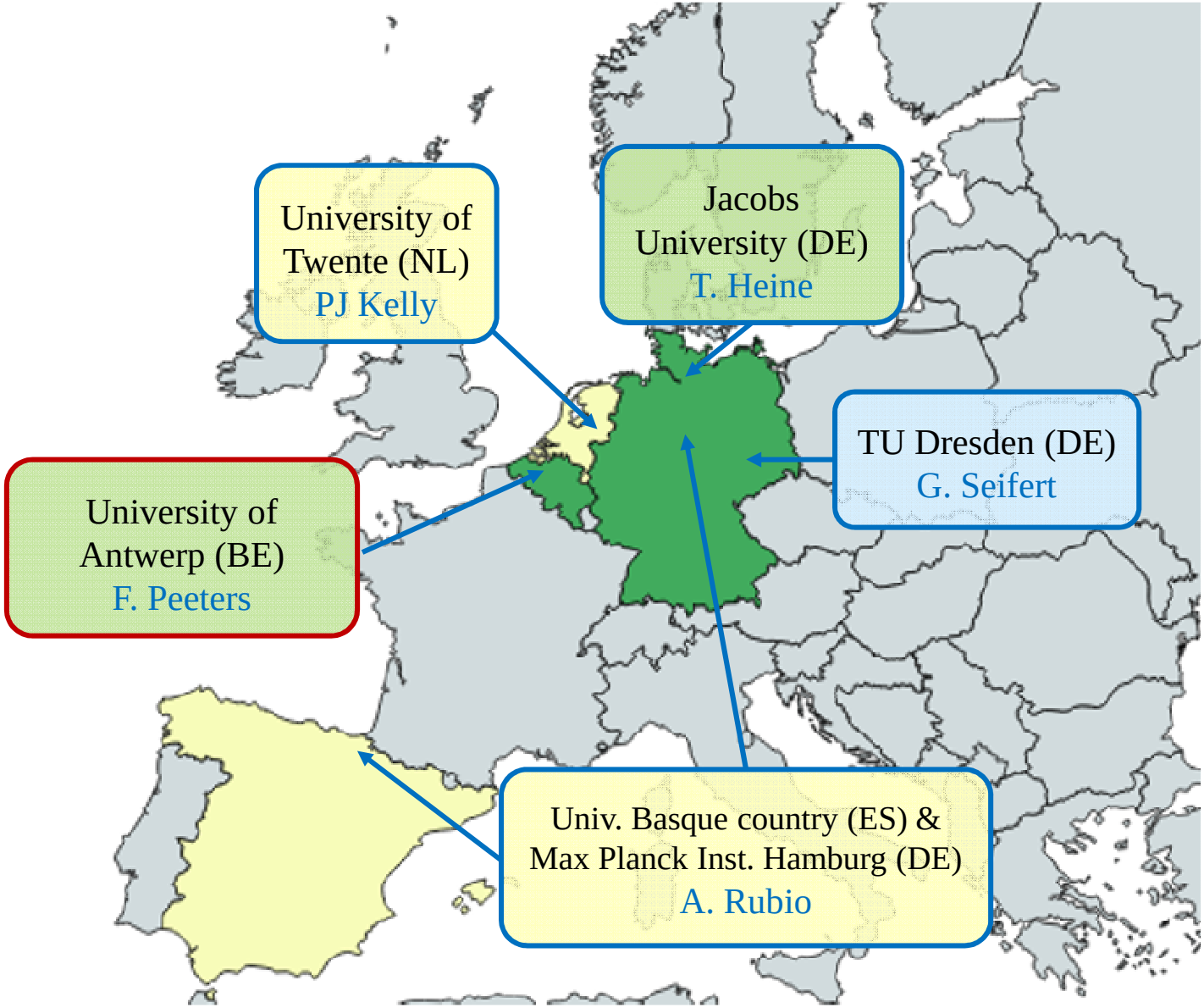
Theoretical investigation of electronic transport in functionalized 2D transition metal dichalcogenides

Trans2DTMD

Trans2DTMD network

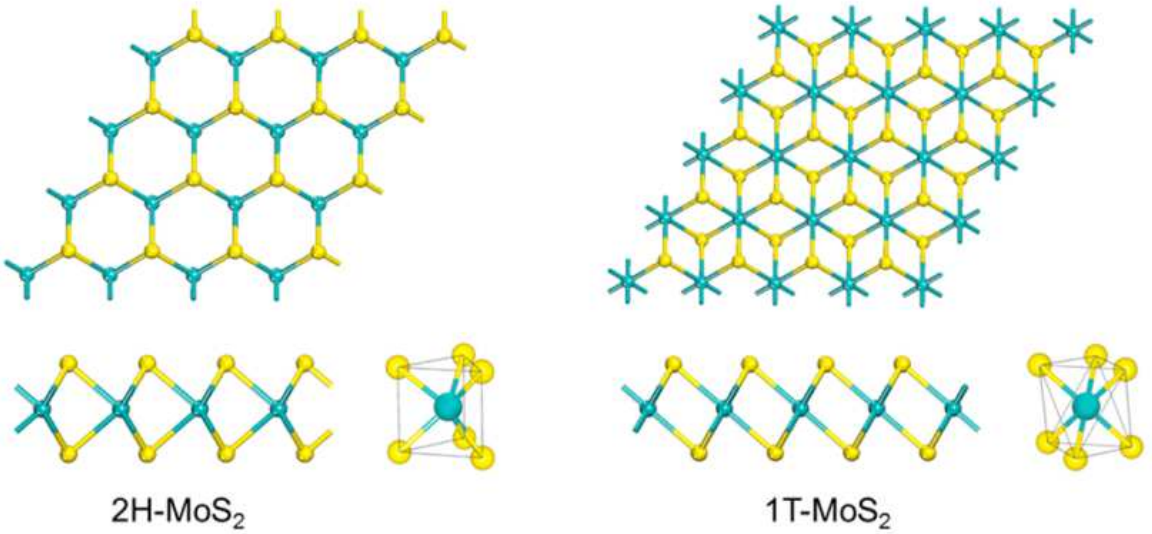


Legend

- Coordinator
- Funded
- Associate
- Flagship

Scientific background

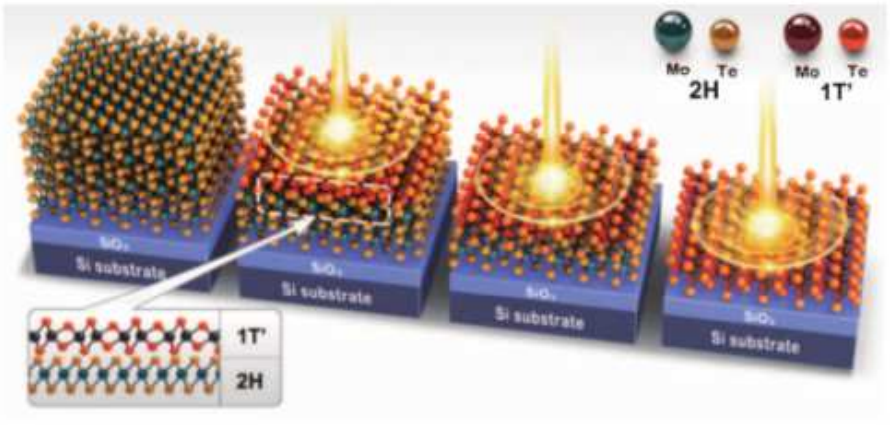
- 2D Transition Metal Dichalcogenides as alternative to graphene
- MX_2 (M = Mo, W; X = S, Se) can be semiconducting (2H) or metallic (1T - metastable)

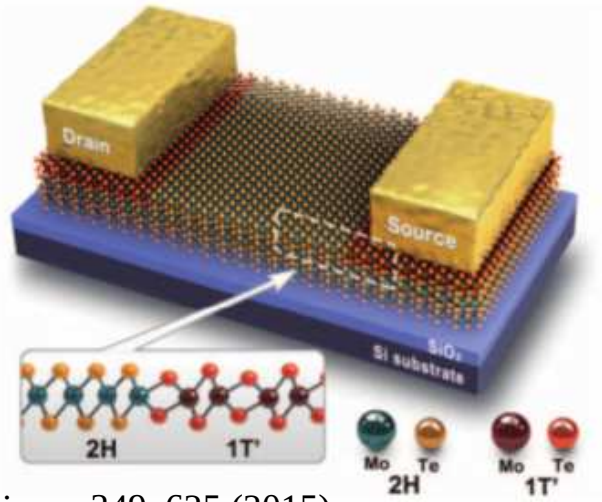


- Some TMDs are intrinsically metallic ($NbSe_2$, TiS_2, etc.)

Scientific background

- How can we stabilize or induce the metallic phases?
  - Topological modifications (at edges of nanoribbons and grain boundaries)
  - Substitutional and defect doping (i.e. replacing metal atoms, substituting chalcogen atoms, vacancy defects)
- Will be used as low resistance contacts to semiconducting TMDs: excellent interface

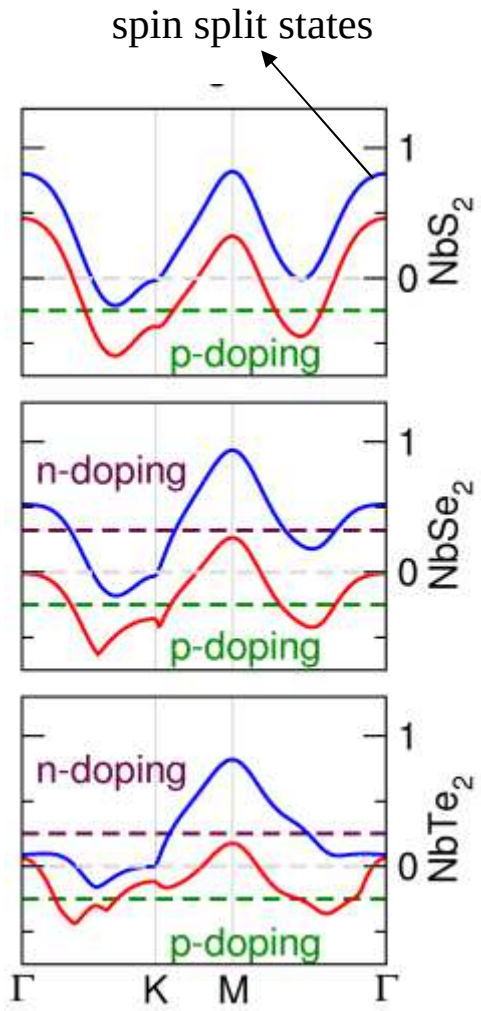




Cho, S et al. Phase patterning for ohmic homojunction contact in $MoTe_2$. Science 349, 625 (2015).

Scientific background

- (Metallic) TMDs are relevant to spintronics applications due to:
  - Enhanced spin-orbit coupling (spin-polarized currents)
  - Appearance of spin quantum Hall effect, spin polarized topological edge states
  - Doping induced ferromagnetism (rich physics due to spin-valley coupling)



Project Objectives

1. Investigate electronic properties and stability metallic TMDs (intrinsic, by doping or by inducing topological surface/interface states).

2. Develop KPM-based O(N) numerical methods for:
   - the conductivity tensor (longitudinal/transverse, spin and valley)
   - non-equilibrium Green's functions approaches

3. Describe electrical and spin-polarized transport through:
   - magnetically doped TMDs,
   - ferromagnetic alloy/graphene interfaces in the presence of spin-orbit couplings.

4. Propose and describe new devices/contact geometries based on TMDs

Work Packages

WP 1. Numerical methods based on Chebyshev polynomial expansions

WP 2. Intrinsic properties of metallic TMDs

WP 3. Controlling metallic phases of intrinsically semiconducting TMDs

WP 4. Spin-polarized current in metallic TMDs

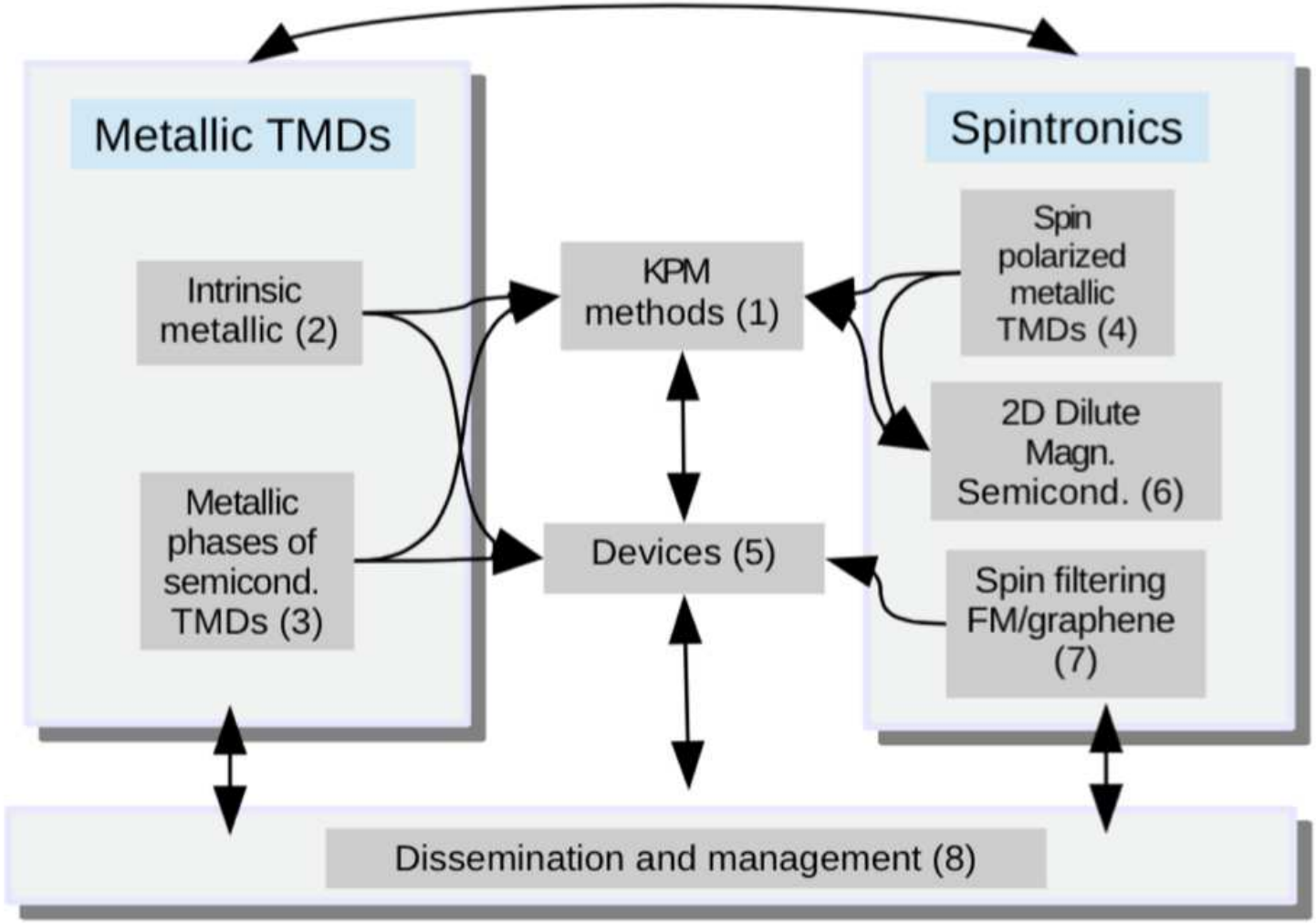
WP 5. Device simulations

WP 6. Proposals for making a 2D diluted magnetic semiconductor

WP 7. Spin filtering with FM alloy| (h-BN) | graphene interfaces

WP 8. Dissemination and management

Work Plan


Metallic TMDs
Intrinsic metallic (2)
Metallic phases of semicond. TMDs (3)
KPM methods (1)
Devices (5)
Spintronics
Spin polarized metallic TMDs (4)
2D Dilute Magn. Semicond. (6)
Spin filtering FM/graphene (7)
Dissemination and management (8)

UAntwerp (F. Peeters)

The Condensed Matter Theory group at UAntwerp:

- theoretical and computational tools for novel meso- and nano-scale materials
- over the last 10 years a lot of effort was devoted to the description of low dimensional materials, like semiconducting and superconducting nanowires, or two-dimensional graphene or TMDs.
- wide variety of numerical approaches: from tight-binding description of model Hamiltonians to pure numerical DFT approaches

Role in project:

- Project coordinator, performs management and monitors dissemination activities
- Development of KPM methods for the linear response conductivity and NEGF methods
- DFT investigations on the stability of the proposed materials using the VASP, SIESTA and QUANTUM ESPRESSO codes
- Investigation of possible structural phase transitions due to doping
- Transport calculations and device modelling using DFT based codes and in-house developed NEGF codes for model Hamiltonians or by using available Transiesta DFT codes

JacobsUni (T. Heine)

The Heine group:

- has worked on transition metal dichalcogenides since about 10 years with many high-impact contributions on TMD nanotubes, layered materials and nanoflakes.
- development of methods and software for materials science, molecular framework compounds, 2D inorganic materials and theoretical spectroscopy.

Role in project:

- Calculation of stabilization of structurally metallic phases by external factors (DFT as in BAND and QUANTUM ESPRESSO codes)
- Study of metallic phases obtained by doping semiconducting TMDs (DFTB and DFTB-NEGF as in ADF/BAND)
- Development of DFTB parameters for calculation of structure, electronic structure and transport in doped TMDs
- Calculation of spin-orbit interactions in metallic TMD states and – if promising – spin- polarized transport calculations

UTwente (PJ Kelly)

The Computational Materials Science group has expertise in:

- first- principles electronic structure theory
- developing algorithms and applying them to a wide variety of condensed matter situations including magnetic materials, which form the basis for magneto-electronics.

Role in project:

- Explore the possibility of making TMDs itinerant ferromagnets by doping (DFT with VASP)
- Calculation of exchange coupling constants and Curie temperatures using Monte Carlo code (to be developed)
- Study the effect of alloy disorder on the "perfect" spin filtering predicted for Ni(Co)|graphene (TB-MTO scattering code)
- Study the deleterious effect of spin-orbit coupling on perfect spin filtering (TB-MTO scattering code)

TUDresden (G. Seifert)

The group of Prof. Gotthard Seifert has:

- extensive experience in application and development of density-functional (DFT) based methods
- well-known expertise in the following fields: electronic, optical and transport properties of nanostructures, especially in 2D carbon-based and inorganic (e.g. MoS_2) structures.

Role in project:

- Development and application of the DFTB method for large-scale atomistic transport calculations as they are planned in the project.
- Support the activities in calculation of transport properties of semiconducting and metallic 2D TMDs and stacks of such materials.
- The group will assist all partners with its experience in the theory and applications of 2D materials.

U Basque Country (A. Rubio)

The group of Prof. Angel Rubio has:

- pioneered new developments within many-body theory and Time Dependent DFT,
- improvements on transport theory, and the theoretical basis of spectroscopic techniques
- studied the electronic response of solids and nanostructures to external electromagnetic fields.
- devoted a lot of efforts in the characterization of new 2D materials (germanene, stanene and silicene) as well as others as BN, graphene and TMDs.

Role in project:

- Provide expertise on the description of topological states
- Provide guidance for the use of the OCTOPUS code in order to perform complementary TDDFT calculations for the proposed structures. This will be the basis to foreseeable continuation of the project, by including the effect of electron-electron interactions, which in 2D materials are expected to be important.

Interactions with the Flagship

High impact applications related to Flagship Work Packages:

- Metallic TMDs offer quasi zero-resistance electrodes for 2D transistors. Relates to device-based FWP 1,5,7 and 8; contacts with van der Zant (TU Delft) and A. Ferrari (Cambridge)
- TMDs have strong spin-orbit coupling and can be tailored to produce spin-polarized currents. Relates to FWP 6 (Spintronics); collaboration with S. Roche (Barcelona)
- Ferromagnetic MX_2 monolayers are 2D ferromagnetic semiconductors. This relates to FWP 6 (Spintronics) and collaborations with the TCD (S. Sanvito, V. Nicolosi, J. Coleman) and EPFL (A. Kis) already exist
- Novel O(N) numerical methods for the modelling of transport relates to FWP 3 (Fundamental science)
- Metallic TMDs show strong catalytic activity and can replace Pt in electrodes. Relates to FWP 9 (Energy); JacobsUni & TU Dresden collaboration

Progress so far

Topological states induced by strong spin-orbit coupling:

- Ma, Y., Kou, L., Li, X., Dai, Y., **Heine, T.**, 2016. Two-dimensional transition metal dichalcogenides with a hexagonal lattice: Room-temperature quantum spin Hall insulators. Phys. Rev. B 93, 035442.
- Ma, Y., Kou, L., Li, X., Dai, Y., Smith, S.C., **Heine, T.**, 2015. Quantum spin Hall effect and topological phase transition in two-dimensional square transition-metal dichalcogenides. Phys. Rev. B 92, 085427.
- Ma, Y., Kou, L., Du, A., **Heine, T.**, 2015. Group 14 element-based non-centrosymmetric quantum spin Hall insulators with large bulk gap. Nano Res. 8, 3412–3420.

Electronic properties of TMDs:

- Island, J.O., Kuc, A., Diependaal, E.H., Bratschitsch, R., van der Zant, H.S.J., **Heine, T.**, Castellanos-Gomez, A., 2016. Precise and reversible band gap tuning in single-layer MoSe2 by uniaxial strain. Nanoscale 8, 2589–2593.
- Bacaksiz, C., Cahangirov, S., **Rubio, A.**, Senger, R.T., **Peeters, F.M.**, Sahin, H., 2016. Bilayer SnS2: Tunable stacking sequence by charging and loading pressure. Phys. Rev. B 93, 125403.
- Ozaydin, H.D., Sahin, H., Kang, J., **Peeters, F.M.**, Senger, R.T., 2015. Electronic and magnetic properties of 1T-TiSe2 nanoribbons. 2D Mater. 2, 044002.
- Levi, R., Garel, J., Teich, D., **Seifert, G.**, Tenne, R., Joselevich, E., 2015. Nanotube Electromechanics beyond Carbon: The Case of WS2. ACS Nano 9, 12224–12232.

Progress so far

Magnetic/transport properties of TMDs:

- Torun, E., Sahin, H., Bacaksiz, C., Senger, R.T., **Peeters, F.M.**, 2015. Tuning the magnetic anisotropy in single-layer crystal structures. Phys. Rev. B 92, 104407.
- Tahir, M., Vasilopoulos, P., **Peeters, F.M.**, 2016. Quantum magnetotransport properties of a MoS2 monolayer. Phys. Rev. B 93, 035406.
- Sivek, J., Sahin, H., Partoens, B., **Peeters, F.M.**, 2016. Giant magnetic anisotropy in doped single layer molybdenum disulfide and fluorographene. Submitted.
- Kamalakar, M.V., Dankert, A., **Kelly, P.J.**, Dash, S.P., 2016. Inversion of Spin Signal and Spin Filtering in Ferromagnet vertical bar Hexagonal Boron Nitride-Graphene van der Waals Heterostructures. Sci Rep 6, 21168.

Co adsorption on graphene
Os doped $MoTe_2$ ⇨ Magnetocrystalline anisotropy ~ electric field
influenced by charging of the system

MoS_2 and fluorographene → replace S/F by transition metal (Os, Ir) ⇨ Strong magnetic anisotropy

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