4.1 Co adatoms on Gr/Ru(0001) and Gr/Ir(111)

Graphene is a 2D material with potential applications in spintronics. As we have discussed in exercise 2.3, the magnetic properties of Co adatoms adsorbed on graphene can be tuned by choosing the appropriate metallic support for graphene.

In exercise 2.3 we have found the energy splitting of the d-orbitals for Co adatoms deposited on Gr/Ru(0001) and Gr/Ir(111) as well as the L_z value of the orbital moment.
Here we want to calculate the MCA for the two systems.

1) Demonstrate that without spin-orbit coupling, L_x =0 for both system.
2) We know that spin-orbit partially restore the orbital moment. Find L_x in the two cases
3) Evaluate the MCA ($K_{MCA} \approx \lambda \boldsymbol{S} \cdot (\boldsymbol{L}_z - \boldsymbol{L}_{x,y})$) for the two systems. Are they equivalent?

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Tailoring the Magnetism of Co Atoms on Graphene through Substrate Hybridization

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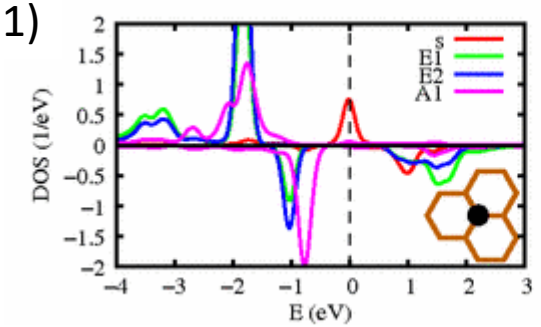
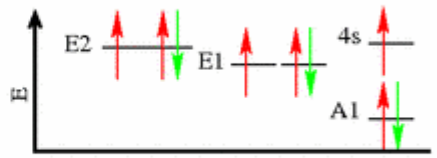
Adapted from: 10.1103/PhysRevLett.113.177201

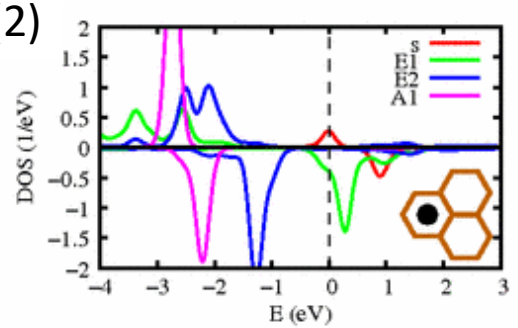
(1)

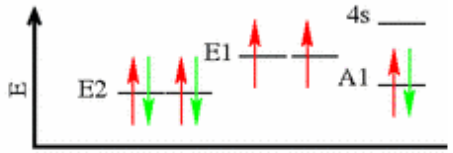




-2.18 eV A_1
0.52 eV E_1
0.57 eV E_2

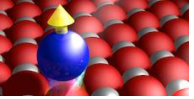
(2)





-0.38 eV A_1
0.65 eV E_1
-0.46 eV E_2

https://doi.org/10.1103/PhysRevB.81.115427



4.1 Co adatoms on Gr/Ru(0001) and Gr/Ir(111) - solution

1) $\langle d_n|L_x|d_{3z^2-r^2}\rangle = 0 \quad \langle d_{yz}|L_x|d_{x^2-y^2}\rangle = 0 \quad \langle d_{xz}|L_x|d_{xy}\rangle = 0$

Matrix elements are zero because the three multiplets are not degenerate

2) Using the matrix element and taking into account the Pauli exclusion principle we have that:

Co/Gr/Ru(0001) d^7: A1 can be transformed in E1 with contribution $\frac{-3\zeta}{\Delta(E_1-A_1)}$; E1 can be transformed in E2 with contribution $\frac{-\zeta}{\Delta(E_2-E_1)}$; thus

$$L_x = \frac{-3\zeta}{\Delta(E_1-A_1)} - \frac{\zeta}{\Delta(E_2-E_1)} = \frac{208}{2700} + \frac{66}{50} \approx 1.4$$

Co/Gr/Ru(0001) d^8: A1 can be transformed in E1 with contribution $\frac{-3\zeta}{\Delta(E_1-A_1)}$; E1 and E2 can be transformed one in the other with identical but opposite contributions $\frac{-\zeta}{\Delta(E_2-E_1)} + \frac{\zeta}{\Delta(E_2-E_1)} = 0$; thus $L_x = 0.08$

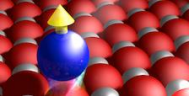
Co/Gr/Ru(0001): Since the total electronic configuration is 50% d^7 + 50% d^8 , $L_x = 0.74$

Co/Gr/Ir(111) d^8: A1 can be transformed in E1 with contribution $\frac{-3\zeta}{\Delta(E_1-A_1)}$; each of the E2 can be transformed in E1 $2*\frac{\zeta}{\Delta(E_2-E_1)}$; thus

$$L_x = \frac{-3\zeta}{\Delta(E_1-A_1)} + \frac{2\zeta}{\Delta(E_2-E_1)} = \frac{-208}{1030} + \frac{122}{1110} \approx 0.09$$

Co/Gr/Ir(111) d^9: A1 can be transformed in E1 with contribution $\frac{-3\zeta}{\Delta(E_1-A_1)}$; E2 can be transformed in E1 $\frac{\zeta}{\Delta(E_2-E_1)}$; thus $L_x = \frac{-3\zeta}{\Delta(E_1-A_1)} + \frac{\zeta}{\Delta(E_2-E_1)} = \frac{-208}{1030} + \frac{66}{1110} \approx 0.14$

Co/Gr/Ir(111): the total electronic configuration is 85% d^8 + 15% d^9 , $L_x = 0.1$



4.1 Co adatoms on Gr/Ru(0001) and Gr/Ir(111) - solution

3) The MCA is defined as $\boldsymbol{K_{mc} = \lambda\, S\cdot(L_z - L_{x,y})}$ with $\lambda = -\zeta/2S$

Co/Gr/Ru(0001) d^7: S = 3/2, $L_z = 1.0,\ \ L_x = 1.4 \Rightarrow K_{mc} = 0.6\,\zeta/3 = 0.2\,\zeta$ meV

Co/Gr/Ru(0001) d^8: S = 1, $L_z = 3,\ \ L_x = 0.08 \Rightarrow K_{mc} = \lambda = -1.46\,\zeta$ meV

Co/Gr/Ru(0001): Since the total electronic configuration is 50% d^7 + 50% d^8 , $K_{mc} = -0.63\,\zeta$ meV

Co/Gr/Ir(111) d^8: S = 1, $L_z = 0\ L_x \approx 0.09 \Rightarrow K_{mc} = 0.09\,\zeta/2 = 0.045\,\zeta$ meV

Co/Gr/Ir(111) d^9: S = 1/2, $L_z = 1, L_x \approx 0.14 \Rightarrow K_{mc} = -0.43\,\zeta$ meV

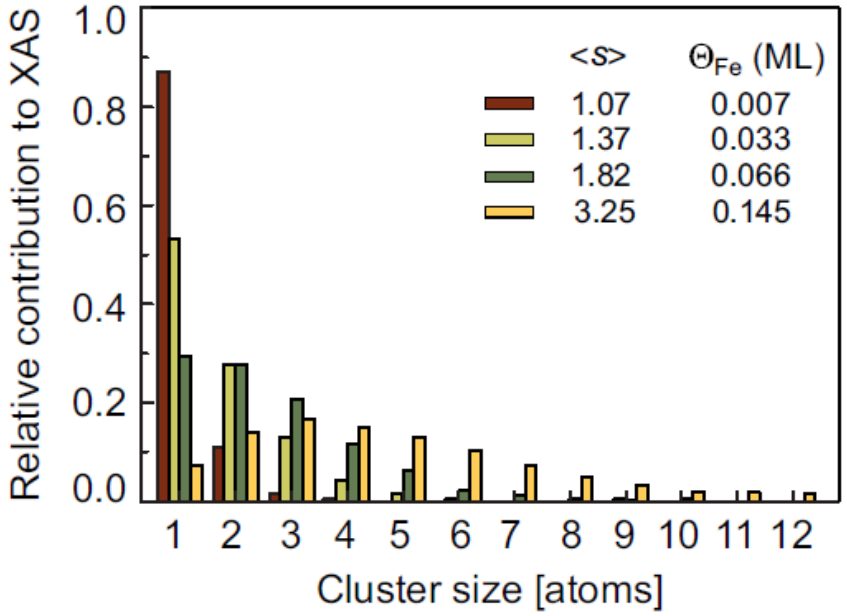
Co/Gr/Ir(111): the total electronic configuration is 85% d^8 + 15% d^9 , $K_{mc} = -0.026\,\zeta$ meV

Including all the electronic details the MCA in both cases is indeed smaller, about 20% of the simplified calculation performed here

4.2 MCA vs ΔL for Fe/Cu(111)

We perform measurements of spin and orbital moments for Fe clusters of different sizes deposited on Cu(111). The clusters are obtained by statistical growth depositing different amount of Fe with the Cu(111) kept at very low T, resulting in a certain dispersion for the cluster size. Values of the out-of-plane moments and clusters size dispersions are shown on the side. Assuming that the in-plane component of the orbital moment is zero, and that clusters with size $i \geq 4$ have the same orbital moment, calculate the MCA ($K_{MCA} \approx \lambda \boldsymbol{S} \cdot (\boldsymbol{L}_z - \boldsymbol{L}_{x,y})$) as a function of cluster size

Coverage (ML)	$\mu_S(\mu_B)$	$\mu_L\ (\mu_B)$	L_3/L_2	$\langle s \rangle$
0.007	2.2 ± 0.1	0.66 ± 0.04	4.2	1.07
0.011	1.95 ± 0.05	0.67 ± 0.05	4.4	1.12
0.033	2.07 ± 0.04	0.55 ± 0.03	4.3	1.37
0.066	2.15 ± 0.04	0.45 ± 0.03	3.9	1.82
0.145	2.12 ± 0.05	0.32 ± 0.02	3.3	3.25
1 [43]	0.7 ± 0.2	0.045 ± 0.015		



Adapted from https://doi.org/10.1103/PhysRevB.91.235426

4.2 MCA vs ΔL for Fe/Cu(111) - Solution

Let introduce the notation L_i for the out-of-plane component of the orbital moment of an object of size *i* such that $L(s) = \sum_i r_i(s) L_i$ where $r_i(s)$ is the relative population of object of size *i*

From the table and figure we deduce that:

1) L(1.07) = 0.88 L_1 + 0.12 L_2 = 0.66
2) L(1.37) = 0.53 L_1 + 0.28 L_2 + 0.12 L_3 + 0.07 L_4 = 0.55
3) L(1.82) = 0.28 L_1 + 0.3 L_2 + 0.22 L_3 + 0.2 L_4 = 0.45
4) L(3.25) = 0.08 L_1 + 0.15 L_2 + 0.18 L_3 + 0.59 L_4 = 0.32

Solving the system we find: L_1 = 0.69, L_2 = 0.47, L_3 = 0.32, and L_4 = 0.23

The MCA is given by $K_{MCA} \approx \lambda \boldsymbol{S} \cdot (\boldsymbol{L}_z - \boldsymbol{L}_{x,y})$ with λ=-ζ/2S where $\zeta_{Fe} = 52\ meV$ Since μ_S = 2S = 2 in the error bar independent on the cluster size, we have that λ = 26 meV; from this we deduce $K_i = \lambda\, S{\cdot}L_i$

K_1 = 18 meV, K_2 = 12 meV, K_3 = 8 meV, K_4 = 6 meV

N.B.: Fitting the hysteresis curves for different orientations of the magnetic field we find $K_1 = 1.8 \pm 0.4$ meV.

Coverage (ML)	$\mu_S(\mu_B)$	$\mu_L\ (\mu_B)$	L_3/L_2	$\langle s \rangle$
0.007	2.2 ± 0.1	0.66 ± 0.04	4.2	1.07
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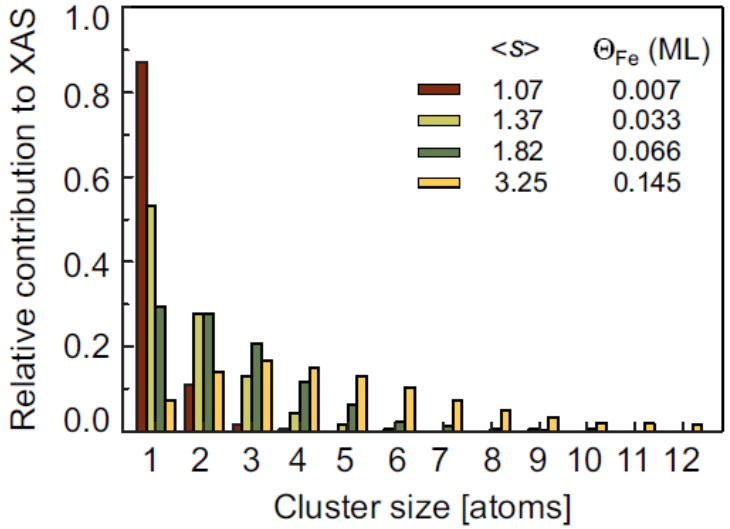
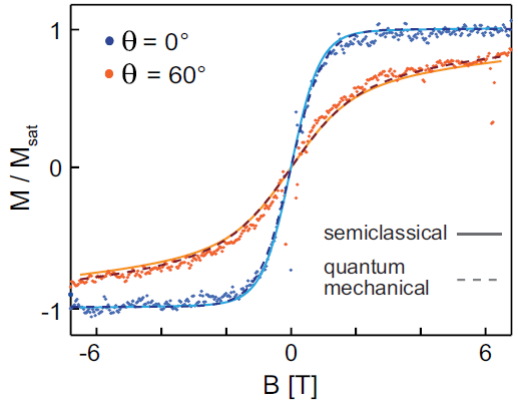
0.066	2.15 ± 0.04	0.45 ± 0.03	3.9	1.82
0.145	2.12 ± 0.05	0.32 ± 0.02	3.3	3.25
1 [43]	0.7 ± 0.2	0.045 ± 0.015		





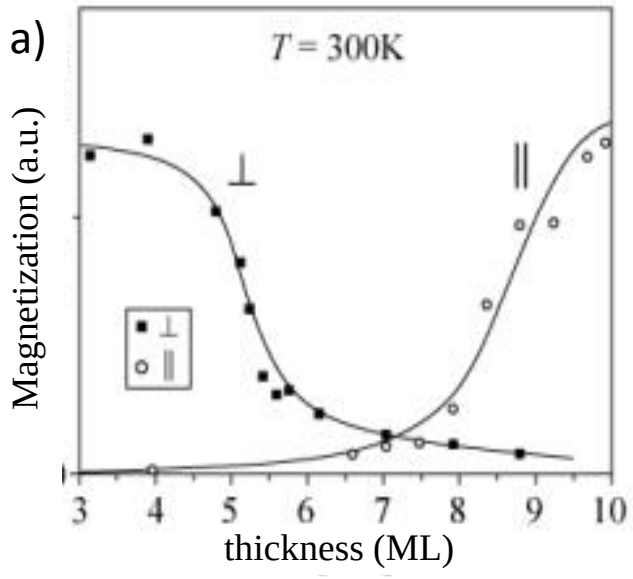
4.3 spin re-orientation transition for Co/Pt(111)

We measure the in-plane ($M_{//}$) and out-of-plane ($M_{\perp}$) component of the remnant (B = 0) magnetization for thin films of Co on Pt(111) as a function of the Co thickness *t*, as reported in figure a). In figure b) we report the magnetization curves measured for a film thickness of 40 ML.

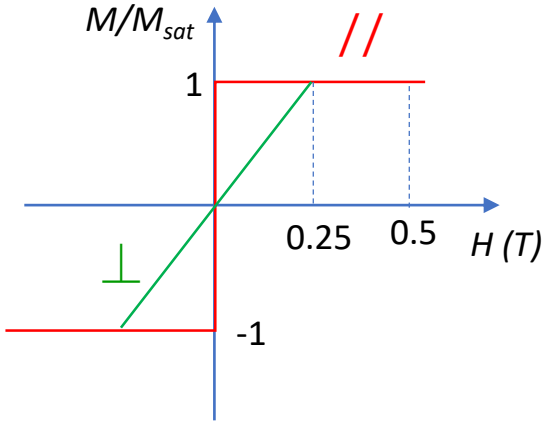
Evaluate the MCA due to the hybridization at the Co/Pt(111) interface, knowing that the magnetic moment of Co at saturation is about $m_S = 1.6\ \mu_B$

Co/Pt(111)

a)



b)



4.3 spin re-orientation transition for Co/Pt(111) - solution

The MAE of Co/Pt(111) is given by $MAE = K_{shape} + K_{MCA} + K_{int}$ where

K_{shape} is the demagnetization term,

K_{MCA} is the contribution due the crystallographic structure of the film, proportional to the Co volume = $surf * t$, and

K_{int} is the contribution due to the Co/Pt interface, proportional to the film surface.

For a thick film, the interface contribution is negligible

From figure b), we deduce that $K_{bulk}(40ML) = K_{shape} + K_{MCA} = E_{//} - E_{\perp} = \frac{1}{2} M H_{sat}$

with $H_{sat} = 0.05T$

$\Rightarrow$ each atom contribute to the bulk MAE with $K_{bulk,\,at} = \frac{1}{2} * 0.25 * 1.6\ \mu_B = 0.012 \frac{meV}{atom}$

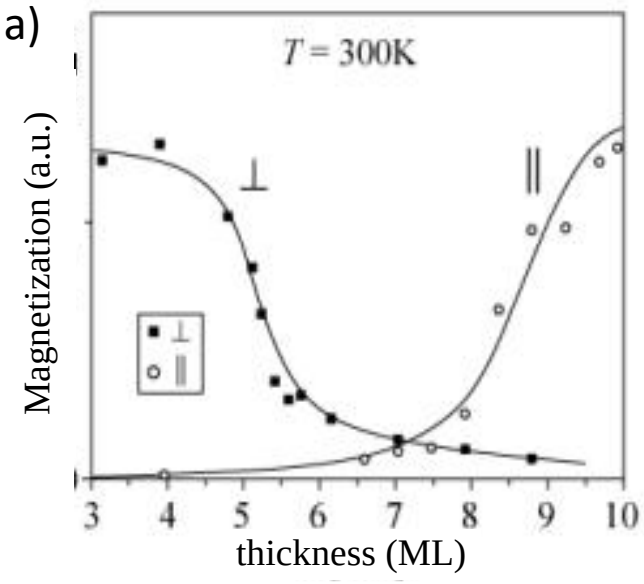
The total bulk contribution is $K_{bulk}(t) = K_{bulk,\,at}(t) * surf * t$

From figure a) we deduce that the magnetization is isotropic for a film thickness of about 7 ML $\Rightarrow K_{int} = -K_{bulk}(7ML)$

$\Rightarrow K_{int} = surf * K_{int,\,at} = K_{bulk,\,at} * surf * 7 \Rightarrow K_{int,\,at} = 7K_{bulk,\,at} \approx 0.084\ meV/atom$

Co/Pt(111)

a)



b)