Hybrid Organic Magnetic Metal Interfaces

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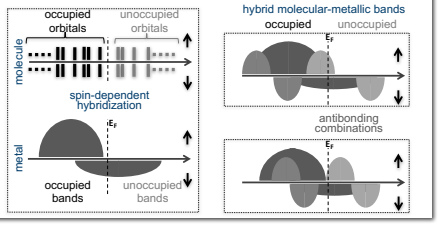
Introduction

The density functional theory provides a framework with predictive power that can be used to describe organic-metal hybrid systems in a realistic manner. In this respect, *ab initio* studies elucidate how the subtle interplay between the electrostatic, the weak van der Waals and the strong chemical interactions determines the geometric, electronic and magnetic structure of hybrid organic-metal interfaces. More precisely, the interaction between the π-like electronic cloud of organic materials with the magnetic states of a metal substrate influences the (i) spin-polarization, (ii) magnetic exchange coupling, (iii) magnetic moments and (iv) their orientation at the hybrid interfaces.

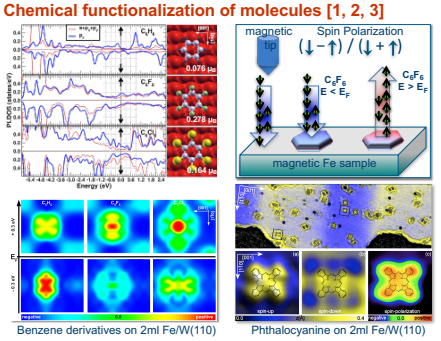
This poster briefly summarizes how first-principles calculations (i) provide the basic insights needed to interpret surface-science experiments and (ii) are a key tool to design novel materials with tailored properties that can be integrated in carbon-based spintronic devices.

Chemisorption: spin-dependent hybridization



Spin-polarization at the interface

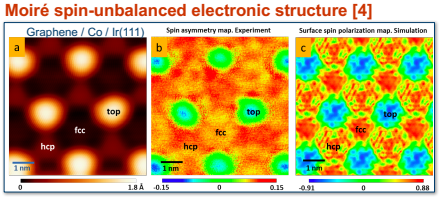
Chemical functionalization of molecules [1, 2, 3]



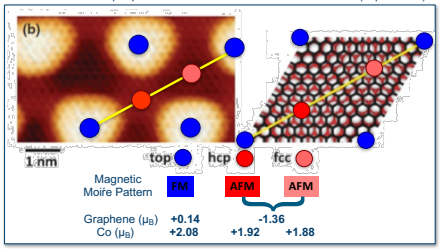
Manipulate the spin-polarized current:

- conventional ferromagnetic surface: apply magnetic field
- molecule / ferromagnetic substrate: change bias voltage

Moiré spin-unbalanced electronic structure [4]

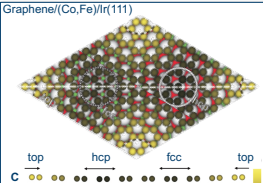


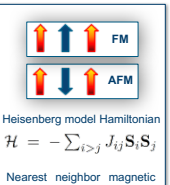
- *spin-unbalanced* π-*d* interaction at the physisorbed sites (top)
- *inversion* of the spin-polarization: ONLY at the chemisorbed sites (hcp and fcc)



Magnetic exchange interactions

Graphene/(Co,Fe)/Ir(111)





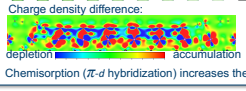
Heisenberg model Hamiltonian

$$\mathcal{H} = -\sum_{i>j} J_{ij}\mathbf{S}_i\mathbf{S}_j$$

Nearest neighbor magnetic exchange coupling J (meV):

	Co	Fe
fcc: J_{fcc} =	41.5	31.7
hcp: J_{hcp} =	37.6	30.3
top: J_{top} =	4.1	13.5

Charge density difference:

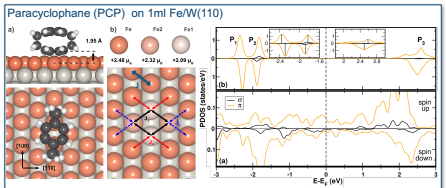


Chemisorption (π-*d* hybridization) increases the magnetic exchange coupling.

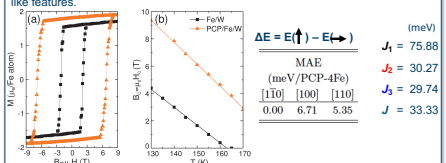
Magnetic hardening & softening with organics

π-*d* hybridization: *intra-layer* magnetic hardening [5]

Paracyclophane (PCP) on 1ml Fe/W(110)



(a) The lower ring binds to substrate via strong chemical bonds and in consequence broad spin-polarized hybrid molecule-surface bands are formed. **(b)** In contrast, the PDOS for the upper ring shows distinct sharp spin-split molecular-like features.



$\Delta E = E(\uparrow) - E(\leftrightarrow)$

MAE (meV/PCP-4Fe)

[1$\bar{1}$0]	[100]	[110]
0.00	6.71	5.35

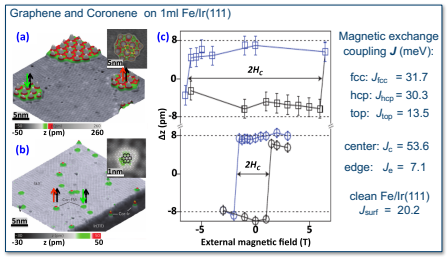
(meV)

J_1 = 75.88
J_2 = 30.27
J_3 = 29.74
J = 33.33

Monte Carlo simulations for the **(a)** magnetization-reversal process **(b)** coercivity field H_c below ordering temperature for a clean and PCP covered 1ml Fe/W(110).

Atomic-like view of the magnetic hardening effect [6]

Graphene and Coronene on 1ml Fe/Ir(111)



Magnetic exchange coupling J (meV):

fcc: J_{fcc} = 31.7
hcp: J_{hcp} = 30.3
top: J_{top} = 13.5

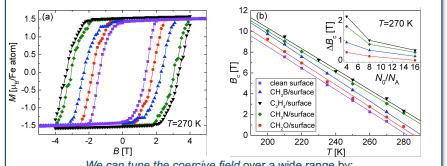
center: J_c = 53.6
edge: J_e = 7.1

clean Fe/Ir(111) J_{surf} = 20.2

Chemical flexibility & magnetic hardening [7]

CH_2BH, C_2H_4, CH_2NH, CH_2O molecules on 1ml Fe/W(110)

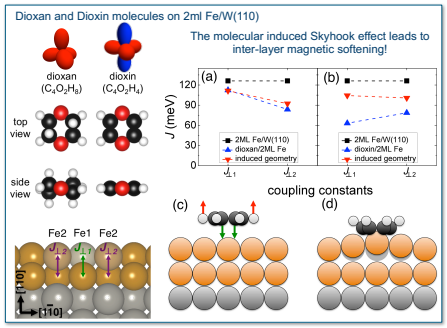
Heteroatom electronegativity increases
Tune the energetic position and spatial extend of the π-states!



We can tune the coercive field over a wide range by:
(i) using different adsorbates and (ii) varying the molecular coverage

Inter-layer magnetic softening with organics [8]

Dioxan and Dioxin molecules on 2ml Fe/W(110)

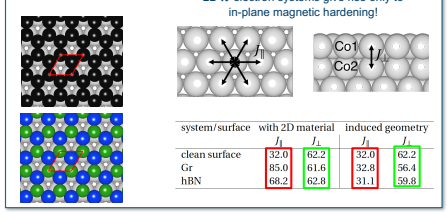


The molecular induced Skyhook effect leads to inter-layer magnetic softening!

Finite size effect: intra-layer magnetic hardening and *inter-layer* magnetic softening [9]

Graphene and hBN on Co(111)

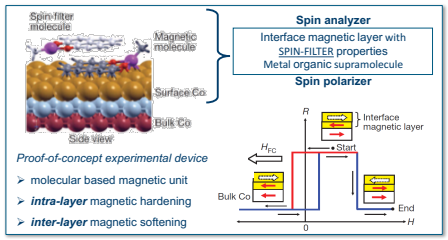
2D π-electron systems give rise only to in-plane magnetic hardening!



system/surface	with 2D material		induced geometry	
	$J_\parallel$	$J_\perp$	$J_\parallel$	$J_\perp$
clean surface	32.0	62.2	32.0	62.2
Gr	85.0	61.6	32.8	56.4
hBN	68.2	62.8	31.1	59.8

A nanomagnet embedded within a magnet

Interface magnetoresistance effect [10]

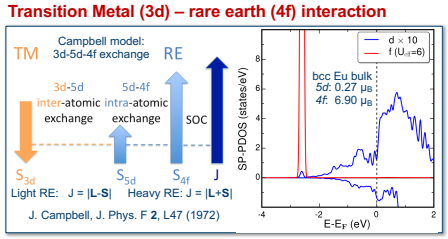


Proof-of-concept experimental device

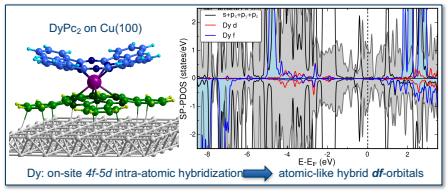
- molecular based magnetic unit
- ***intra-layer*** magnetic hardening
- ***inter-layer*** magnetic softening

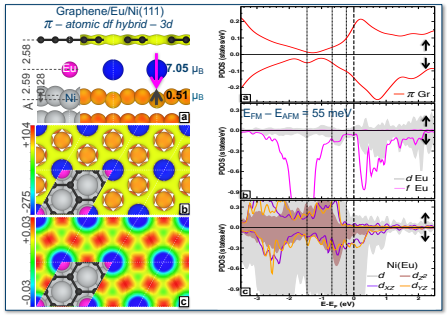
Present and future investigations

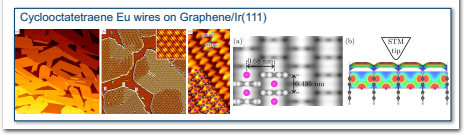
Transition Metal (3d) – rare earth (4f) interaction



On-site rare earth *4f* - *5d* hybridization [11, 12, 13]







Acknowledgments

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