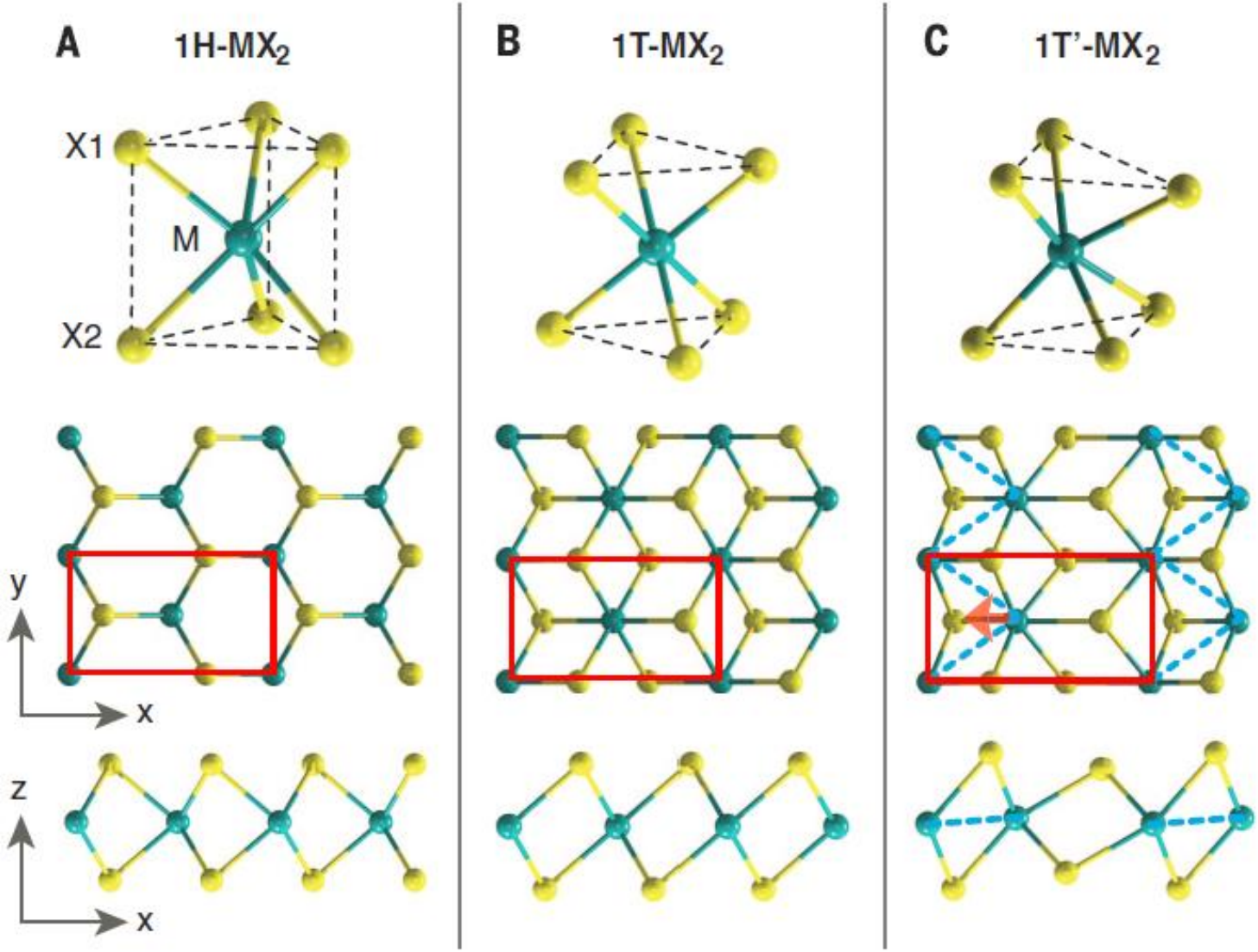
For materials with inversion symmetry the topological index $\nu$ can be calculated as

$$(-1)^{\nu} = \prod_{\Gamma_a} \prod_{m} \xi_m(\Gamma_a)$$



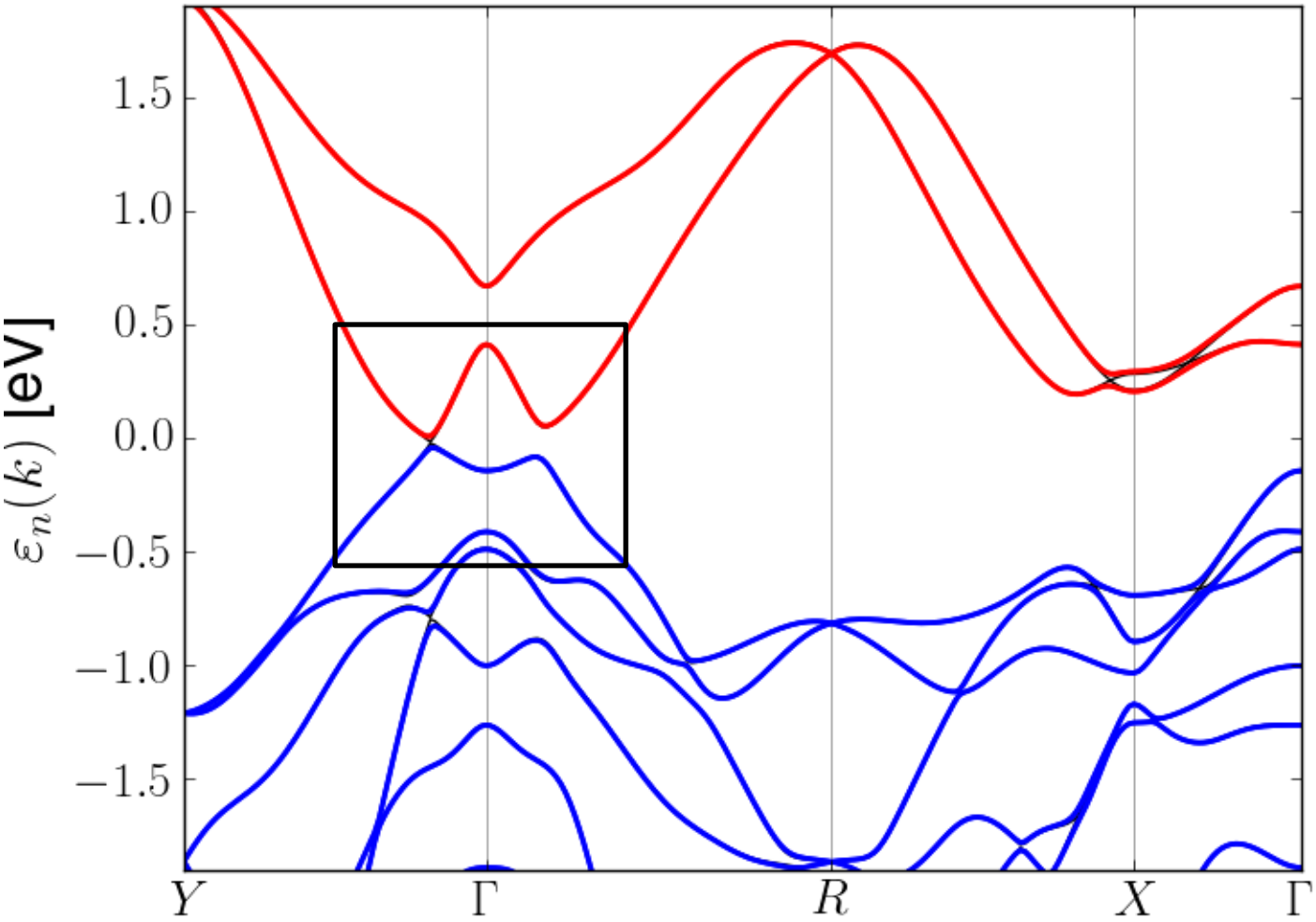

Where the product is over occupied Kramers pairs of inversion eiegnvalues at the TR invariant points – implemented in GPAW in pw mode

# 1T'-$MoS_2$

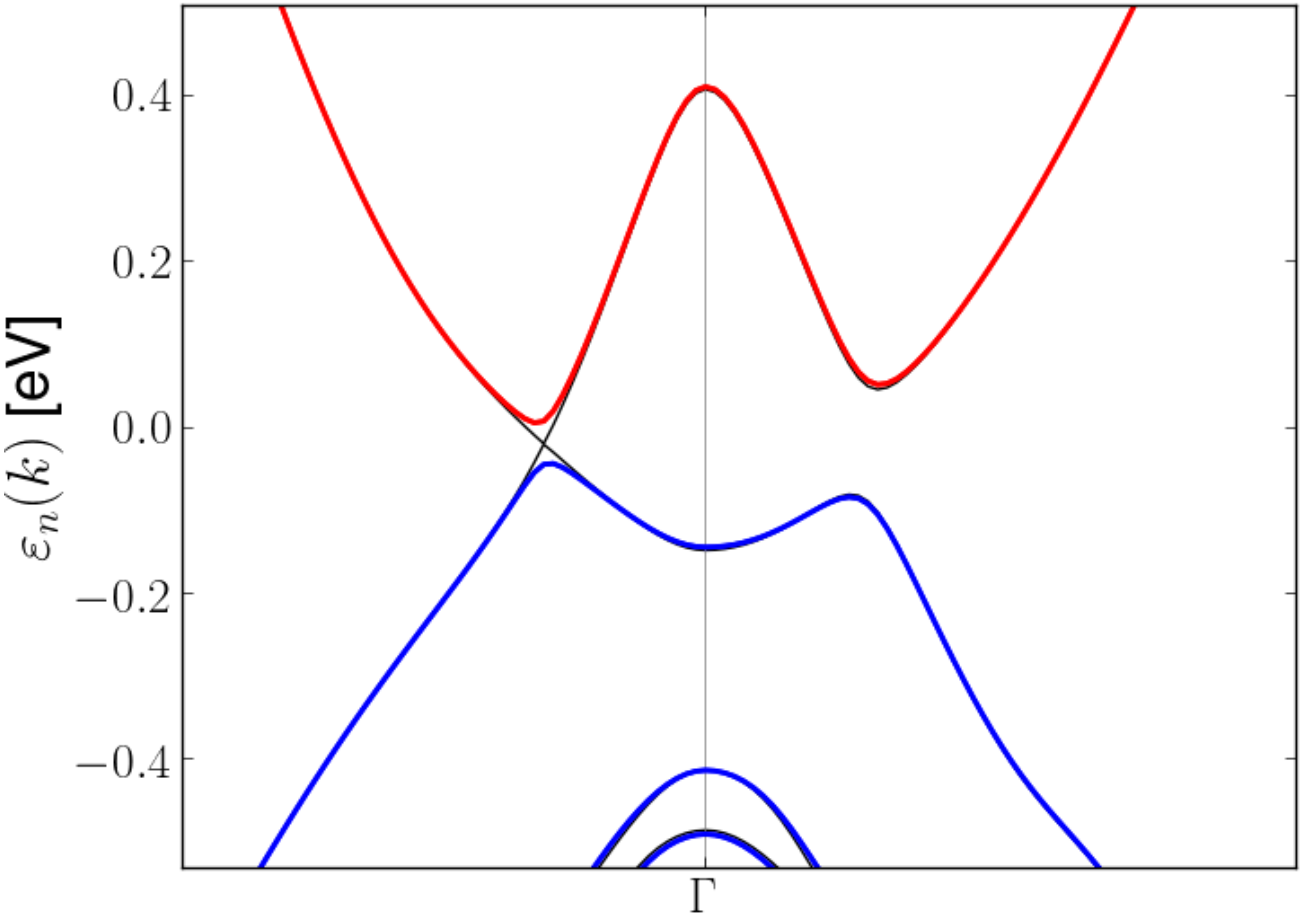



Qian et al. *Science* **346**, 1344 (2014)

# 1T'-$MoS_2$



# 1T'-$MoS_2$




$$\prod_{\Gamma_a} \prod_{m} \xi_m(\Gamma_a) = -1$$

# Edge states

Edge states can be obtained by calculating the spectral function in a local basis set:

$$A_S(k_\parallel, \omega) = -\frac{1}{\pi}\sum_{i \in S} Im\, G^R_{ii}(k_\parallel, \omega\,)$$

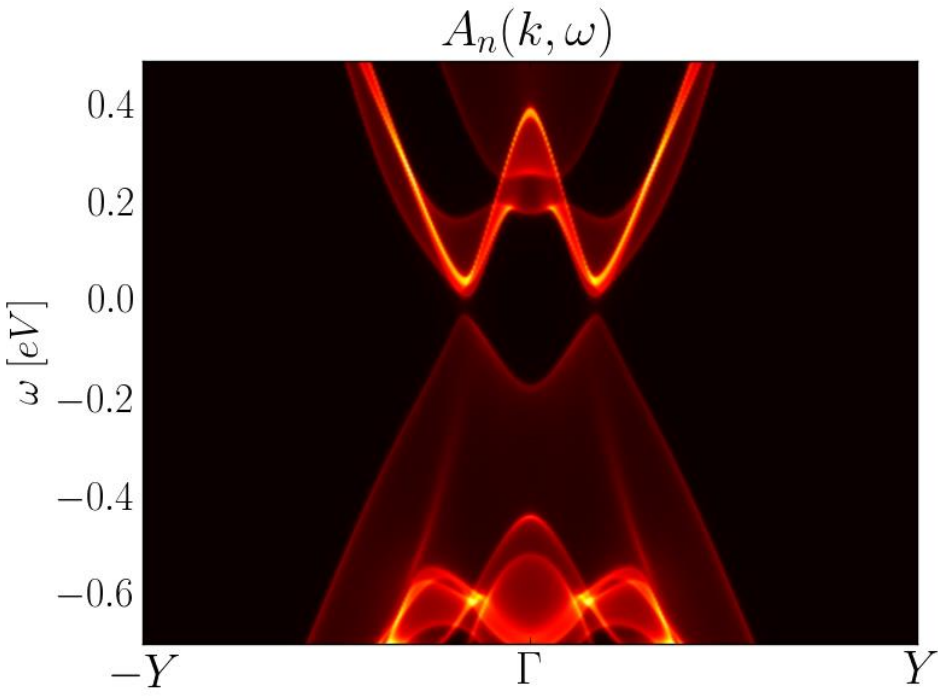
$$G^R_{ij} = (\omega + i\eta - H_{ij})^{-1}$$


The Greens functions can be obtained iteratively once the Hamiltonian is transformed to a local basis

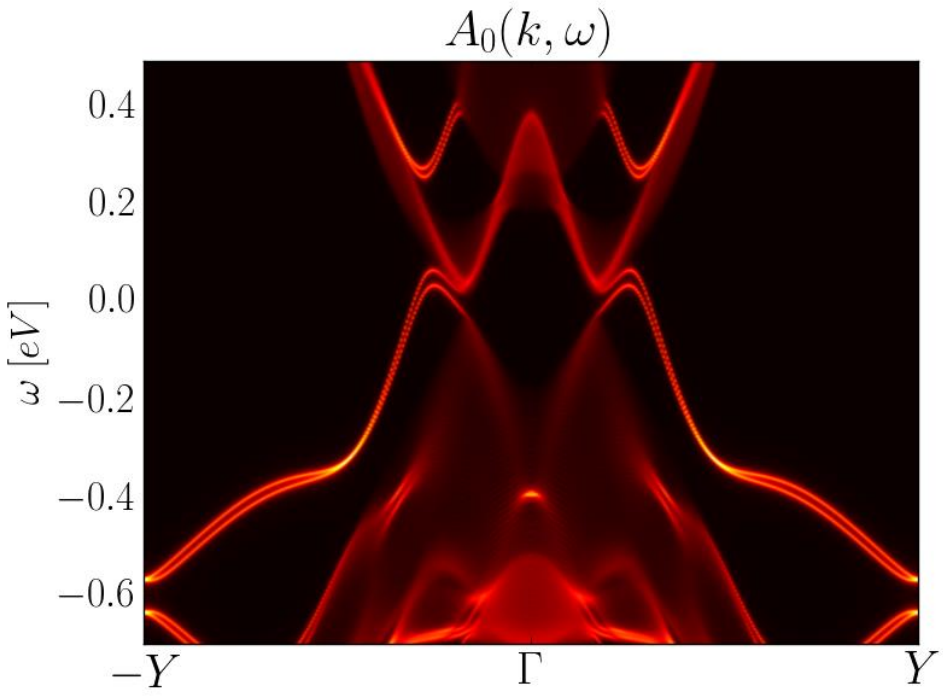
Sancho et al. *J. Phys. F* **15**, 851 (1985)

The transformation is accomplished with Wannier functions using Wannier90.
Interface recently been implemented in GPAW – see tutorial for details

# 1T'-$MoS_2$

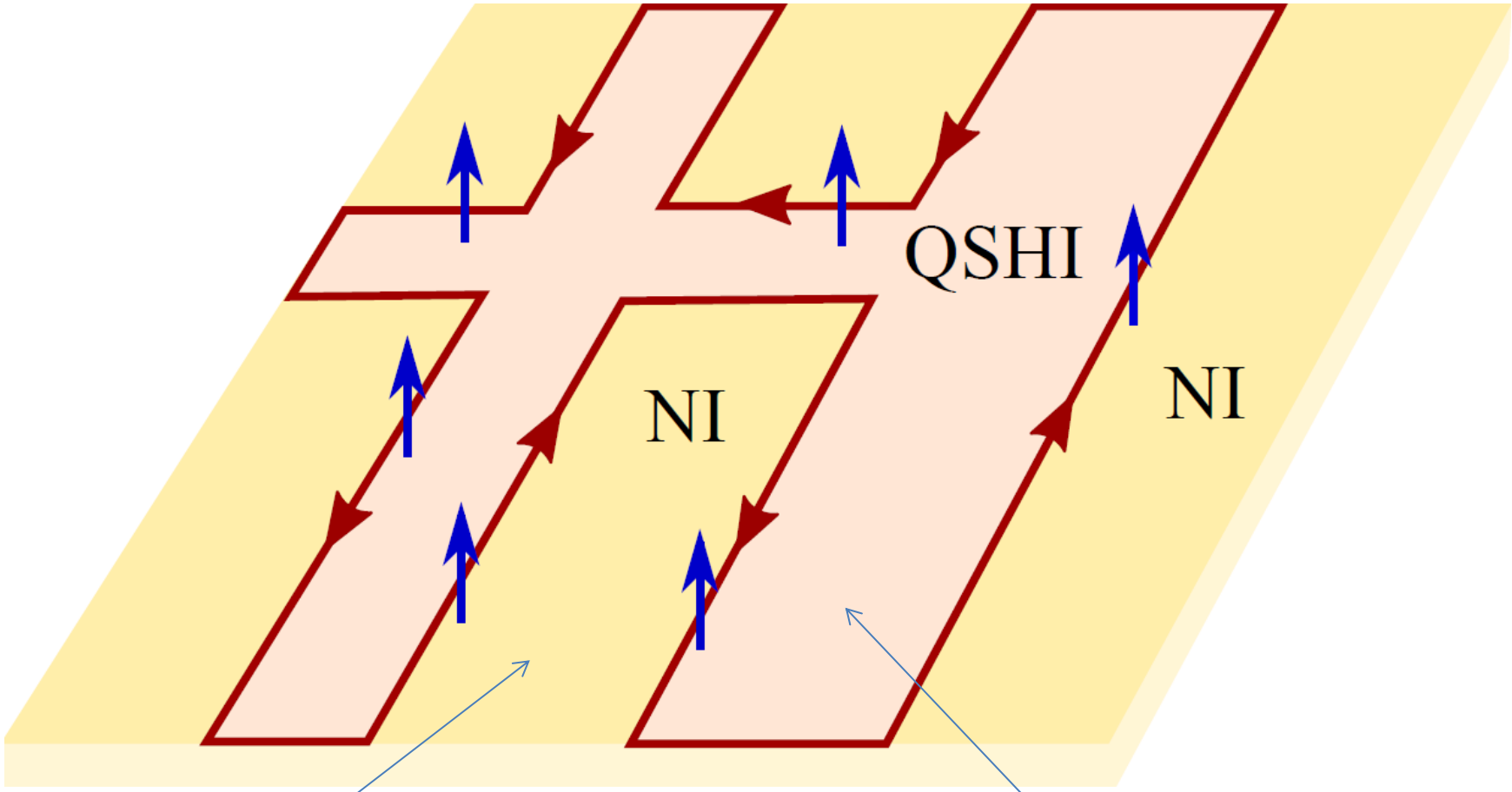



Bulk spectral function

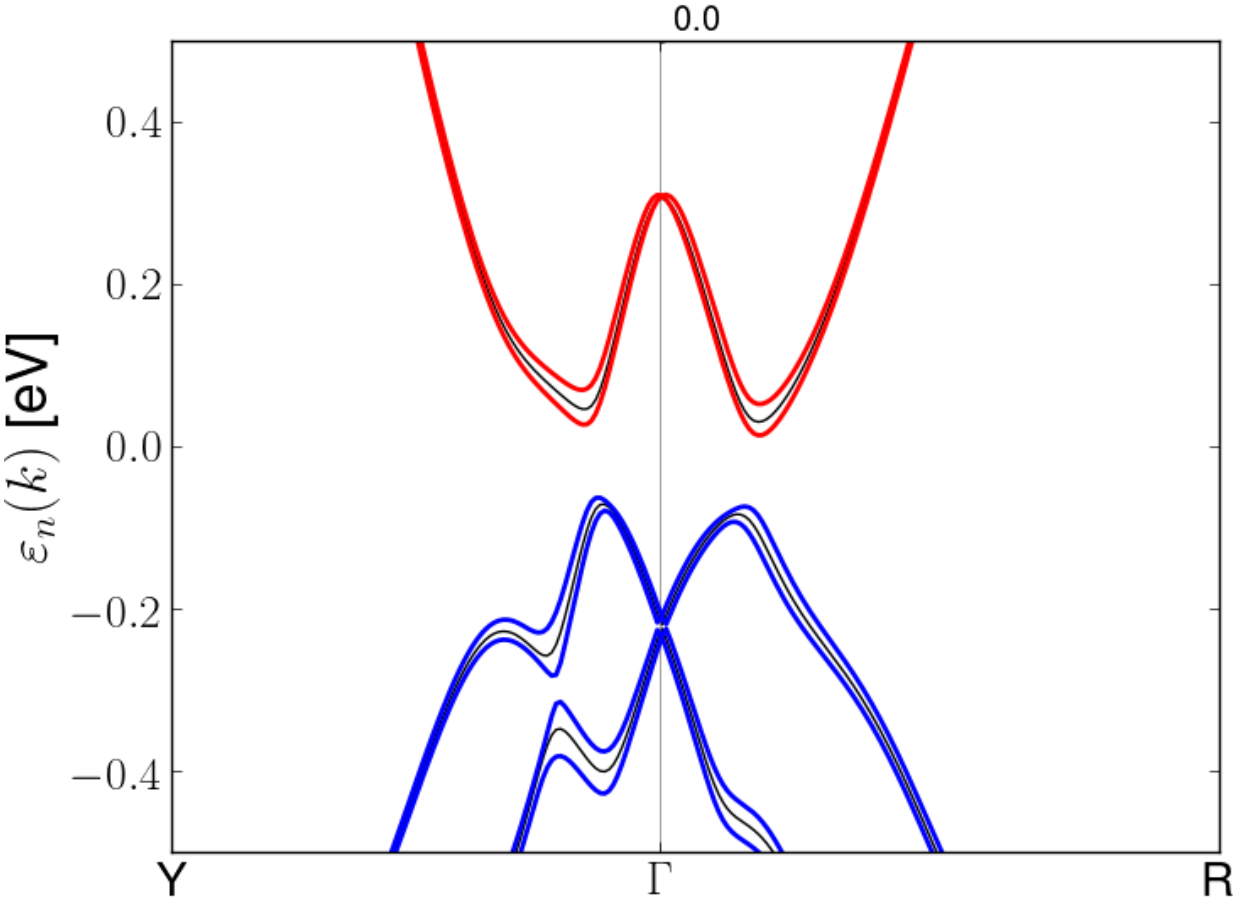



Surface spectral function

# In-plane topological heterostructures

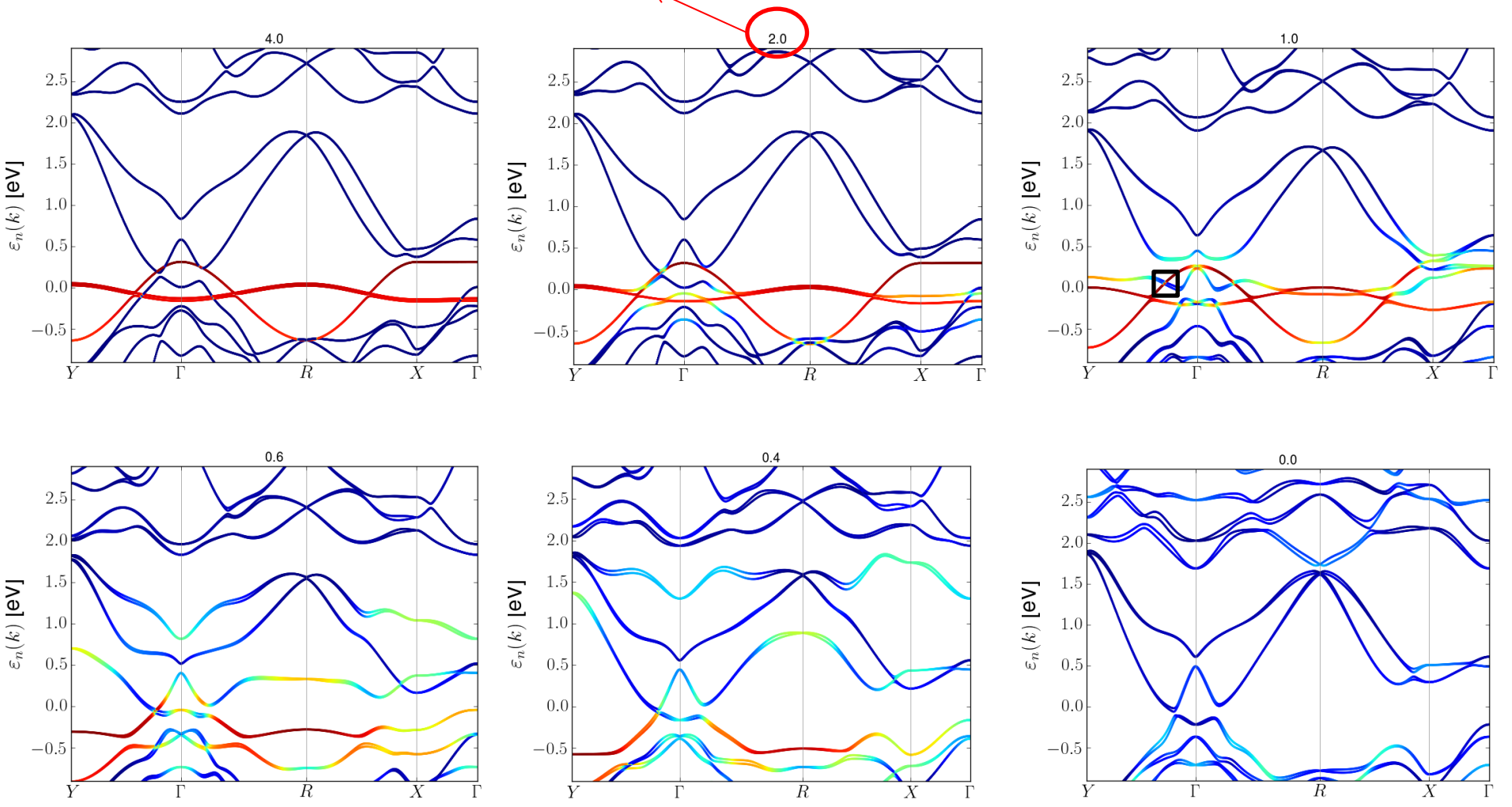


# 1T'-$MoS_2$ – Adsorbed O

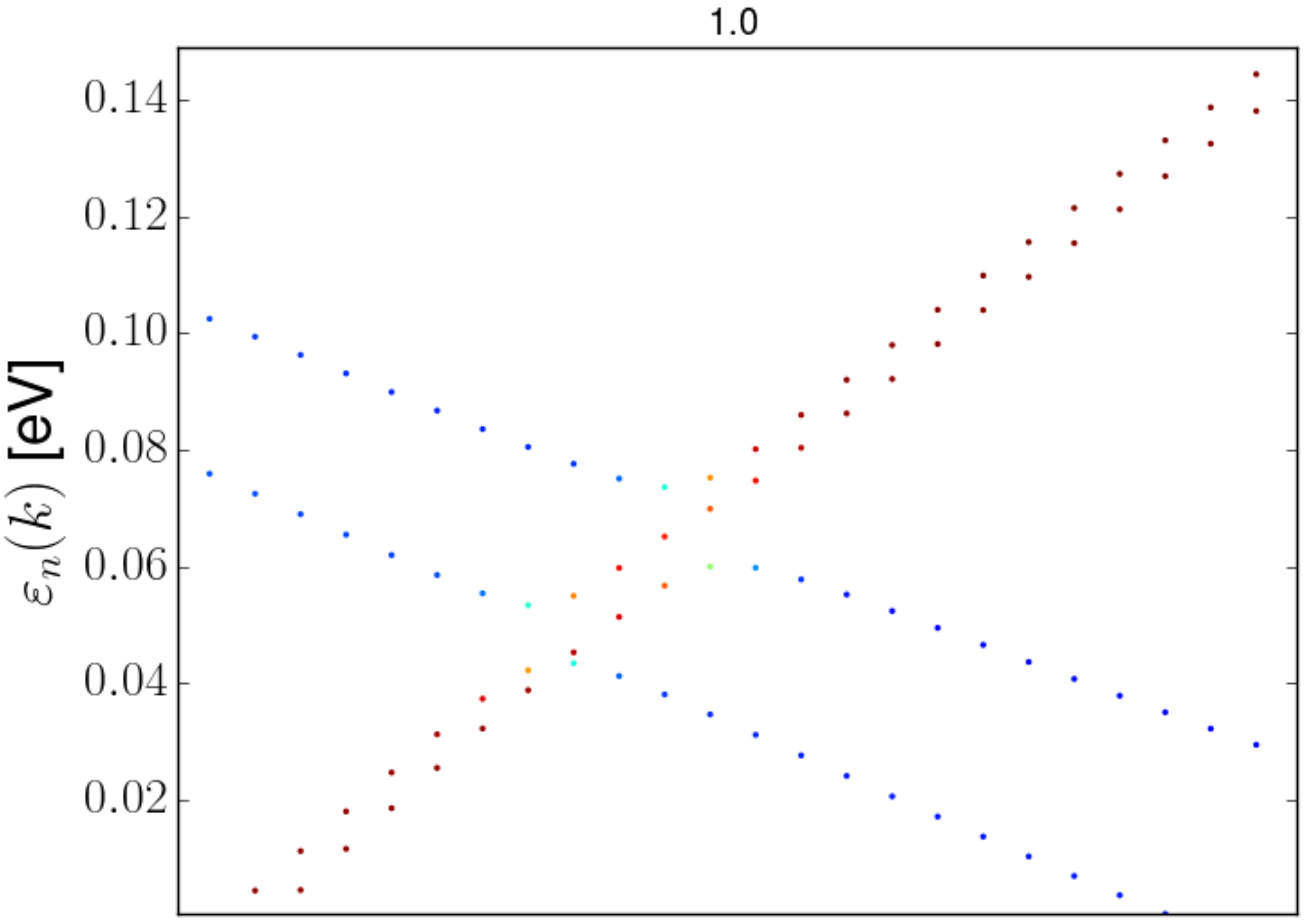


# 1T'-$MoS_2$ – Adsorbing O

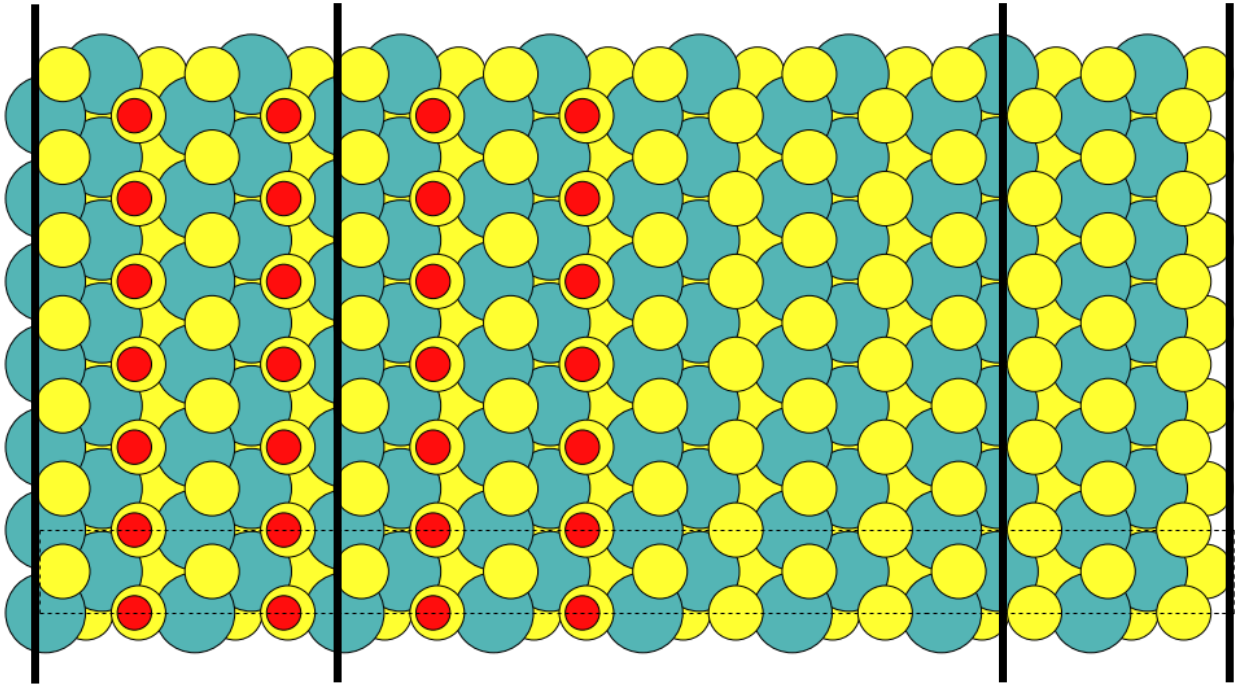
Distance from equilibrium



# 1T'-$MoS_2$ – Adsorbing O



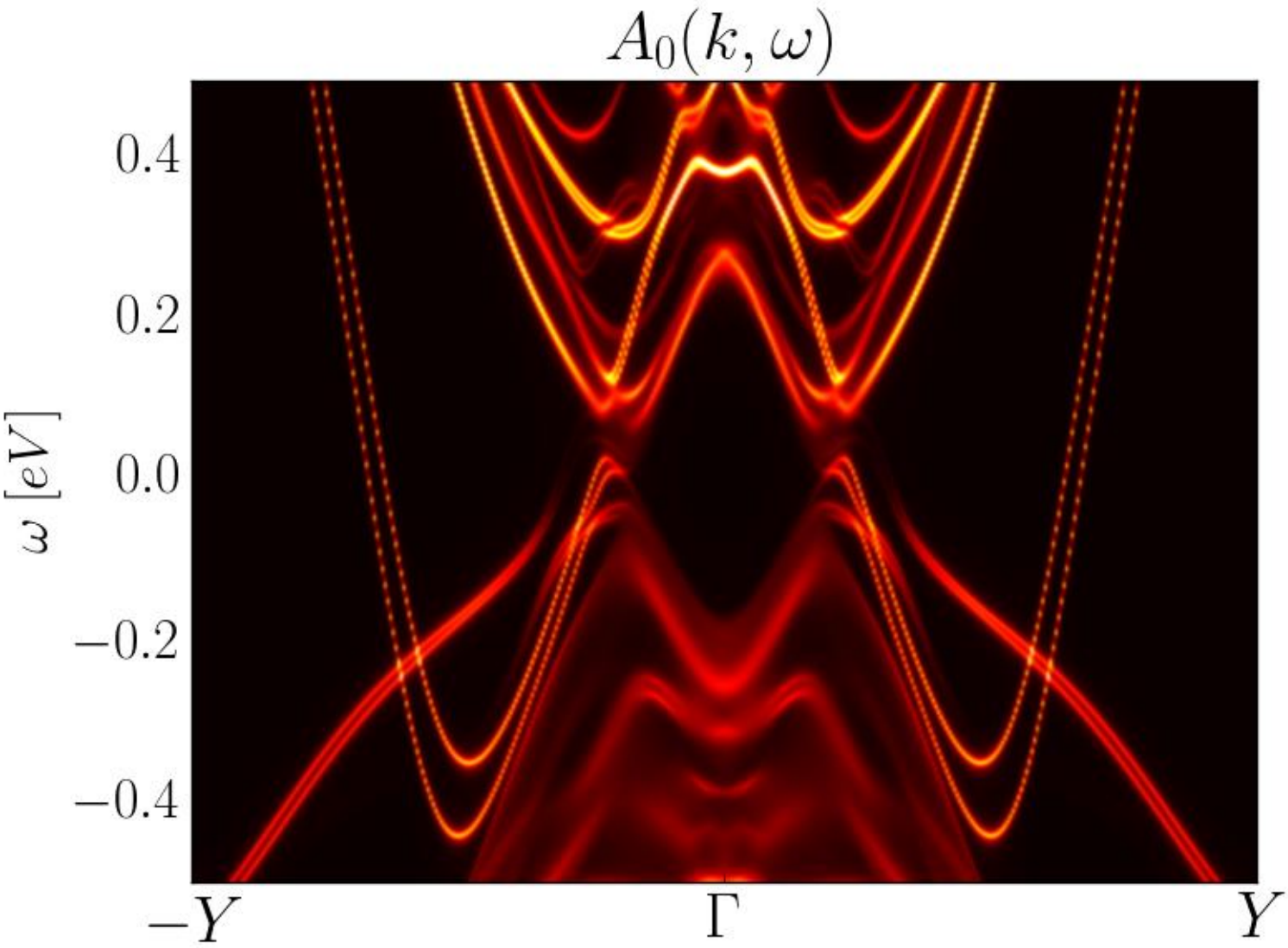
# 1T'-$MoS_2$ – boundary with NI



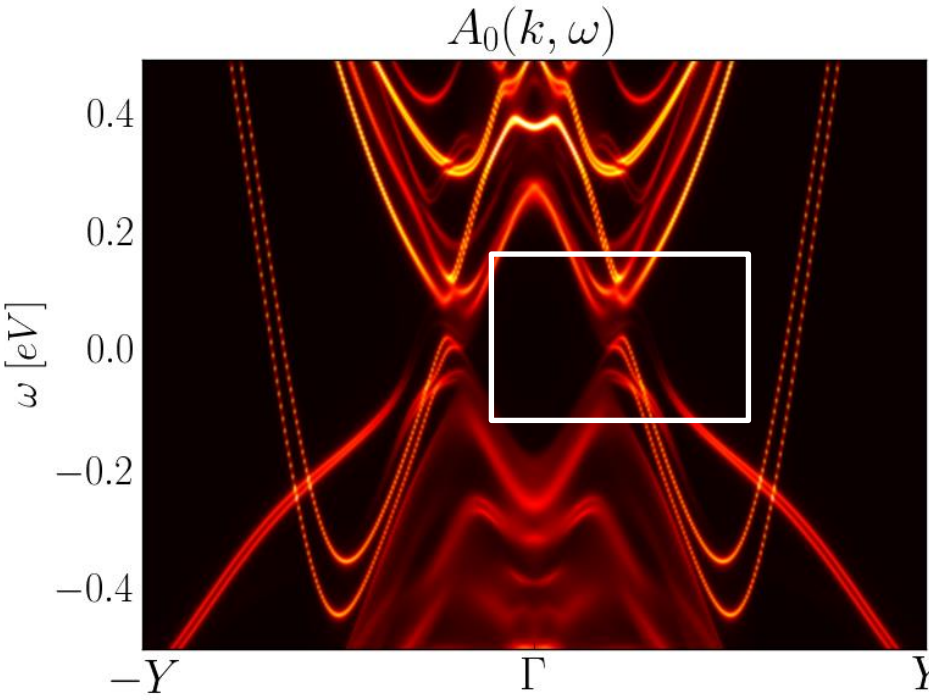
$$\ldots H_{11} \quad H_{11} \quad H_{22} \quad H_{33} \quad H_{33}\ldots$$
$$\ldots H_{01} \quad H_{01} \quad H_{12} \quad H_{23} \quad H_{34} \quad H_{34}\ldots$$

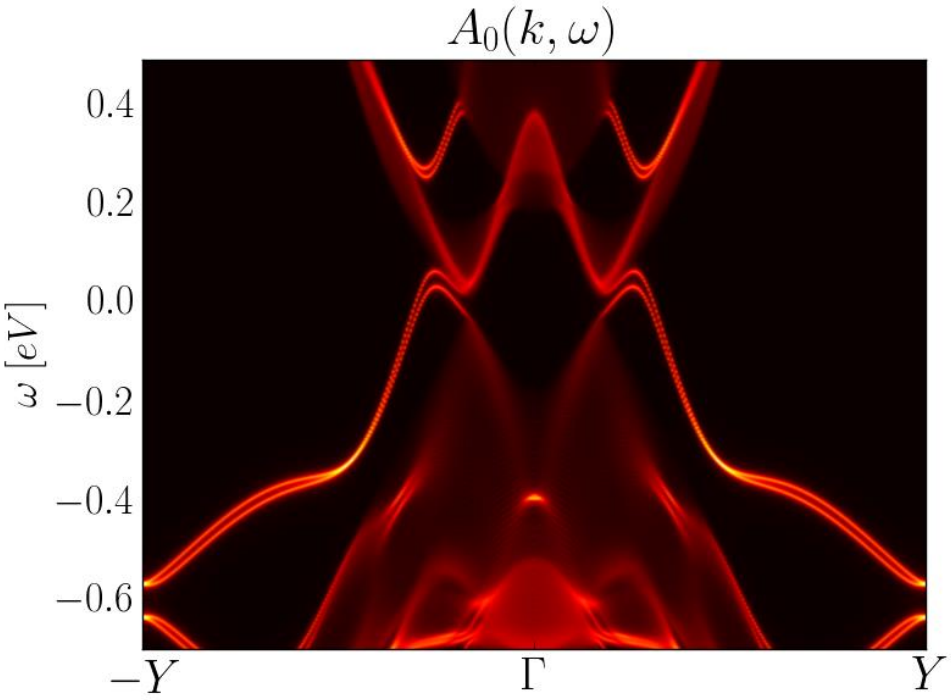
Retarded Greens function at interface can be obtained iteratively

# 1T'-$MoS_2$ – partly adsorbed O



# 1T'-$MoS_2$ – partly adsorbed O

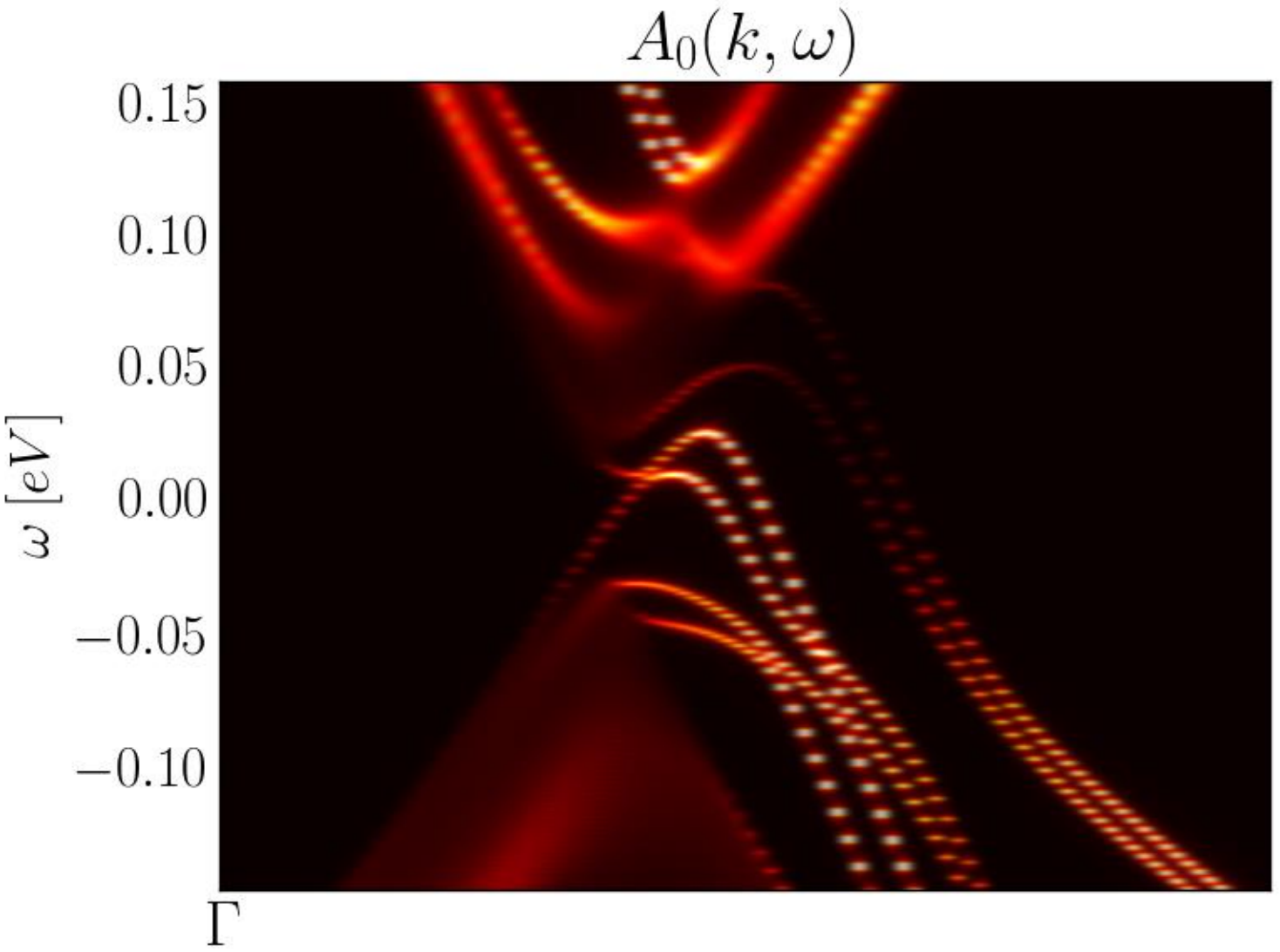



Interface with oxygen adsorbate region

Bare edge

# 1T'-$MoS_2$ – partly adsorbed O



# Outlook

- Topological in-plane heterostructures could comprise a clever way of designing 1D electronics
- Many other adsorbates are possible
- The topological boundary states needs to be analyzed further. What about other edges?
- The structure is hardly thermodynamically stable!

Thank you for the attention