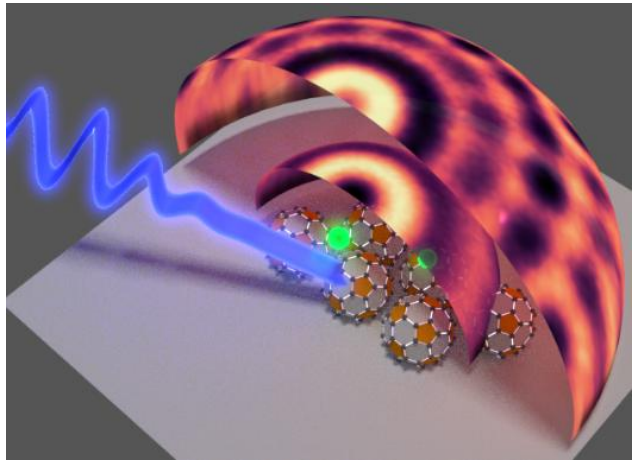


Imaging Charge and Spin Carriers of Hybrid Systems in Momentum Space

Benjamin Stadtmüller, University of Augsburg

Universität Augsburg University

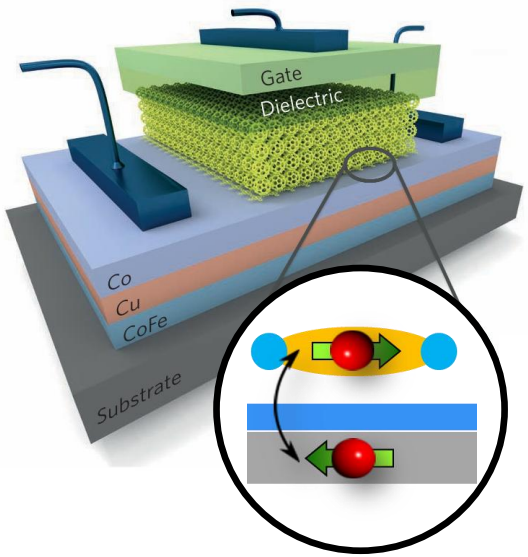
RPTU Rheinland-Pfälzische Technische Universität Kaiserslautern Landau

eli attosecond

SPIN+X SFB/TRR 173 Kaiserslautern • Mainz

Functionalization of Interfaces for Applications

Prototypical Device Structure



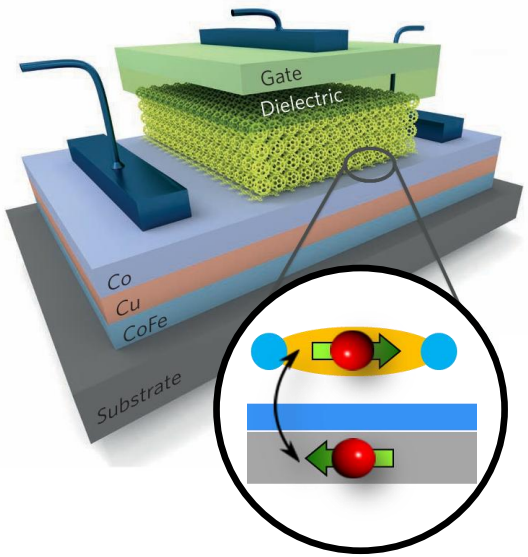
Hybrid interfaces

Interfacial functionalities:

- Spin filtering
- Charge and spin separation
- Charge and spin current generation
- …

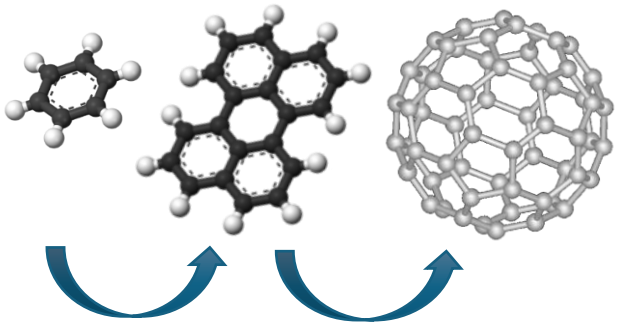
Functionalization of Interfaces for Applications

Prototypical Device Structure

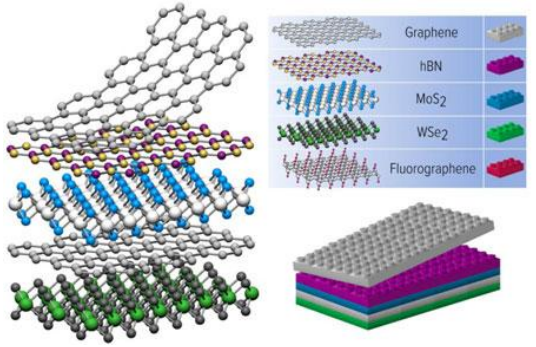


Hybrid interfaces

The manifold of molecular and 2D materials



Size Shape



Nature **499,** 419–425 (2013)

Interfacial functionalities:

- Spin filtering
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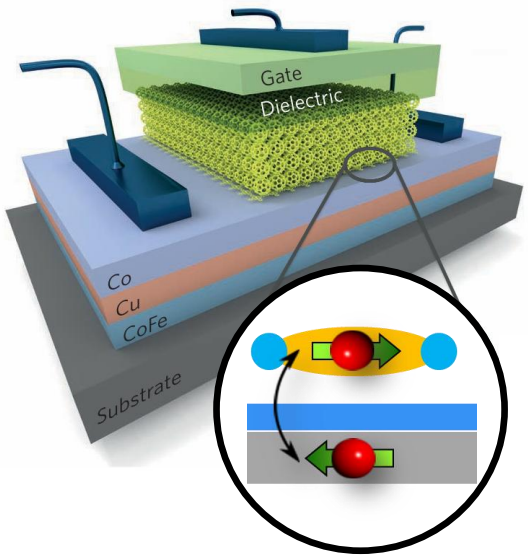
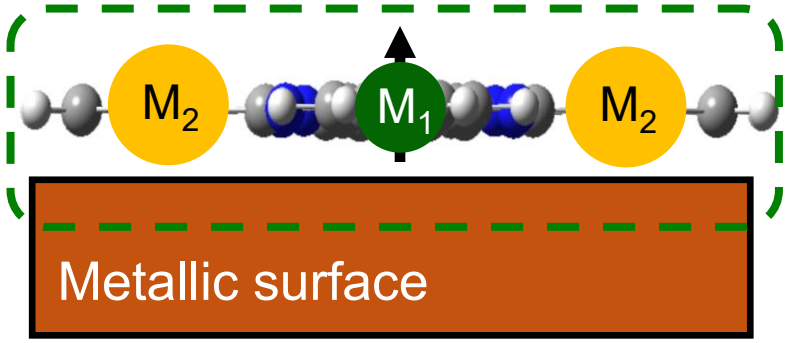
Functionalization of Interfaces for Applications

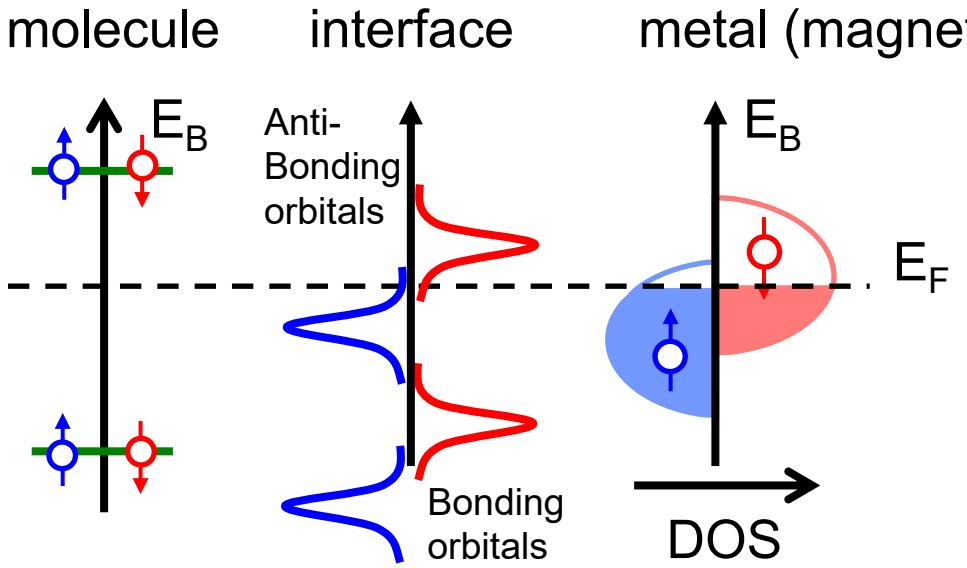
Prototypical Device Structure



Hybrid interfaces

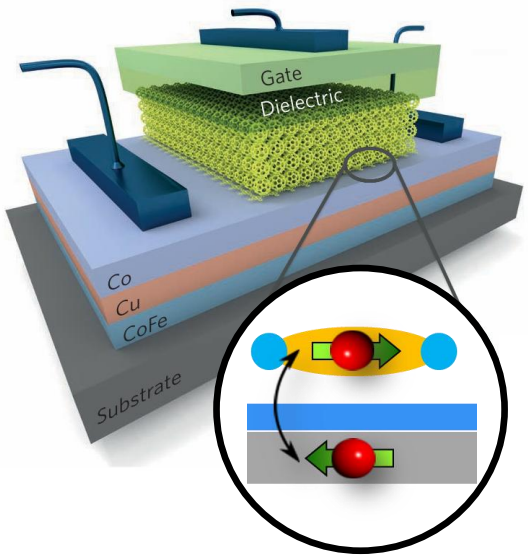


Chemical interaction at hybrid interfaces

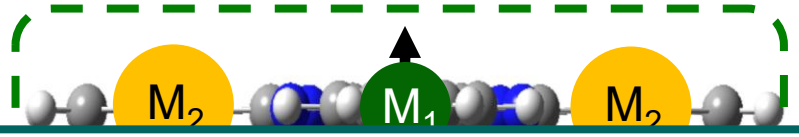


Functionalization of Interfaces for Applications

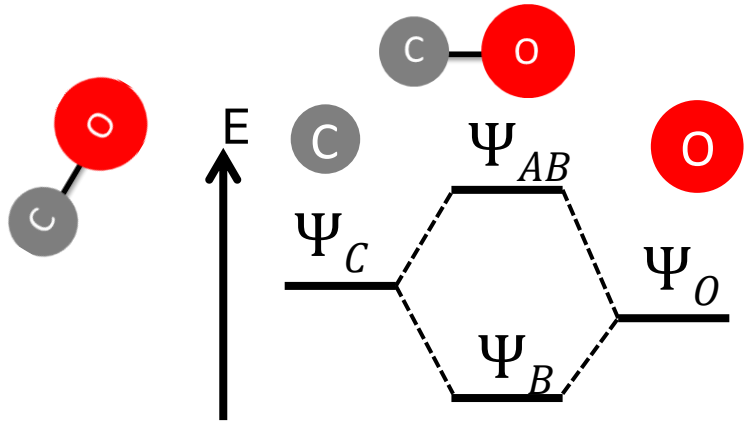
Prototypical Device Structure

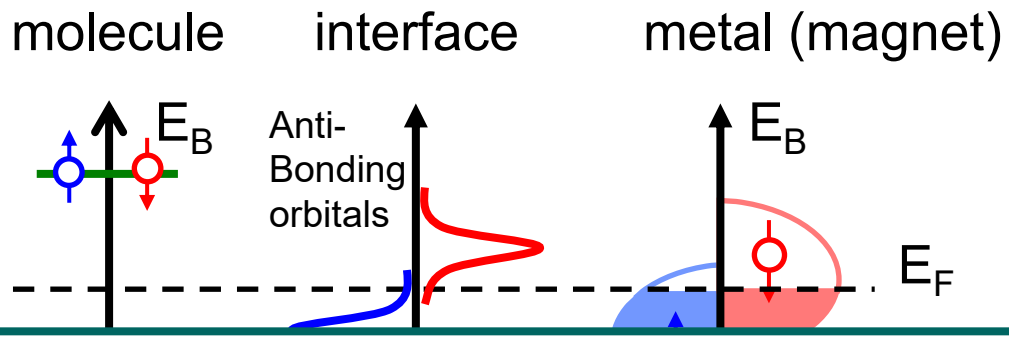


Hybrid interfaces



Chemical interaction at hybrid interfaces

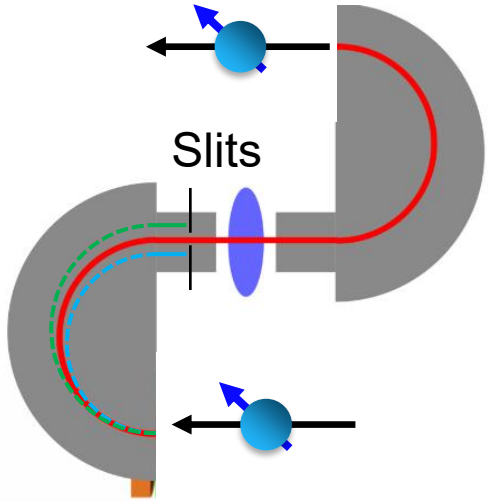


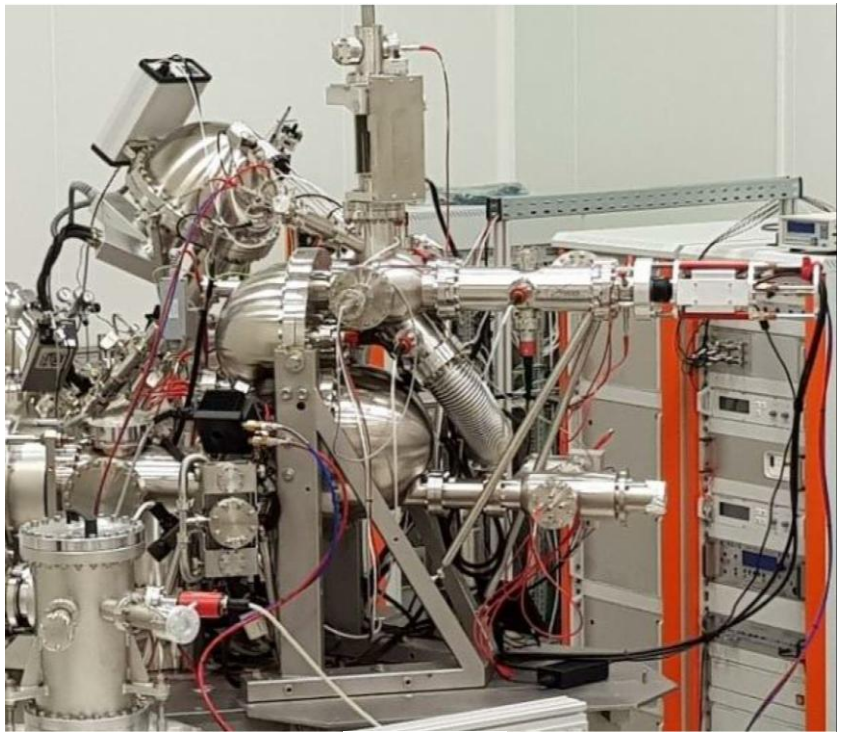


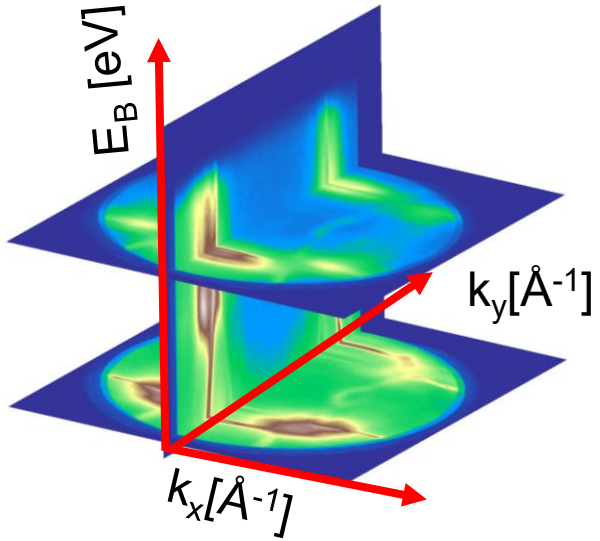
Can we reveal signatures of chemical interaction in the interfacial band structure?
Can we tune and control the interfacial band structure chemically or optically?

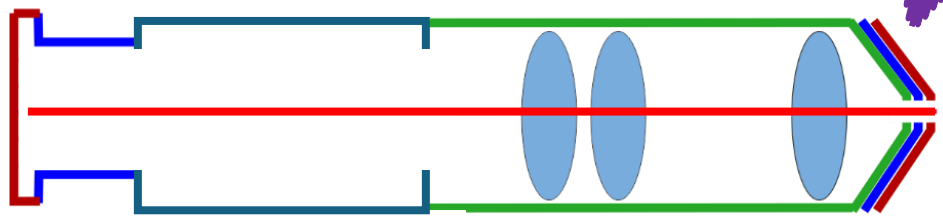
The NanoEsca Instrument @ ELI-ALPS

Dispersive
energy analyzer

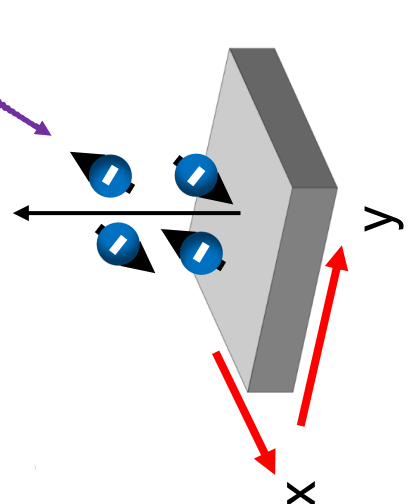






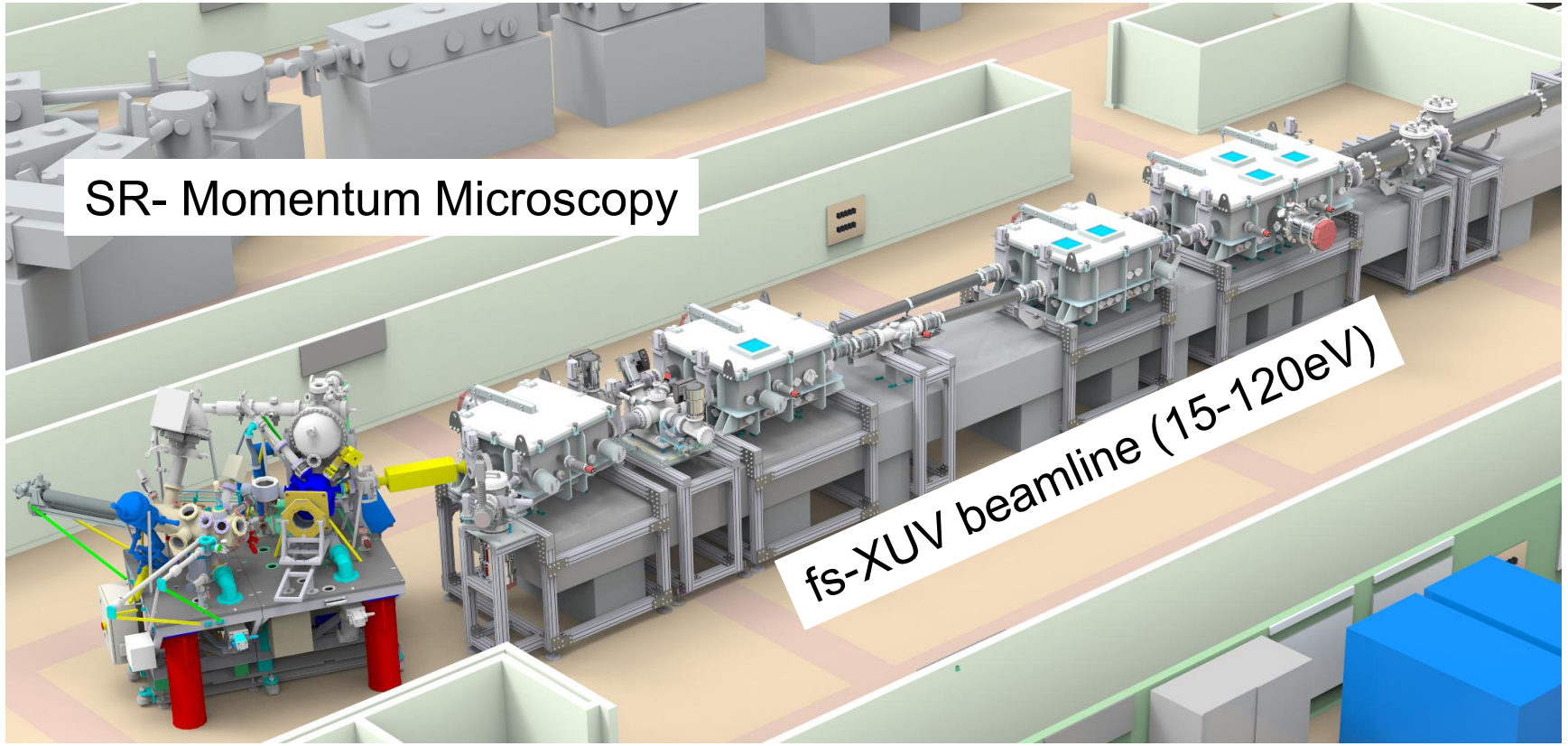


Energy analyzer Electron optics



The NanoEsca Instrument @ ELI-ALPS

Multidimensional (time, spin, momentum, energy) photoemission spectroscopy with *momentum microscopy @ ELI-Alps: NanoEsca End station*

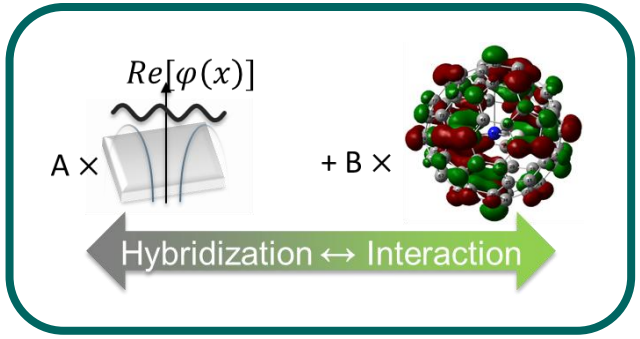


ELI-ALPS
Szeged, Hungary:
EU research facility

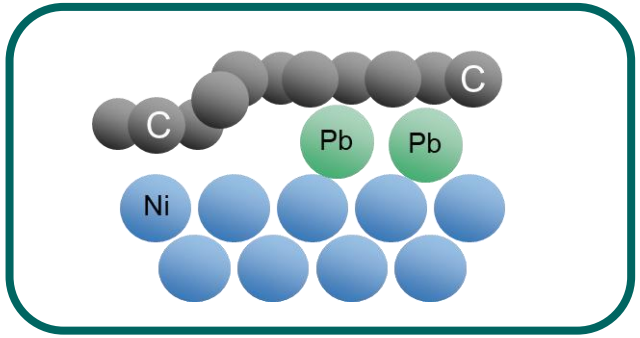
100kHz, IR fiber-based, 3fs, 5mJ

sub 30-fs XUV source (15 - 120 eV) for ultrafast time-resolved surface science studies

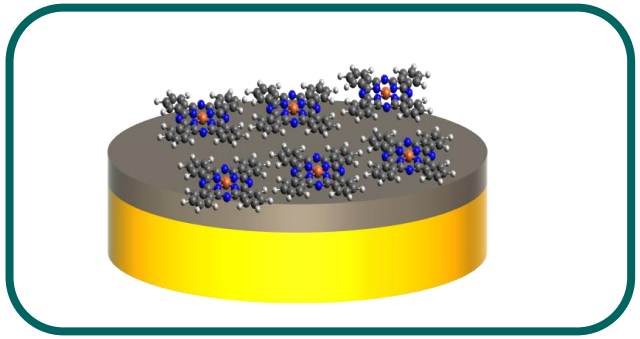
Outline



Chemical interaction at hybrid interfaces
Formation of new hybrid interface states and bands



Ultrafast interfacial charge carrier dynamics:
Charge and energy transfer at a functionalized graphene/Ni interface



Spin-polarized hybrid interface states:
Spin-polarized molecular orbitals at a FePc/Co interface

The Team

RPTU Kaiserslautern:
M. Aeschlimann,
C. Schott, M. Mitkov, E. Walther, M. Anstett,
S. Hedwig, B. Arnoldi






University of Augsburg

T. Eul, L. Lu, A. Schmid, R. Hemm,
F. Müller, G. Zinke






ELI-ALPS NanoEsca team
L. Ovari, G. Halasi, C. Vass,

University of Graz:
P. Puschnig,
A. Windischbacher, C Kern

University of Göttingen
S. Mathias, M. Reutzel,
M. Jansen, W. Bennecke

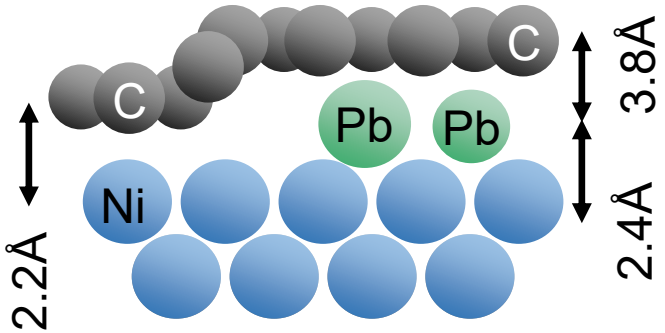
FZ Jülich, PGI-6 and Elettra Synchrotron

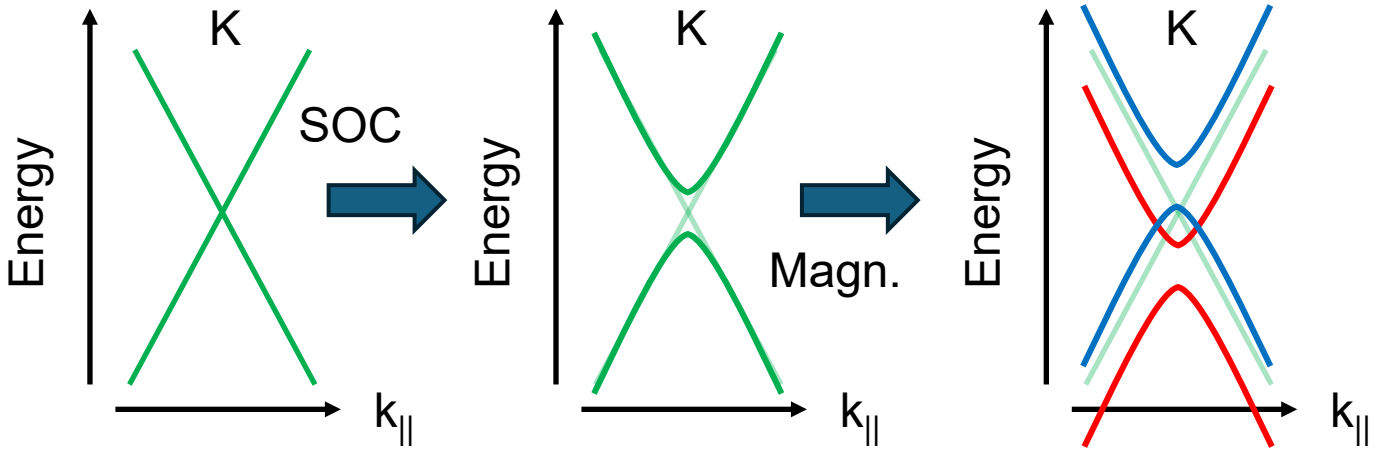
V. Feyer and team

University of Arizona
O. L.A. Monti, S. Zachritz

Functionalizing graphene by intercalation

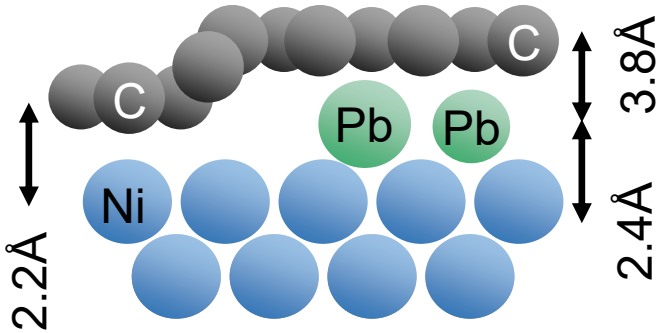
Vertical adsorption geometry



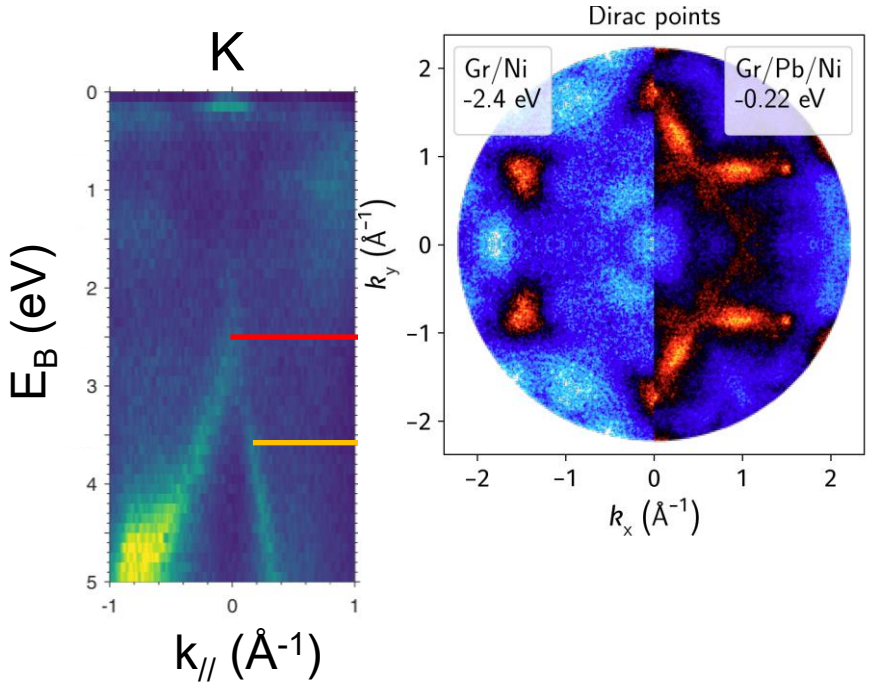


Functionalizing graphene by intercalation

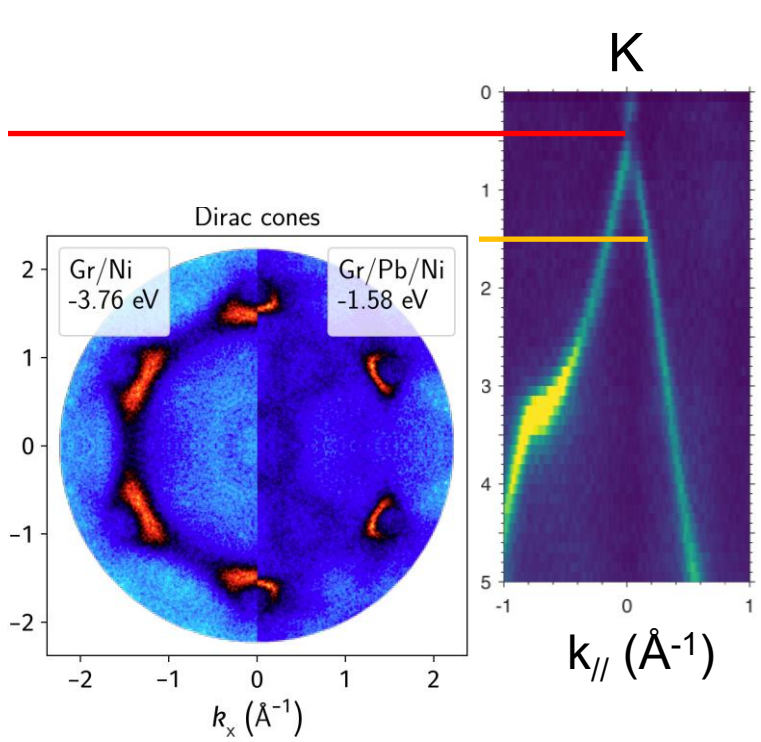
Vertical adsorption geometry

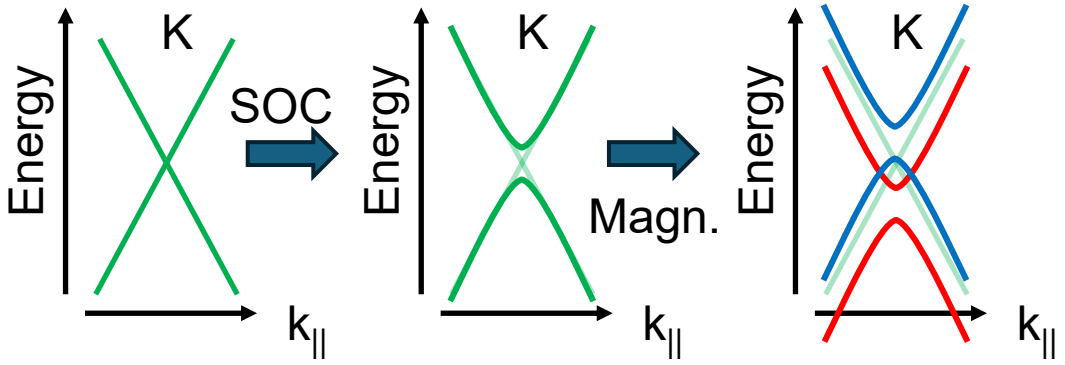


Gr/Ni



Gr/Pb/Ni



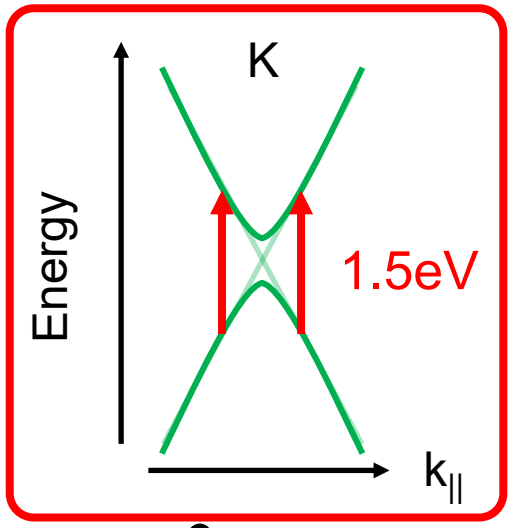


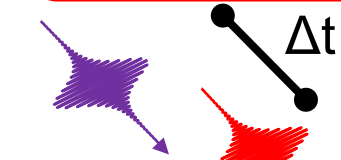
Pb intercalation leads to

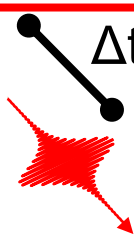
- Recovery of quasi-free-standing Dirac cone dispersion by chemical decoupling
- Opening of a small band gap by proximity effects

Ultrafast Charge Carrier Dynamics of Functionalized Graphene

Gr/Pb/Ni: Ultrafast electron dynamics



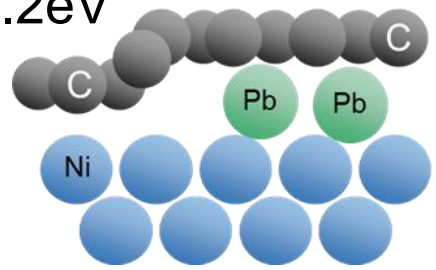


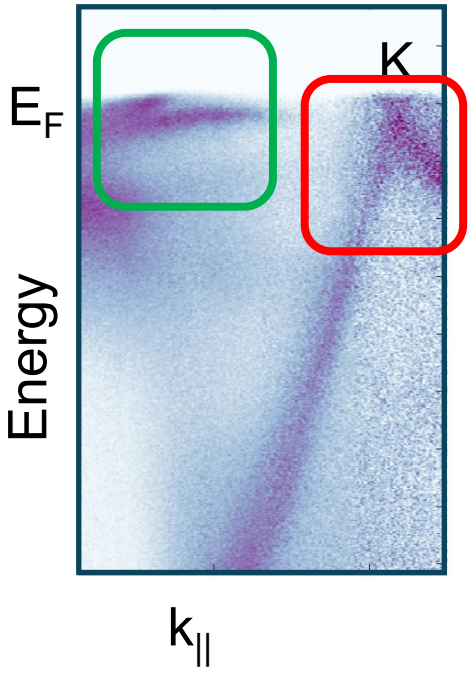


Δt

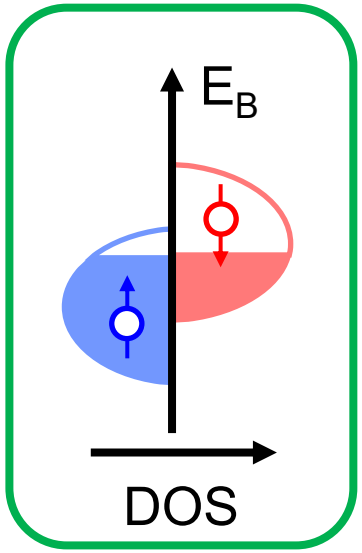
Pump:
1.5eV

Probe:
22.2eV



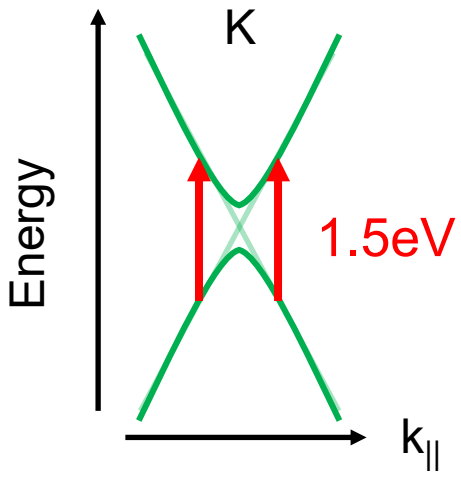


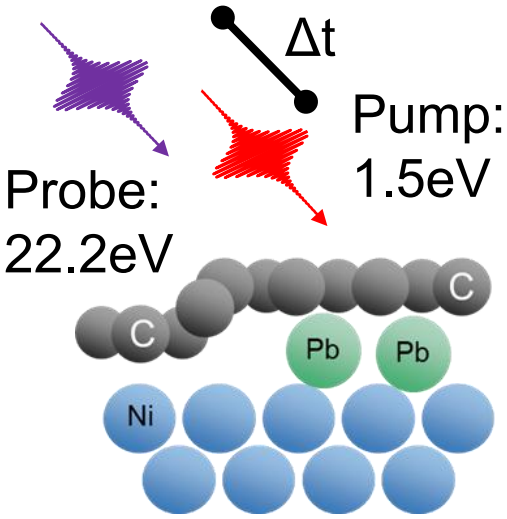
Exchange splitting
of Ni bands

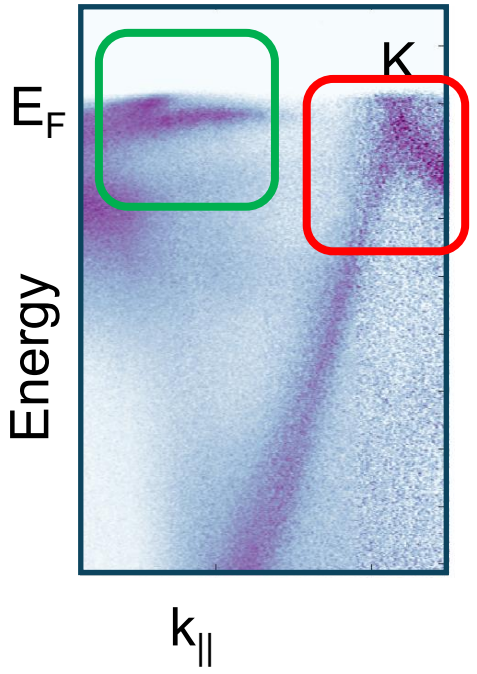


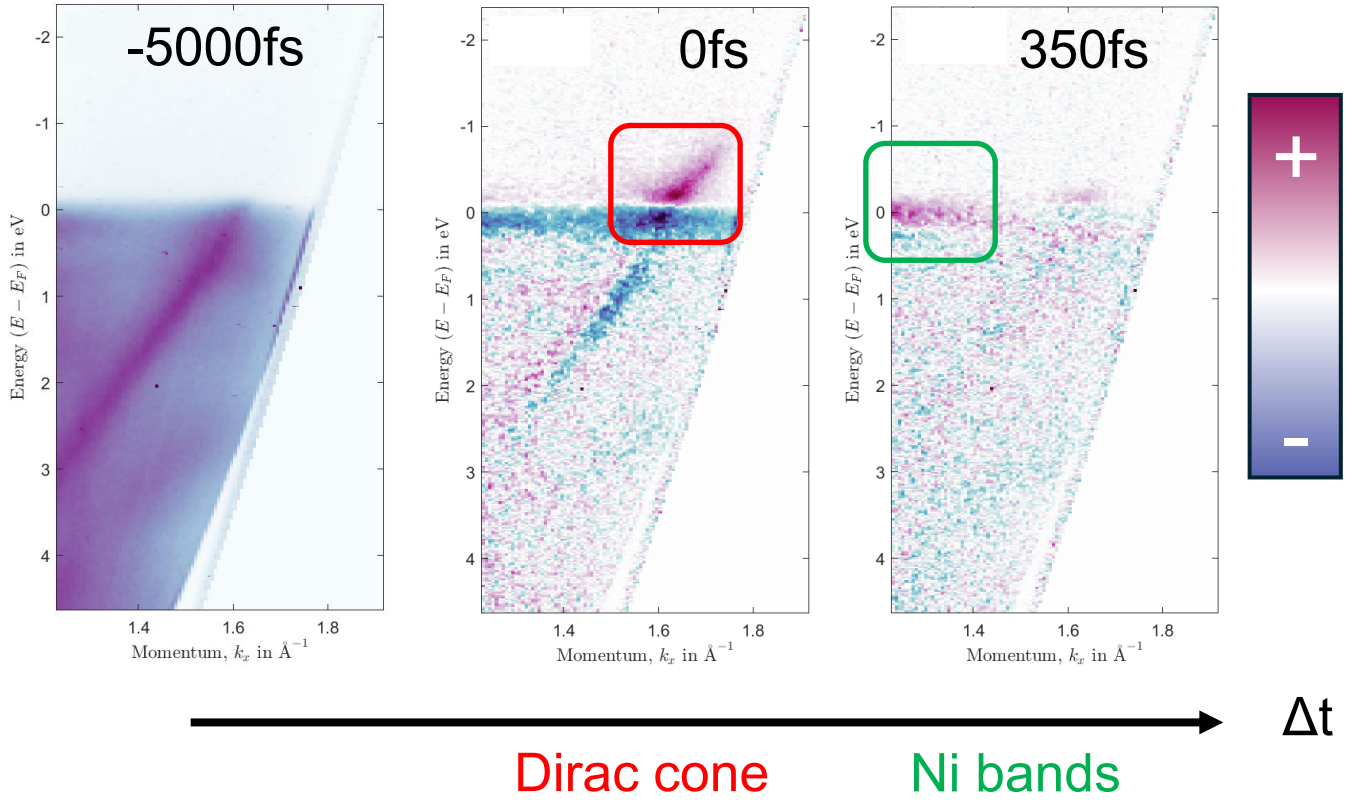
Ultrafast Charge Carrier Dynamics of Functionalized Graphene

Gr/Pb/Ni: Ultrafast electron dynamics





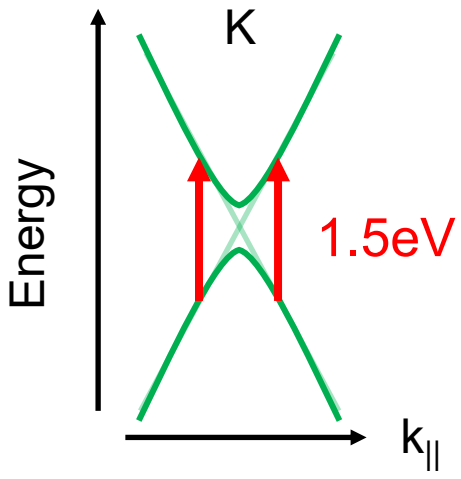


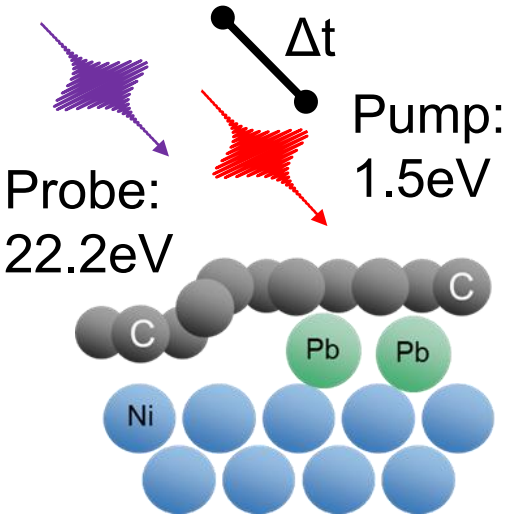


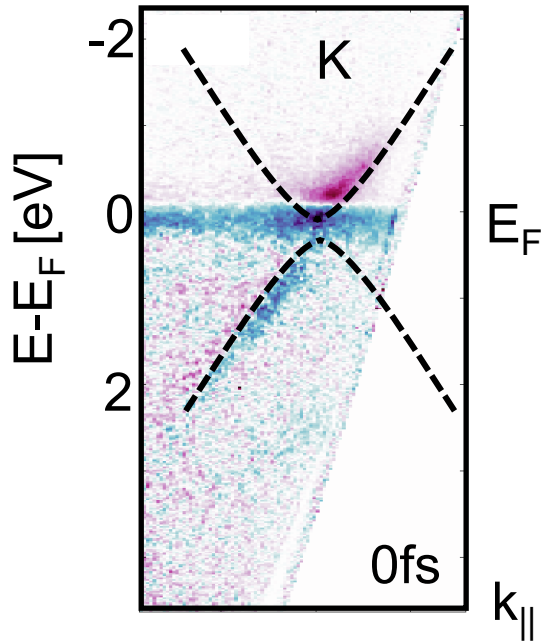
- Optical occupation of upper part of Dirac cone
- Transient band structure renormalization of Ni d-bands

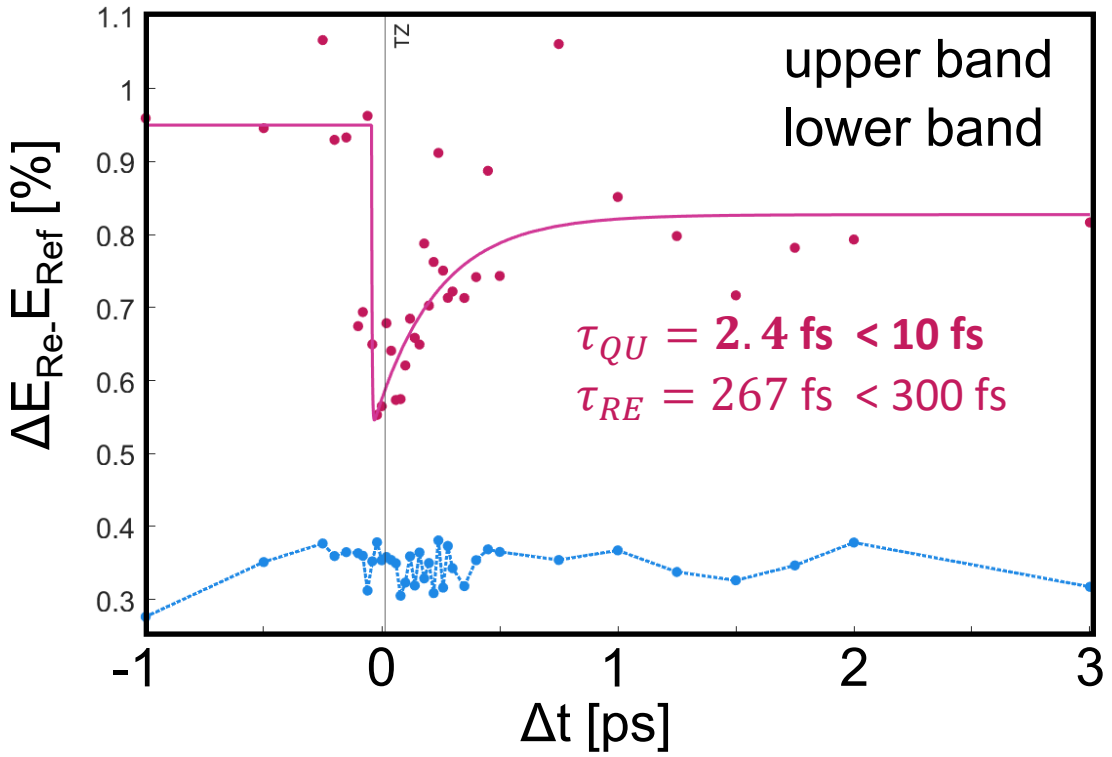
Ultrafast Charge Carrier Dynamics of Functionalized Graphene

Gr/Pb/Ni: Ultrafast electron dynamics of the Dirac cone





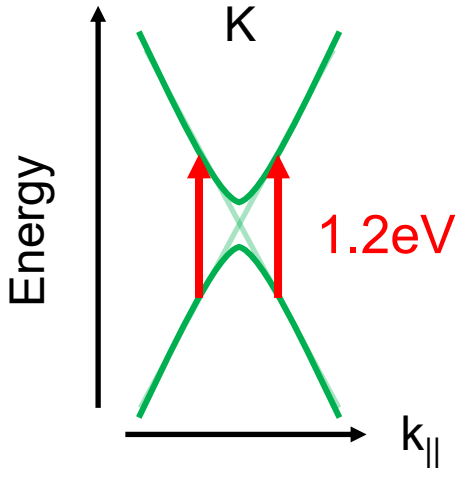


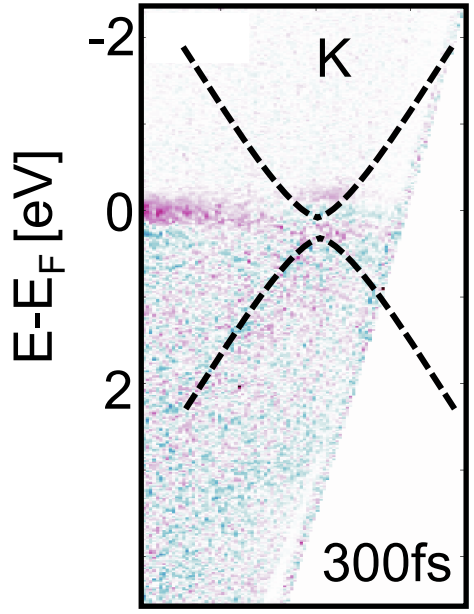


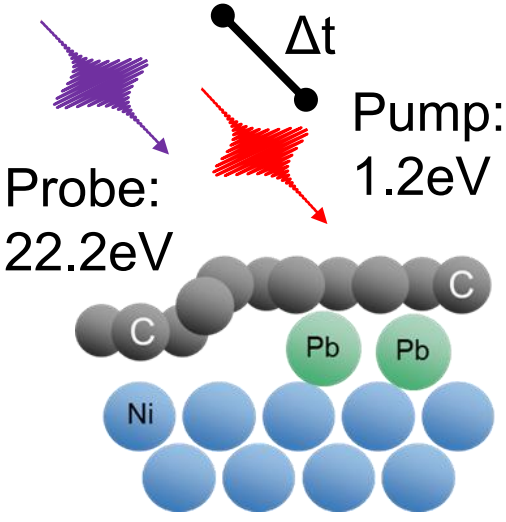
- Substantial renormalization of gap due to photodoping
- Relaxation within electron thermalization time

Ultrafast Charge Carrier Dynamics of Functionalized Graphene

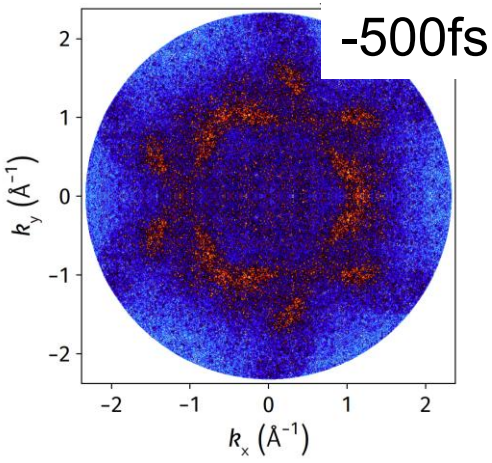
Gr/Pb/Ni: Ultrafast electron dynamics of the Dirac cone



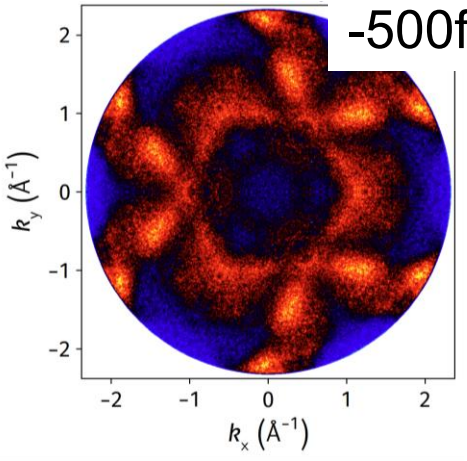




Excited States: E_F+0.18eV

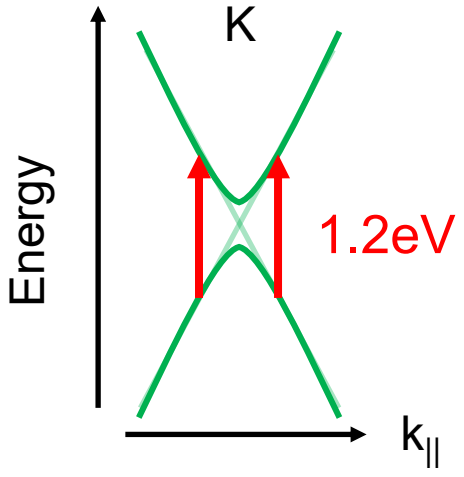


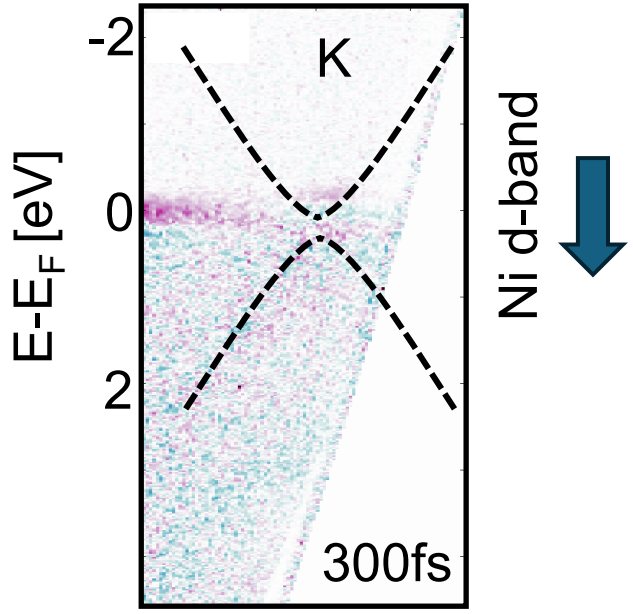
Valence States: E_F-0.14eV

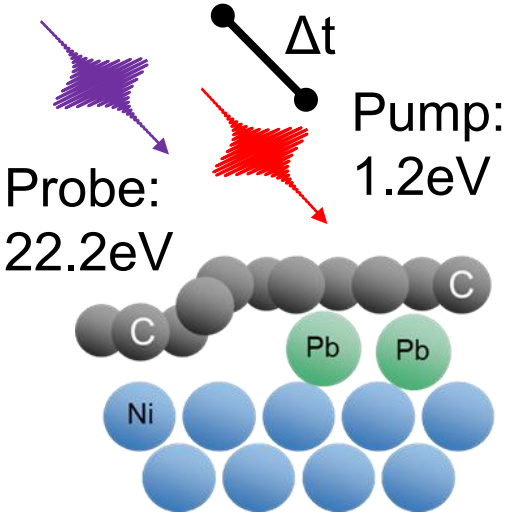


Ultrafast Charge Carrier Dynamics of Functionalized Graphene

Gr/Pb/Ni: Ultrafast electron dynamics of the Dirac cone

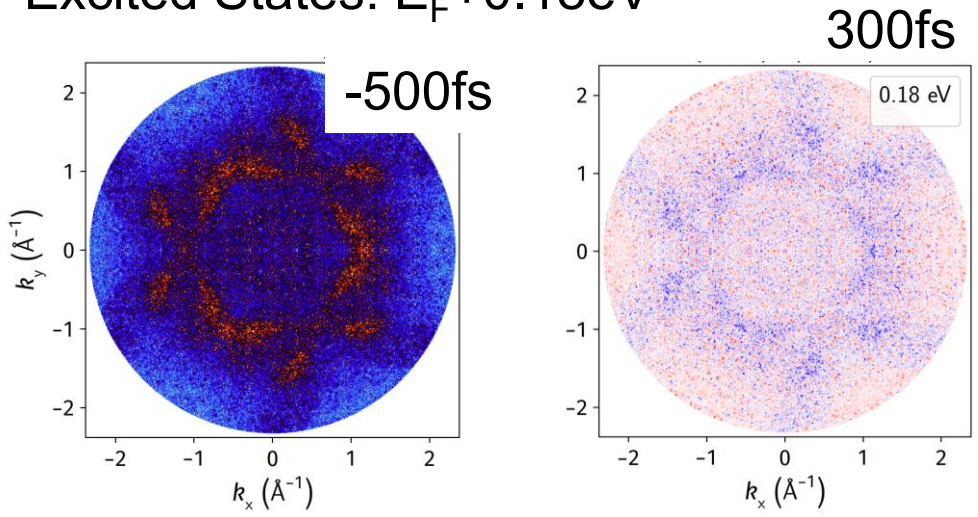




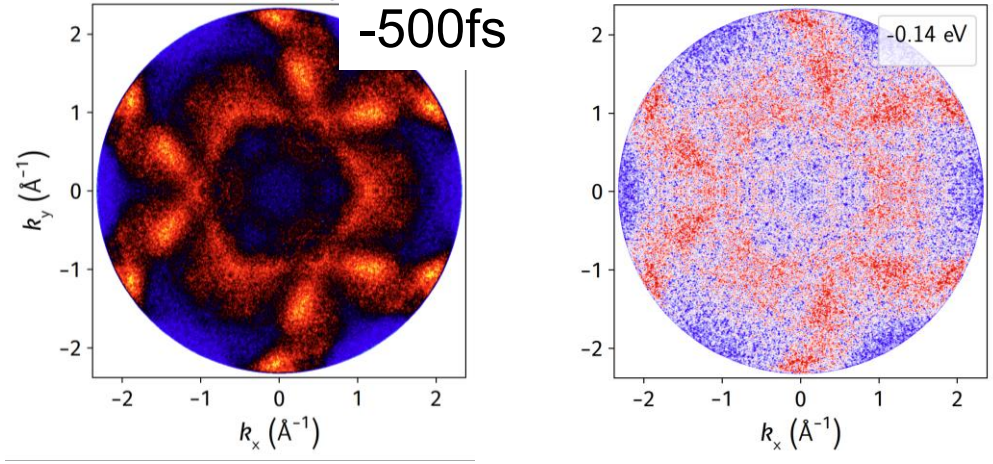


- Shift of Ni d- bands
- Sign of collapse of exchange splitting?

Excited States: E_F+0.18eV

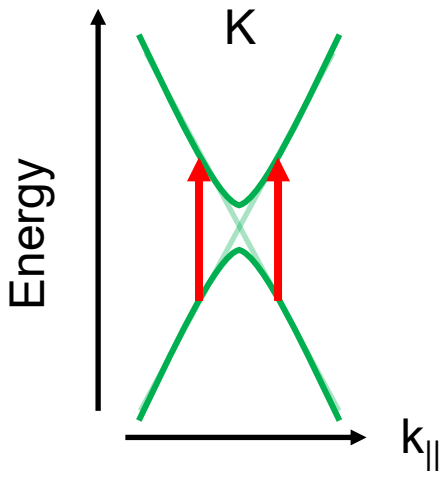


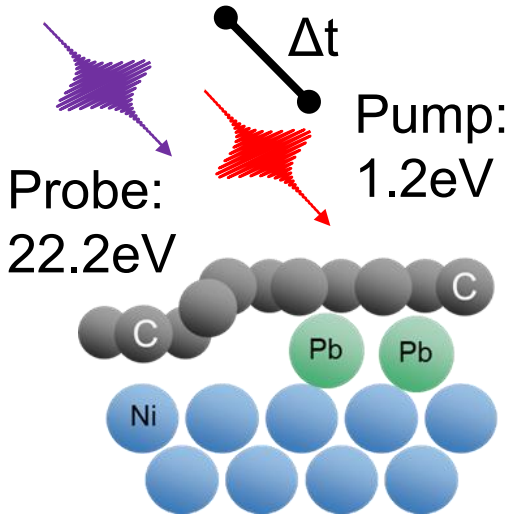
Valence States: E_F-0.14eV

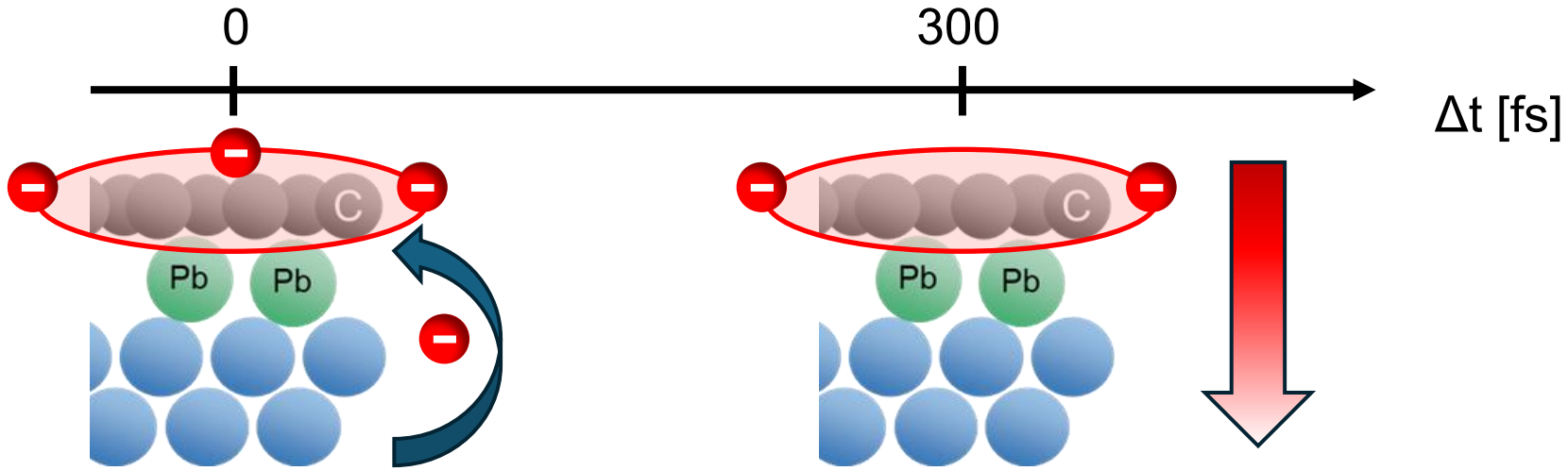


Ultrafast Charge Carrier Dynamics of Functionalized Graphene

Gr/Pb/Ni: Ultrafast electron dynamics of the Dirac cone







Transient charging of the graphene layer:

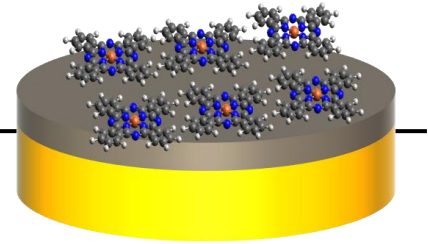
- Band gap renormalization
- Depletion of Ni states around E_F

Thermalization of electrons:

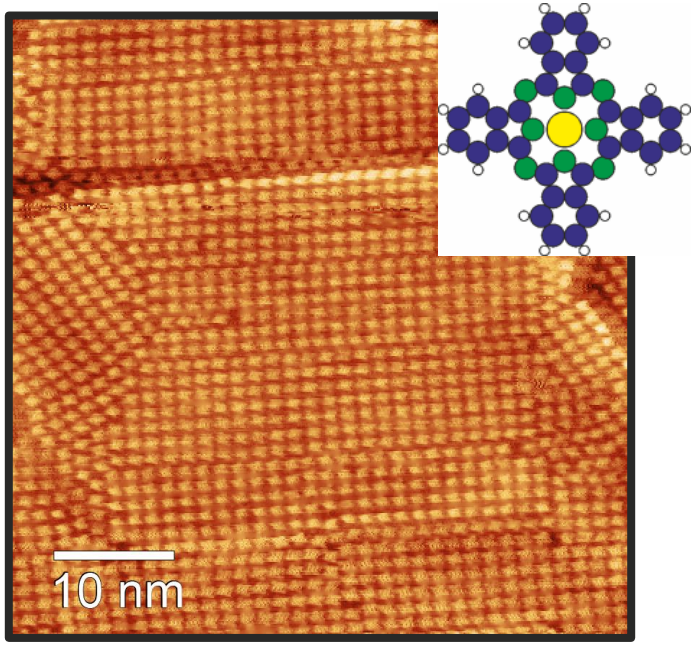
- Energy transfer from graphene into Ni
- Transient band renormalization of Ni bands

→ Demagnetization of Ni?

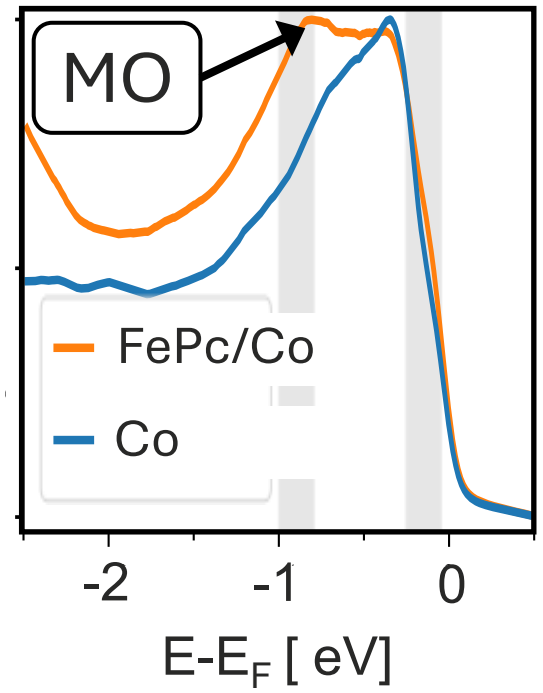
Formation of Spin-Polarized Hybrid Interface States



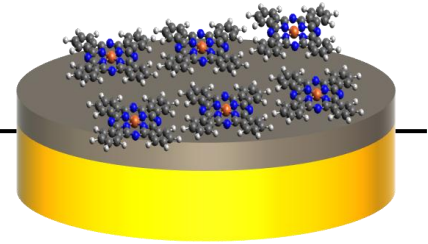
FePc on Co/Au(111) – The model system



Valence band structure



Formation of Spin-Polarized Hybrid Interface States



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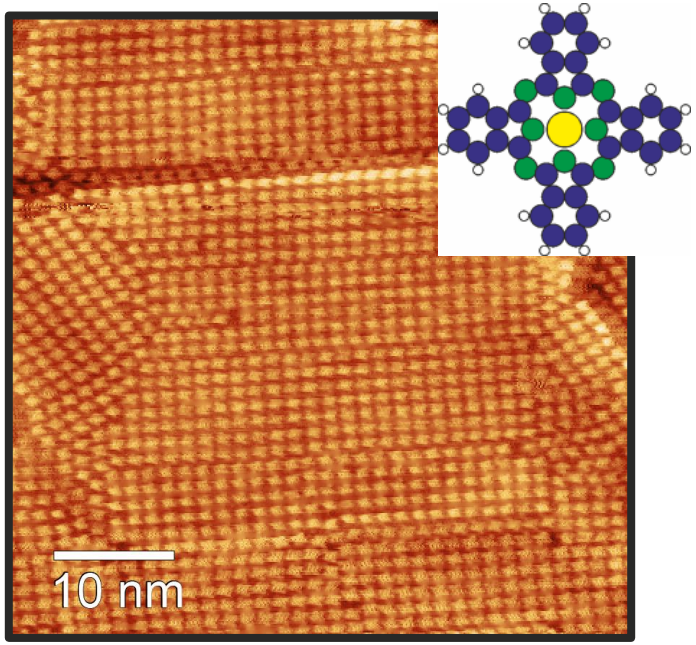

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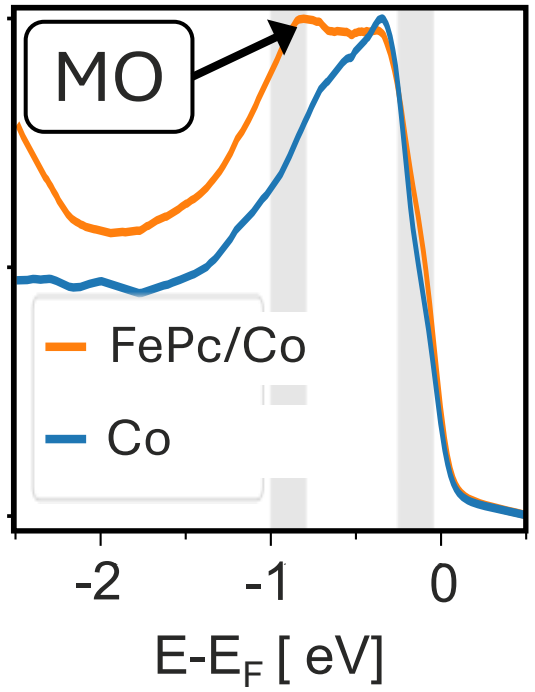
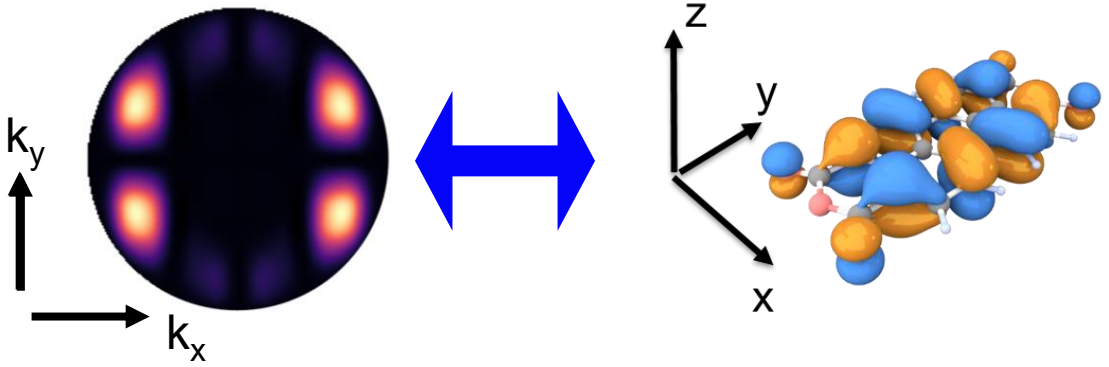


Photoemission Orbital Tomography (POT)

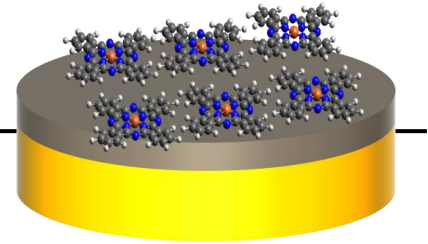
Constant Energy Map

HOMO



P. Puschnig, M. Ramsey, Science **326**, 702 (2009)

Formation of Spin-Polarized Hybrid Interface States



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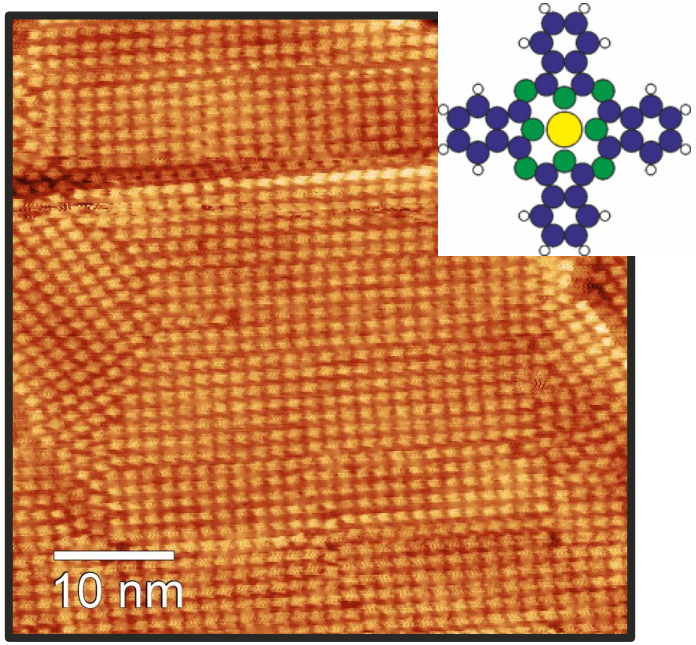

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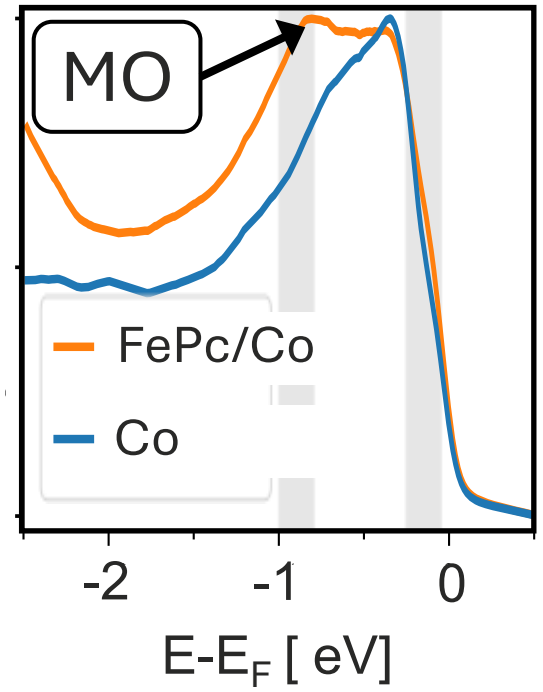

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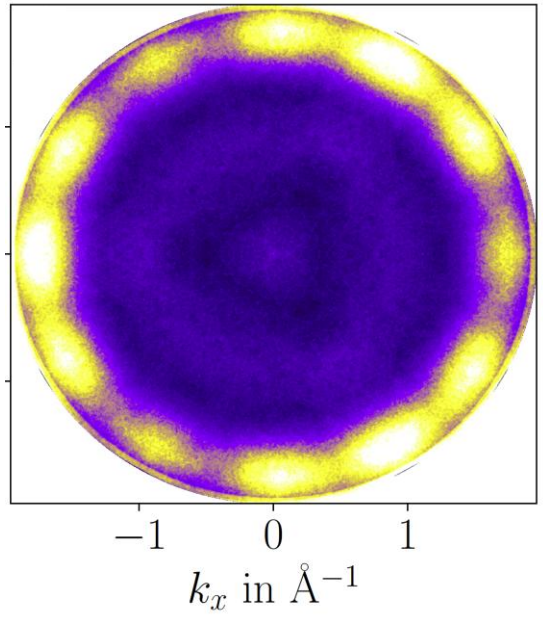
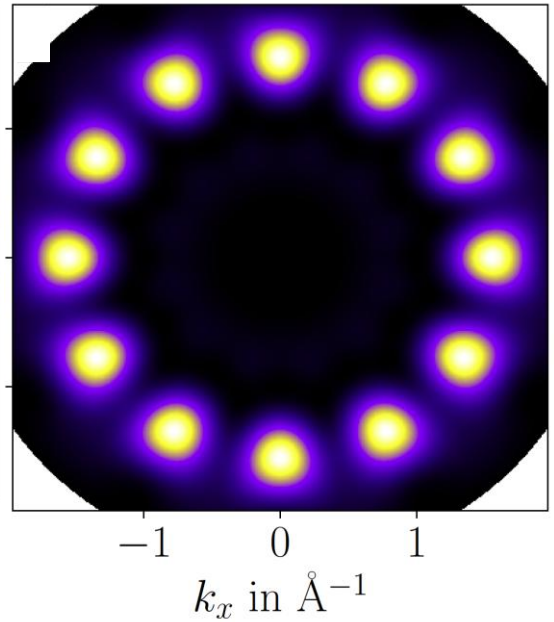


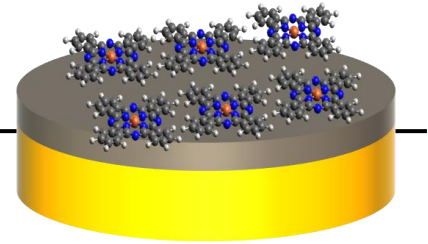
POT: HOMO



Emission pattern: Interface state is derived from HOMO

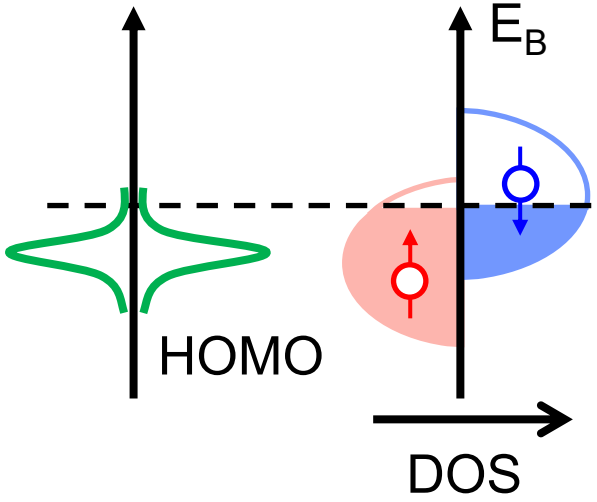
Linewidth: Comparable to adsorption on Cu

Formation of Spin-Polarized Hybrid Interface States

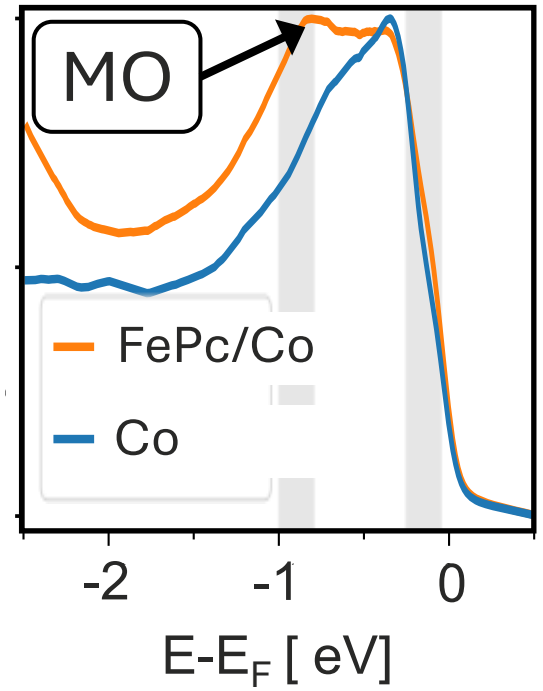


FePc on Co/Au(111) – The model system

Energy level alignment



Valence band structure



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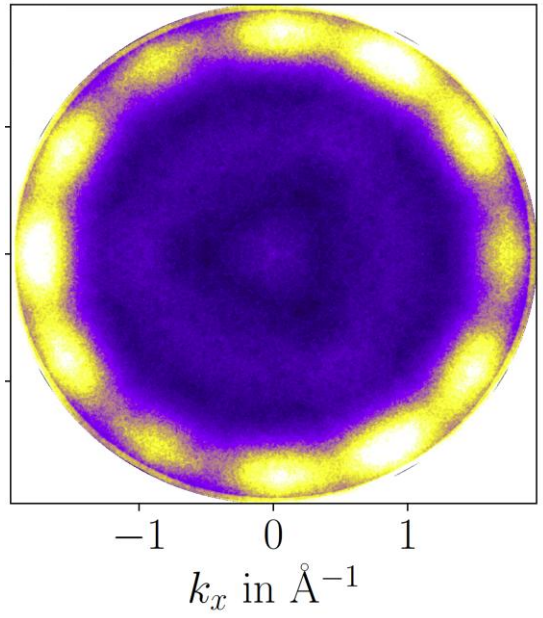

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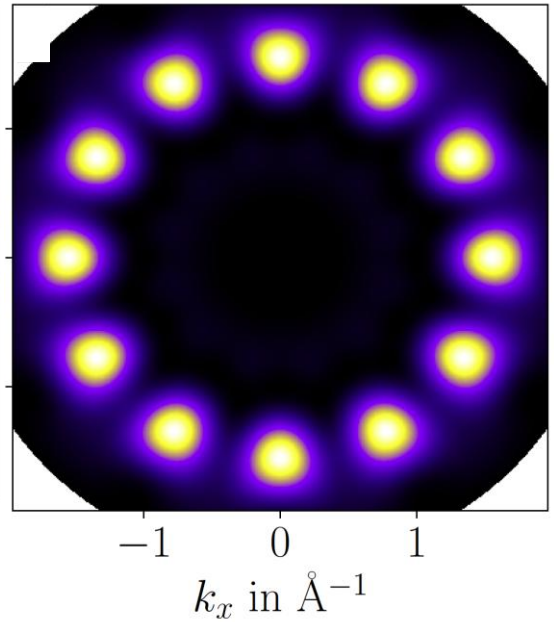

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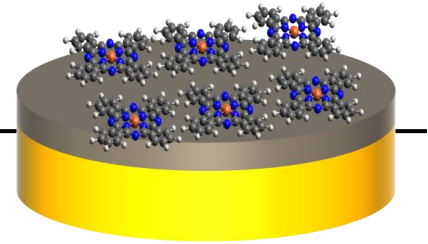
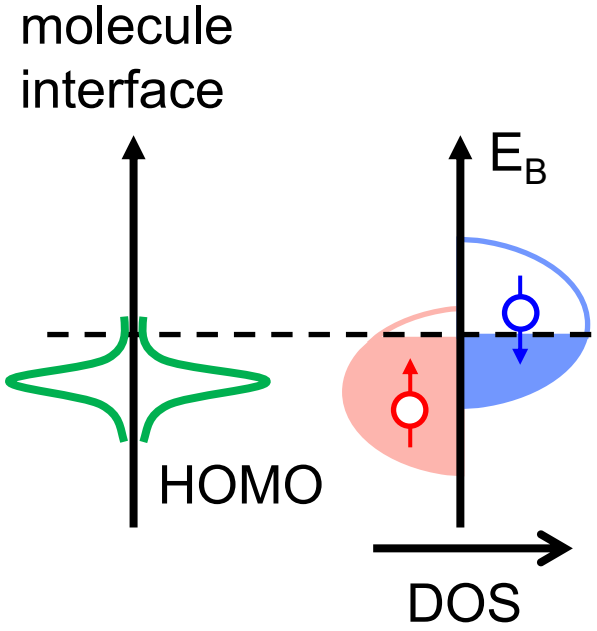


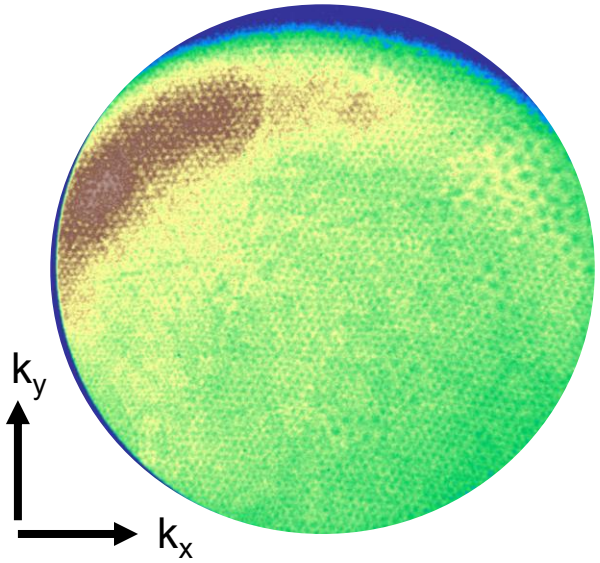
Valence band structure: **Spin-resolved POT** of FePc HOMO

Energy level alignment



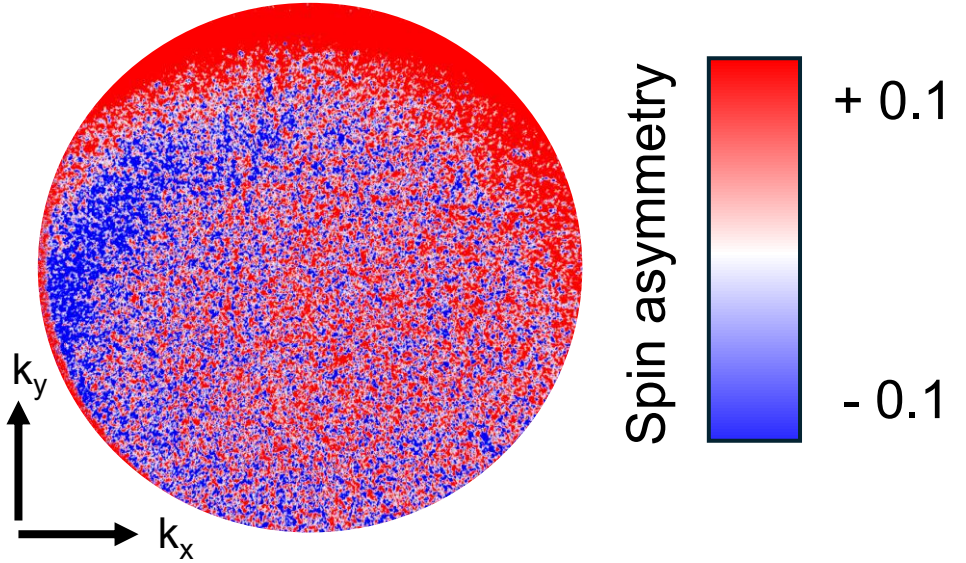
Spin-Sum:

$$R_{E1} \cdot \mathrm{N}^{\uparrow} + R_{E2} \cdot \mathrm{N}^{\downarrow}$$

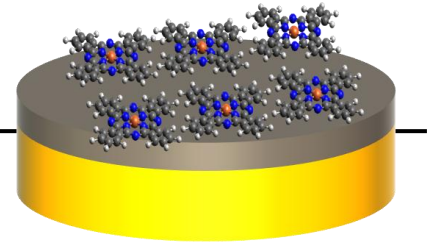


Spin asymmetry

$$\frac{R_{E1} \cdot \mathrm{N}^{\uparrow} - R_{E2} \cdot \mathrm{N}^{\downarrow}}{R_{E1} \cdot \mathrm{N}^{\uparrow} + R_{E2} \cdot \mathrm{N}^{\downarrow}}$$



Formation of Spin-Polarized Hybrid Interface States



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Spin-dep. level alignment

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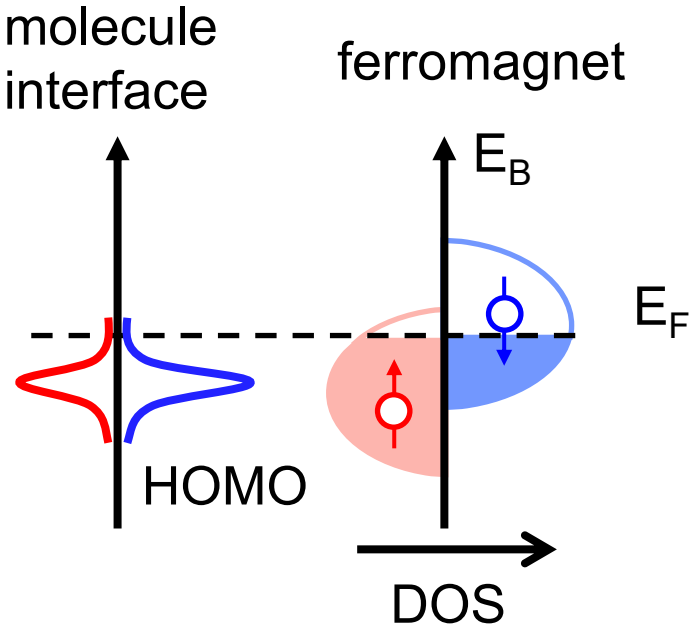
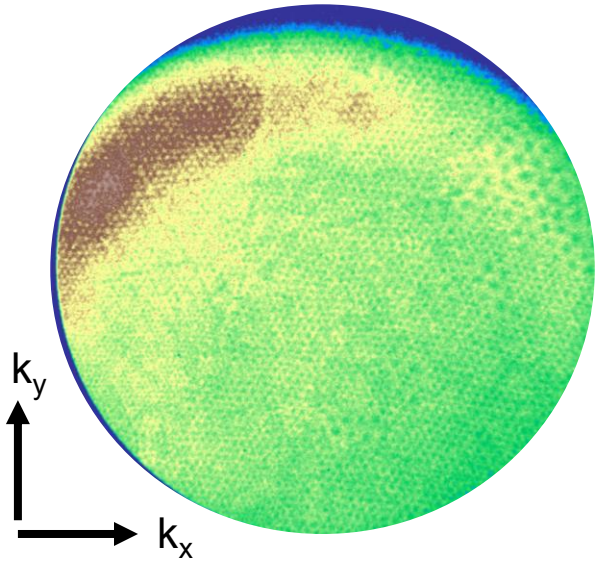
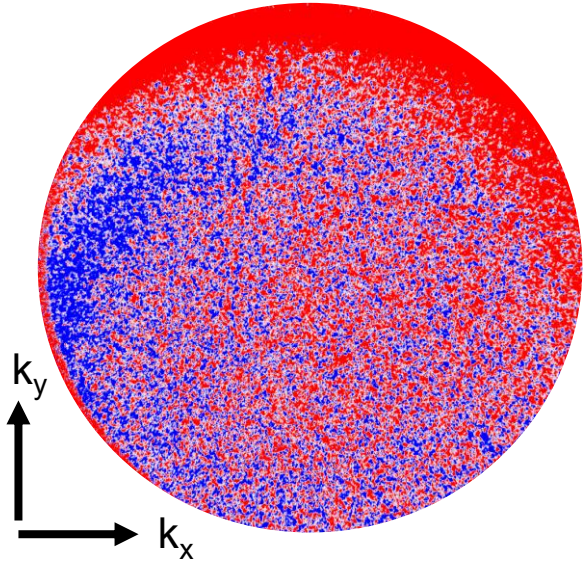
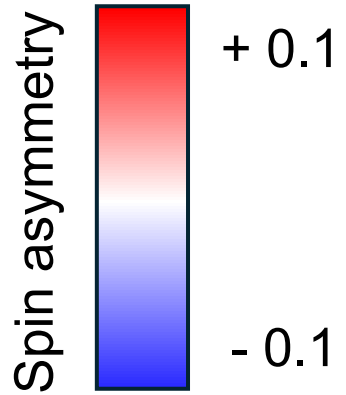








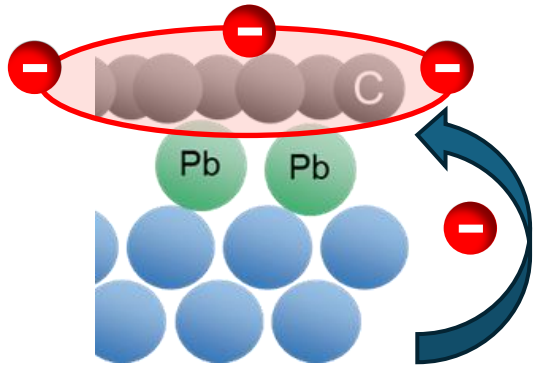
HOMO: **negative minority** spin polarization

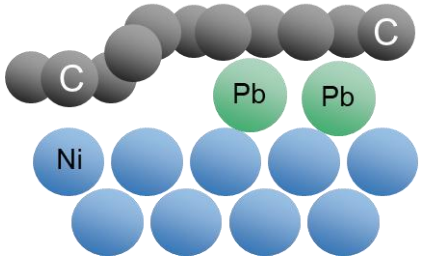
→ Hybridization of HOMO orbitals with **minority** Co states with **negative** spin polarization

Summary

Ultrafast interfacial charge carrier dynamics:
Pb-intercalated graphene/Ni interface

- Recovery of quasi-free-standing Dirac cone dispersion by chemical decoupling
- Ultrafast band gap renormalization due to transient charging of graphene





Spin-polarized hybrid interface states:
FePc/Co interface

- Spin-resolved POT:
  Hybridization of HOMO with **minority** Co states with **negative** spin polarization