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
The Dirac equation is

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

where $\alpha$ and $\beta$ 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 $\eta$ and expanding to second order in $(E - V)/mc^2$ yields

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

where $\xi$ is a two-component spinor

where spherical symmetry

$$- \frac{S \cdot L}{2m^2c^2} \frac{1}{r} \frac{dV}{dr}$$
Implementation of SO in GPAW

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

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

$$|\psi_{n\sigma}\rangle = \sum_{ai} \langle \tilde{p}_i^a | \tilde{\psi}_n \rangle |\phi_i^a \sigma\rangle$$

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

$$H_{n_1n_2\sigma_1\sigma_2} = \delta_{\sigma_1\sigma_2} \delta_{n_1n_2} \varepsilon_{n_1\sigma_1} + \langle \psi_{n_1\sigma_1} | H^{SO} | \psi_{n_2\sigma_2} \rangle$$

$$\langle \psi_{n_1\sigma_1} | H^{SO} | \psi_{n_2\sigma_2} \rangle = \sum_{ai_1i_2} \langle \tilde{\psi}_{n_1} | \tilde{p}_{i_1}^a \rangle \langle \phi_{i_1}^a \sigma_1 | H^{SO} | \phi_{i_2}^a \sigma_2 \rangle \langle \tilde{p}_{i_2}^a | \tilde{\psi}_{n_2} \rangle$$
Implementation of SO in GPAW

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

\[ \phi^a_i(r) = f_i(r)Y_i(\phi, \theta) \]

We thus have to evaluate

\[ \langle \phi^a_i \sigma_1 | H^{SO} | \phi^a_j \sigma_2 \rangle = \langle f^a_i Y^a_i \sigma_1 | H^{SO} | f^a_j Y^a_j \sigma_2 \rangle \]

\[ = - \frac{1}{2m^2c^2} \langle Y^a_i \sigma_1 | \mathbf{S} \cdot \mathbf{L} | Y^a_j \sigma_2 \rangle \langle f^a_i | \frac{1}{r} \frac{dV}{dr} | f^a_j \rangle \]

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

\[ \langle p_i | L_x | p_j \rangle = \begin{bmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad i, j \in \{y, z, x\} \]
Spin degeneracy is protected by inversion symmetry
The double degenerate $d$ states carry a two-dimensional irreducible representation of $C_{4v}$.

Symmetry is reduced to $C_4$ by spin-orbit coupling.
Color scale denotes spin character: \( S_n(k) = \langle \psi_{kn} | \sigma_z | \psi_{kn} \rangle \)
The band structure depends on the direction relative to the spin projection
Bulk MoS$_2$ is composed of stacked two-dimensional sheets bound by van der Waals interactions.

The individual sheets have a honeycomb lattice similar to graphene or hBN.

Brillouin zone
Monolayer MoS$_2$

Lack of inversion symmetry

\[ \tilde{\epsilon}_n(k) \text{ [eV]} \]

Color scale denotes spin character:

\[ S_n(k) = \langle \psi_{kn} | \sigma_z | \psi_{kn} \rangle \]

\[ \tilde{\epsilon}_n(k) = 0.149 \text{ eV} \]
Topology

Number of holes in two-dimensional manifolds
Topology

Winding number around "stick"

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

Winding number of occupied Bloch states around Brillouin zone edge

Edges of two-dimensional insulator

Bulk 2H-MoS$_2$

Time-reversal invariant points

$k_x$

$k_y$

BZ
Edges of two-dimensional insulator

Two things may happen at the edge:

- Kramers degenerate pairs

![Diagram showing the edges of a two-dimensional insulator with band structures and k-space representation.](image)
Edges of two-dimensional insulator

Or:

Kramers degenerate pairs
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

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

Where the product is over occupied Kramers pairs of inversion eigenvalues at the TR invariant points — implemented in GPAW in pw mode.
1T’-MoS$_2$
$1T'$-MoS$_2$

\[ \prod_{\Gamma_a \Gamma_m} \xi_m(\Gamma_a) = -1 \]
Edge states

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

\[ A_S(k_\parallel, \omega) = -\frac{1}{\pi} \sum_{i \in S} \text{Im} \ G_{ii}^R (k_\parallel, \omega) \]

\[ G_{ij}^R = (\omega + i\eta - H_{ij})^{-1} \]

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


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

Topological Insulator (Quantum Spin Hall Insulator)

Normal Insulator

Topological Insulator (Quantum Spin Hall Insulator)
1T'-MoS$_2$ – Adsorbed O
$1T'$-MoS$_2$ — Adsorbing O

Distance from equilibrium
1T’-MoS$_2$ — Adsorbing O
Retarded Greens function at interface can be obtained iteratively
1T’-MoS$_2$—partly adsorbed O

$A_0(k, \omega)$
1T’-MoS$_2$—partly adsorbed O

Interface with oxygen adsorbate region

Bare edge
$1T'$-MoS$_2$—partly adsorbed O

$A_0(k, \omega)$

$\omega$ [eV]

$\Gamma$
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