Self-Assembled Structures towards Nanospintronic Devices

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11June 2014

Comprendre le monde, construire l'avenir®

Wednesday, 11 June 14

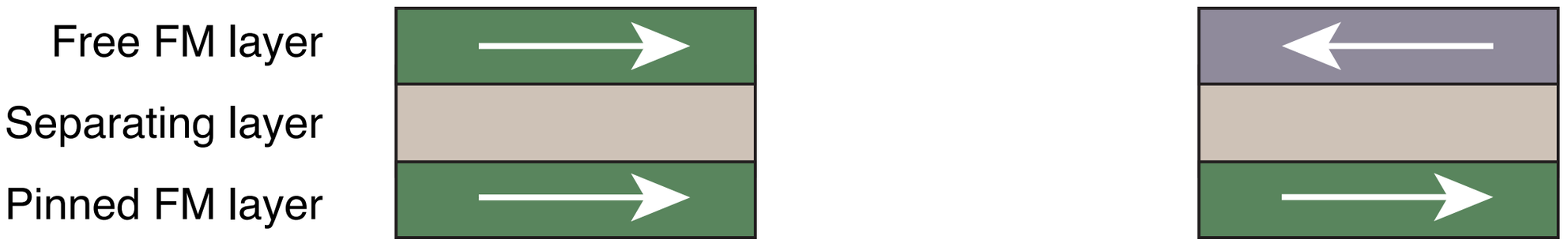
Bonjour je vais vous presente le theme de recherche nanosystemes magnetique et plus particulairement mes avtivitees de recherche lie a la spintronique moleculaire

Introduction

Spin Electronic (Spintronic)

- An emerging discipline that utilizes the spin properties of the electron

Giant Magneto Resistance Device / Spin Valve



Parallel Mode

Antiparallel Mode

P. Grünberg, et. al *Phys. Rev. B*, **1989**, *39*, 4828-4830

A. Fert, et. al *Phys. Rev. Lett.*, **1988**, *61*, 2472-2475

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Pour comprendre que est la spintroniqye je vais vous explique comme marche un dispositif spintronique et ou sont les limitations des dispositifs actuelements employees
tout d'abord Un dispositif spintronique doit génère une courante qui est polarise en spin et il doit détecte cette curante. Le modèle de dispositif le plus simple est le "Giant Magneto Resistor" ou GMR. Le dispositif est characterise par deux couche ferromagnétique séparé par une couche separatrice métallique.
la couche separatrice est cruciale parce que si n'y a pas cette couche on ne peux pas separe les deux electrodes.
cette technologie était employé dans les disk dure a partir de 1997 et en 2007 le prix nobel était attibue a albert fert et péter grunberg pour leur découverte

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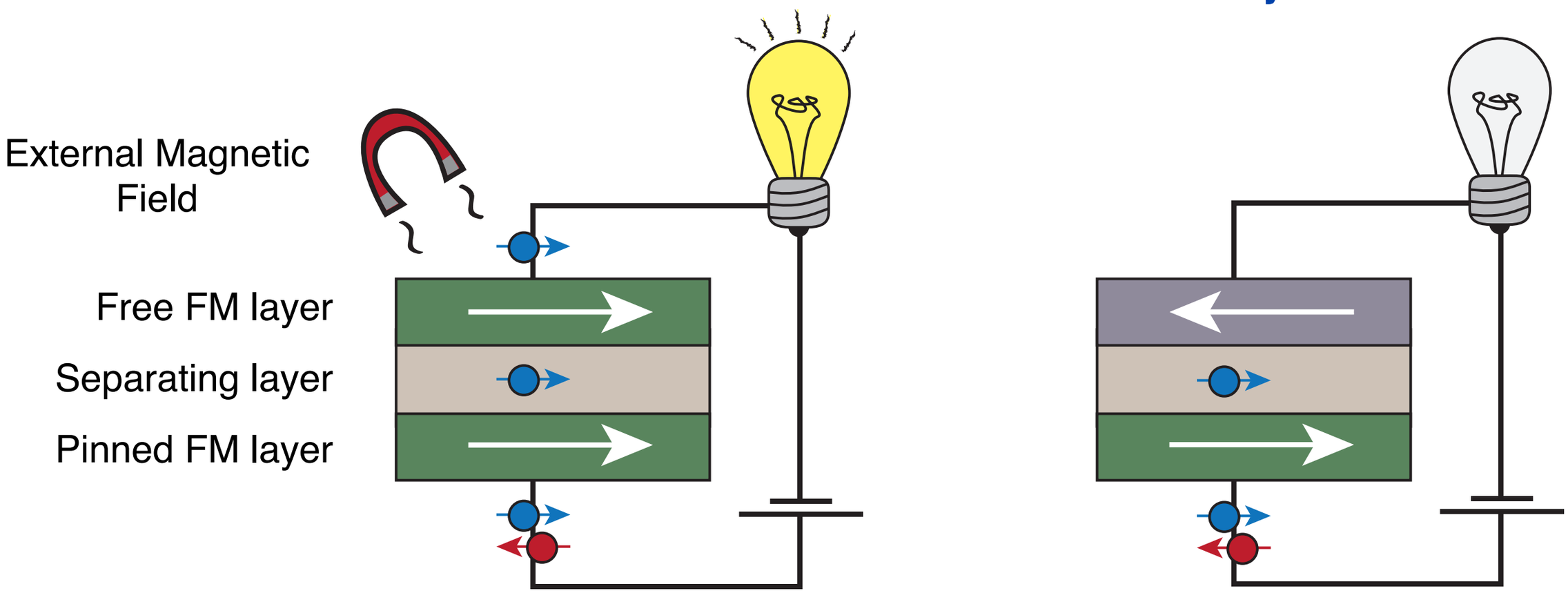
Giant Magneto Resistance Device / Spin Valve

Used since 1997 by IBM in read heads



Parallel Mode

Small Resistance

Antiparallel Mode

Large Resistance

2007 Physics Nobel Prize

Albert Fert et Peter Grünberg

P. Grünberg, et. al *Phys. Rev. B*, **1989**, *39*, 4828-4830

A. Fert, et. al *Phys. Rev. Lett.*, **1988**, *61*, 2472-2475

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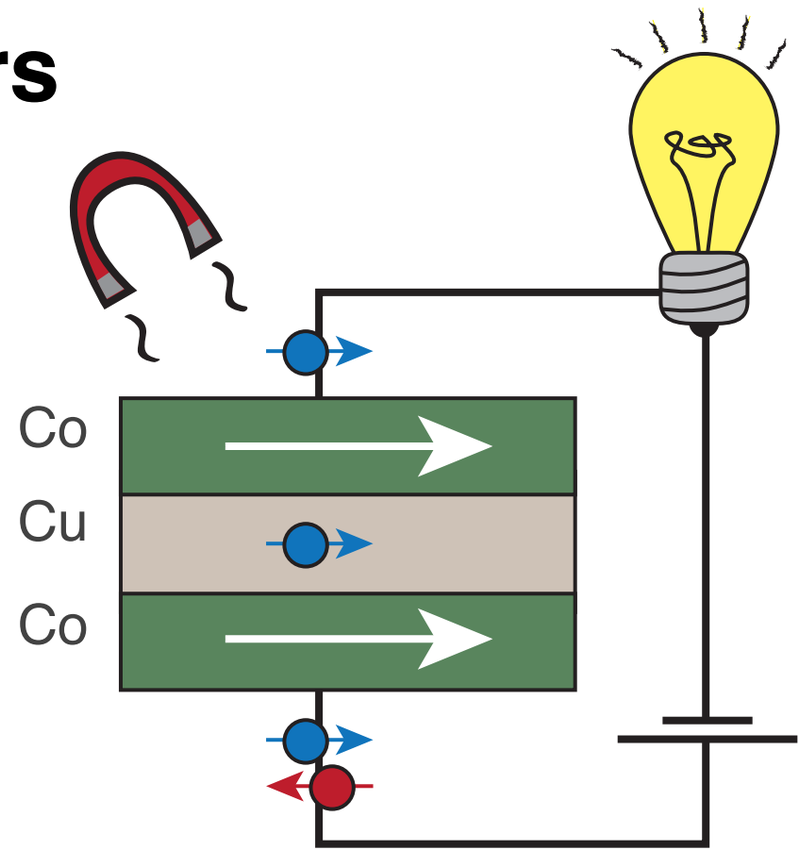
quand le dispositif est dans le mode antiparalleele, les deux electrodes FM sont polarise de facon opposee, donc les electron polarise en spin comme l'electrode du bas peuvent passe, mais ils sont bloquee par l'electrode du haut et le courrant ne passe pas. quand le dispositif est dans le mode parallel les deux electrodes sont polarise de la meme facon donc les electrons avec le meme spin que les electrode donc il passe pas seulment a travers l'electrode du bas, mais ausse il passe par l'electrode du haut, donc le courrant polarise en spin passe par le dispositif.
il est possible de controle l'etat de polarisation de l'electrode FM par des champ magnetique exterieurs et faibles, d'ou le terme magnetoresistance.
cette technologie était employé dans les disk dure a partir de 1997 et en 2007 le prix nobel était attibue a albert fert et péter grunberg pour leur découverte

Introduction

Spin transport through semi-conductors

- Metal based devices do not amplify the signal
- Fabricate spin-based transistors



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Les dispositif actuellement employes sont fabriques à paritre de metaux. quand la couche separatrice est un metal ou un isolant, il n’est pas possible de module le courant. Les dispositif à base de metaux sont des excellent transporteur de courant, mais ils ne amplifient pas le signal. Donc si on peuvait incorpore des semi-conducteurs dans ces dispositifs, il sera possible de amplifie le signal, et de genere des transitors à base de spin.

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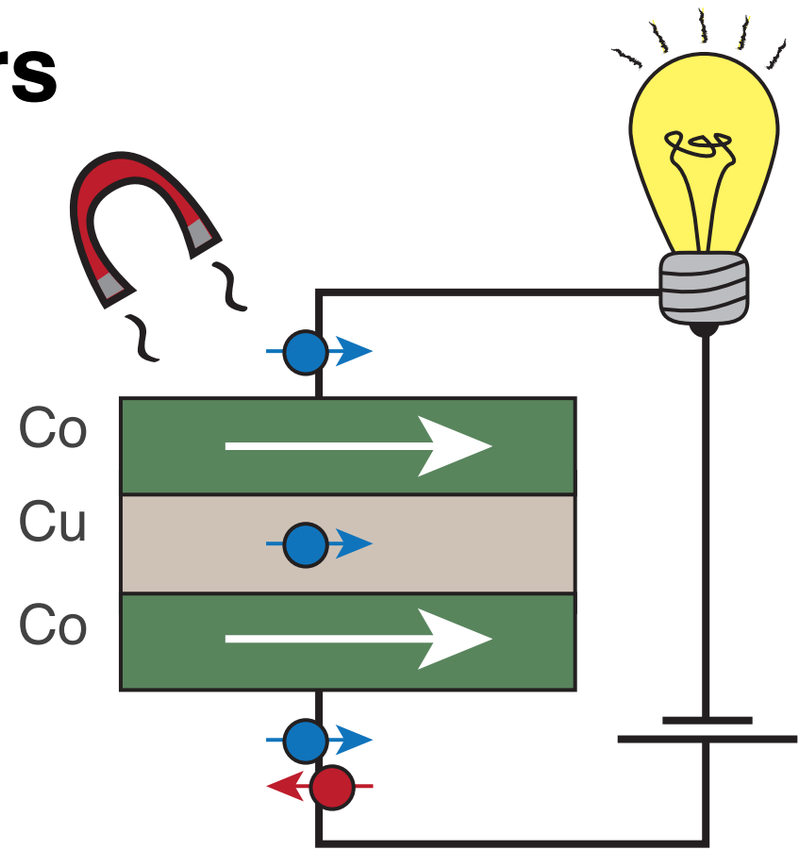
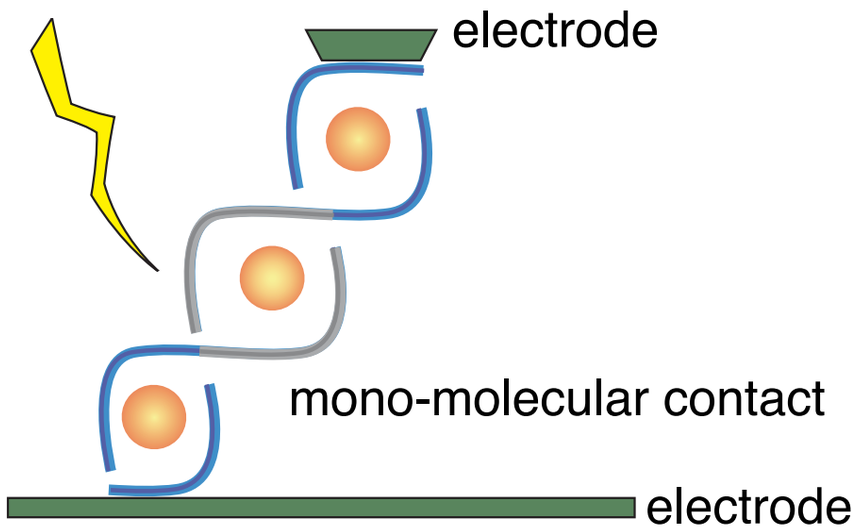


Spin transport through molecular components

- Smaller devices
- Functionalize molecules ⟹ **MODULATE**
- Long spin relaxation time



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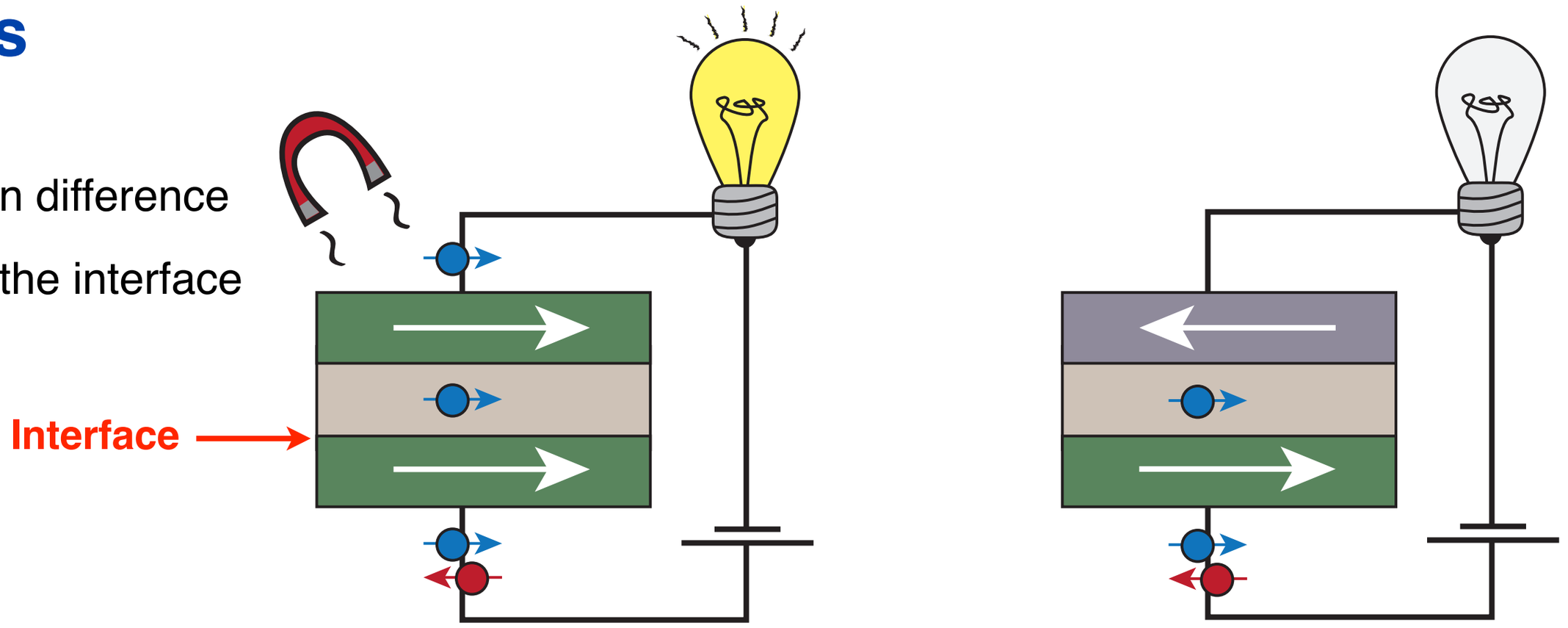
L'idee du projet est de fabrique des dispositifs à partir de semi-conducteurs moleculaires. pour quoi moleculaires? parce que ca permet de fabriquer des dispositif plus petit. parce que on peut functionalize les molecules donc on peurra controller le dispositif par des perturbations autre que un champ magnetique. et parce que dans les materiaux organique le temp de spin-relaxation tres long donc les electron n'on pas le temp de changer de polarization dans la couche separatrice ce qui donne un meilleure rendement au dispositif.

Introduction

Problems

- Conduction difference
- Quality of the interface



V. A. Dediu *Nat. Mater.*, **2013**, *9*, 210-211

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Mais quand on veux utilise des composant moleculaire dans des dispositifs spintronique on rencontre des problèmes: les plus importants est que le mechanism de injection des spins dans les couches semiconductrices n'est pas bien compris, que il y a une grand difference de conduction entre le metal ferromagnetique et la couche semiconductrice. et que l'interface entre le FM et la couche separatrice est crucial pour le bonne fonctionnement du dispositif. les deux solutions trouve jusqu'a present sont: l'introduction de une couche isolant qui permet au dispositif de fonctionner par effet tunnel. et l'utilisation de demi-metaux comme le LSMO au lieux de métaux semiconducteurs, qui permet une meilleure superposition avec une couche separatrice semiconductrice.
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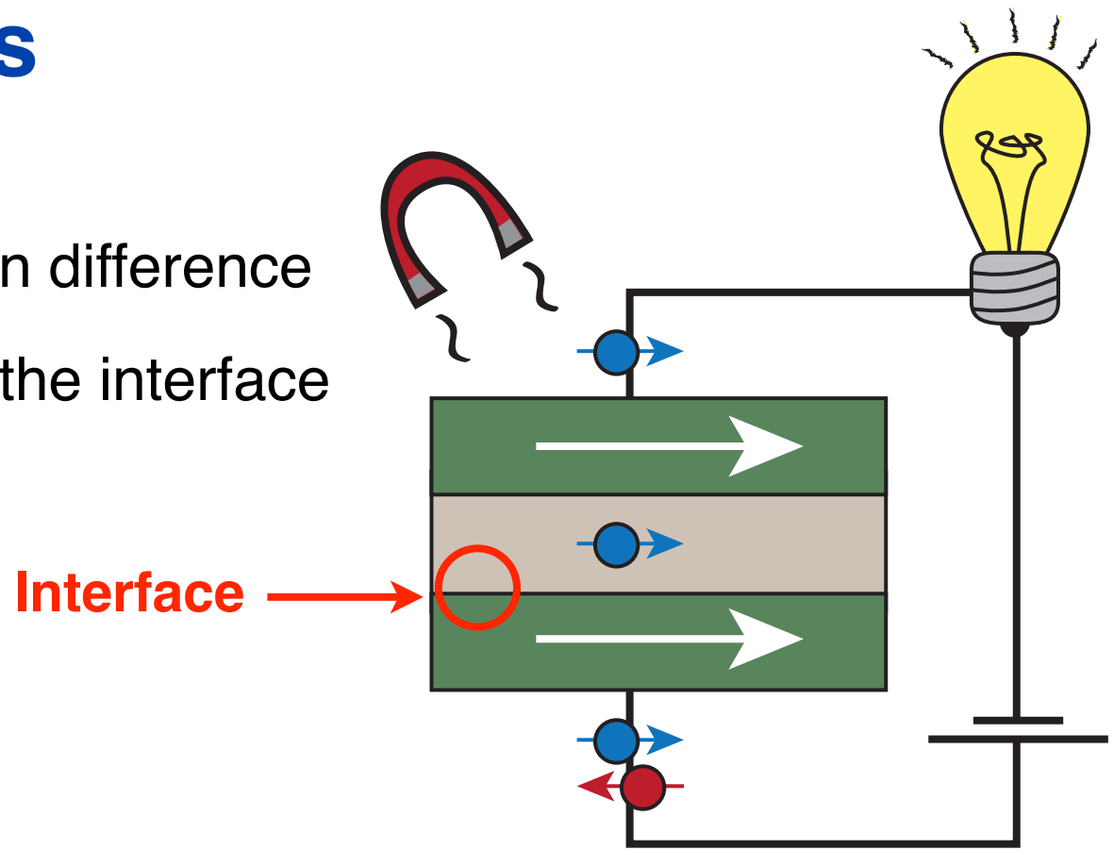

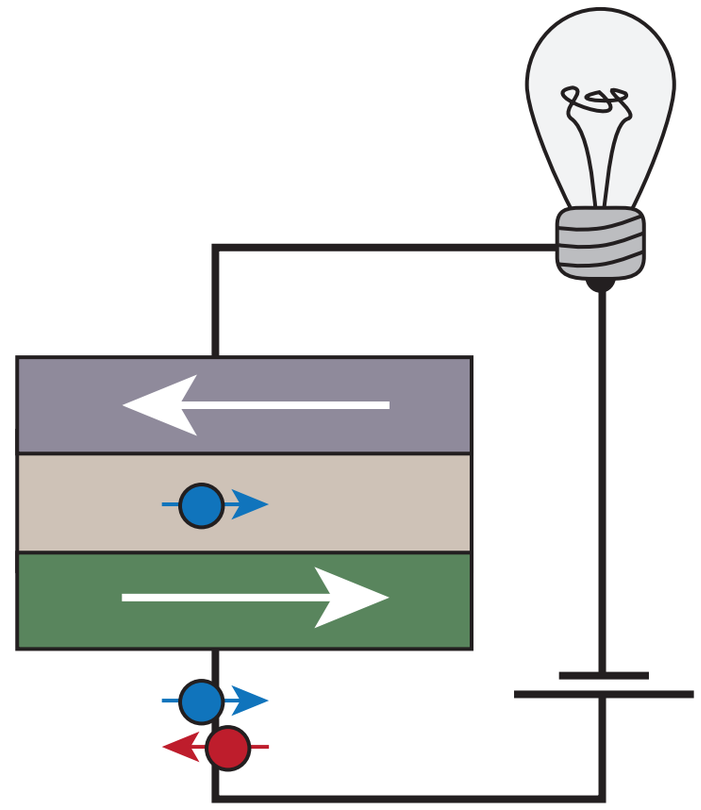

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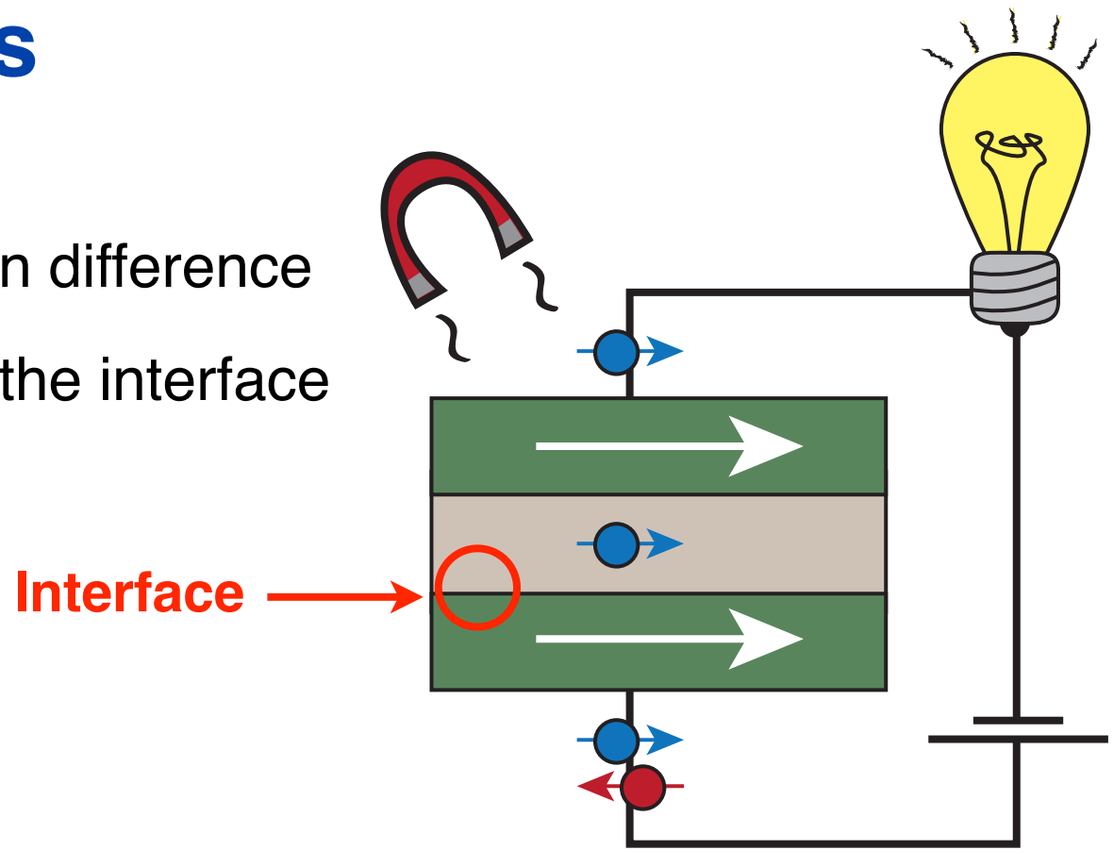

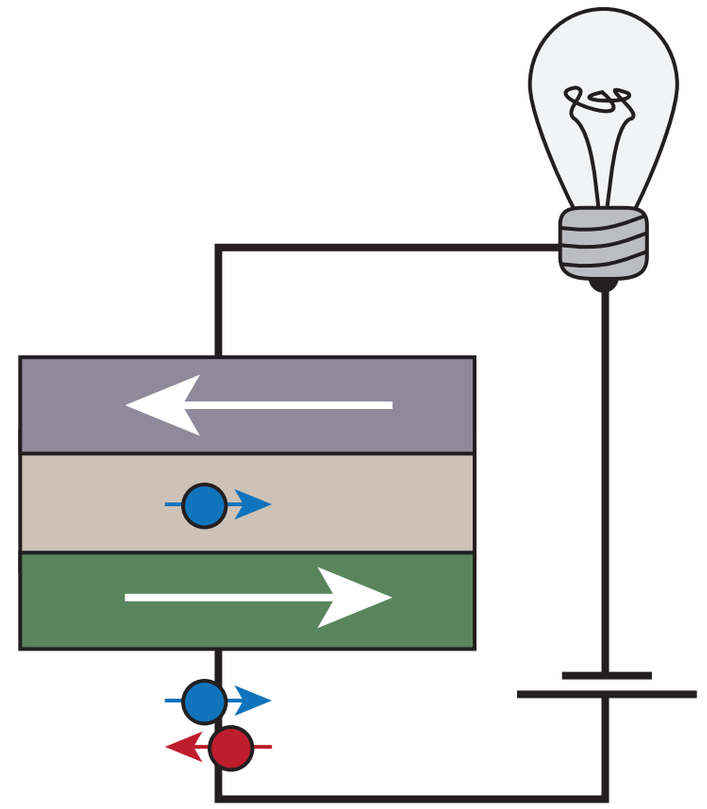

Solution

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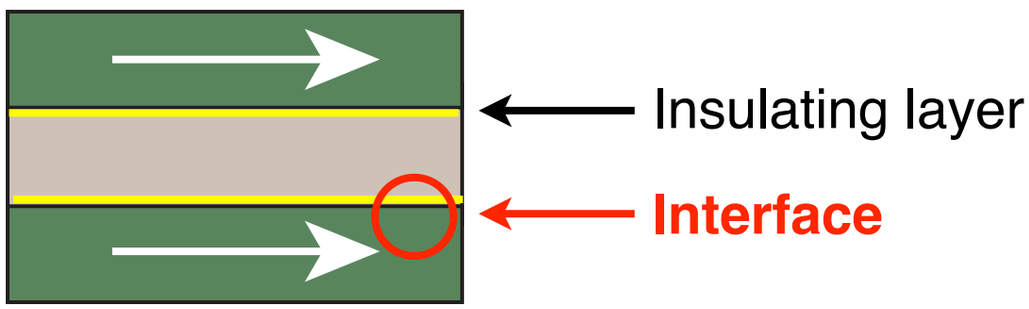

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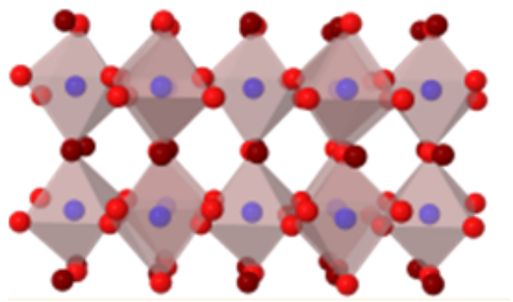
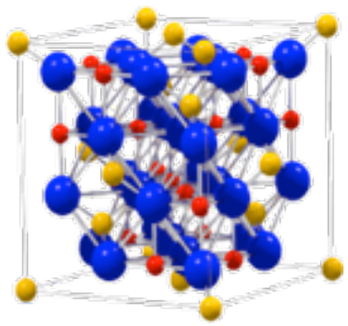


$LaSrMnO_3$ (LSMO)



Fe_3O_4

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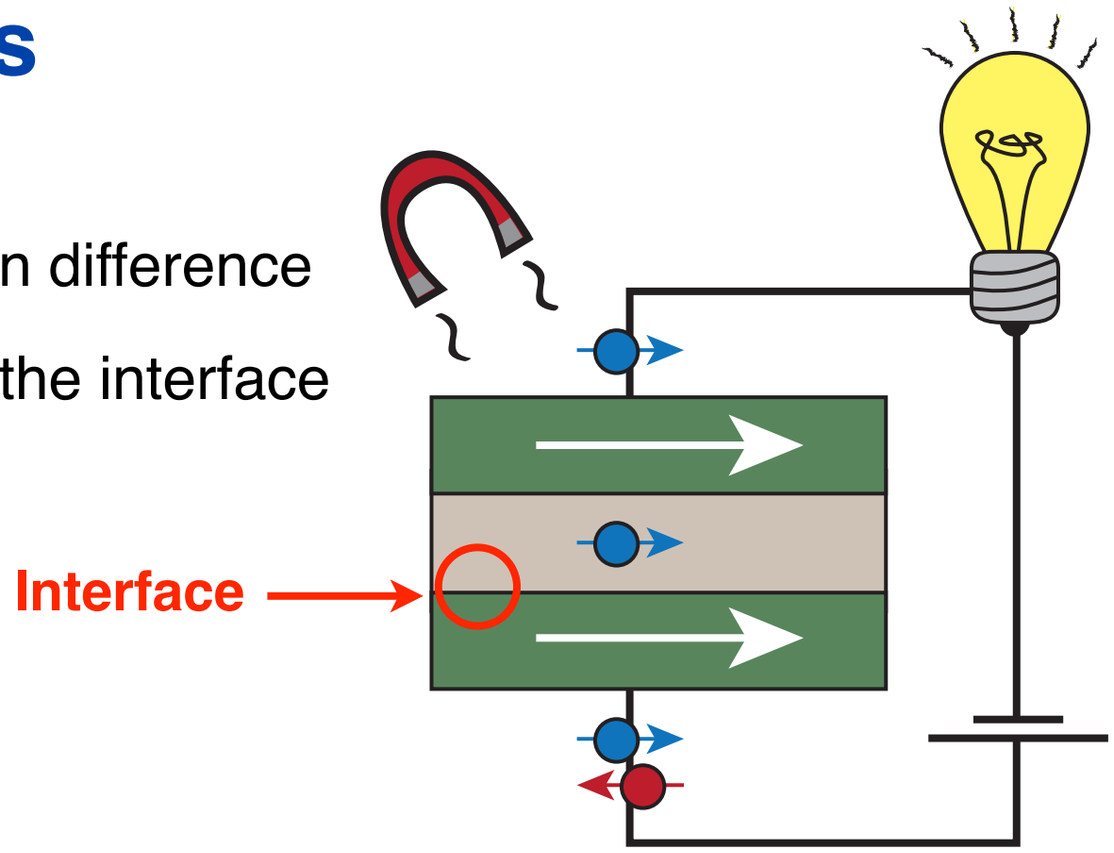

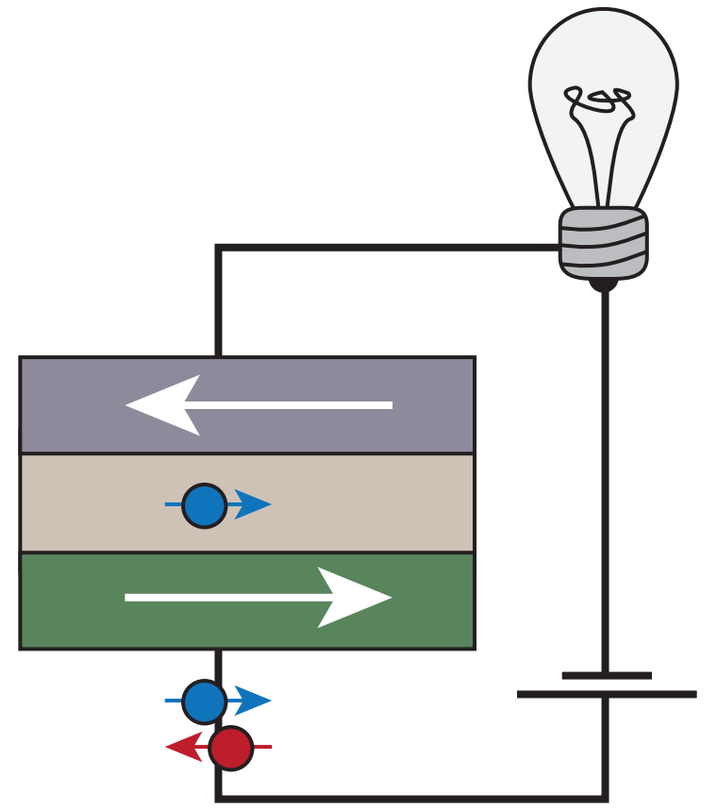

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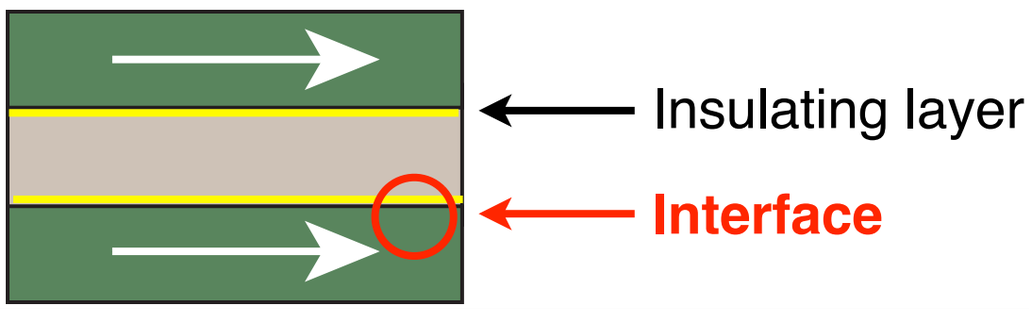

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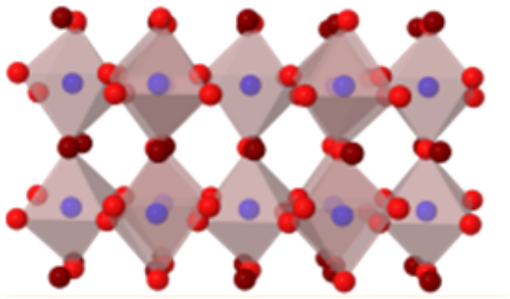

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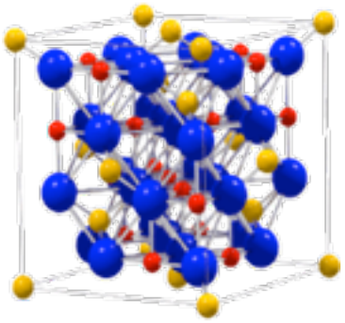

New Functional Materials
control over the growth of the separating layer

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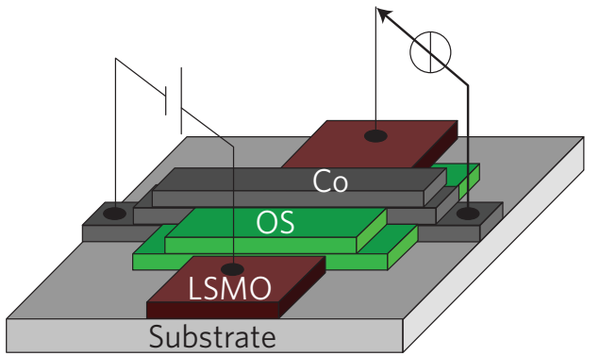
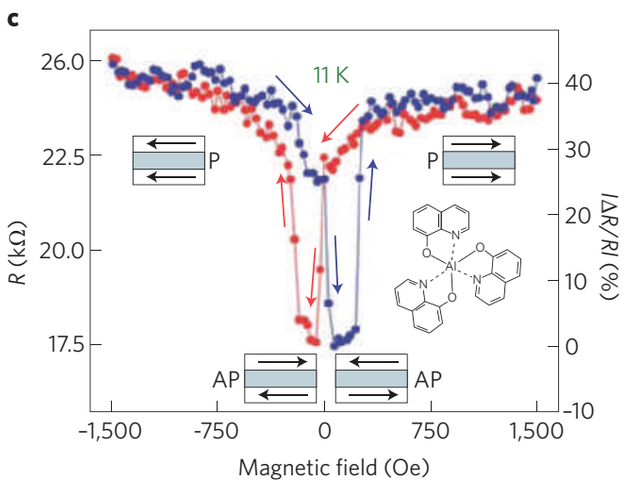
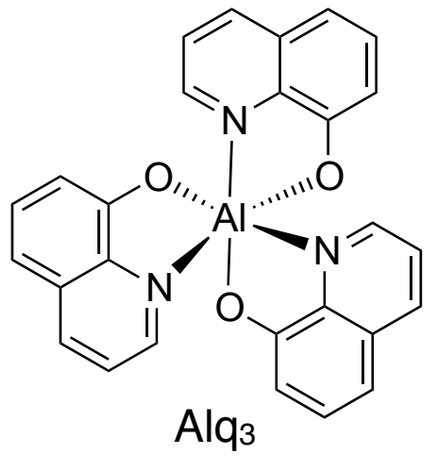
Towards molecular spintronics



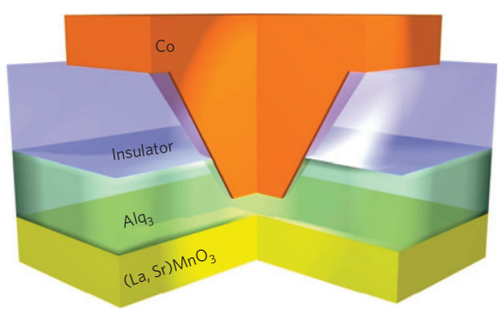


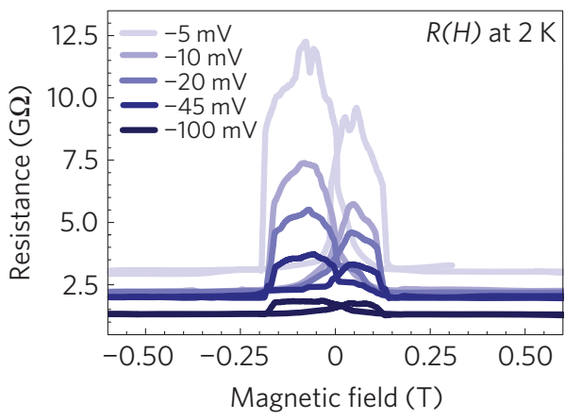


Alq_3

not reproducible

V. A. Dedieu et al., *Nat. Mater*, **2009**, *8*, 707-716





C. Barraud, et al., *Nat. Phys*, **2010**, *6*, 615-620

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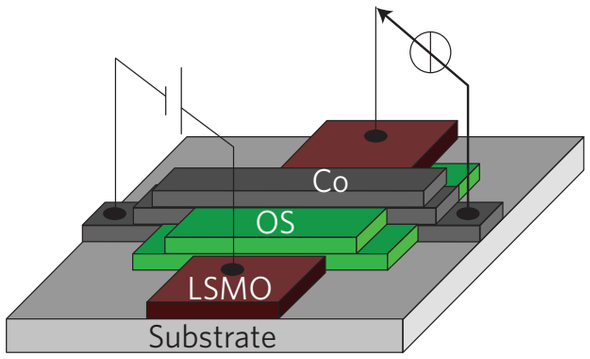
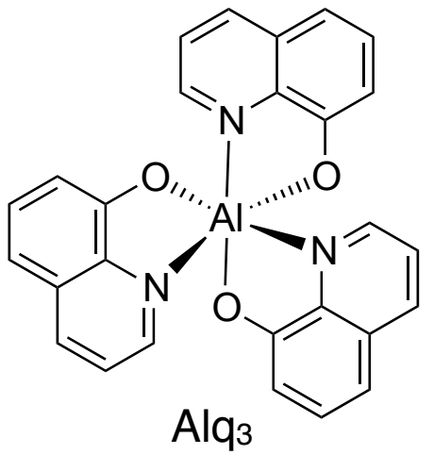
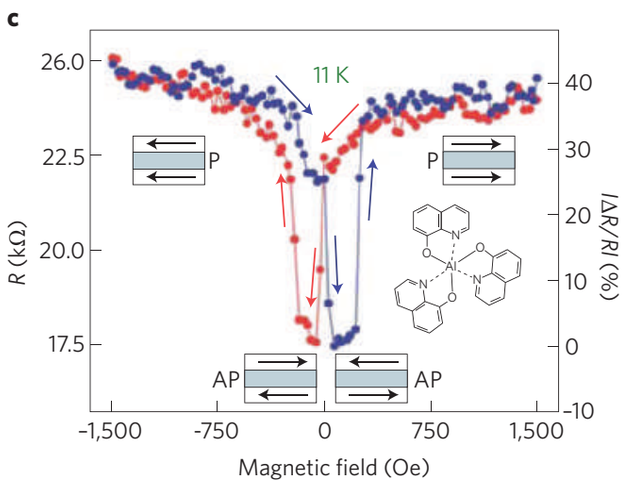
in this slide I will show you some examples of efforts undertaken towards molecular spintronic devices
here on the left is probably the first example of a hybrid spin valve. in both studies the spin valve is composed of an lsmo bottom electrode, an cobalt top electrode and an Alq3 separating layer
similar experimental setups were used but in one case a negative magnetoresistance was seen and in the other a positive one
this phenomena was attributed to the quality of the interface, which dictates the device response

Introduction

Towards molecular spintronics







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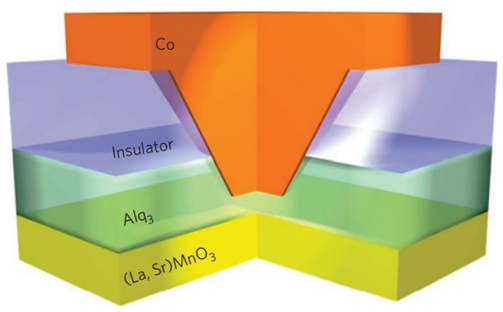

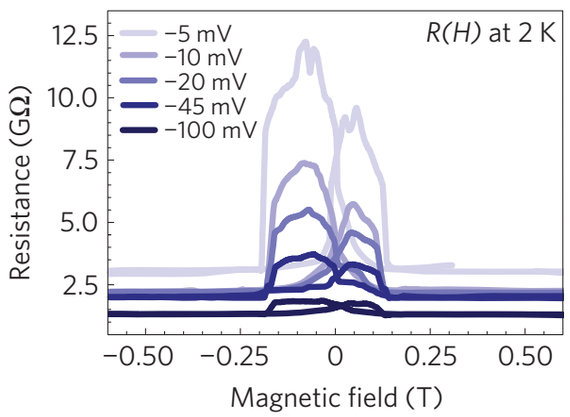

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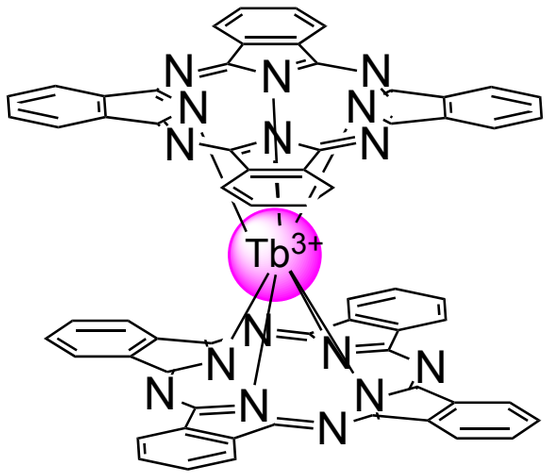
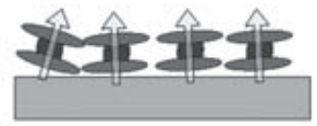
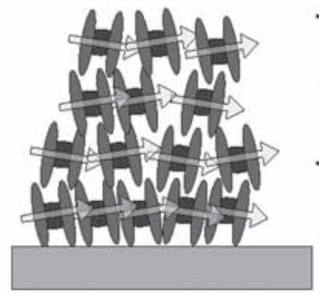


different orientation
different response





L. Margheriti et al.
Adv. Mat., **2010**, *22*, 5488-5493

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in this example a single molecule magnet was used. the authors showed that the orientation of the molecule varied according to the evaporation condition used.
a different response was obtained when the molecules were in plane or out of plane

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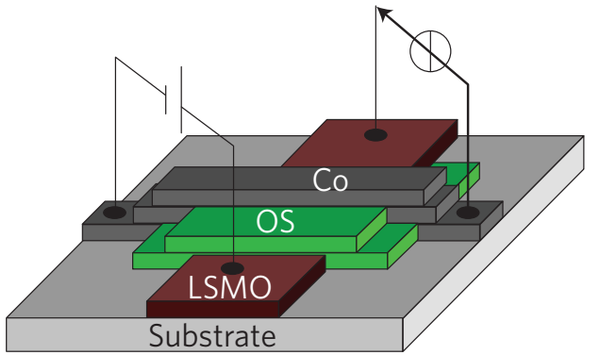

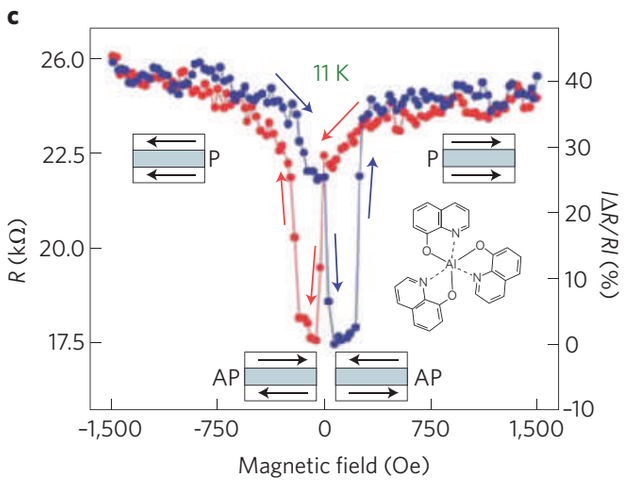

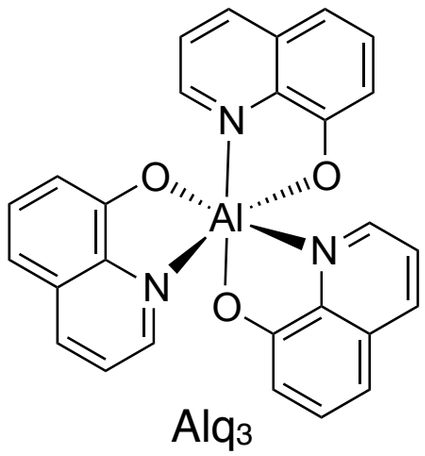

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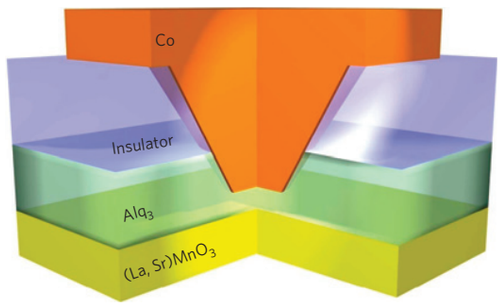

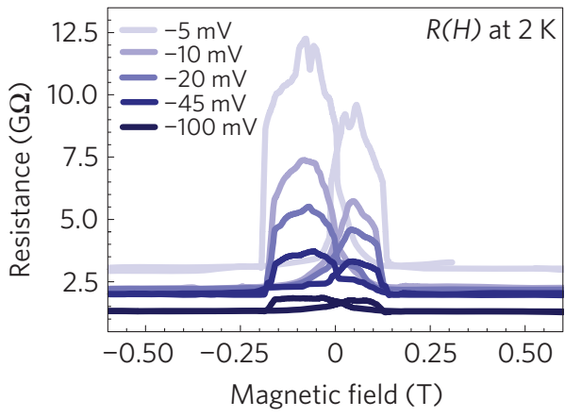

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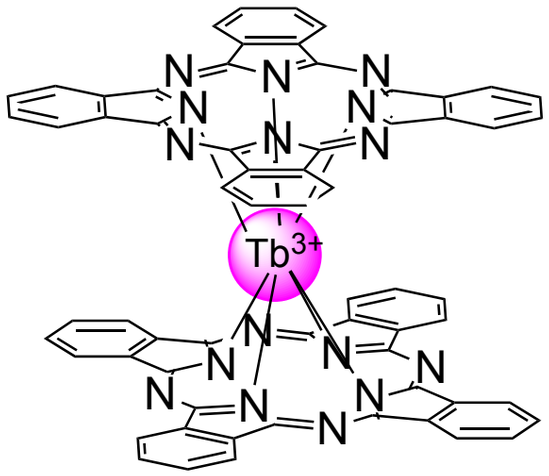

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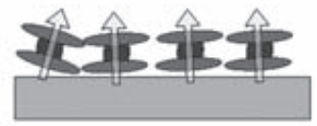

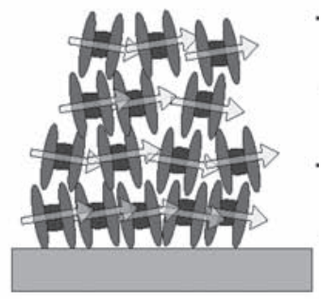

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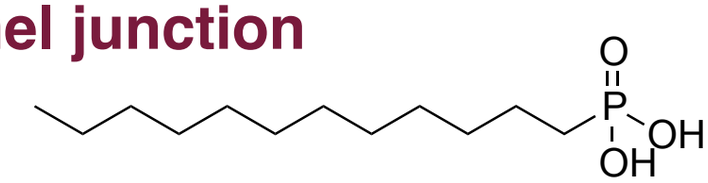
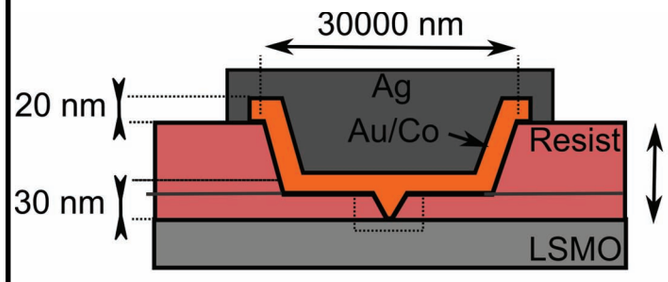
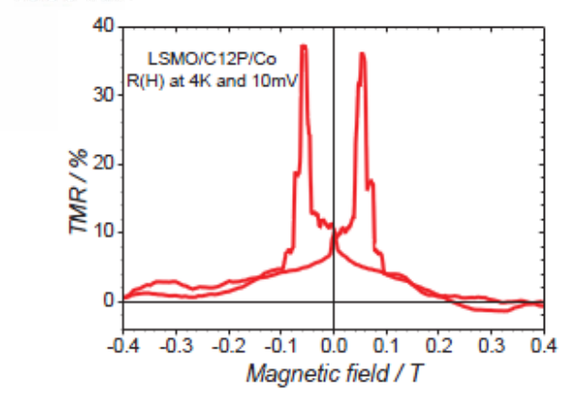
magnetic tunnel junction







1000 nm

S. Tatay et al.
ACS Nano, **2012**, *6*, 8753-8757

M. Galbiati et al.
Adv. Mat., **2012**, *24*, 6429-6432

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in this last example a self-assembled monolayer was grafted directly onto an lsmo substrate since the SAM is not conducting the device functions as a magnetic tunnel junction

First Objective

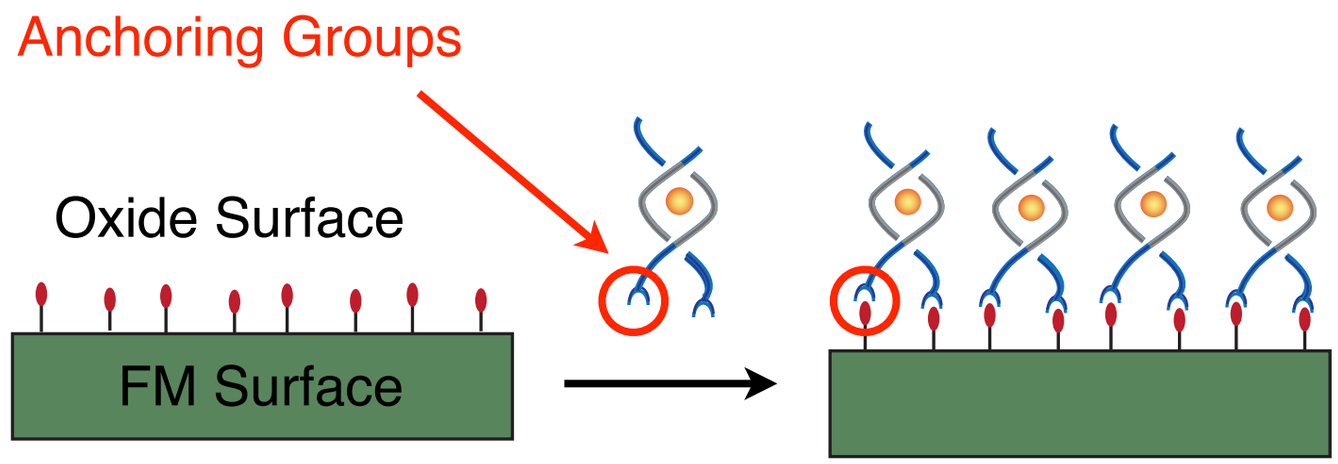
Control the magneto-resistance by controlling the interface

Wednesday, 11 June 14

le premiere objectif etait donc de controler la Magnetoresistance par le controle de l’interface

First Objective

Control the magneto-resistance by controlling the interface



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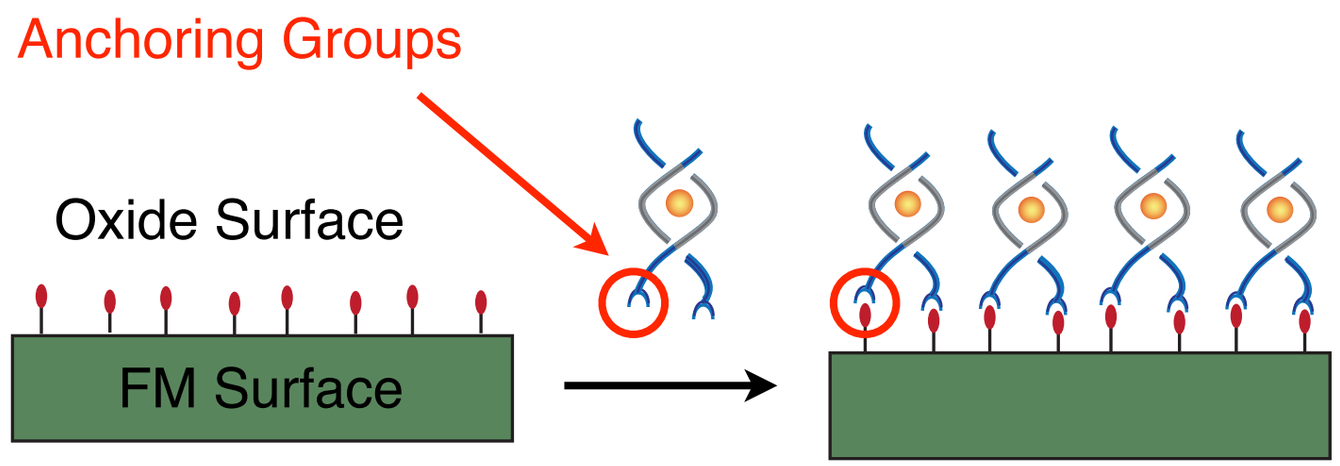
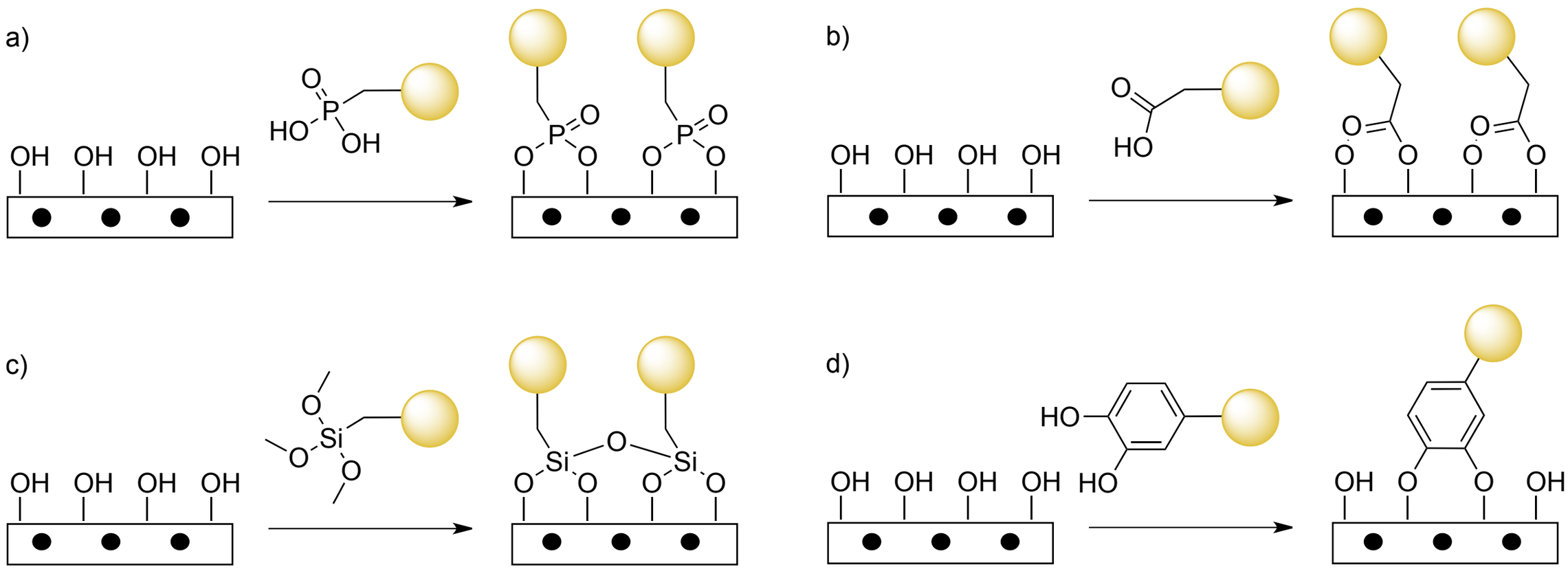
l'idee est de employer une monocouche de molecules qui pouront se lie directement sur la surface par une lieson chimique

First Objective

Control the magneto-resistance by controlling the interface



- Mn for LSMO
  Fe for Fe_3O_4



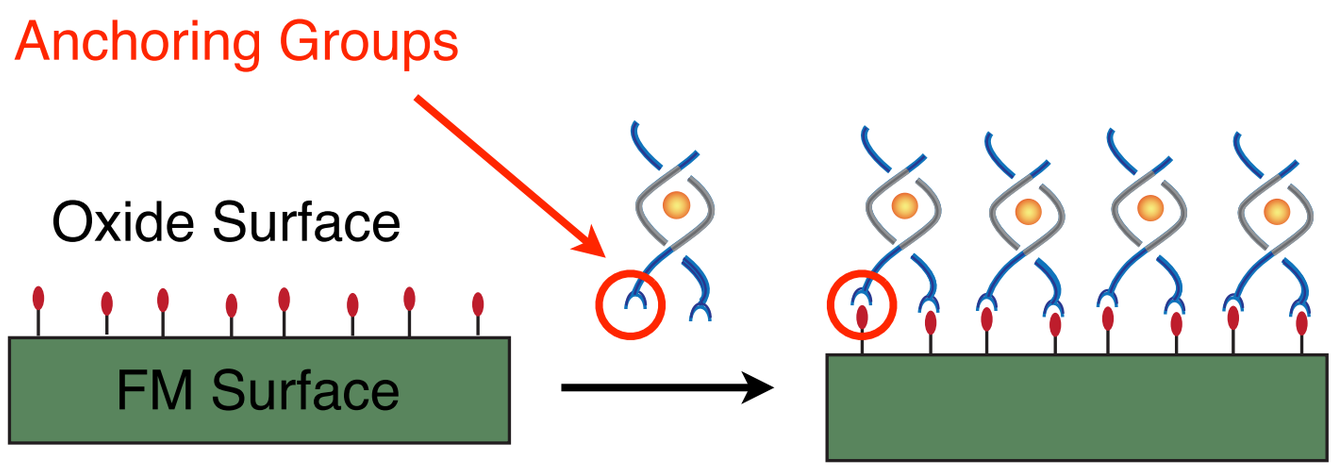
Wednesday, 11 June 14

pour ce la nous avons a disposition numbreux groups d’anchrache. nous avons coisi de comince nos etudes pas des phosphonates car il est connue de la litterature que les phosphonates se lie avec differents types d’oxydes

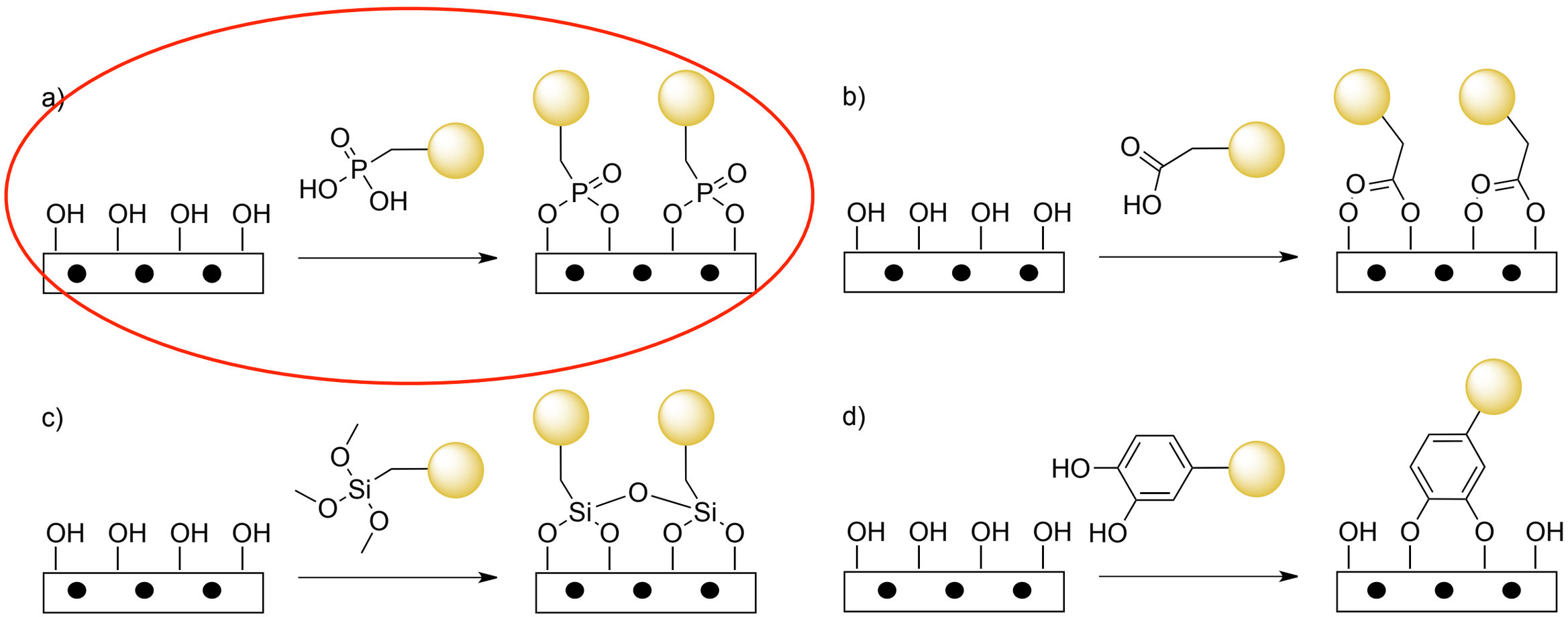
First Objective

Control the magneto-resistance by controlling the interface

ref for oxide surfaces



- Mn for LSMO
  Fe for Fe_3O_4

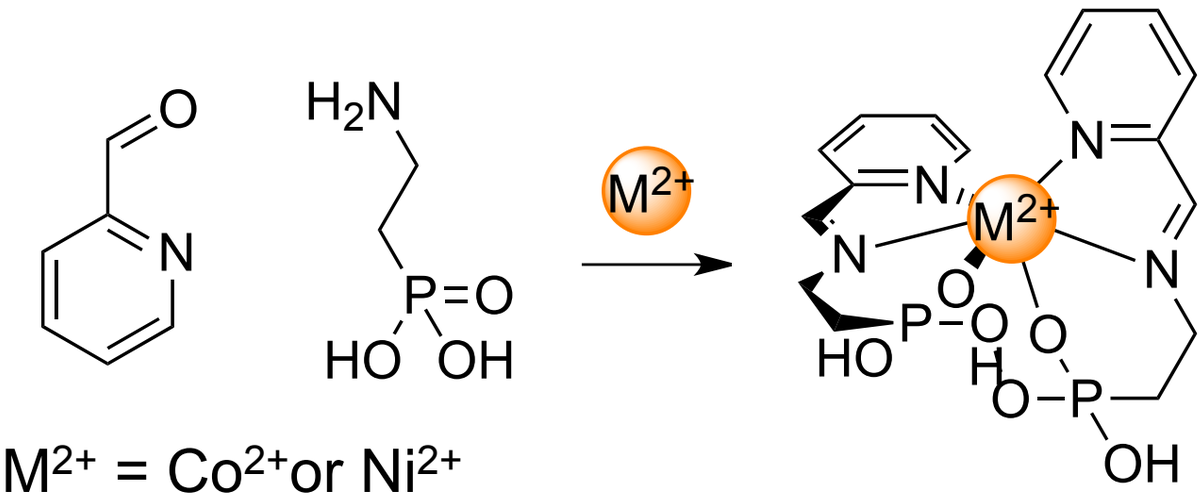


Wednesday, 11 June 14

pour ce la nous avons a disposition numbreux groups d'anchrache. nous avons coisi de comince nos etudes pas des phosphonates car il est connue de la litterature que les phosphonates se lie avec differents types d'oxydes

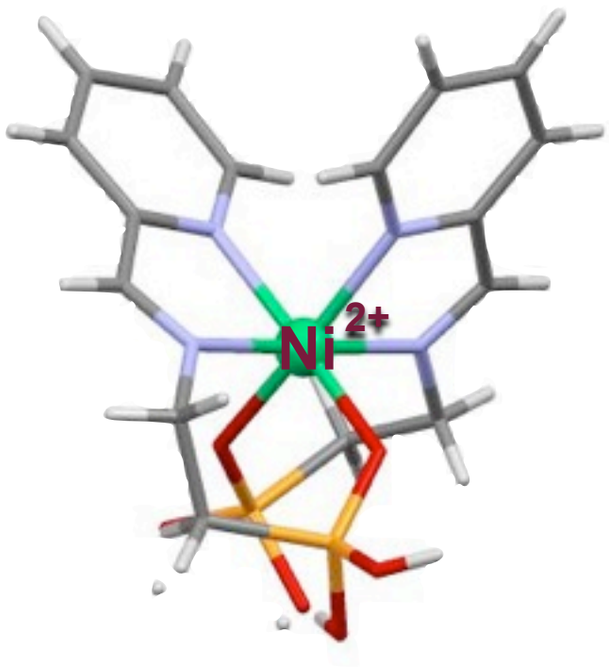
Molecular Spintronics

Synthesis

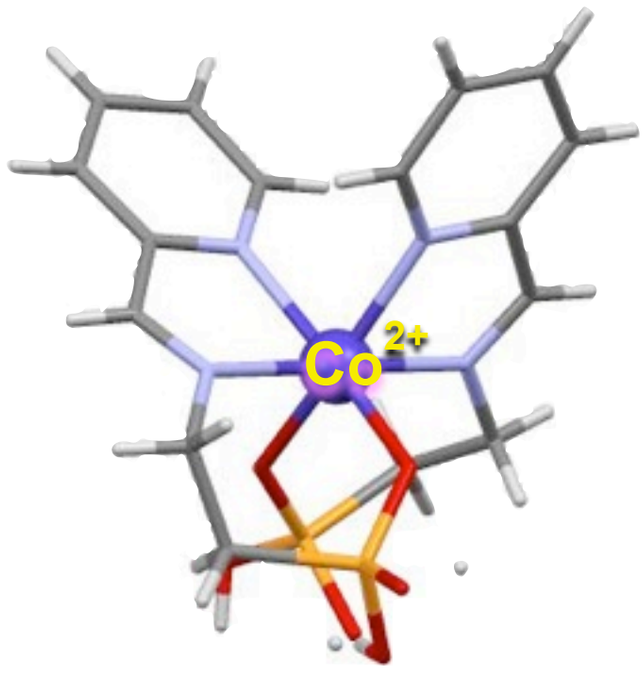


M^{2+} = Co^{2+}or Ni^{2+}

- Mild conditions
- High yields
- Stable in solution



Ni-pyPhos



Co-pyPhos

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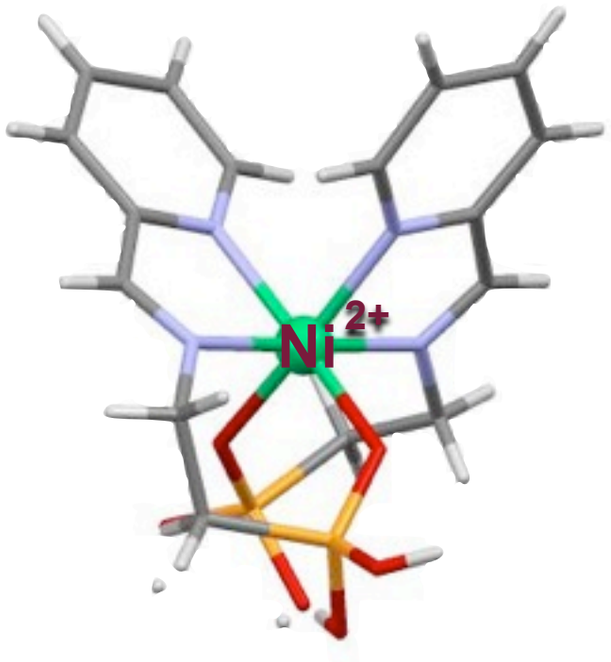
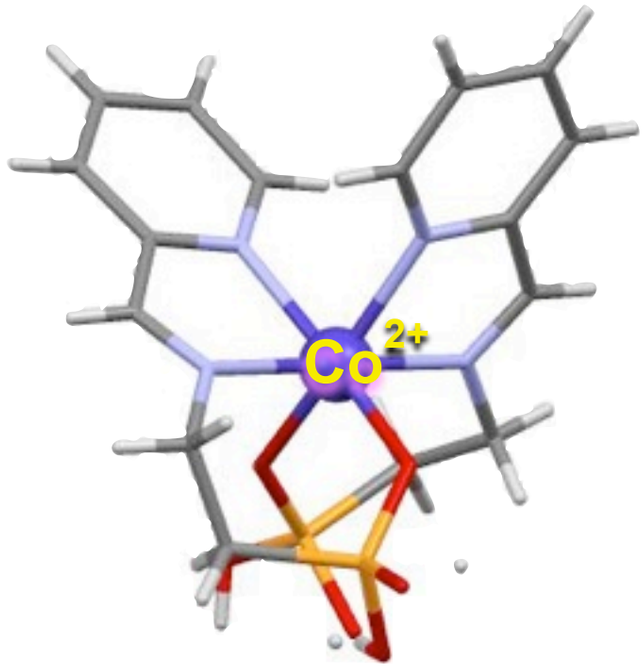
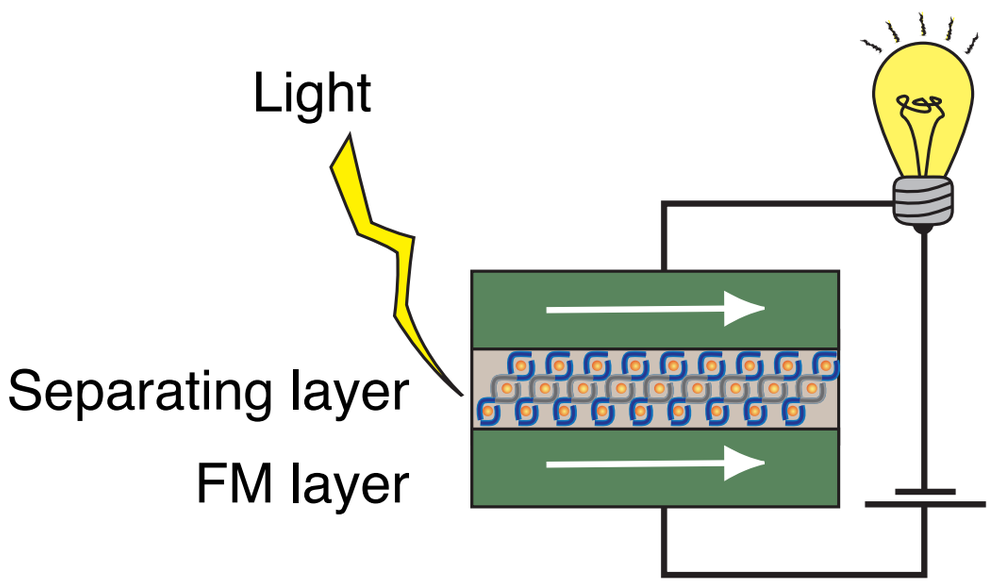
The first molecules synthesized were constructed via the subcomponent self-assembly technique
the simultaneous condensation in methanol of pyridine aldehyde with this phosphonate amine around a metal ion gave complexes of this type
shown here are x-ray crystal structures of the nickel and the cobalt complexes
the two complexes are isostructural

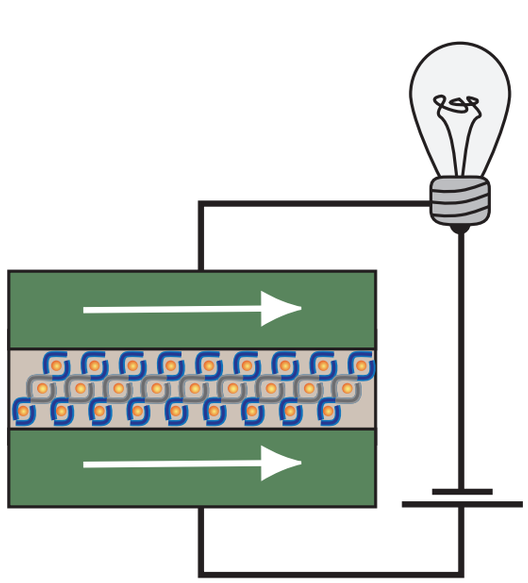
Molecular Spintronics





Why transition metal complexes?





- ***New Devices***
- ***Tuneable***

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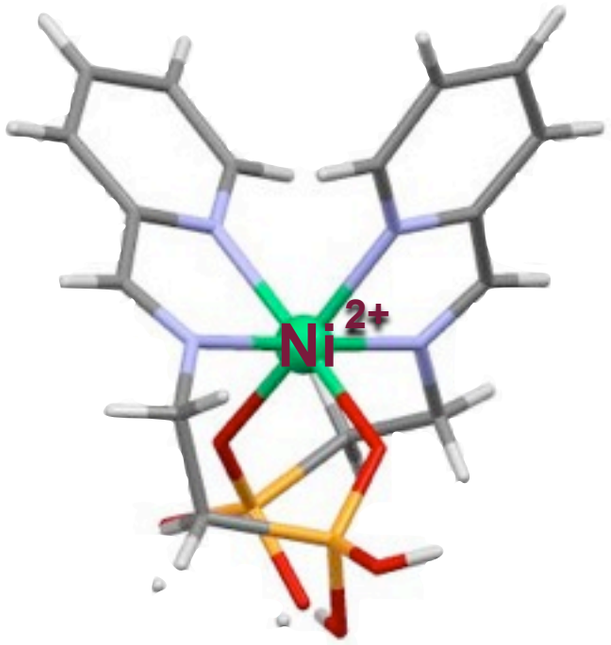
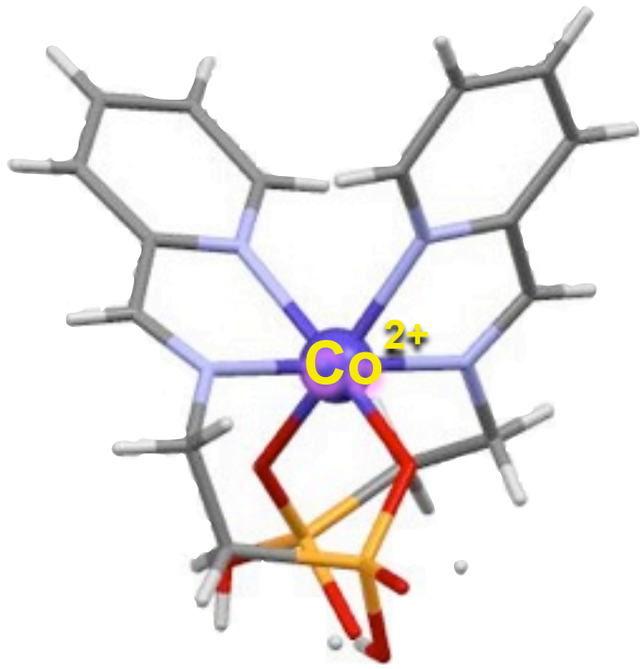
Pourquoi les métaux des transition? parce-que 1) dans les complex paramagnetic les orbitaux semi-occupe et les orbital les baisse vide sont des orbitaul d qui permet une meilleure superposition avec les électrodes ferromagnetiques. 2) il est possible de contrôler l'état de spin avec une perturbation extérieur comme par exemple la lumière ou le champ electrique 3) on a aces a un grand nombre des complex avec des propriétés électroniques différents, donc on peut contrôler la courant en spin en fonction de la couche séparatrice,

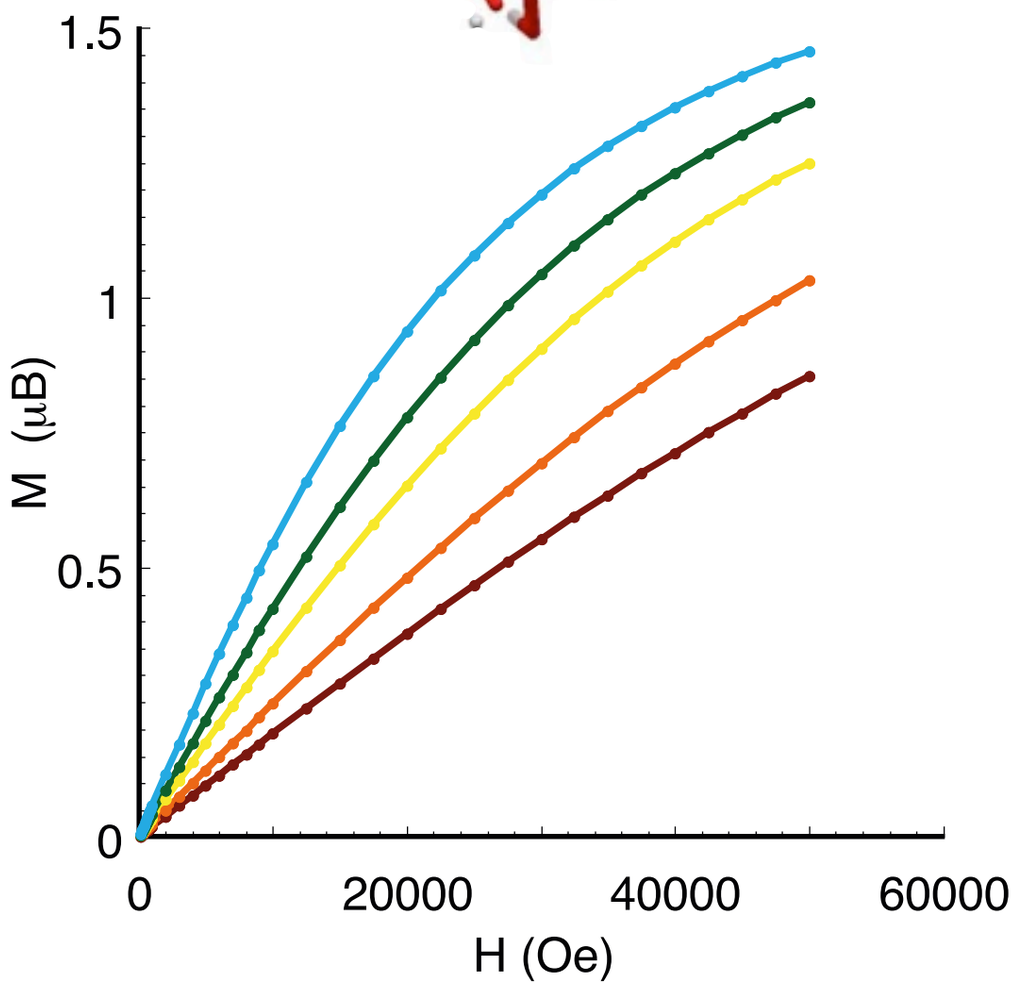
1) large number of molecules
2) semi-ocupied and lowest unocupied are usually d orbitals better overlap with fm contact
3) possible to tune by use of an external stimuli such as light

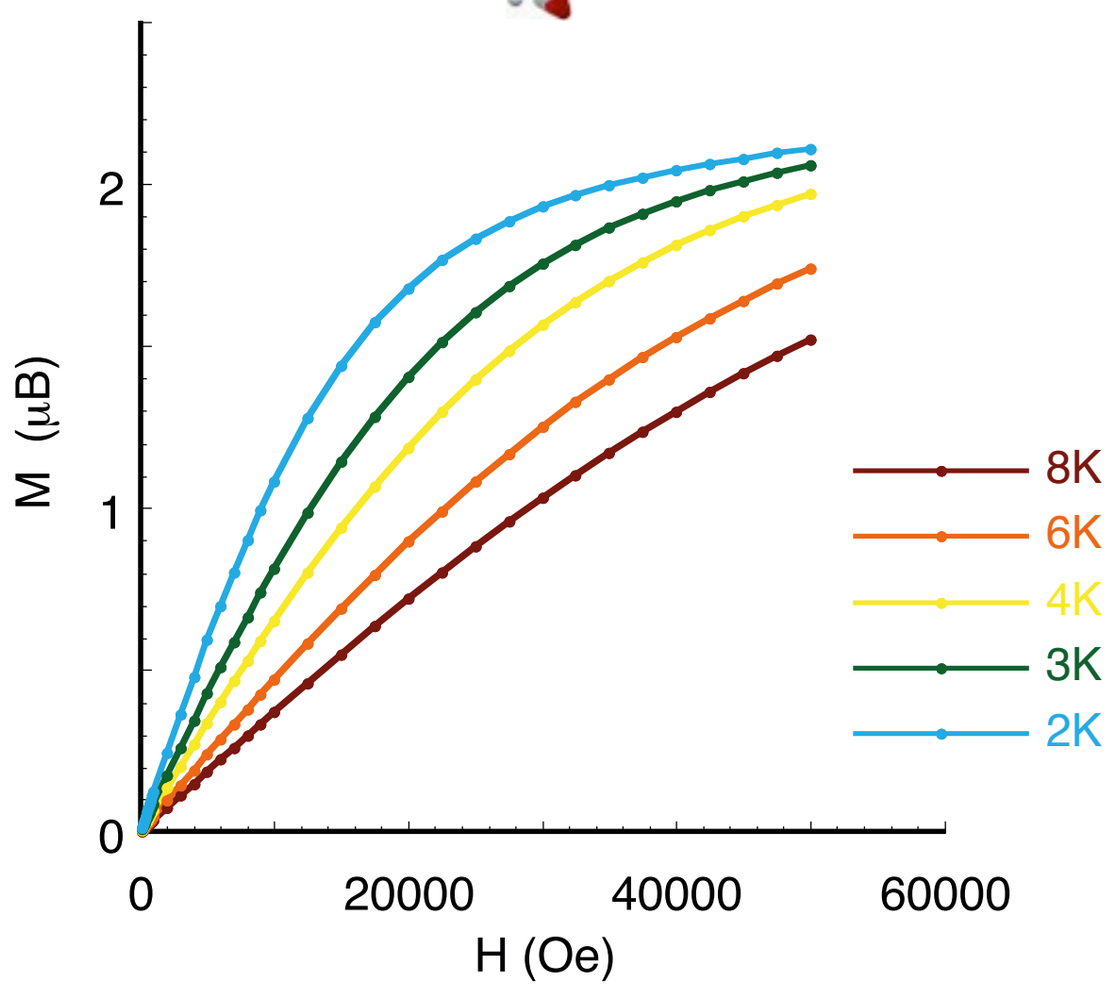
Molecular Spintronics

Magnetic Data







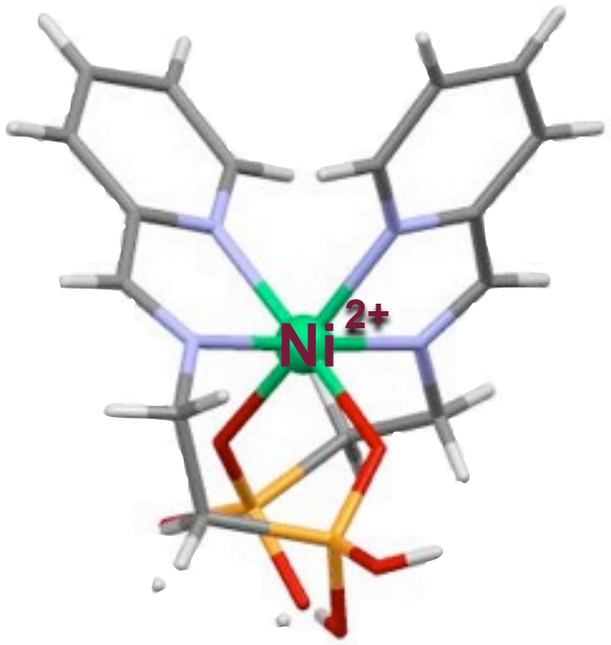


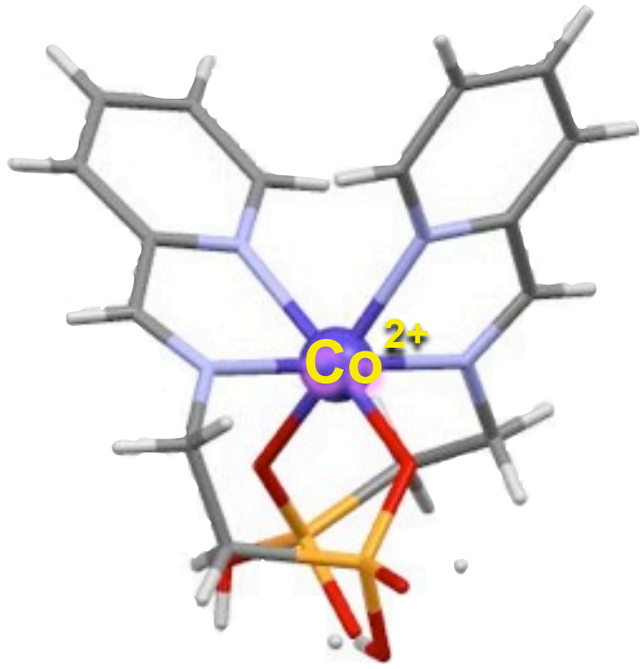
Wednesday, 11 June 14

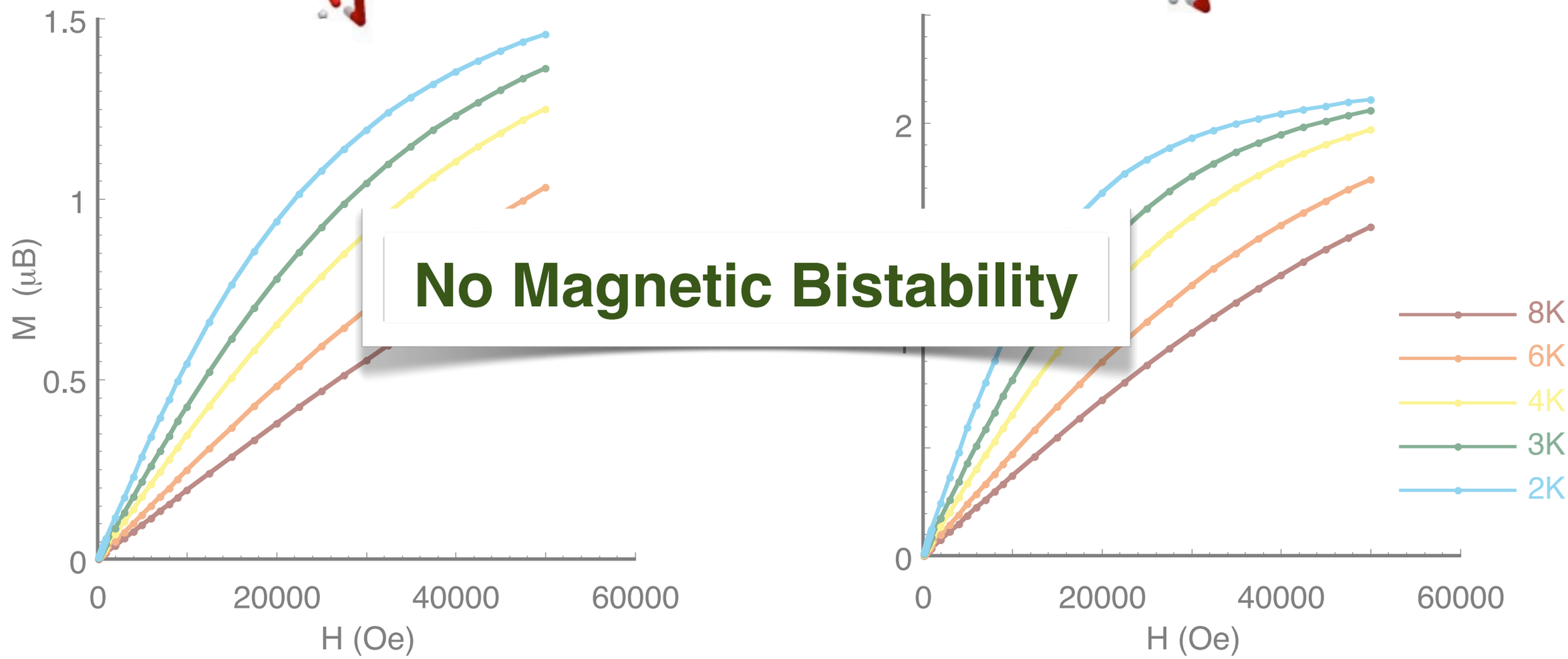
The different transition metal centers result in differenr magnetic properties.
shown here are the magnetization as a function of field plots taken at different temperature
these data tell us that these complexes are paramagnetic

Molecular Spintronics

Magnetic Data







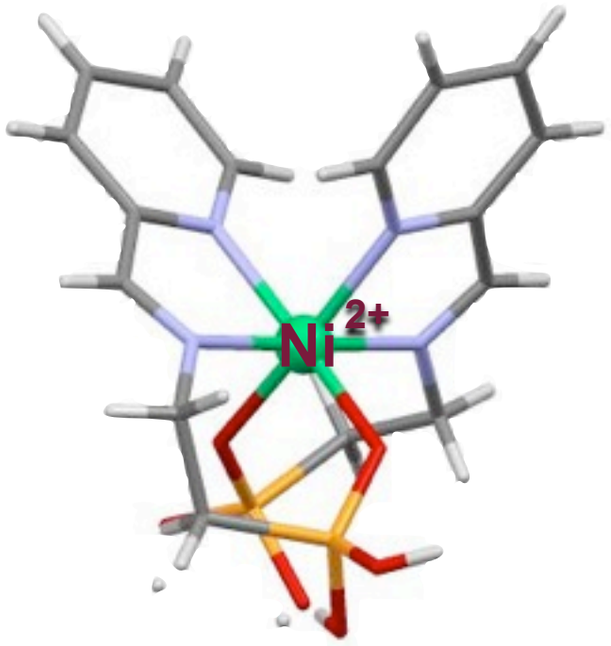
No Magnetic Bistability

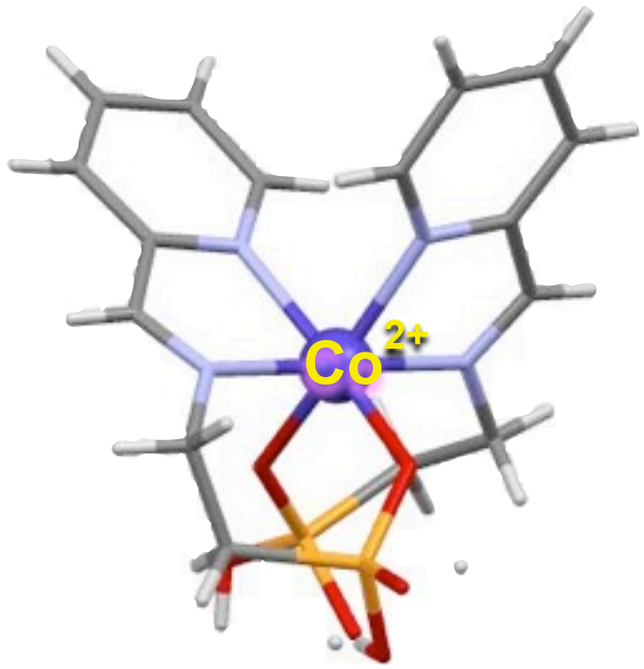
Wednesday, 11 June 14

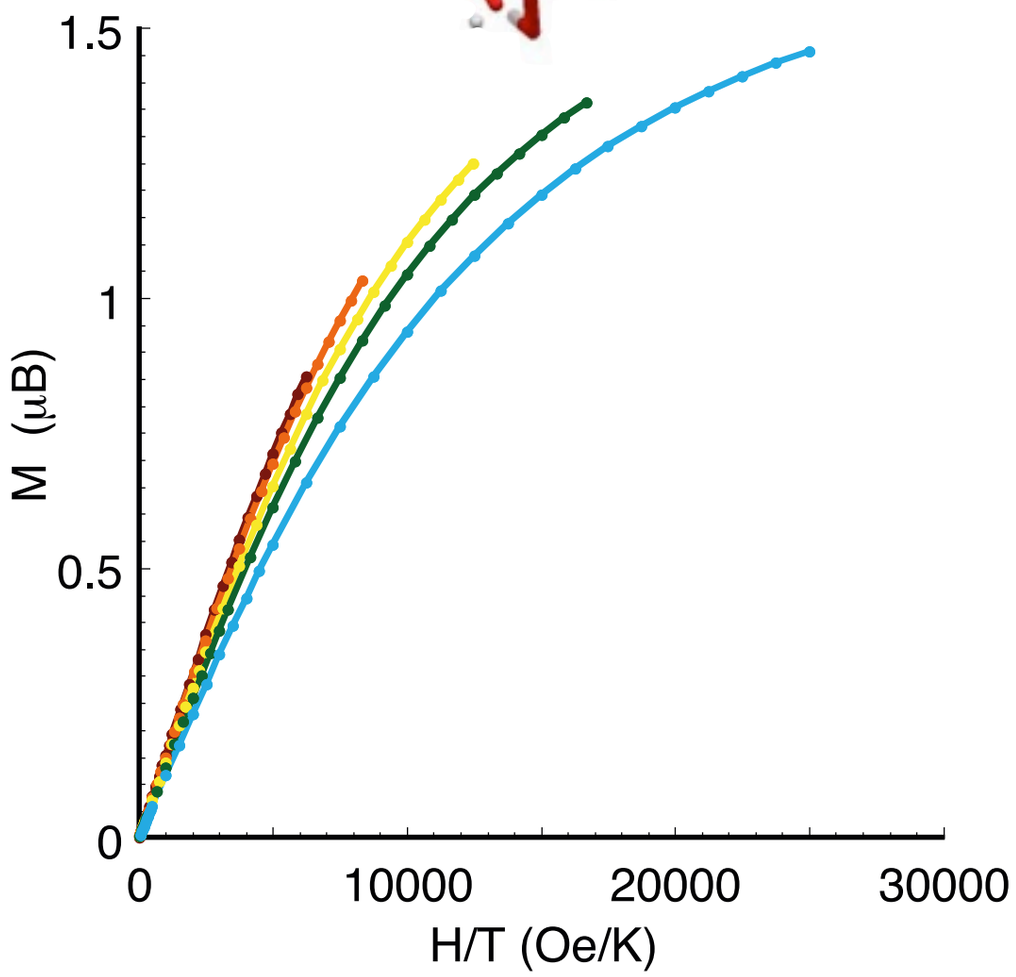
The different transition metal centers result in differenr magnetic properties.
shown here are the magnetization as a function of field plots taken at different temperature
these data tell us that these complexes are paramagnetic

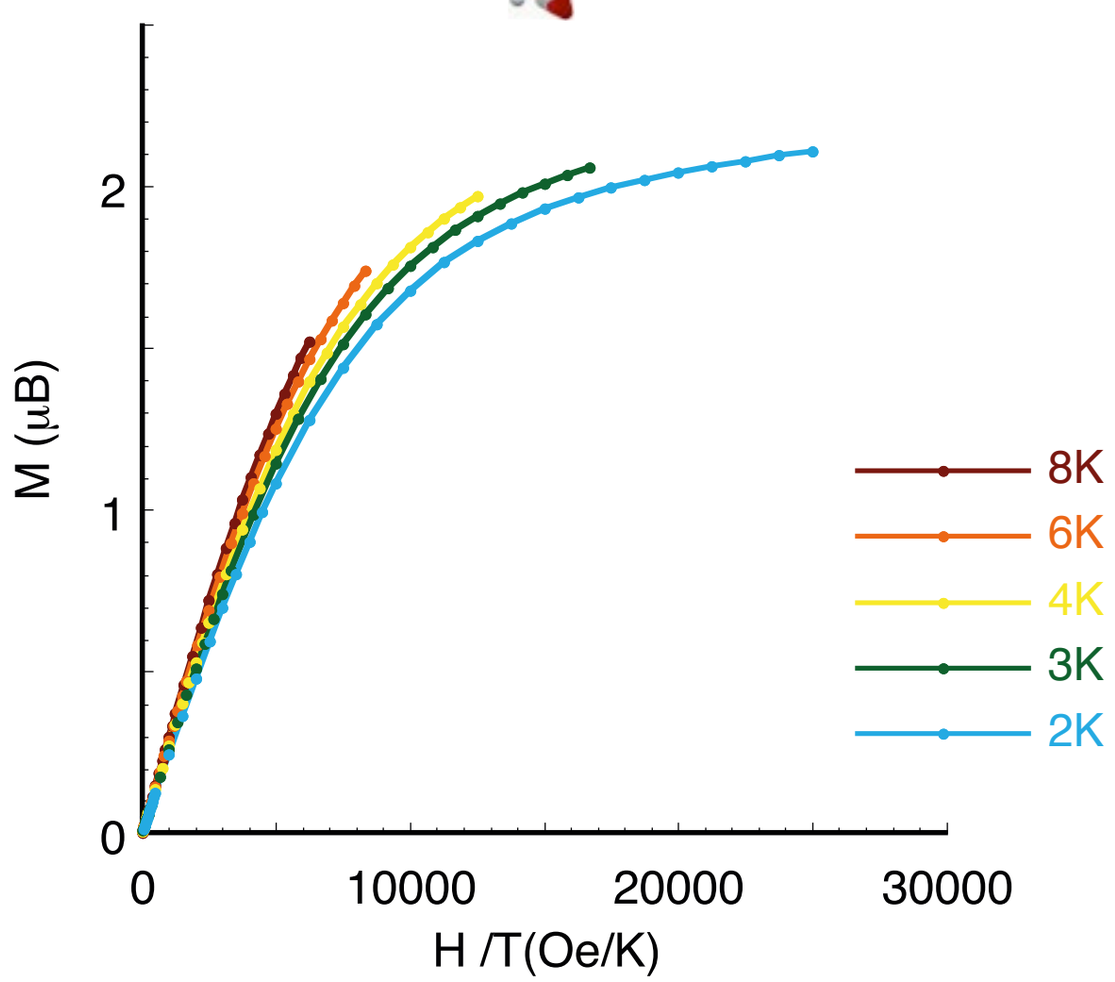
Molecular Spintronics

Magnetic Data









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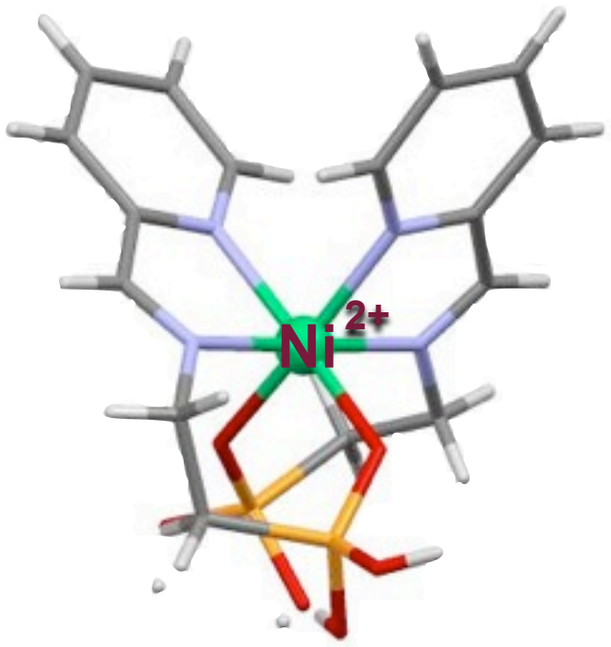
It is possible to get a preliminary sense of the nature of the anisotropy of mononuclear transition metals by plotting the magnetization as a function of the field divided by the temperature at different temperatures

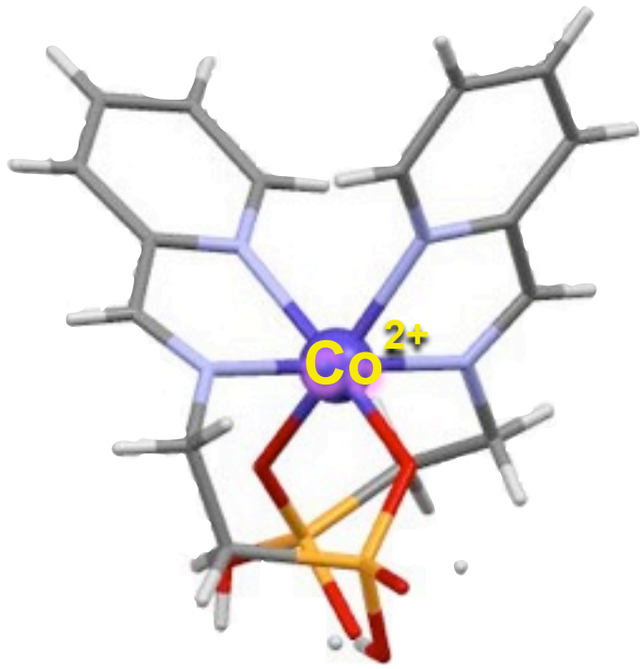
if the plots do not overlap the complexes have a magnetic anisotropy axes

Molecular Spintronics

Magnetic Data



Magnetically Anisotropic



D = 3 cm^{-1}

D > 30 cm^{-1}

ab initio calculations

LCPQ, Université Paul Sabatier, Toulouse France

- Dr. Nathalie Guihery

SQUID

Université Paris Sud, Orsay France

- Dr. Eric Riviere (SQUID)

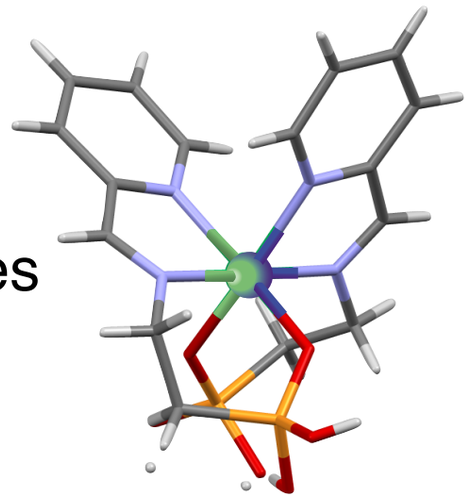
Wednesday, 11 June 14

it is possible to extrapolate the zero field splitting parameter for the Ni complex and it was found that the D value was around -6
this result was further confirmed by high field EPR

Molecular Spintronics

Self-Assembly on Surfaces

Co- or Ni-complexes



+

LSMO or Fe_3O_4

concentration of sulutions

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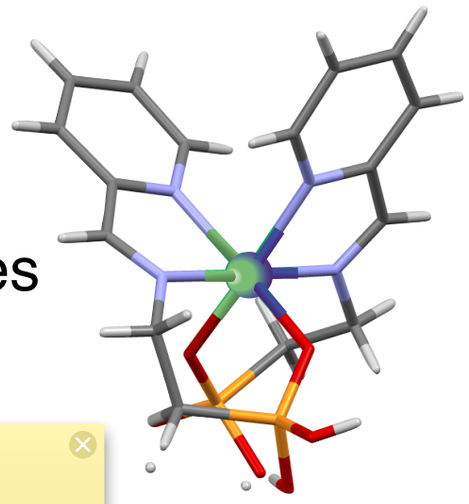
l'etape suivant etait l'etude de l'auto assemblage des molecules sur les surfaces d'oxydes de fer ou lsmo
le greffage ce fait par voi liquide dans une solution de methanol et eau

Molecular Spintronics

Self-Assembly on Surfaces

Co- or Ni-complexes

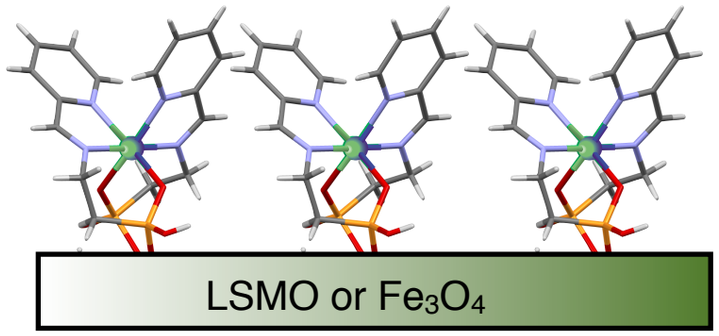


\+

LSMO or Fe_3O_4

add refs
add characterization

Monolayer



AFM
XPS
SQUID
DFT

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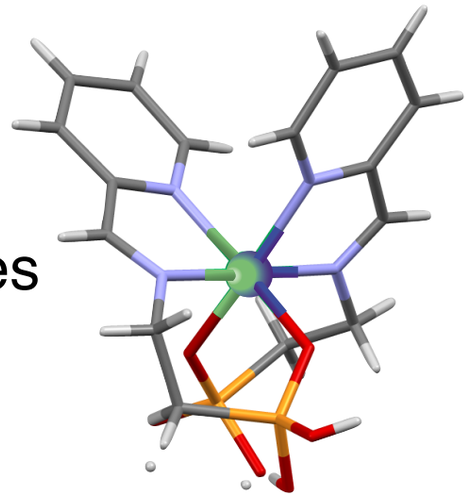
la monochouche forme etait chararterize par IR, XPS et AFM et nous avons constate que nous avons bien greffe la molecule sur la surface

Molecular Spintronics

Self-Assembly on Surfaces

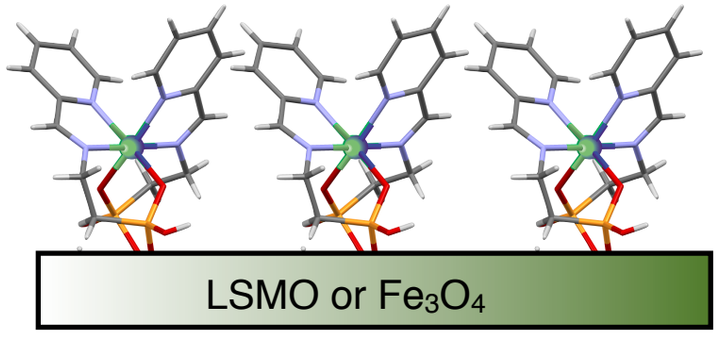
Co- or Ni-complexes



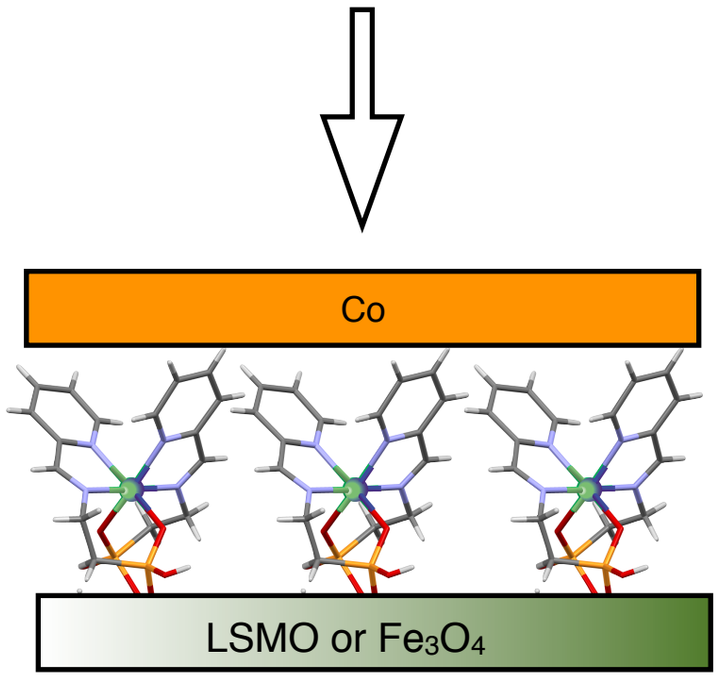
+

LSMO or Fe_3O_4

Monolayer



AFM
XPS
SQUID
DFT



Heterojunction

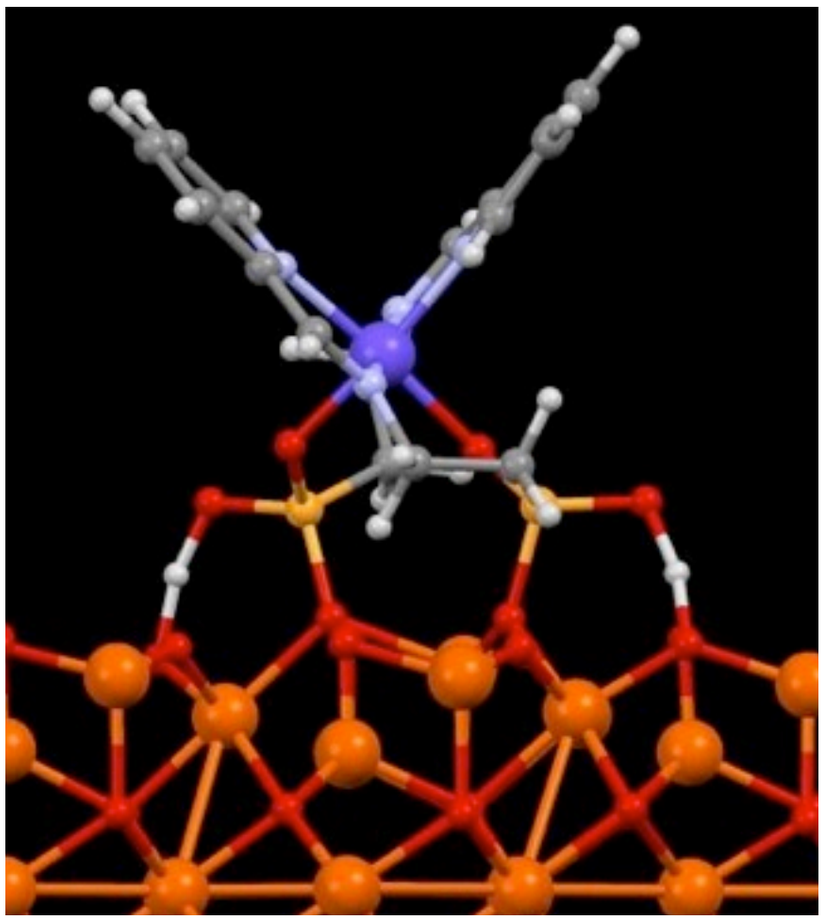
Wednesday, 11 June 14

as i had showed you in a previous slide a spin valve is composed of two electrodes a bottom and a top one
in order to construct a heterojuncion a top cobalt electrode was deposited on top of the monolayer
I would just like to point out that in order for the cobalt not to oxidize a gold layer capped the heterojunction

Molecular Spintronics

Co-pyPhos / Fe_3O_4

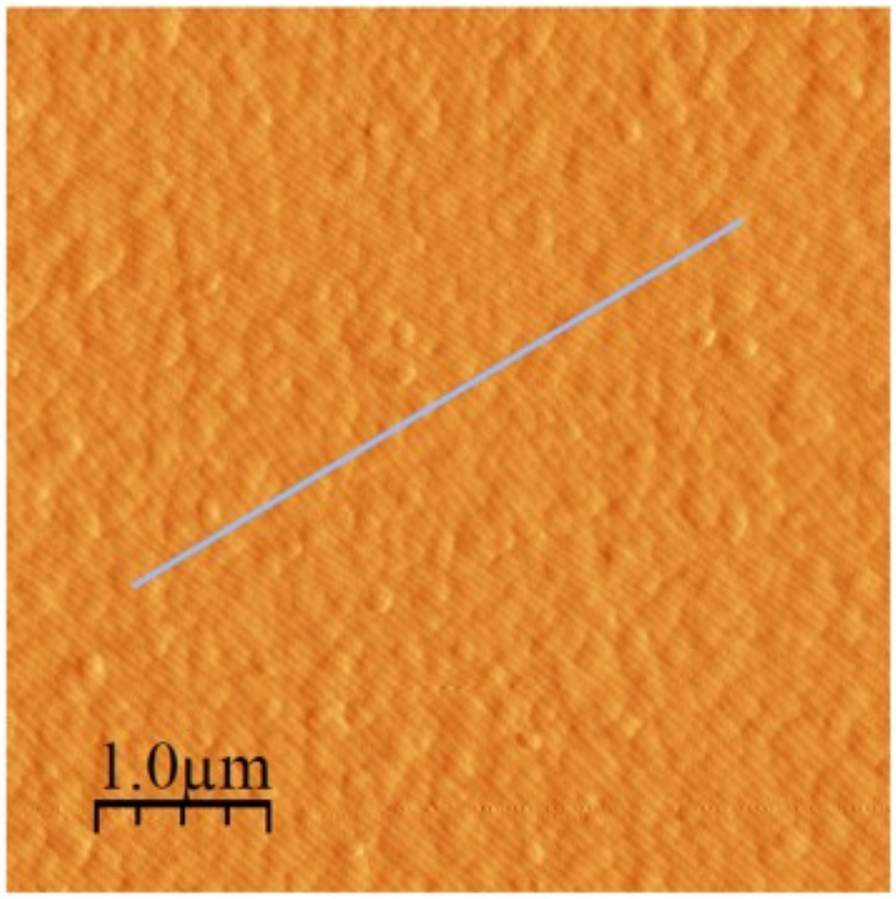
DFT



SPCSI, CEA-Saclay, Saclay France

- Dr. Yannick Dappe

AFM



SPCSI, CEA-Saclay, Saclay France

- Dr. Ludovic Tortech

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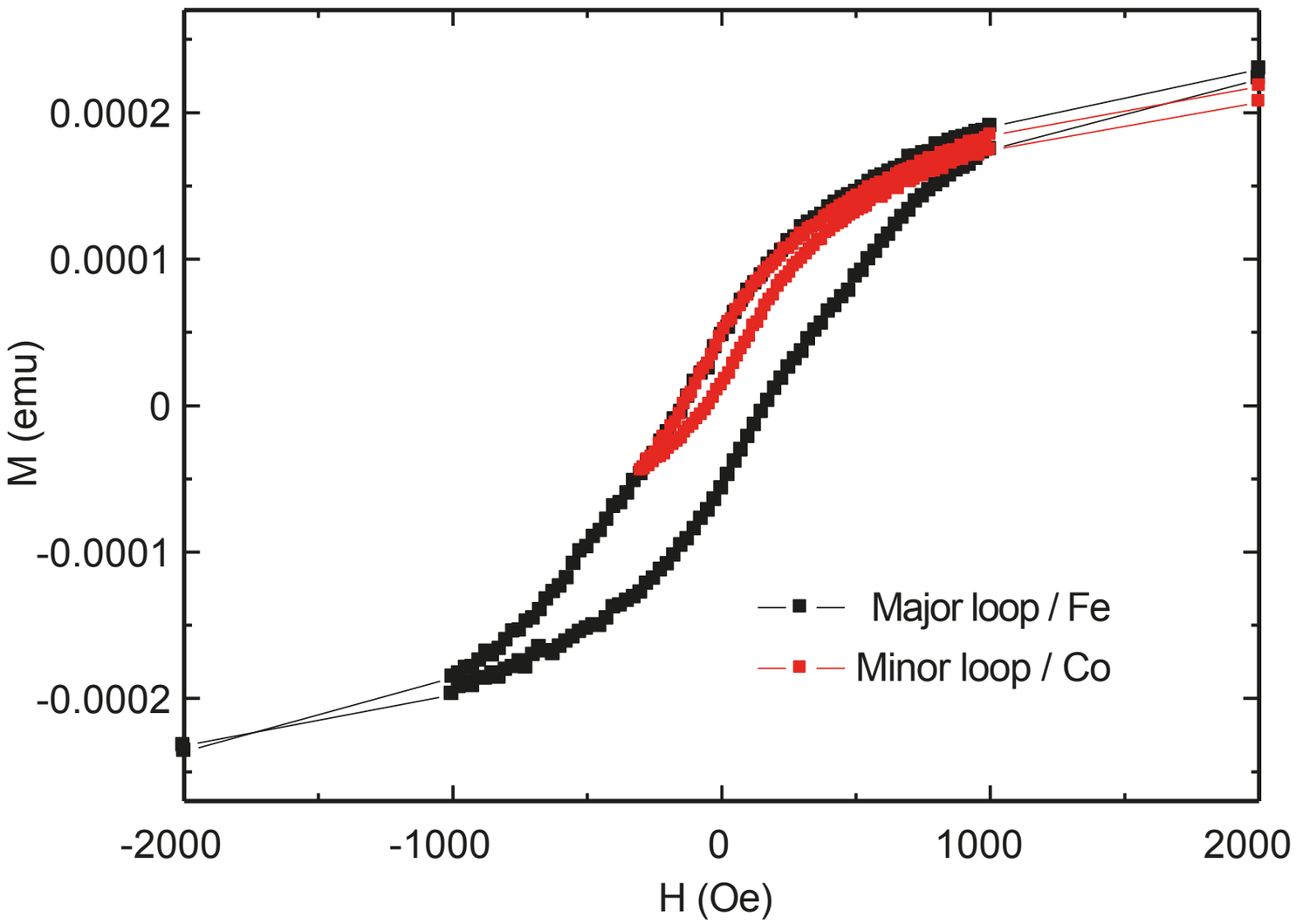
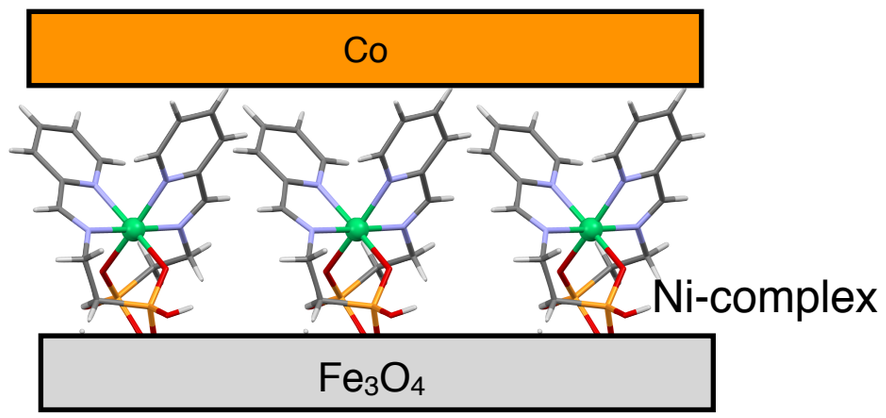
to afford a monolayer of molecules
the monolayers were characterized by AFM and XPS to ensure deposition of the molecules onto the surface

Molecular Spintronics

Ni-pyPhos / Fe_3O_4





SPCSI, CEA-Saclay, Saclay France

- Dr. Jean-Baptiste Moussy

Wednesday, 11 June 14

at each step of the way the magnetic properties of the layers were studied by SQUID magnetometry
here I show you the hysteresis loops of the final heterojuntion.
we was clearly see the major hysteresis loop which comes from the iron here in black and we can see the minor hysteresis loop which comes from the cobalt here is red
here is the expansion of the cobalt loop.
I would like to point out that the electrodes are separated by a monolayer of molecules and that they are indeed decoupled
because if there was a short circuit, meaning that the cobalt is in direct contact with the iron oxide we would not see a major and a minor hysteresis loop

since the thickness of the monolayer is much much smaller than that of the electrodes, it is not possible to zoom in onto the signal of the molecule with this technique

Molecular Spintronics

X-ray Magnetic Circular Dichroism

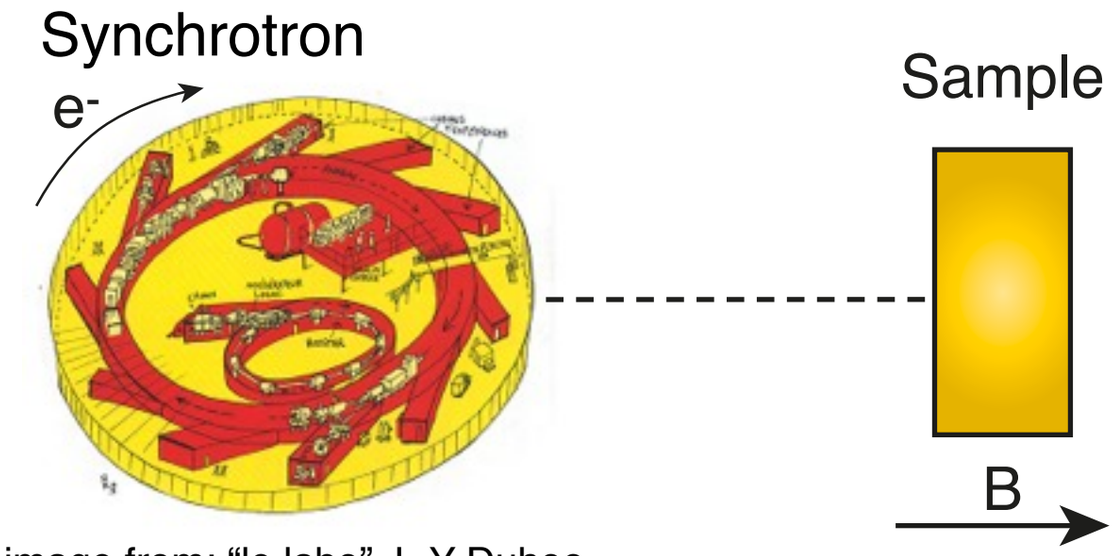
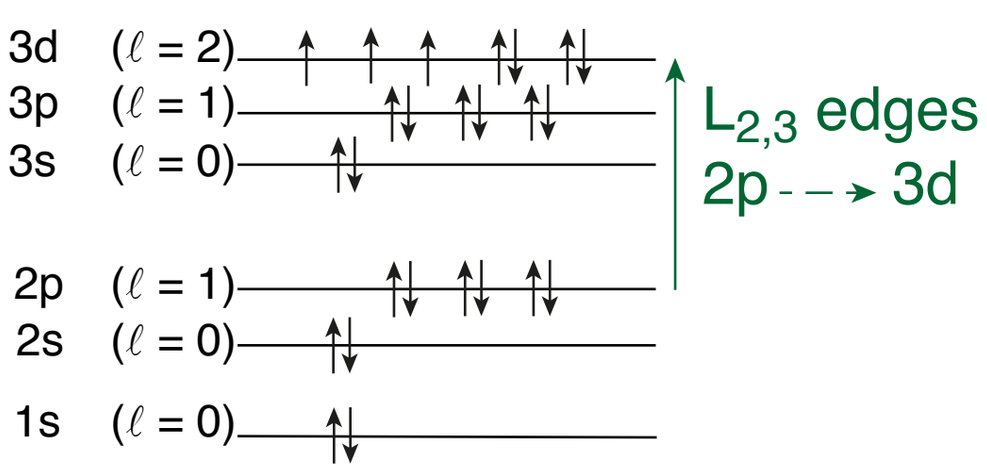


image from: “le labo” J.-Y Duhoo

Co^{2+} ($3d^7$)



Electric Dipole Selection rules

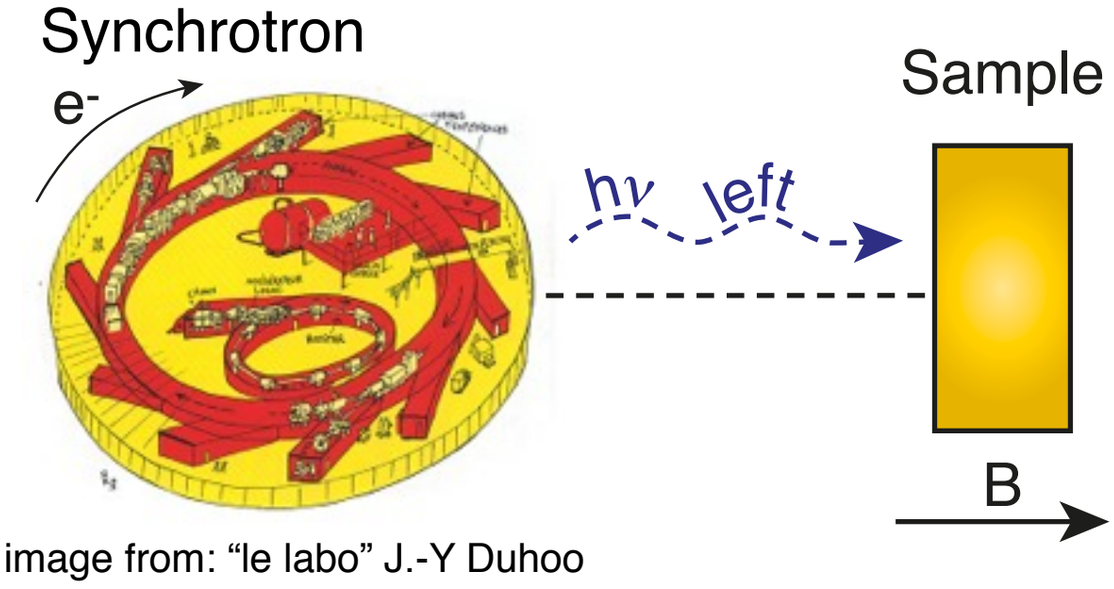
(ε.r) : $\ell = \pm 1$

Wednesday, 11 June 14

for that we need to rely on another technique called X-ray magnetic circular dichroism (XMCD)
In order to do XMCD measurements we need a few important things
1) we need a synchrotron radiation source
2) we need a paramagnetic sample
3) we need to place the sample in a magnetic field

Molecular Spintronics

X-ray Magnetic Circular Dichroism



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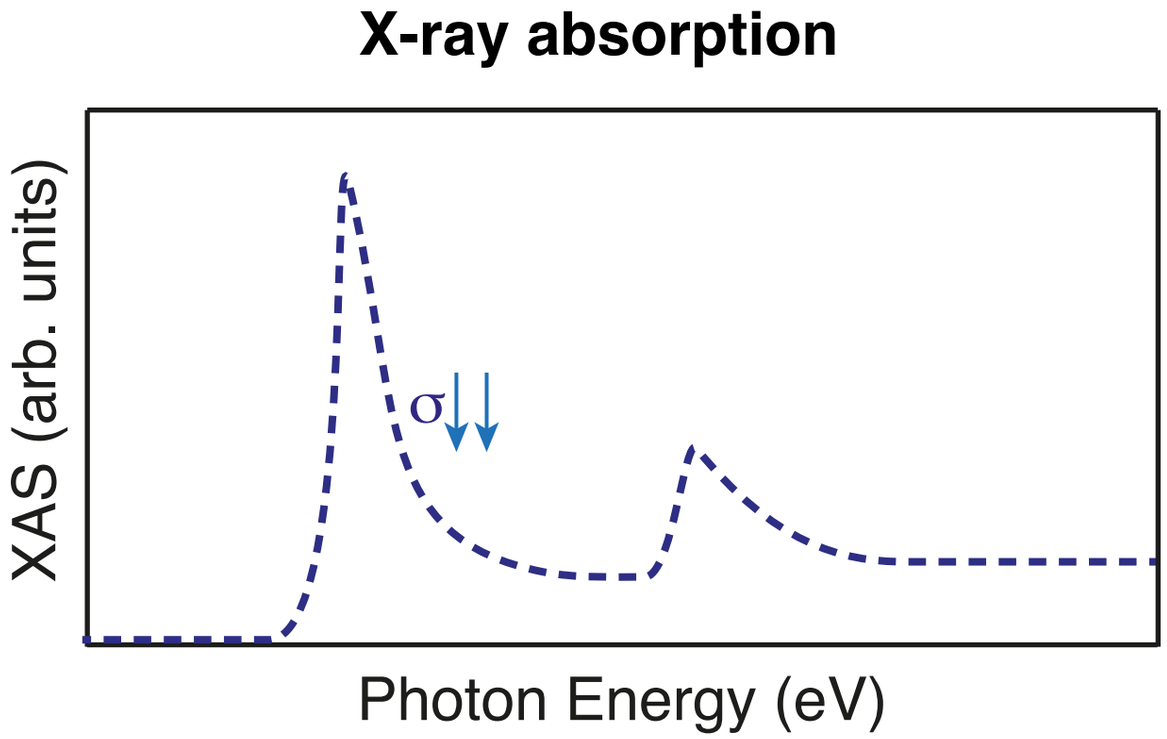
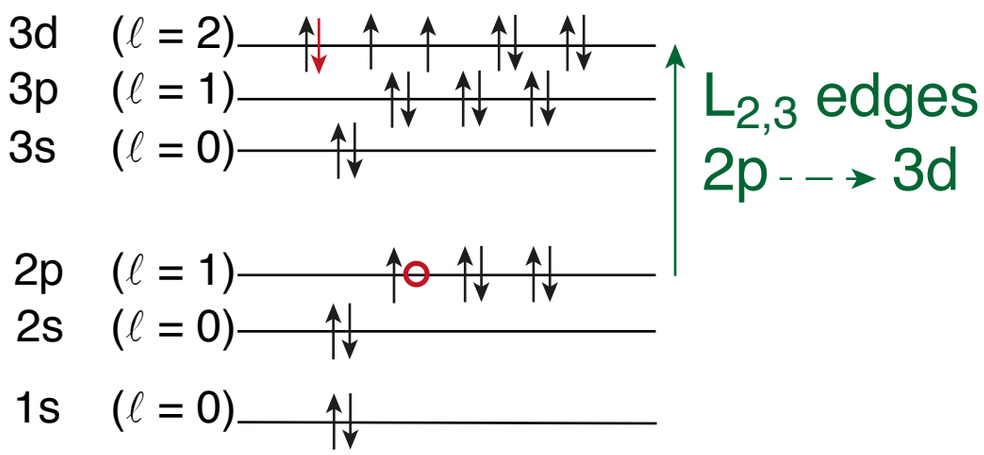


Co^{2+} ($3d^7$)



$L_{2,3}$ edges
$2p \dashrightarrow 3d$

Electric Dipole Selection rules
(ε.r) : $\ell = \pm 1$

Wednesday, 11 June 14

Then the sample is irradiated first with left circularly polarized light
one electron is excited from the 2p shell to an empty 3d shell

Molecular Spintronics

X-ray Magnetic Circular Dichroism

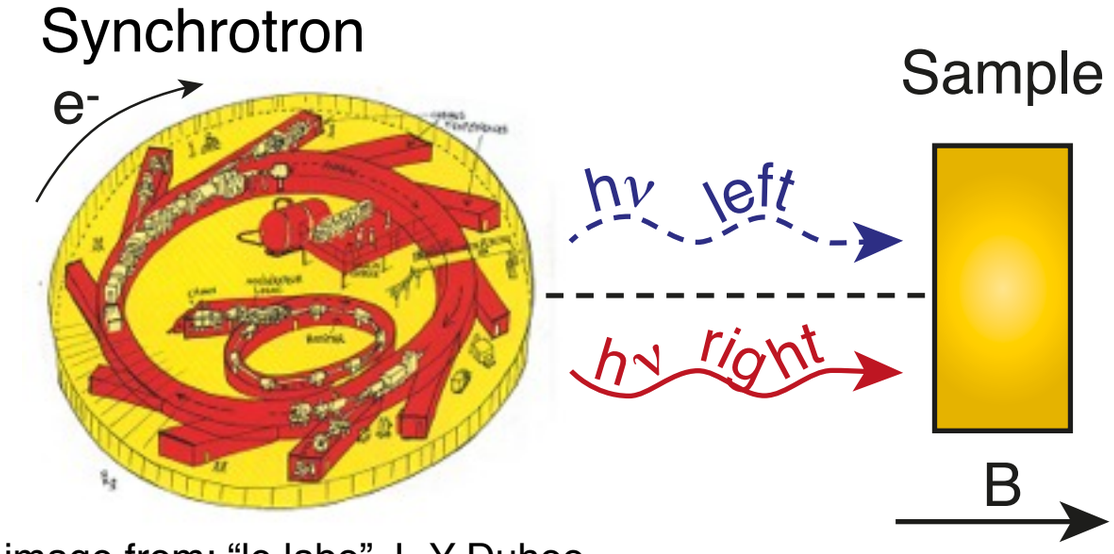
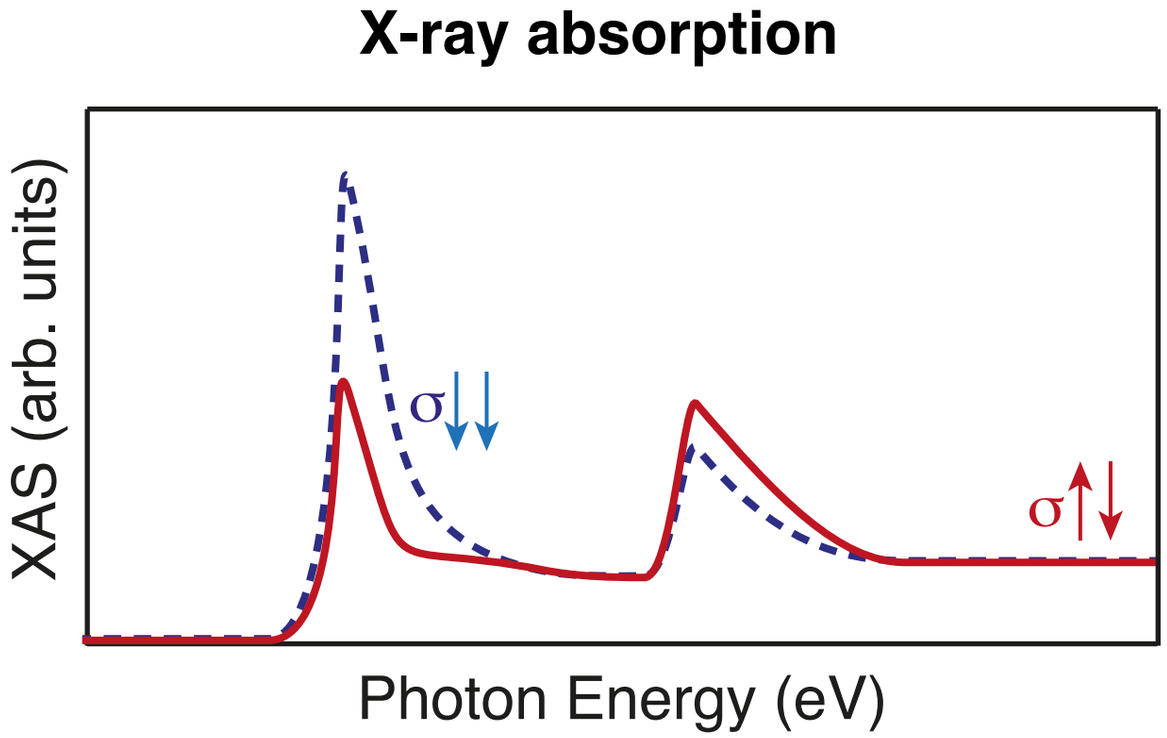
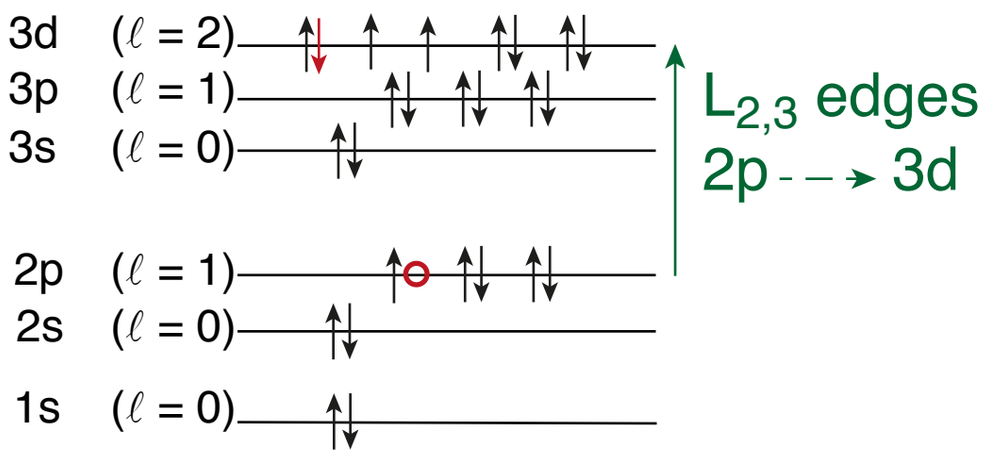


image from: “le labo” J.-Y Duhoo



Co^{2+} ($3d^7$)



$L_{2,3}$ edges
$2p \dashrightarrow 3d$

Electric Dipole Selection rules
$(\varepsilon.r) : \ell = \pm 1$

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then the sample is irradiated with right circularly polarized light
because of the selection rules the two absorption spectra are not entirely the same

Molecular Spintronics

X-ray Magnetic Circular Dichroism

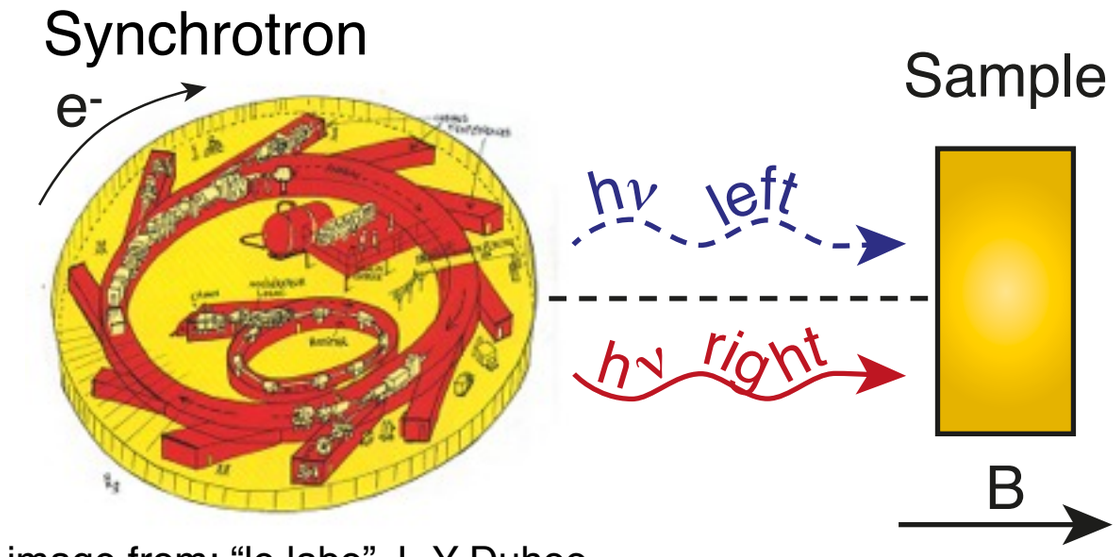
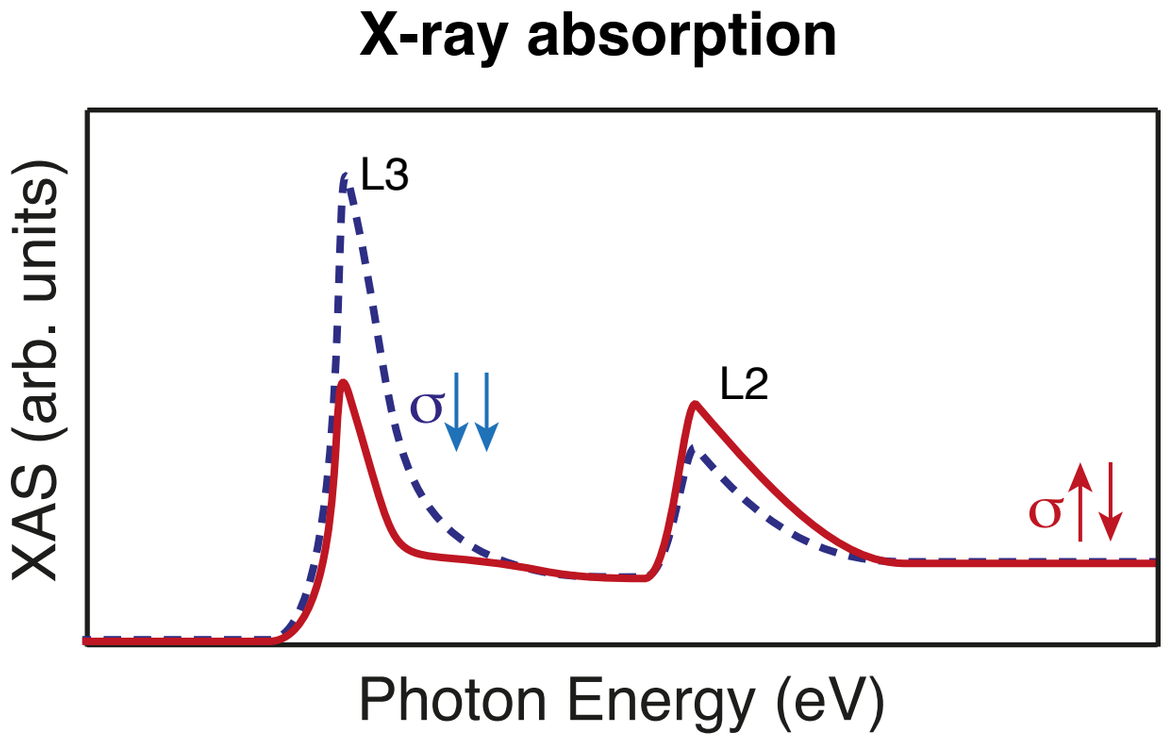
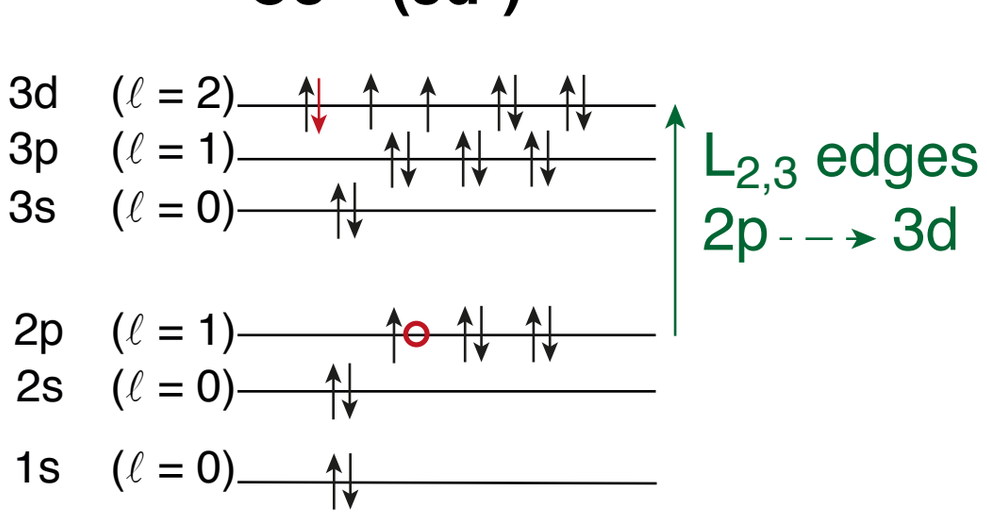
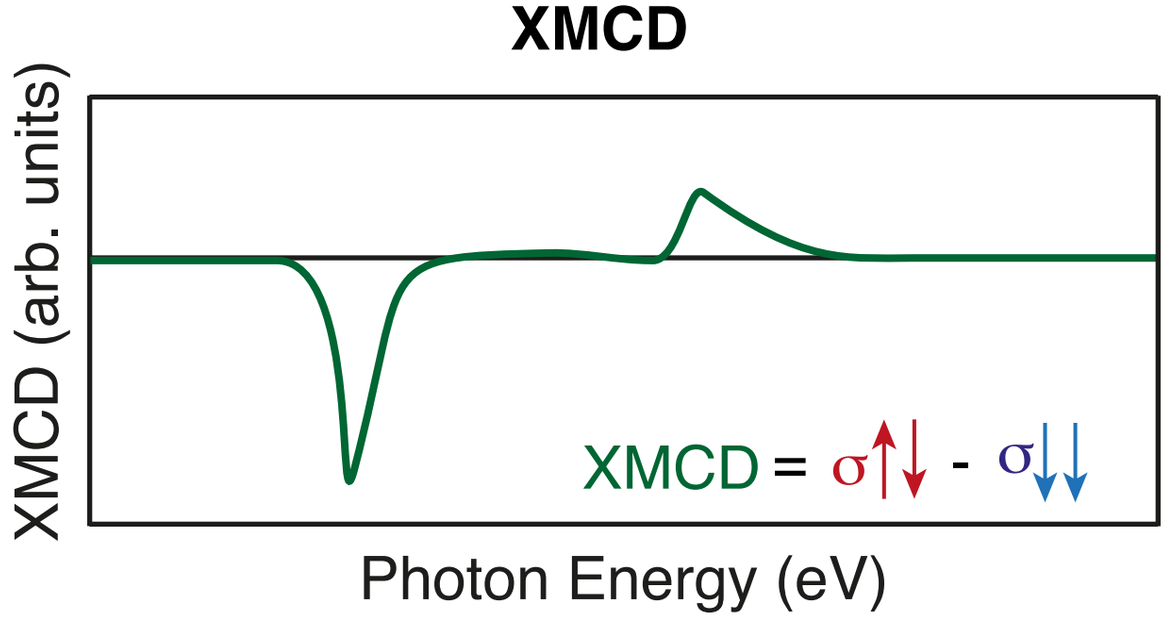


image from: “le labo” J.-Y Duhoo







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By taking the difference of the right and left spectra it is possible to obtain the XMCD spectrum
we can in such manner obtain information on the local magnetic moment of the ion
since each element absorbs at different energies we can zoom in on the element of choice

for that we need to rely on another technique called X-ray magnetic circular dichroism (XMCD)
XMCD is a difference spectrum of two x-ray absorption spectra (XAS) taken in a magnetic field, one taken with left circularly polarized light, and one with right circularly polarized light. By closely analyzing the difference in the XMCD spectrum, information can be obtained on the magnetic properties of the atom, such as its spin and orbital magnetic moment.
In the case of transition metals such as iron, cobalt, and nickel, the absorption spectra for XMCD are usually measured at the L-edge. This corresponds to the process in the iron case: with iron, a 2p electron is excited to a 3d state by an x-ray of about 700 eV. Because the 3d electron states are the origin of the magnetic properties of the elements, the spectra contain information on the magnetic properties.
one draw back of this technique is that it may only be done using a synchrotron radiation light source

XMCD is the difference, for a magnetic material, between the absorption of left and right circularly polarized X-rays. In X-ray absorption, the atom absorbs a photon, giving rise to the transition of a core electron to an empty state above the Fermi level. The absorption cross-sections are large, especially in the soft X-ray range (500-2000 eV). The absorption edges have energies which are characteristic for each element and, due to the dipole selection rules, final states with different symmetries can be probed by choosing the initial state. (http://neel.cnrs.fr/spip.php?article798&lang=fr)
http://140.110.201.35/djhuang/research_xas.html

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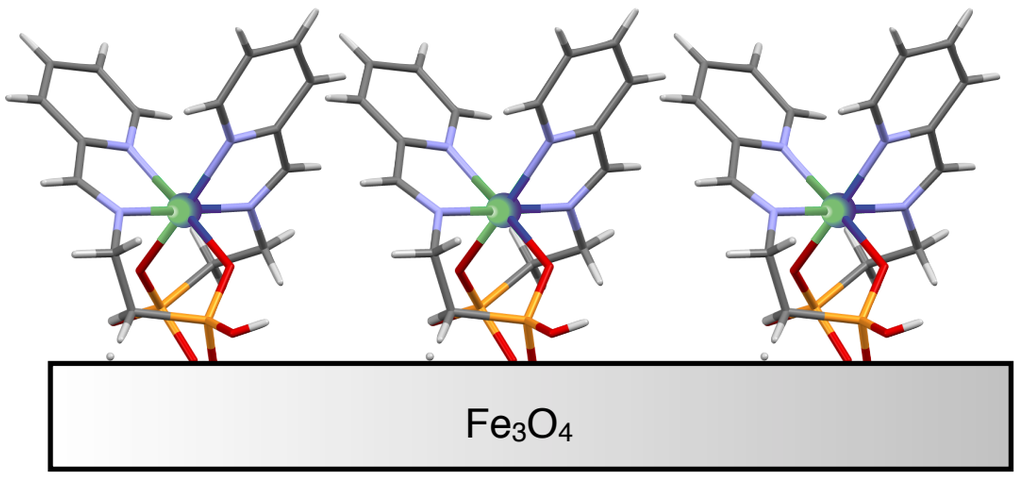

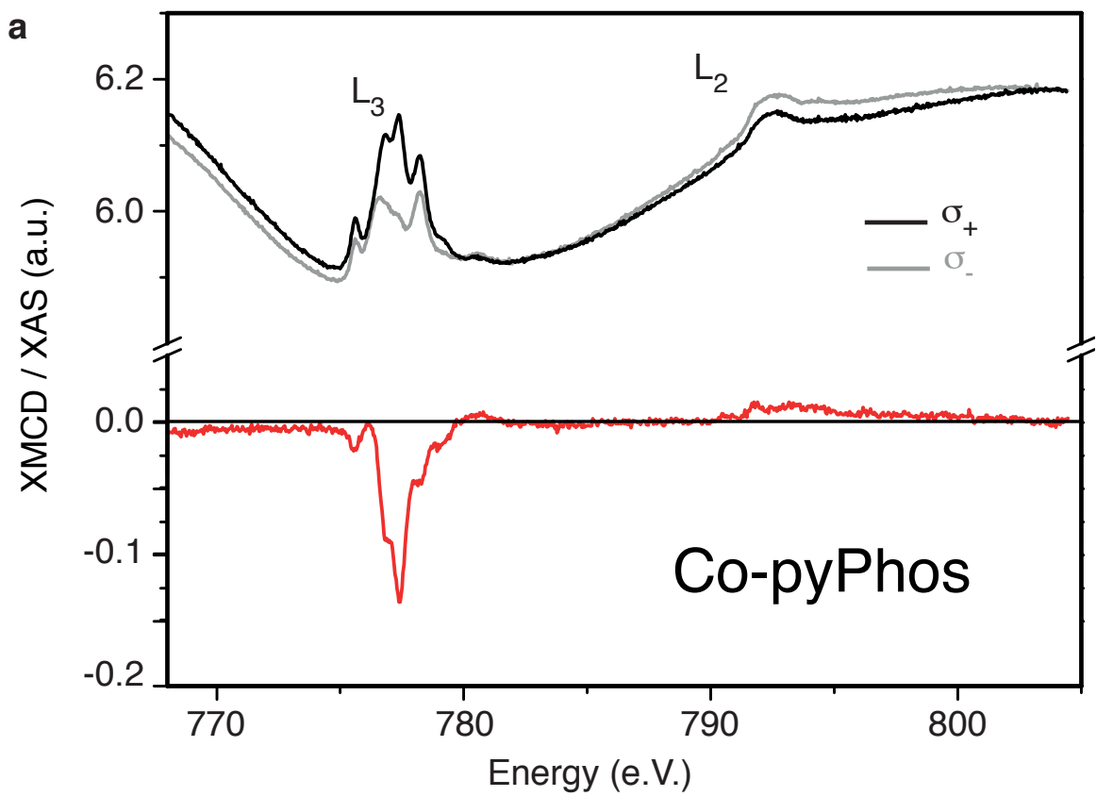

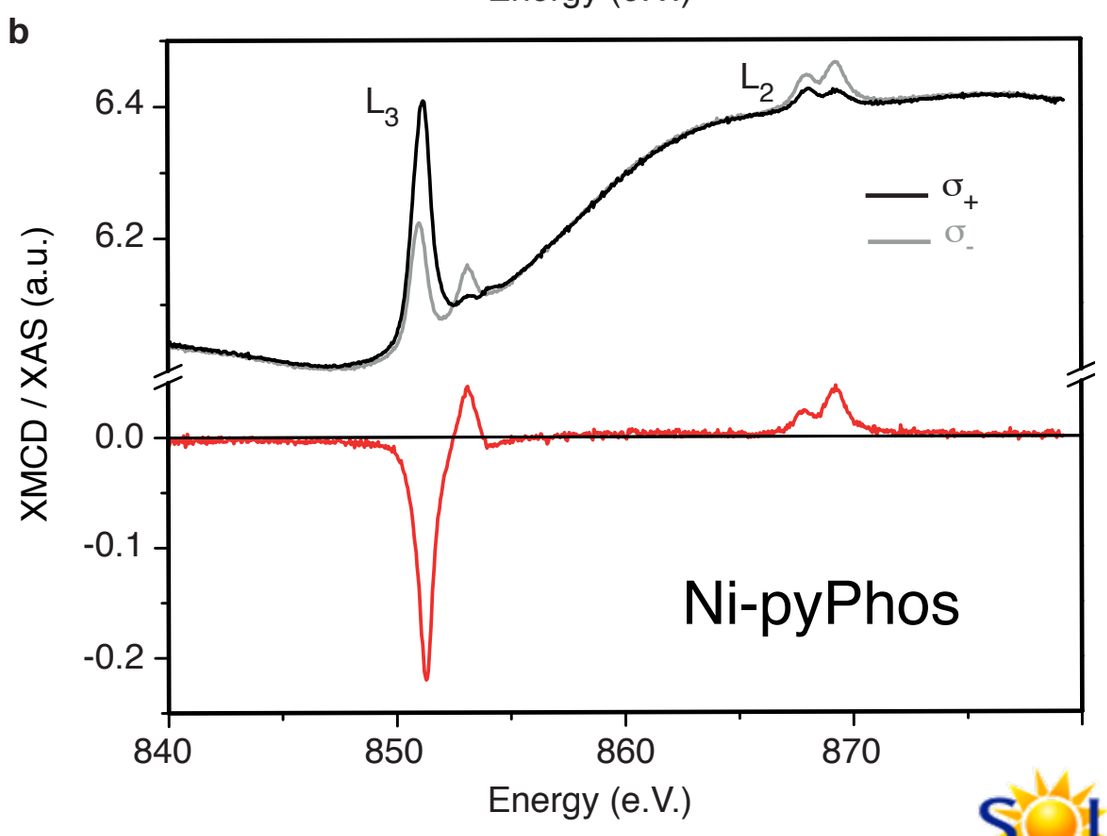

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SOLEIL SYNCHROTRON

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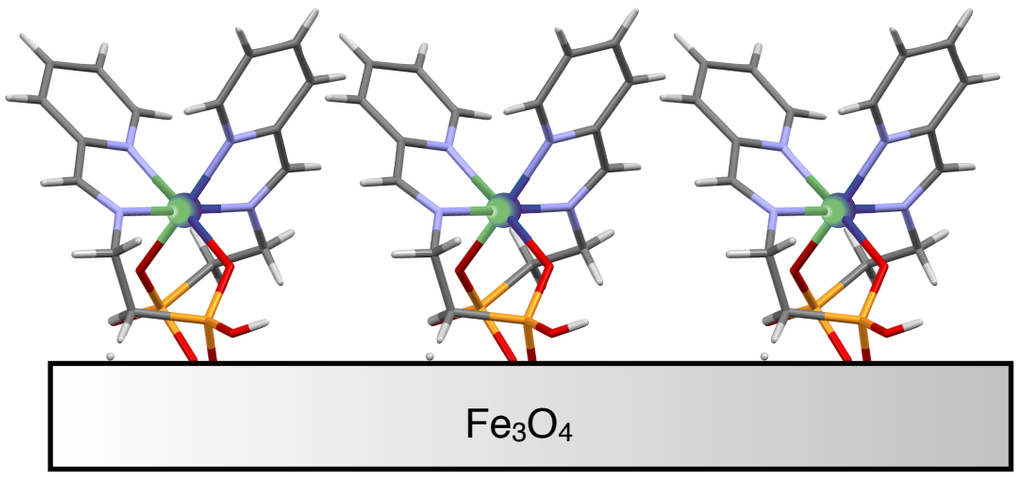
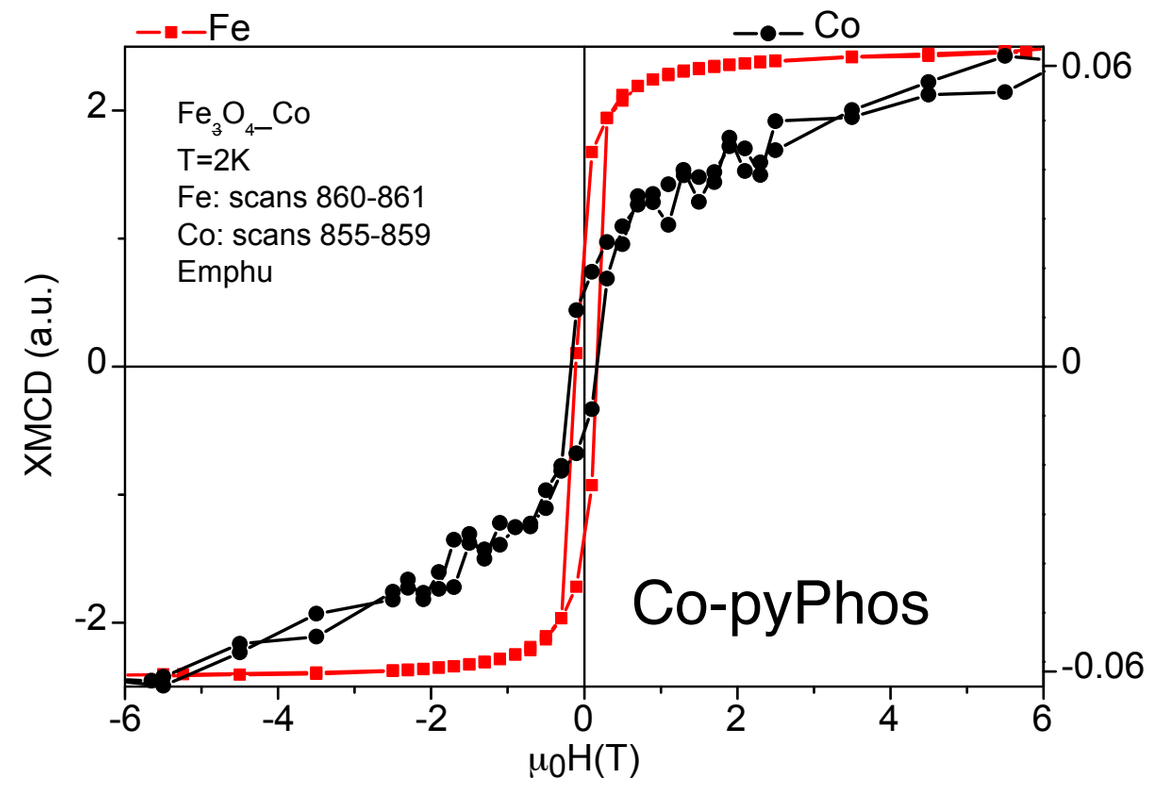
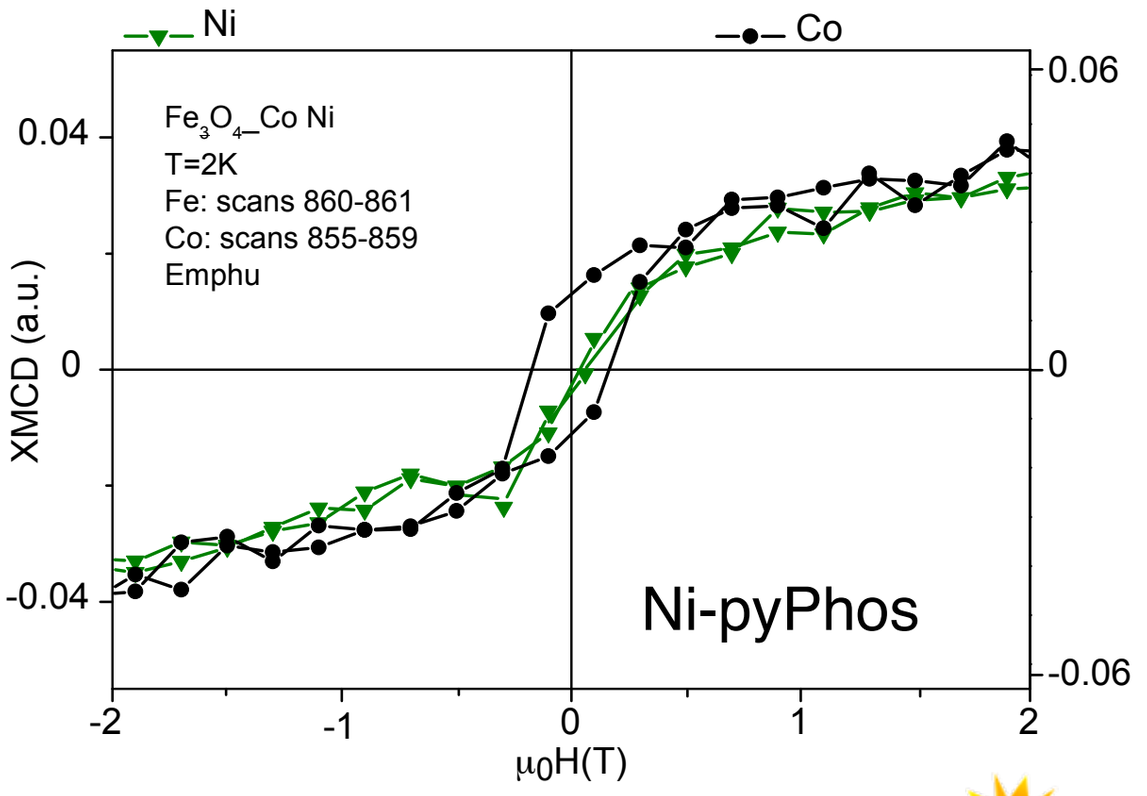
in this slide I will show you recent results obtained in Paris at synchrotron Soleil on the DEIMOS line.
the data i will show was taken on the cobalt complex grafted onto iron oxide
here is the x-ray absorption and the xmcd taken at the L3 edge of cobalt where it is apparent that the cobalt is present
what was remarkable was that it was possible to obtain a hysteresis loop of the cobalt complex at 2K. to the best of our knowledge this is the firs example of a molecular hysteresis loop indicating bistability of transition metal complexes onto an iron oxide.
we were also able to see a very small signal in the x-ray natural linear dichroism experiment. this

Pour mieux etudier l'interaction de ces molecule avec les surfaces de Fe3O4 et LSMO nous avons fait des mesure XMCD a Soleil sur la ligne DEIMOS. Ici je vous montre des resultats preliminaires qui date de debut decembre. Avec cette technique c'est possible de obteneir des informations sur une molecule isolee. Ici je vous montre les resultats obtenue pour le complexe de Co(II). On vois le signal XMCD au suil L2 ce molecules sont structuralment orientees sur la surface ce qui conduit a l'observation de bistabilite des molecules isolees.

Molecular Spintronics







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SOLEIL SYNCHROTRON

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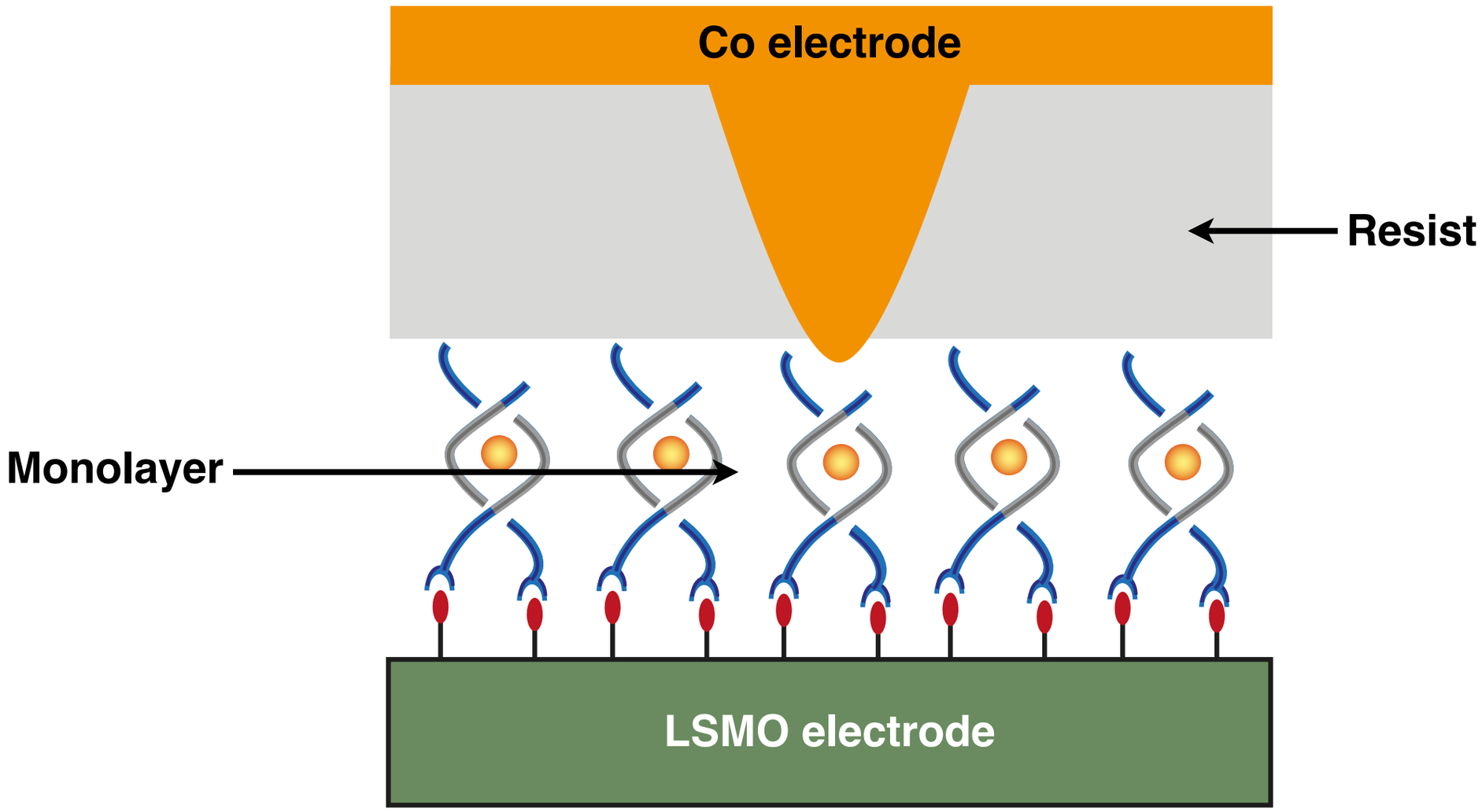
exchange coupling is different in the two complexes leading to an openin of the the magnetization loop for co and no opening for ni

Molecular Spintronics

Spin transport

Fundamental investigation of the spin transport capabilities of these materials



Spin Valve

C. Barraud et al., *Nat. Phys*, **2010**, *6*, 615-620

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Utlimately the goal is to construct a spin valve and to test the spin transport properties of these complexes
this task is in progress the spin valve will have the following architecture
an lsmo electrode, a monolayer of complexed followed by a monomolecular contact fabricated by nanolitography

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in red are people who have worked on the projects discussed today and in green and people i collaborate with on other projects
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