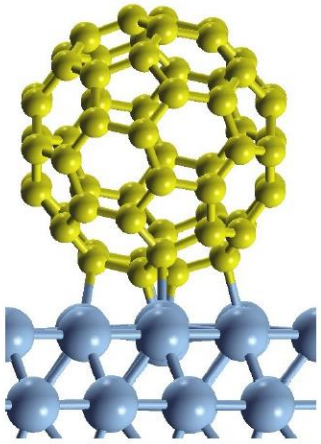
DE LA RECHERCHE À L'INDUSTRIE

MODELLING OF THE MAGNETIC ANISOTROPY OF MOLECULE/FERROMAGNETS INTERFACES



Cyrille Barreteau

Kaushik Bairagi, Vincent Repain, Amandine Bellec, Dongzhe Li, Alexander Smogunov, Ludovic Le Laurent

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INTRODUCTION

ANISOTROPY IN MAGNETIC STUDIES

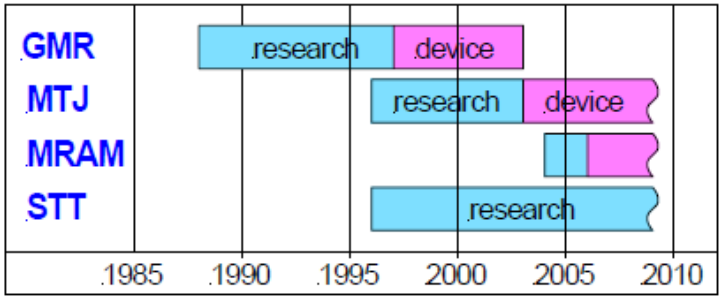
Search for permanent hard magnets (large Anisotropy) without rare earth

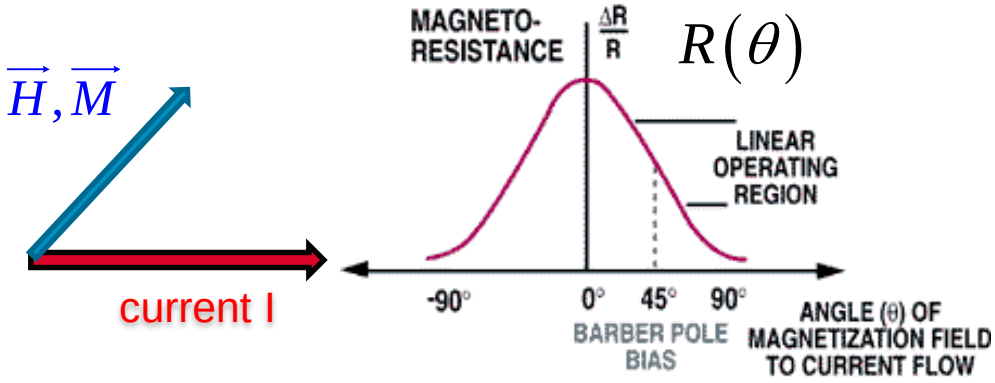
Replace Samarium-Cobalt, Neodyum by nanostructured « cheap » TM magnets

Spintronics

From fundamentals to applications

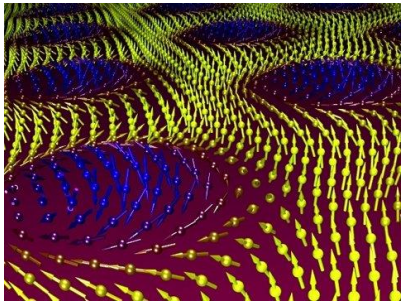
AMR



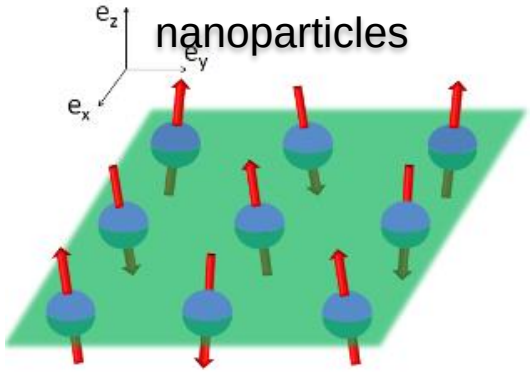


Ultimate data storage

skyrmions

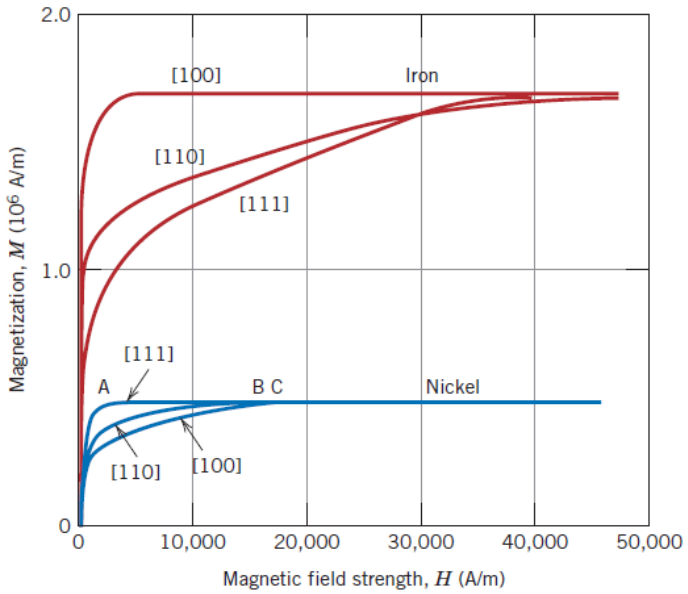


nanoparticles

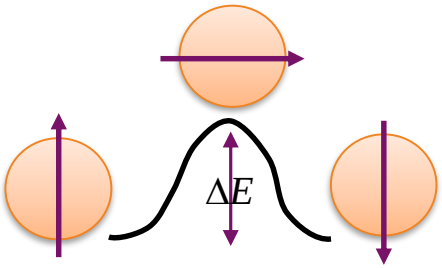


MANIFESTATION OF MAGNETIC ANISOTROPY

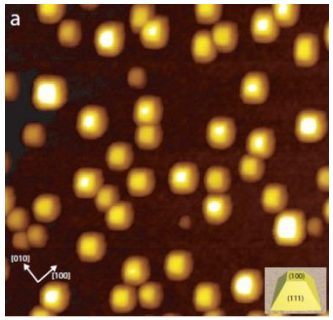
Magnetization curve of single crystals



Thermal stability of magnetic nanoparticles



$$\nu = \nu_0 e^{-\frac{\Delta E}{k_B T}} \qquad \tau = \tau_0 e^{\frac{\Delta E}{k_B T}}$$



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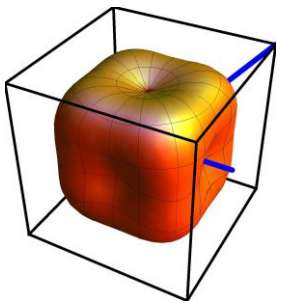
τ	1s	1mn	1h	1day	1year	10years
$\frac{\Delta E}{k_B T}$	23	27	31	34	40	43

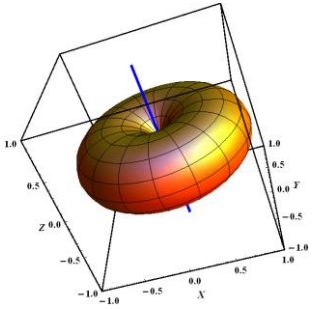
A question of energy

$$E = E(\theta, \varphi)$$

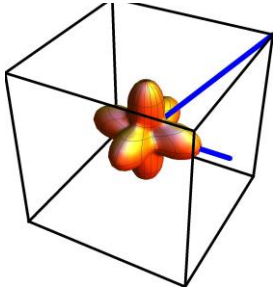
bccFe



hcpCo



fccNi



$\Delta E \sim 10^{-2} - 10^{-3} meV$ / atom in bulk Fe, Co, Ni

Much larger in nanostructures

$\Delta E \sim meV$ / atom in bulk FePt L10

ORIGIN OF THE MAGNETIC ANISOTROPY

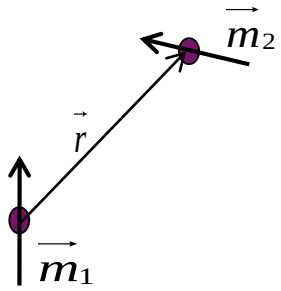
Shape anisotropy

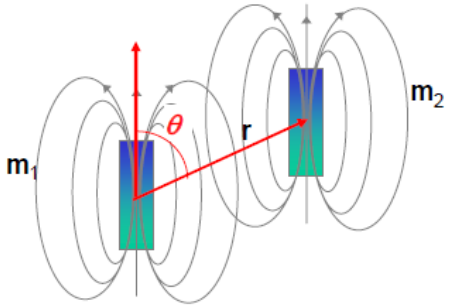
•dipole-dipole interaction

$$E_{dip}(\vec{r}) = \frac{\mu_0}{4\pi r^3}\left[\vec{m}_1.\vec{m}_2 - \frac{3}{r^2}\left(\vec{m}_1.\vec{r}\right)\left(\vec{m}_2.\vec{r}\right)\right]$$

Collinear magnetism

$$E_{dip}(\theta) = \frac{\mu_0}{4\pi r^3} m_1 m_2 \left[1 - 3\cos\theta\right]$$





For θ<54.74° FM For θ>54.74° AF

Favors elongated direction

Magnetocrystalline anisotropy

•Spin-orbit coupling

Dirac $\xrightarrow[\frac{v}{c}\ll 1]{}$ Schrödinger + Zeeman + Mass velocity + Spin-orbit + Breit

$$V_{SOC} = \underbrace{\frac{1}{2m^2c^2}\frac{1}{r}\frac{dV}{dr}}_{\xi(r)}\mathbf{L.S}$$

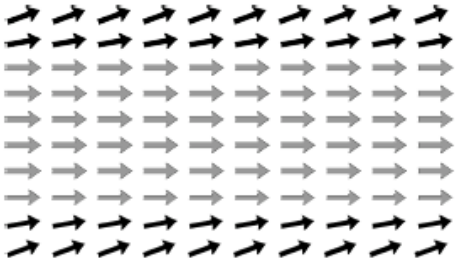
- $\xi(r)$ Very localized potential
- $\xi(r) \sim Z^4$ Increases drastically with atomic number

$$\xi_d \sim 0.05eV \text{ for } 3d \qquad \xi_d \sim 0.5eV \text{ for } 5d$$

- $L.S$ Couples spin and orbital moment → generates anisotropy

SHAPE ANISOTROPY VERSUS MAGNETOCRYSTALLINE ANISOTROPY

Shape anisotropy	Magnetocrystalline anisotropy
Long range interaction $\frac{1}{r^3}$	short range interaction
Very little dependent on crystallographic structure and local atomic environment	Strongly dependent on crystallographic structure and local atomic environment
Very little dependent on symmetry	Strongly dependent on symmetry
Very little dependent on chemical nature of atoms	Strongly dependent on chemical nature of atoms
Average quantity (proportional to the volume of the sample)	Local quantity dominated by surface and interfaces



$E^{\mathrm{MCA}} \sim \mathrm{Surface}$

$E^{\mathrm{shape}} \sim \mathrm{Volume}$

At the end the shape anisotropy always wins!

MODEL

MODELLING

DFT

Plane wave basis set

Ultra-soft pseudo-potentials

Collinear and non-collinear magnetism

Spin-orbit coupling

TB

spd tight-binding

Semi-empirical model fitted on ab-initio

Collinear and non-collinear magnetism

Spin-orbit coupling

HOW TO CALCULATE MCA

Brute force method (self-consistent)

$$E^{MCA} = E^{\mathrm{tot}}_{\mathbf{n}_1} - E^{\mathrm{tot}}_{\mathbf{n}_2}$$

where $E_{\mathbf{n}_1}$ and $E_{\mathbf{n}_2}$ are obtained from SCF calculation including SOC

In principle « exact » but very time consuming and hard to converge
One should use penalization techniques to obtain E_n for any direction

Force Theorem method

$$E^{MCA} \approx \Delta E_{\mathrm{band}} = \int^{E_F^1} En^1(E)dE - \int^{E_F^2} En^2(E)dE \approx \int^{E_F} (E - E_F)\Delta n(E)dE$$

Initial density is obtained from a SCF spin-collinear calculation and spin-moment further rotated to appropriate spin direction and one step SOC calculation.

Very « fast » and numerically stable but cannot be applied to systems with too large SOC.

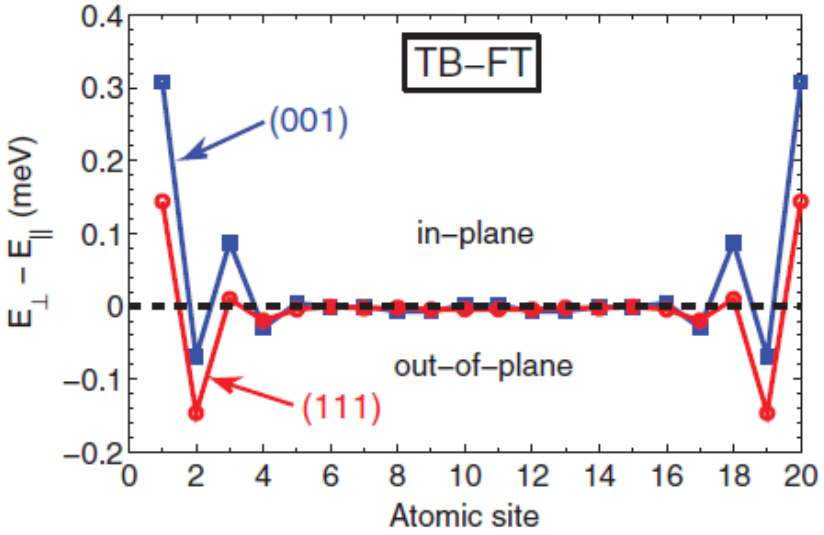
Local « site » and orbital decomposition

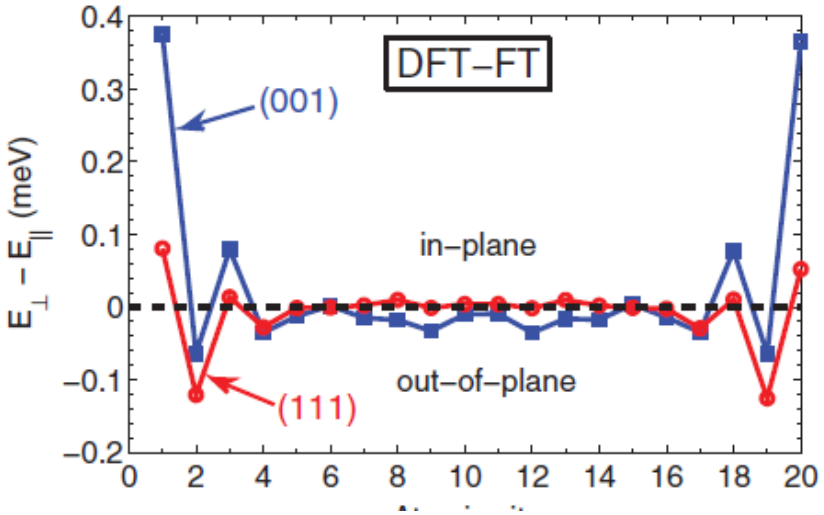
$$E^{MCA} \approx \sum_{i\lambda} \int^{E_F} (E - E_F)\Delta n_{i\lambda}(E)dE$$

VALIDATION OF TB MODEL

Co fcc « slabs »

TB





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MCA
OF MOLECULE/FERROMAGNETS INTERFACES

Co/Au(111) EXPERIMENT

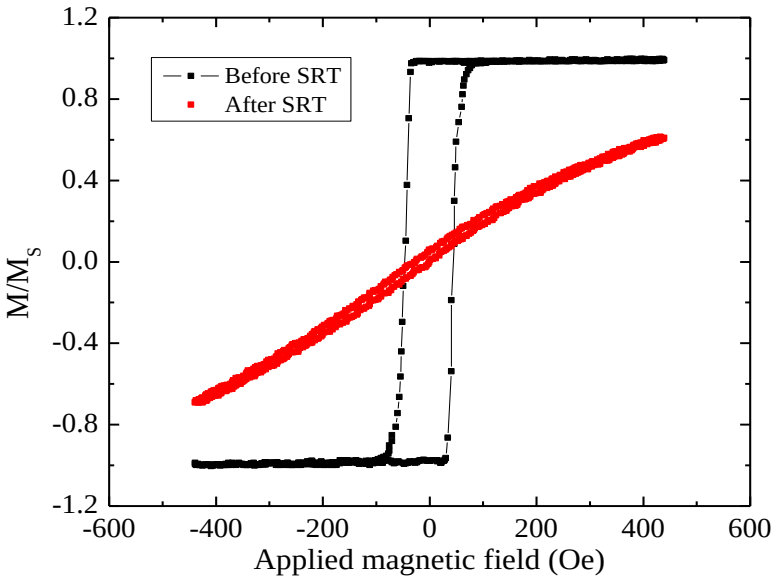
substrate Co/Au(111)

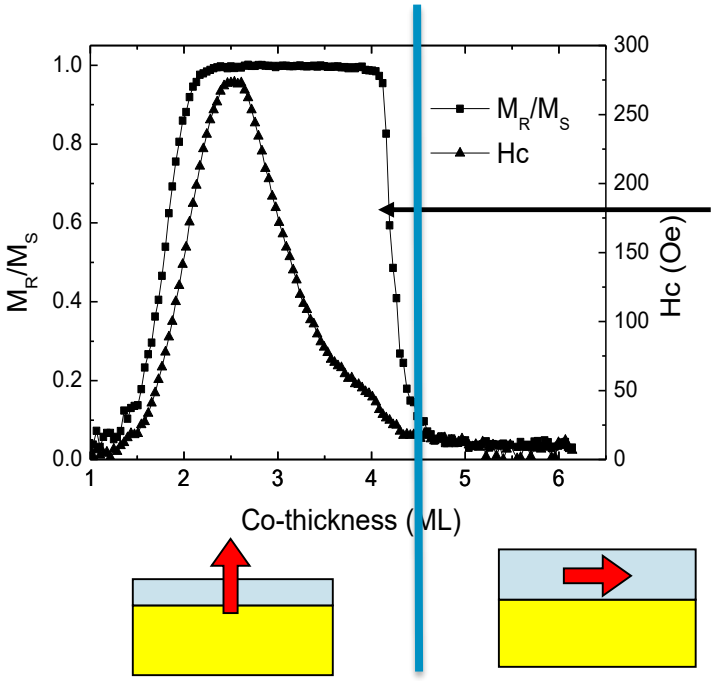
$$K^{eff} = \frac{E^{MCA} + E^{shape}}{V} = K^{v} + \frac{K^{s}}{t}$$

Co/Au(111) K^s= 0.75 mJ.m^{-2} and K^v= -900 kJ.m^{-3}

Spin Reorientation Transition (SRT) at t*=4.2 ML

Polar MOKE



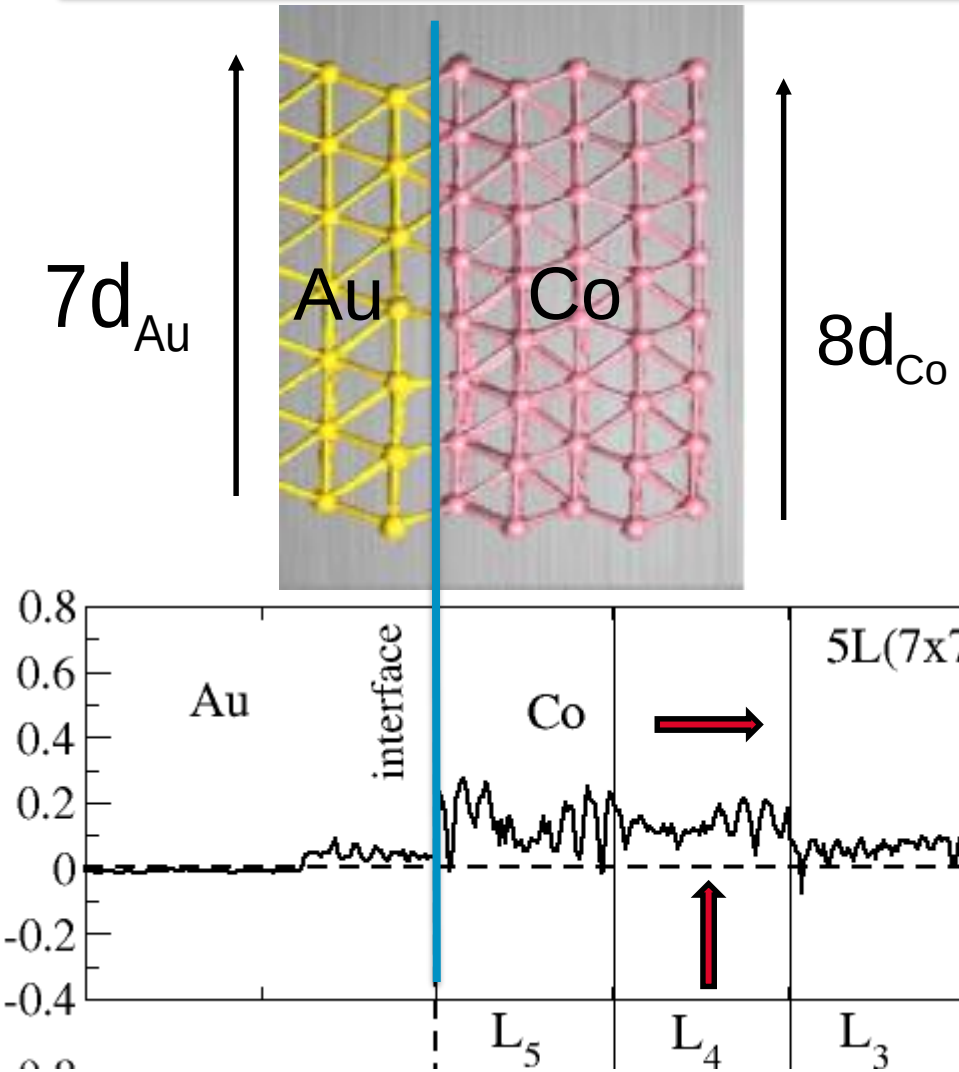


SRT

Ultra sensitive to magnetic anisotropy change close to the SRT

Co/Au(111): TB MODEL

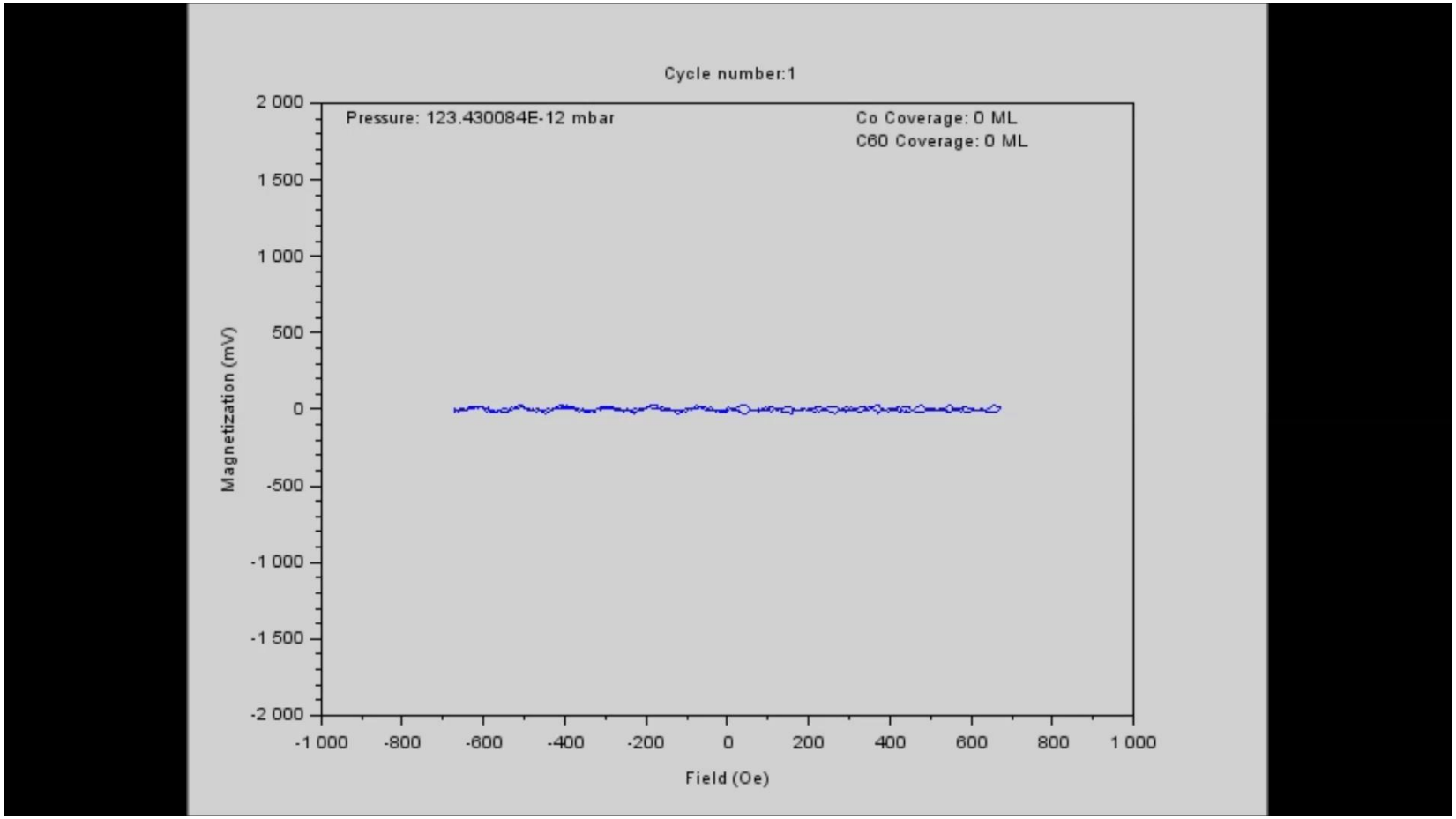
•MCA of Co(0001)/Au(111) from a realistic TB modelling



Gold interface favors « out-of-plane » magnetization

C60/Co/Au(111) EXPERIMENT

•Adsorption of C_{60} on Co(0001)/Au(111): Experiment

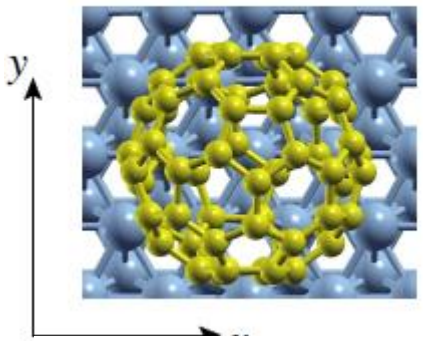


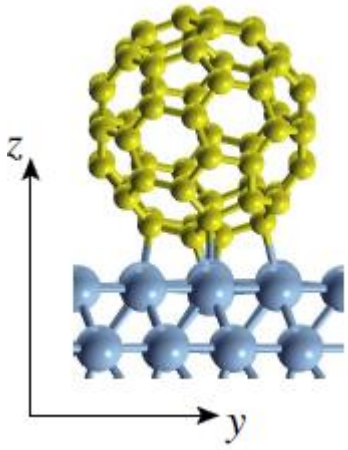
C60 favors « out-of-plane » magnetization

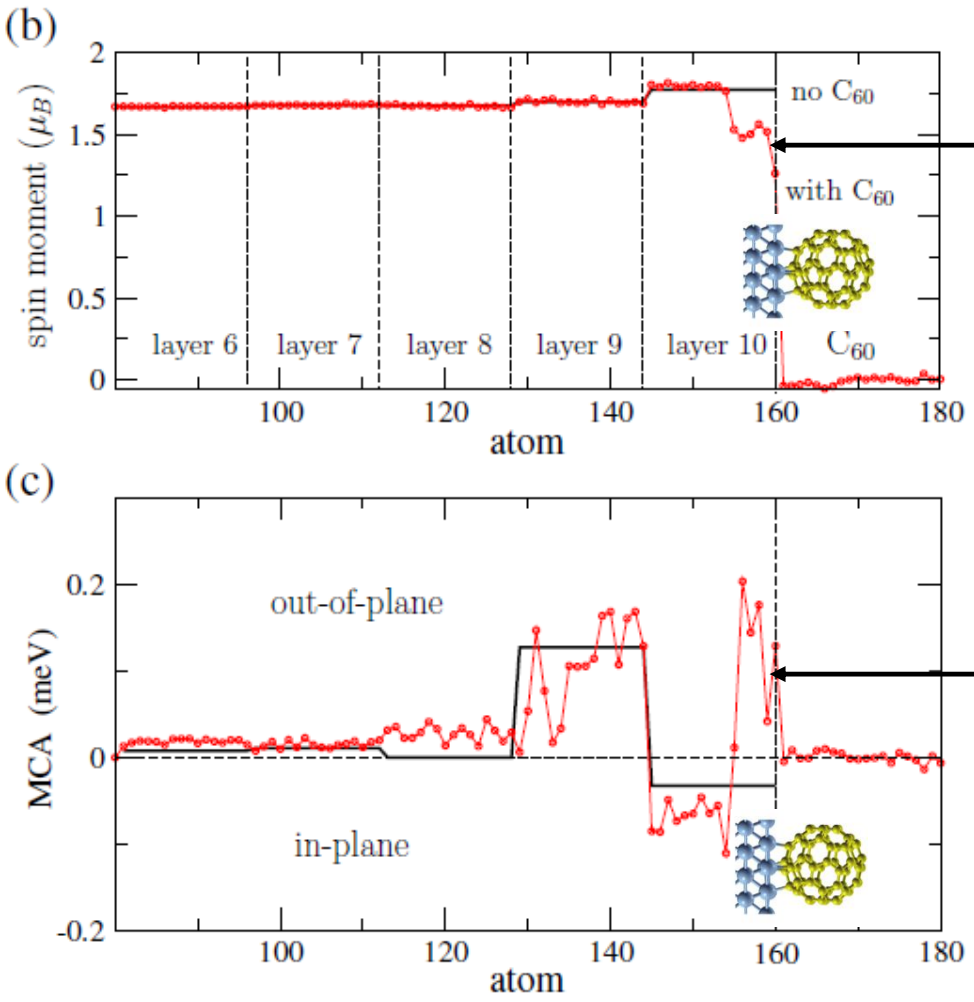
C60/Co MODELLING

•Adsorption of C_{60} on Co(0001)/Au(111): Theory

DFT calculation:
C60/10L Co (4x4)





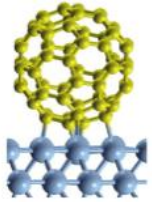


Decrease of magnetization

increase of "out-of-plane" MCA

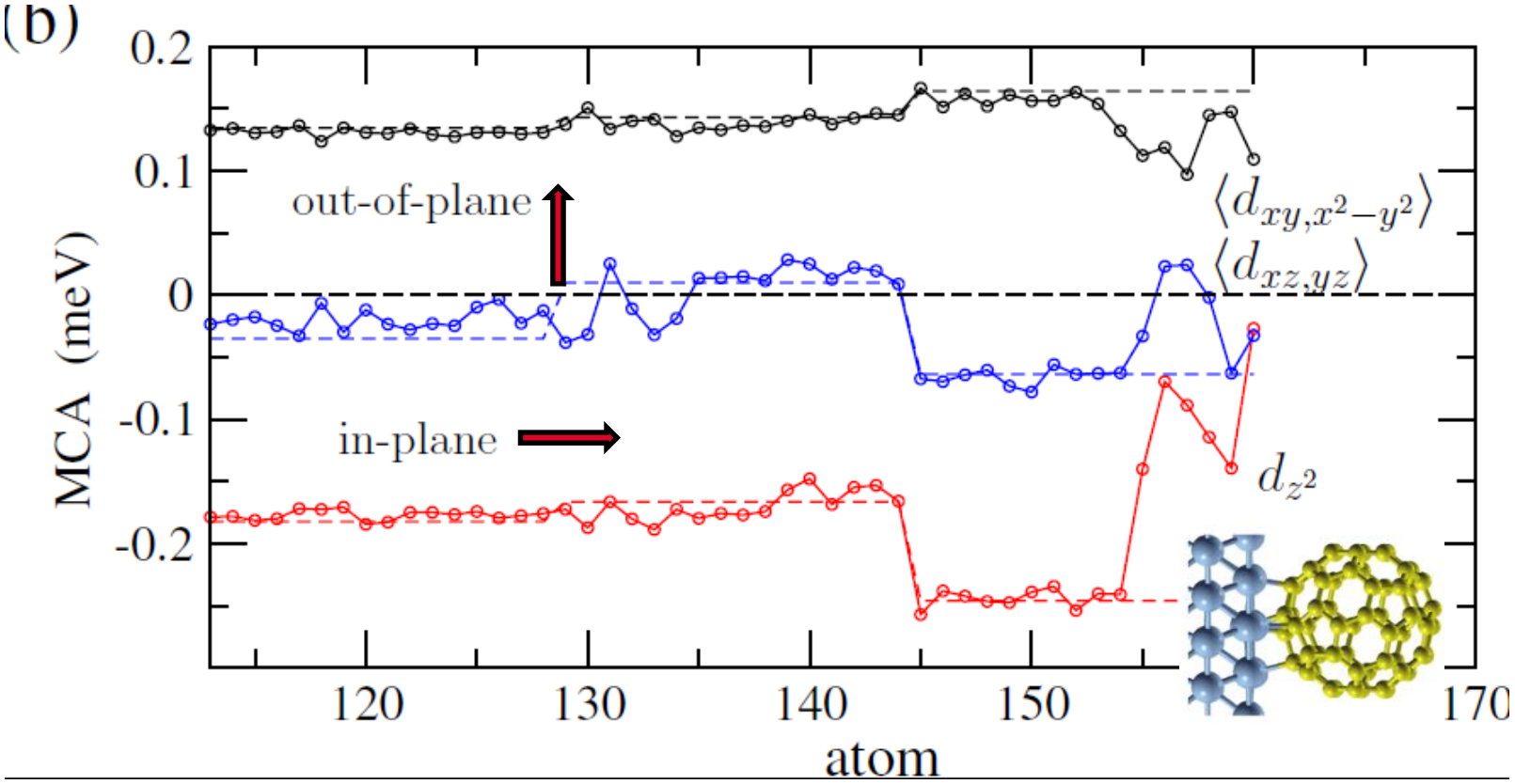
$$\Delta E^{ani} = \underbrace{\Delta E^{MCA}}_{0.9} + \underbrace{\Delta E^{shape}}_{0.6} = 1.5meV$$

For 5x16 Co atoms $\Delta E^{ani} = 19\mu eV / atom$



C60/Co TB MODEL

Orbital-resolved MCA



Experiment: *C60 favors « out-of-plane » magnetization*

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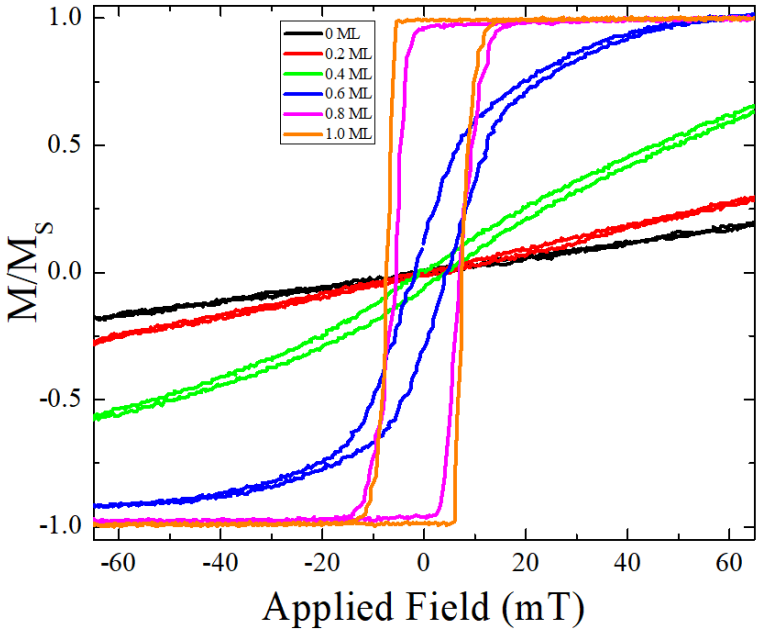
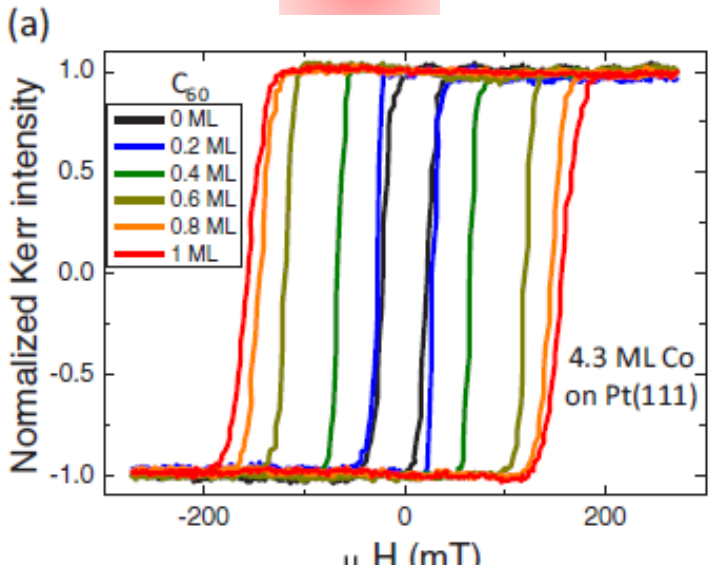
Theory: *C60 kills « in-plane » dz^2 component of MCA*

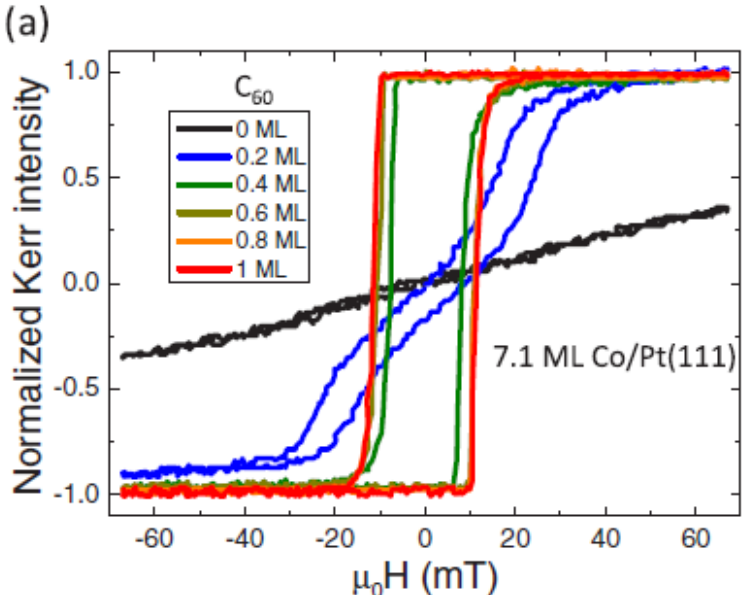
C_{60}/Co/Au(111) VERSUS C_{60}/Co/Pt(111) EXPERIMENT

Co/Au



Co/Pt



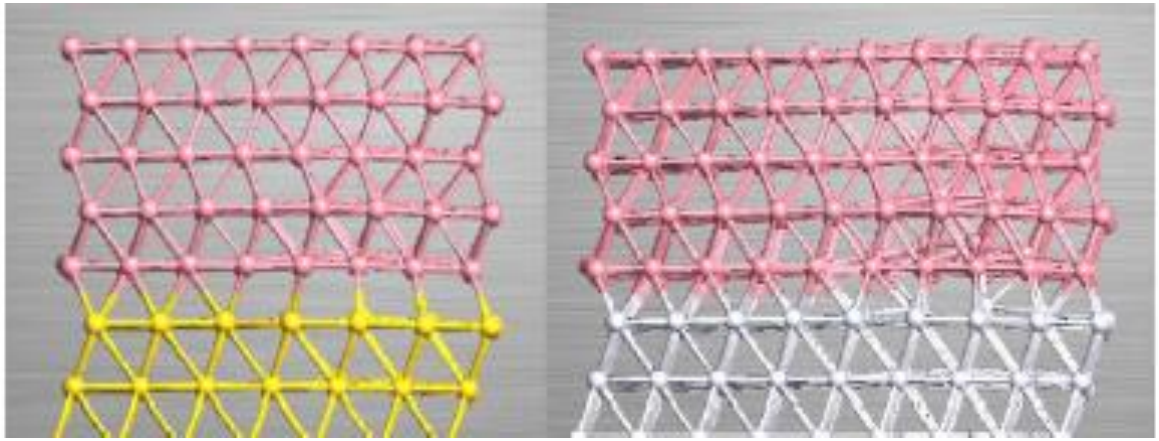


Increase of coercive field

100% for C_{60}/Co/Au
700% for C_{60}/Co/Pt

C_{60}/Co/Au(111) VERSUS C_{60}/Co/Pt(111) MODEL

Co/Au(111) Co/Pt(111)



3 possible impact of Co thin film structure

- Co Stacking: fcc vs hcp

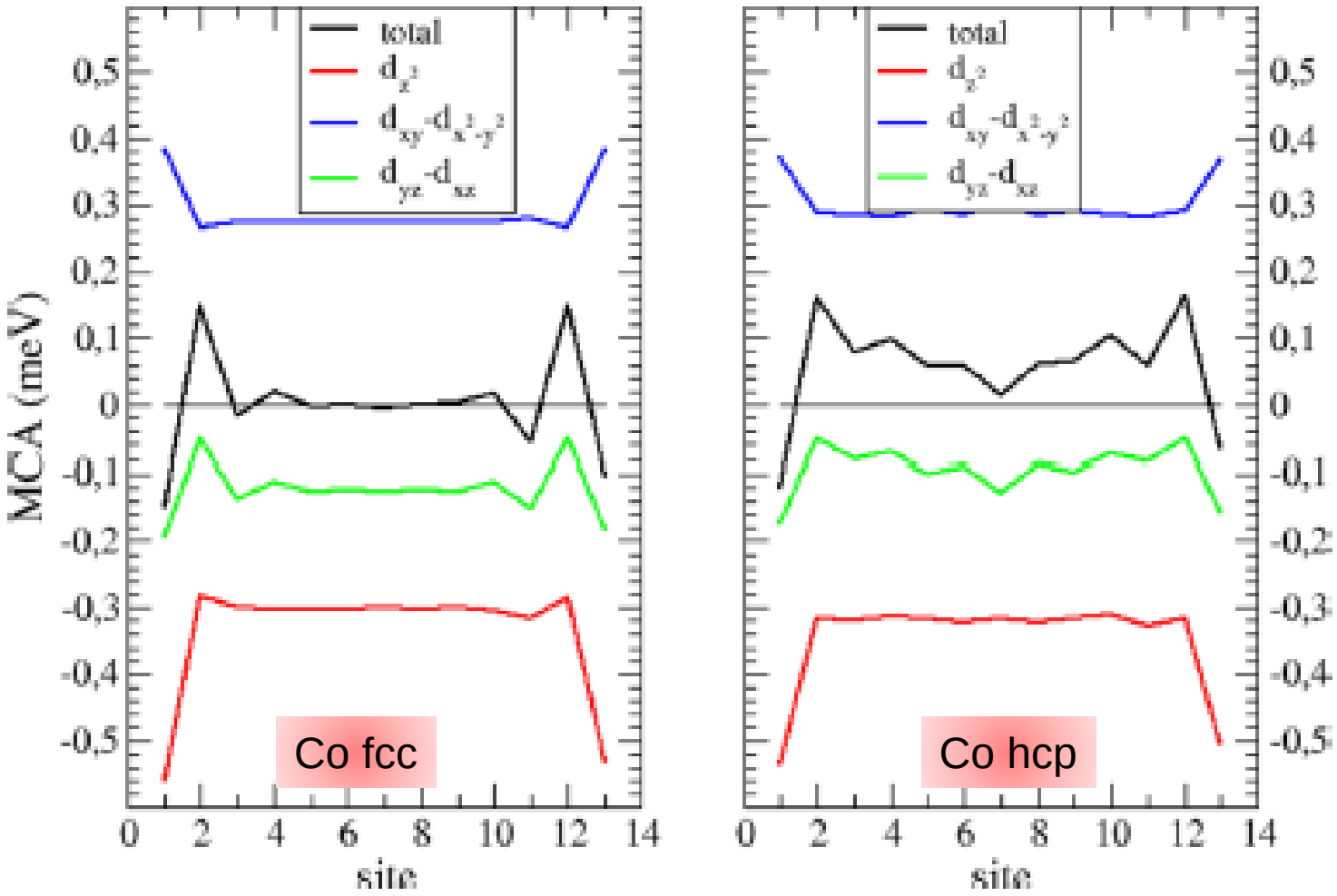
  fcc favored on Pt(111)

- ≠ C_{60} Adsorption geometry

  ≠ corrugation could induce
  ≠ adsorption site and geometry

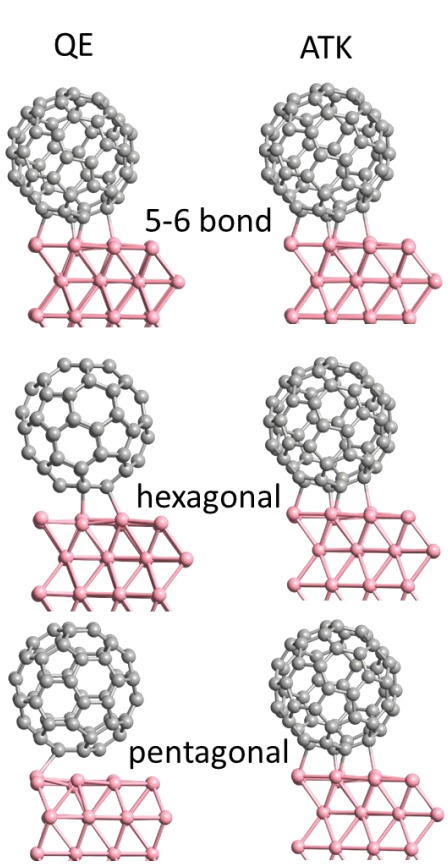
- ≠ Strain

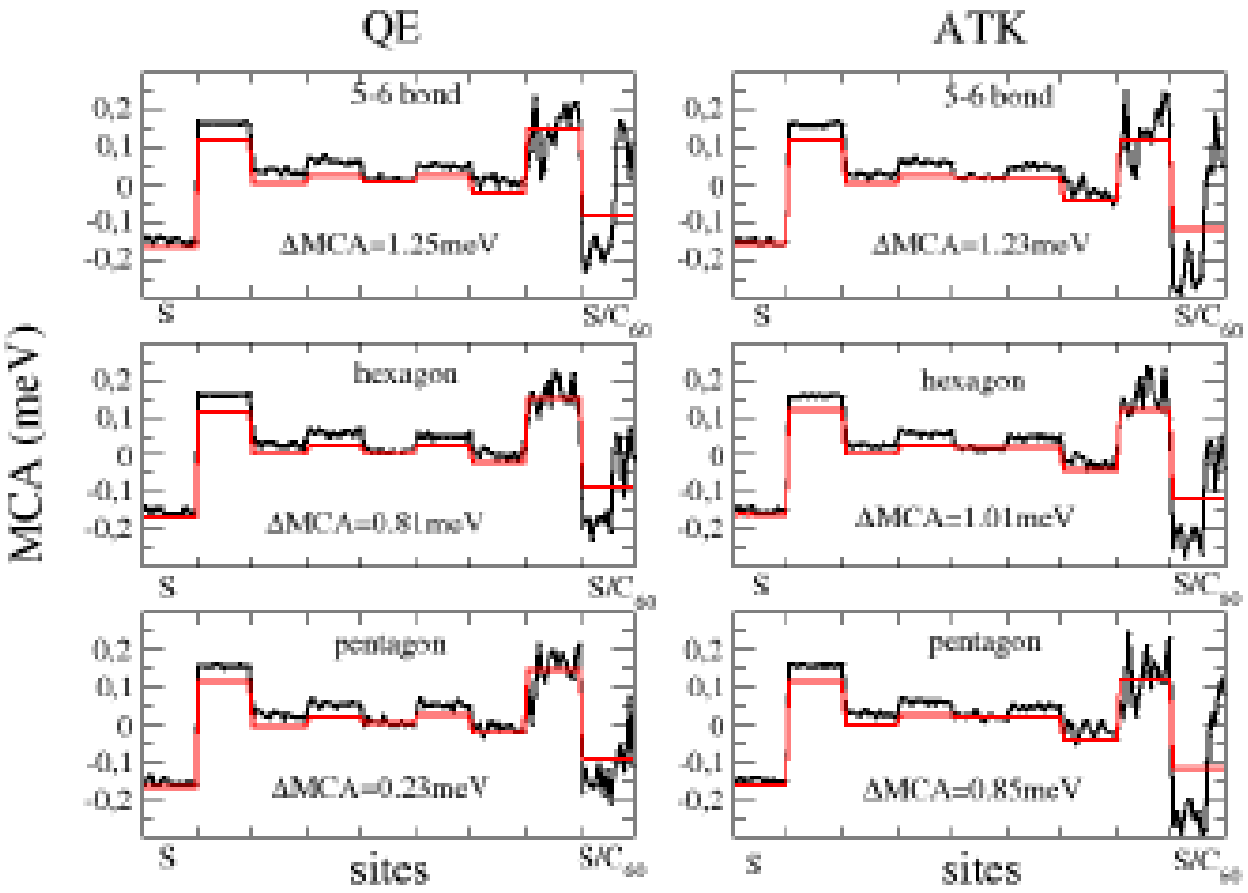
FCC VS HCP



Rather modest effect

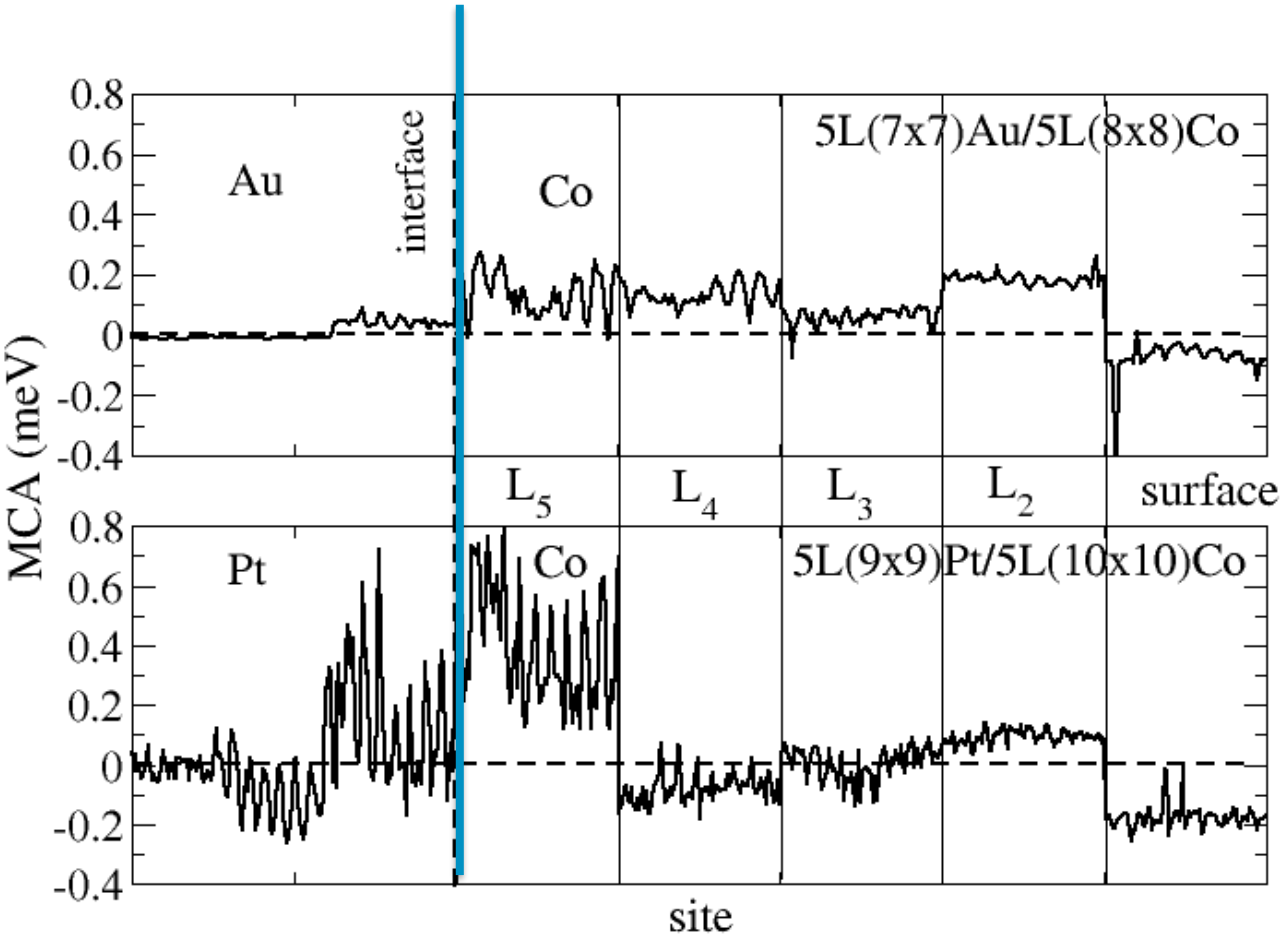
ADSORPTION GEOMETRY





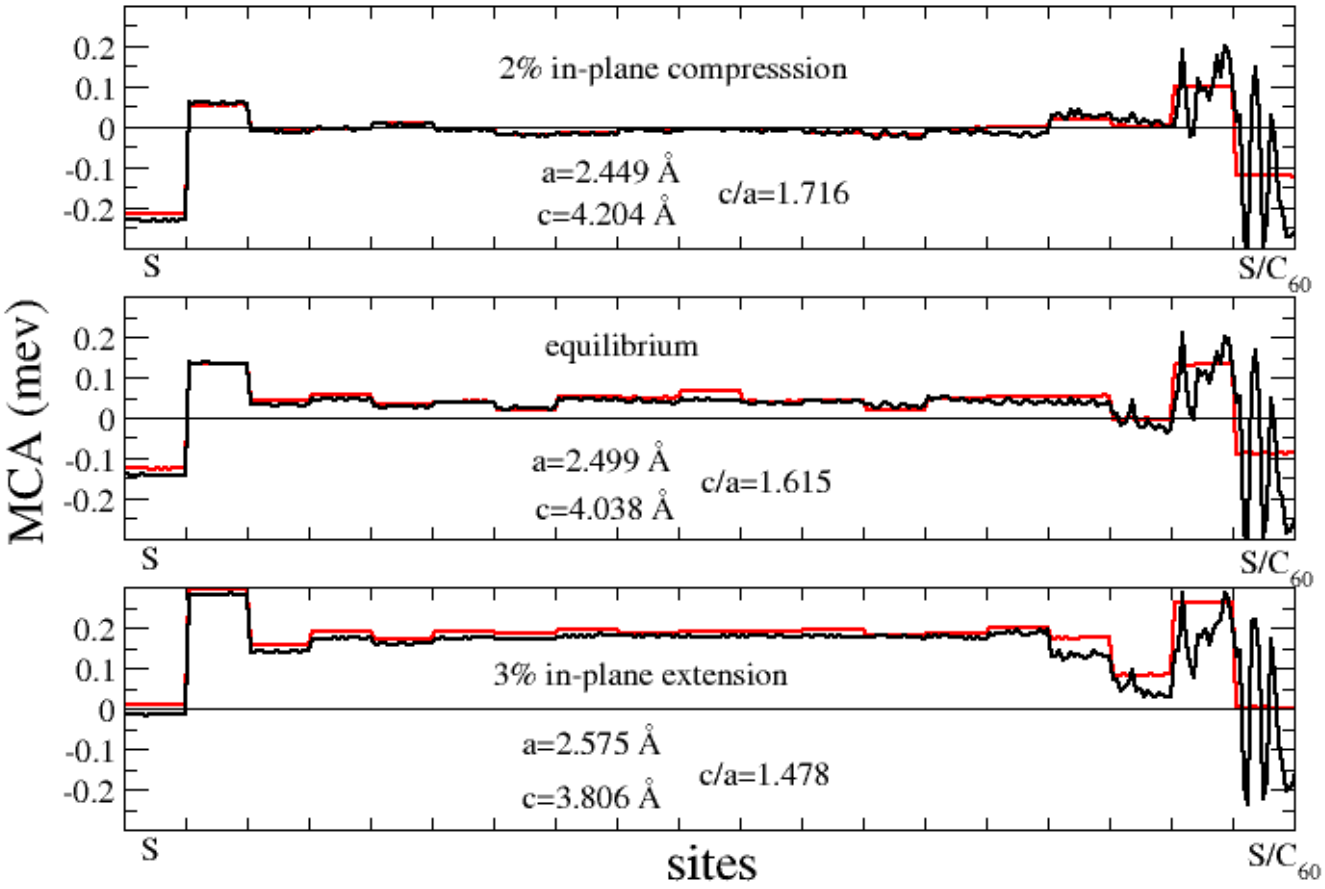
Rather important influence but cannot be correlated to STM observations.

STRAIN
Co/Au(111) VERSUS Co/Pt(111)



strain induced by interface can affect the surface MCA

STRAIN C_{60}/Co



$\Delta MCA > 1.5 meV$

Variation of MCA due to C60 adsorption is also very much influenced by strain

CONCLUSION
&
PERSPECTIVE

CONCLUSION & PERSEPCTIVES

- Possible tuning of magnetic anisotropy

- Optimal choice?

  - Adequate combination molecule/substrate based on orbital engineering

  - Critical influence of strain → **straintronics!**

  - Optimal substrate: thin film thickness..

- Reversible tuning?

  - Adequate switchable molecule?

ACKNOWLEDGEMENTS

ANR, SPIROU

SPIn **R**esistance **O**n **U**ltimate molecular devices

FET open Grant No. 766726

COncepts and tool**S** in **M**olecular spintron**ICS**

THANK YOU FOR YOUR ATTENTION

QUESTIONS?

COMMENTS?