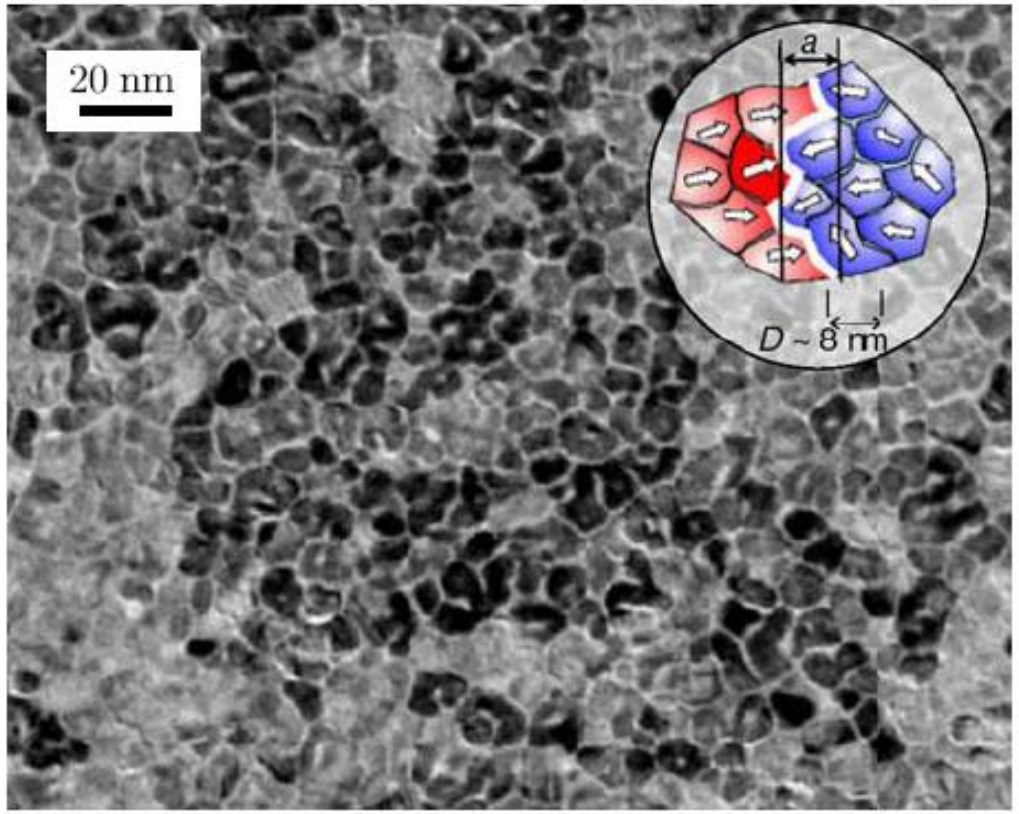
Some basic questions …..
… that need an answer



CoCrPt recording layer

1) Why every grain has a magnetic moment?
2) How the material is chosen?
3) Why every grain is magnetically decoupled from the neighbors
4) Why the grain magnetization is pointing in one specific direction?
5) Why the magnetization direction changes from grain to grain?
6) Why the grain magnetization does not fluctuate with time?
7) …

Magneto-crystalline anisotropy energy (MAE)

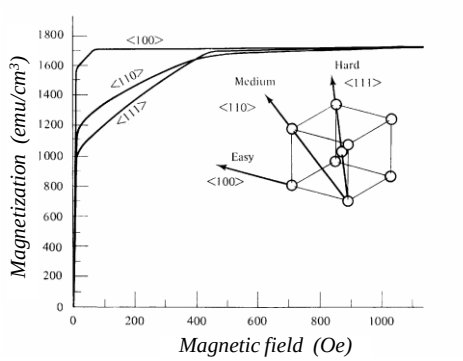
Define the space direction/s along which the magnetic moment prefers to align (together with the shape)

Bulk: MAE (K) depends on the crystal structure

Fe bcc

$K_1 = 4.8\times10^4\ J/m^3$

$= 2.4\ \mu eV/atom$

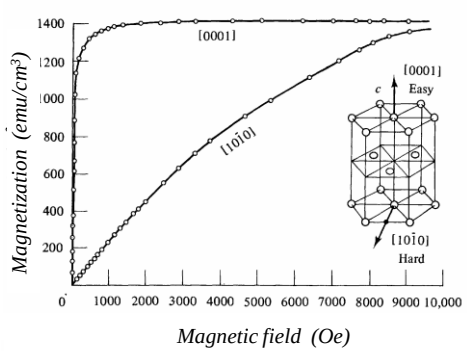


Co hcp

easy axis: (0001)

$K_1 = 4.1\times10^5\ J/m^3$

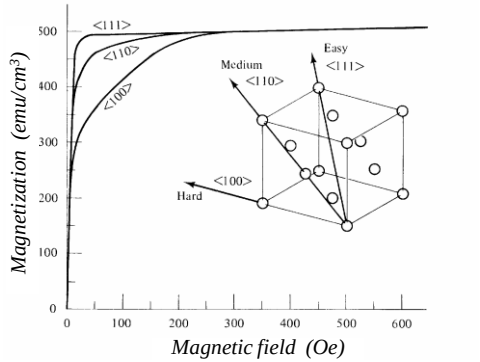
$= 45\ \mu eV/atom$



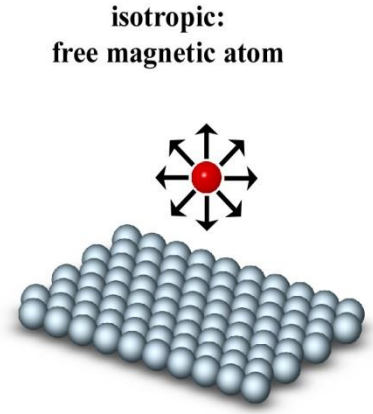
Ni fcc

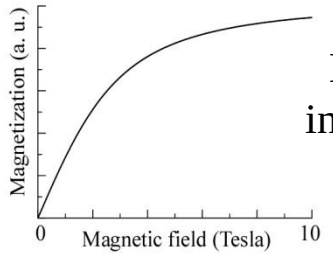
$K_1 = -5.5\times10^3\ J/m^3$

$= -0.3\ \mu eV/atom$

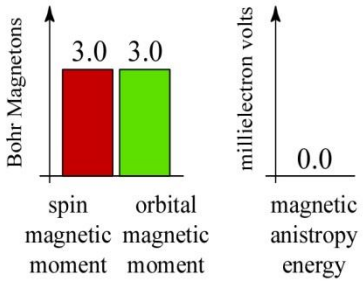


Free magnetic atom: spatially isotropic ($K = 0$)



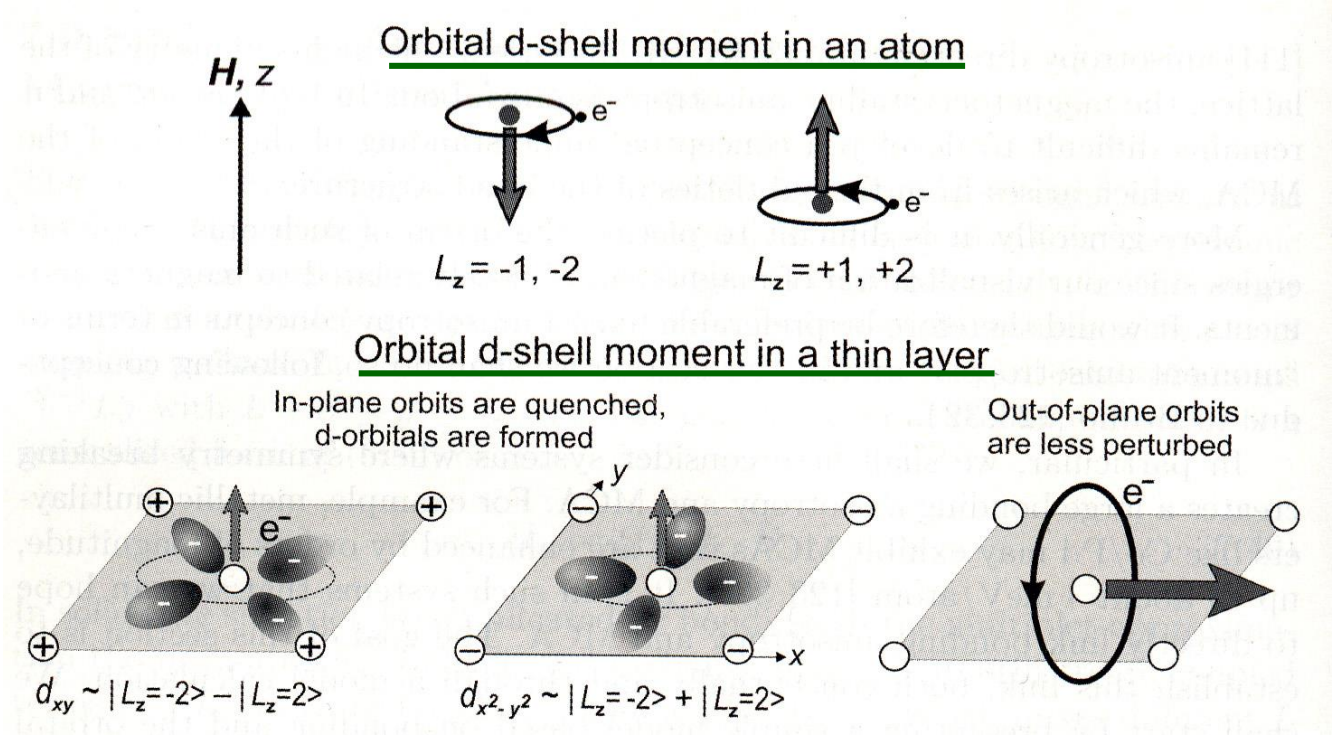


Direction independent



Co atoms

Origin of the MAE: Orbital moment and anisotropic bonds



Qualitative argument

A strong directional bond (crystal field) generates a reduction in the component of L perpendicular to the bond direction

- d electron in a free atom. Free orbital motion -> atom with maximum L_z due to Hund's rules

- atom in a plane forming bonds with neighbours atoms .

-a) in-plane orbital motion frozen by bond formation -> out-of-plane orbital moment is quenched

-b) out-of-plane orbital motion unperturbed by bonds -> in-plane orbital moment stays unquenched

⟹ symmetry breaking implies anisotropic orbital moments

-The spin moment S is isotropic.

However, spin-orbit (λ S·L) **locks** the spin along the spatial direction having maximum L

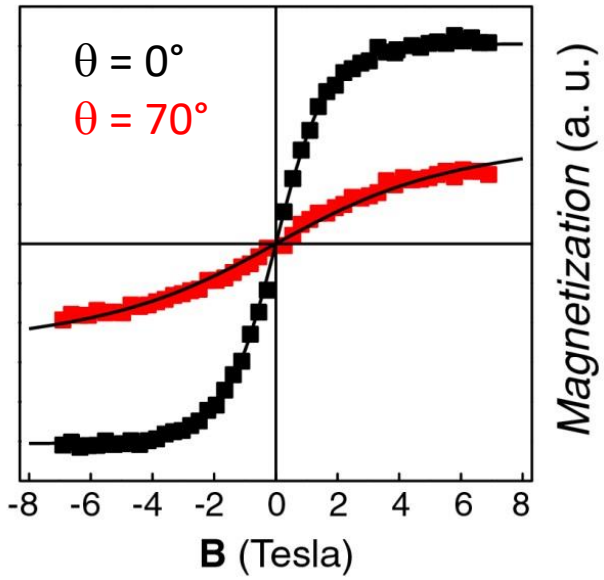
⟹ **easy axis of magnetization**

Magneto-crystalline anisotropy energy (MAE) = λ S·($L_z - L_{x,y}$) = λ S·ΔL

MAE (K) and orbital moment (L) *vs.* cluster size

MAE originates from spin orbit coupling and anisotropic orbital bond: MAE = λ S ΔL

Spin does not depend too much on size



θ is the angle between the normal to the surface and the field B

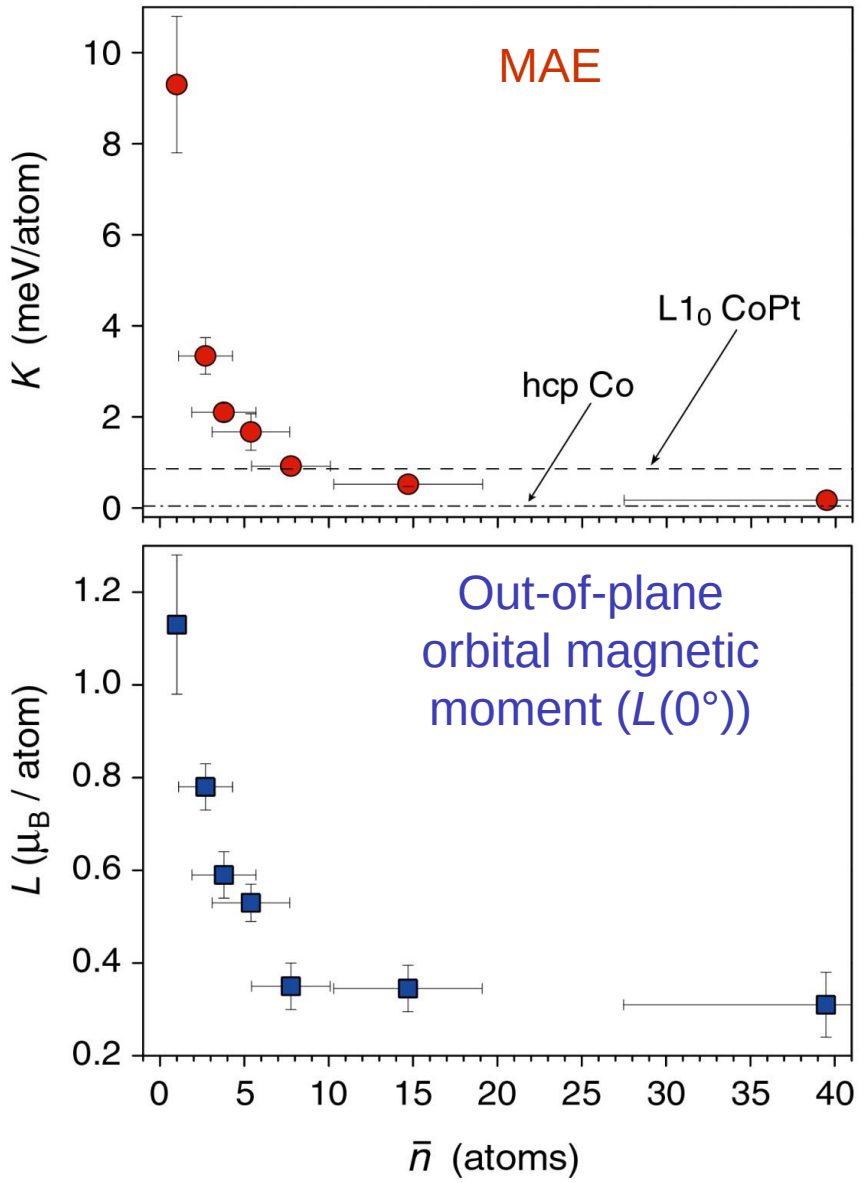
Using the XMCD (x-ray magnetic circular dichroism) technique, it is possible to measure L, S.

Fitting the magnetization curve knowing the adatom magnetic moment provides the MAE

What is the maximum theoretical value of the MAE for a Co atom?
Co: S ≈ 3/2; L ≈ 3; λ ≈ 20 meV
MAE ≈ 90 meV

Hybridization reduces L and thus K

Co clusters on Pt(111) as a function of the size ***n***

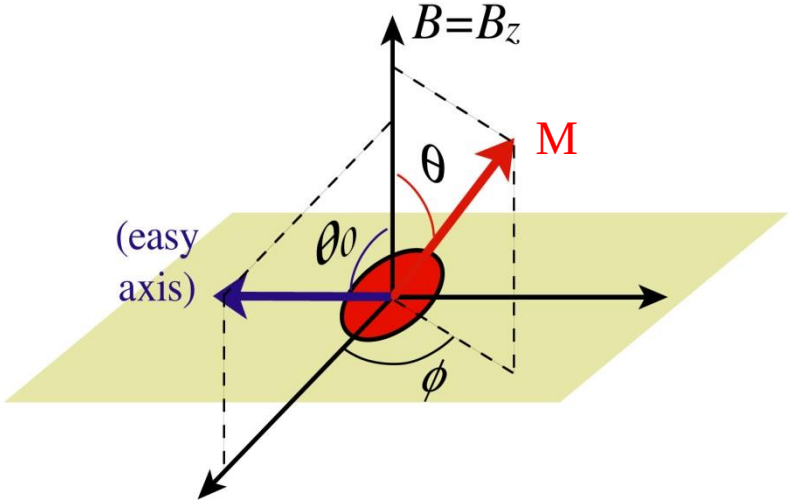


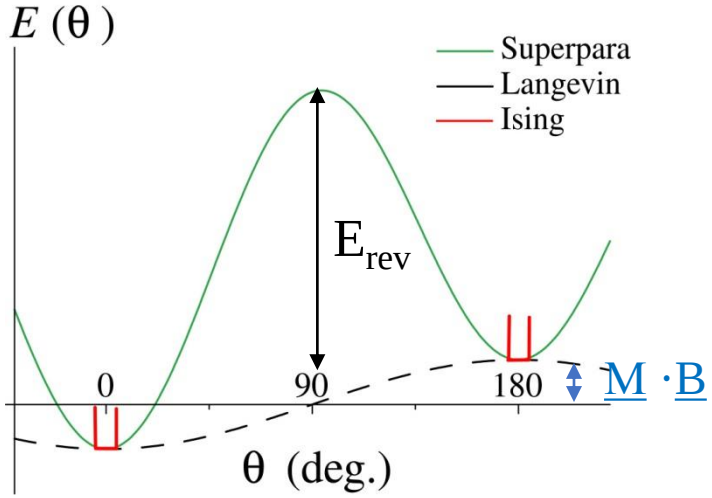
P. Gambardella *et al.*, Science **300**, 1130 (2003)

Bit magnetization thermal stability

A bit is a binary system where 1 and 0 correspond to the magnetization being up or down

Magnetization along a defined axis: easy magnetization axis





$$E(\theta,\theta_0,\phi) = -\mathbf{M}\cdot\mathbf{B} - E_{rev}\cos^2(\mathbf{easy\ axis}\cdot\mathbf{M})$$

Simplified phenomenological expression for the MAE

E_{rev} is the magnetization reversal energy barrier

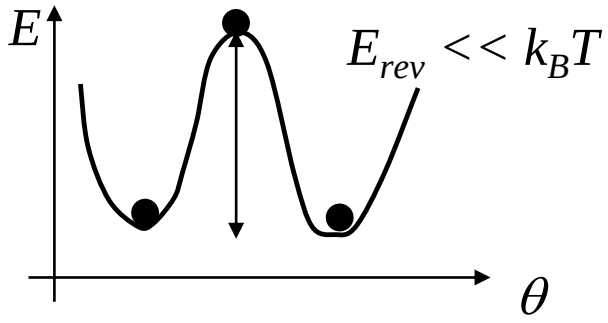
E_{rev} = MAE (crystal field) + shape anisotropy ($=\frac{1}{2}\mu_0 M^2$) assuming:

a) a single magnetic domain bit

b) coherent magnetization reversal (i.e. all spins in the bit turn at the same time)

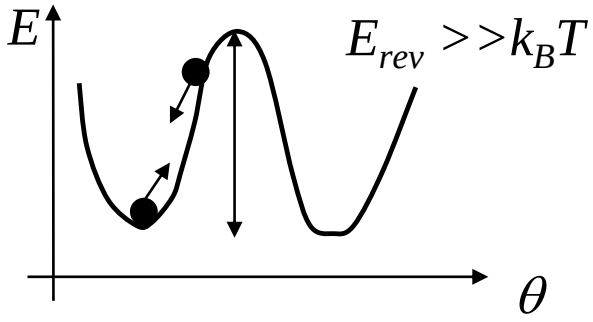
Bit magnetization thermal stability

B = 0 $E(\theta,\theta_0,\phi) = -E_{rev}\cos^2(\textbf{easy axis} \cdot \textbf{M})$



$E_{rev} << k_BT$
the magnetization vector isotropically fluctuates in the space.

Information **can not** be stored



$E_{rev} >> k_BT$
the magnetization vector can not switch the direction

Information **can** be stored

Relaxation time = Avg. time needed to jump from one minimum to the other:

$\tau = \tau_0 \exp(E_{rev}/k_BT)$ $\tau_0 \approx 10^{-10}$s

τ = 1 year → E_{rev} = 40 k_BT
τ = 1 second → E_{rev} = 23 k_BT

E_{rev} determines the thermal stability of the magnetization direction

The superparamagnetic limit

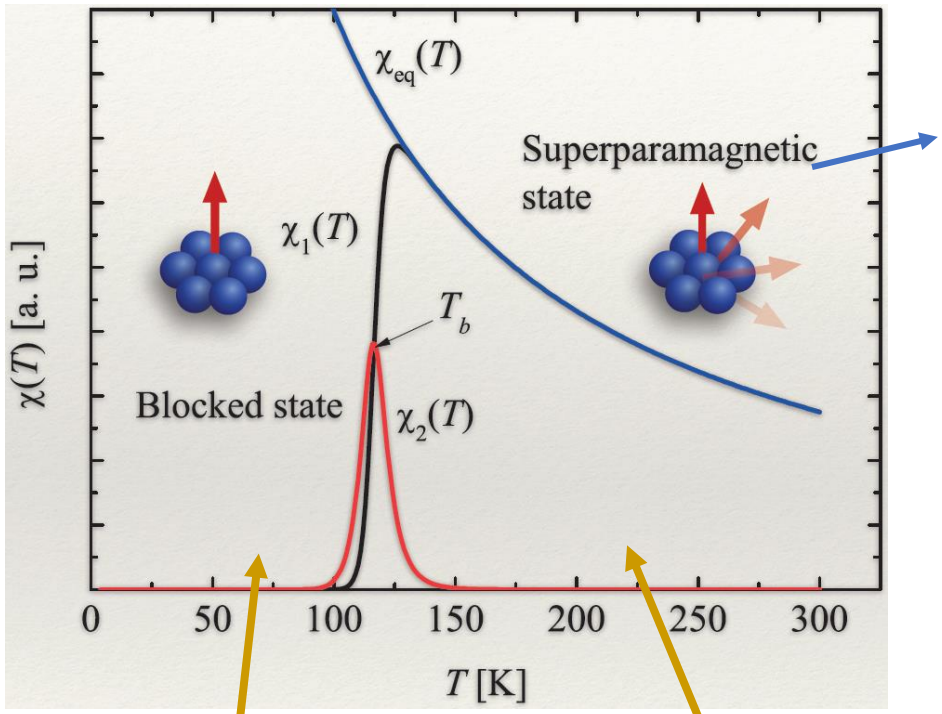
Blocking temperature T_b -> a way to measure E_{rev}

T_b is defined by the peak position in the susceptibility *vs* T curve

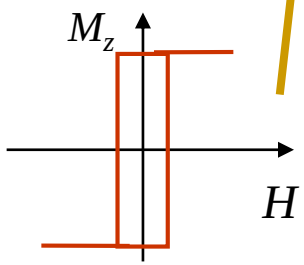
The macrospin feels:
1) Thermal agitation $\tau = \tau_0 \exp(E_{rev}/k_B T)$
2) External field $B = B_0 \sin(\omega t)$

The susceptibility peak is observed when the thermal agitation equals the oscillations induced by the external field:
$1/\omega \approx \tau_0 \exp(E_{rev}/k_B T_b)$

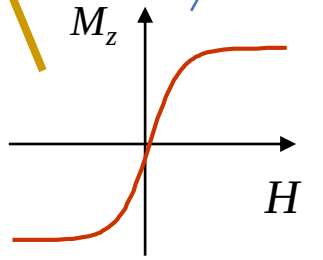
$T_b = E_{rev}/k_B \ln(1/\omega\tau_0)$,



Superparamagnetism:
- The atom spins in the clusters are still coupled by exchange to form a macrospin.
- The macrospin fluctuates similarly to the spin of a paramagnetic atom



Blocking: $E_{rev}/kT >> 30$



Superparamagnetic: $E_{rev}/kT << 30$

Writing field: Coherent magnetization reversal at T= 0 K

Single-domain particles: the Stoner-Wohlfart model

Magnetization of a single-domain particle in an external field.

$E = E_{Zeeman} + E_{mc} + E_{dm}$ E_{dm} = shape anisotropy

Suppose $\mu = MV$ = const. for any H value (coherent rotation) and, for simplicity, $E_{dm} = 0$. Φ = const.

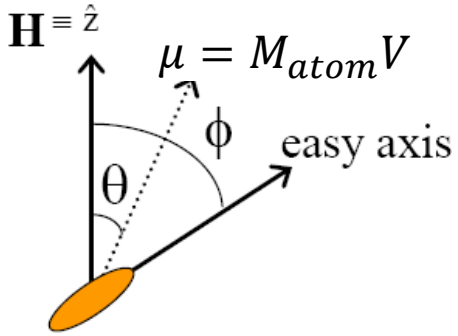
$E = -\mu H\cos\theta - K_1 V \cos^2(\theta-\phi)$ $E_{rev} = K V$

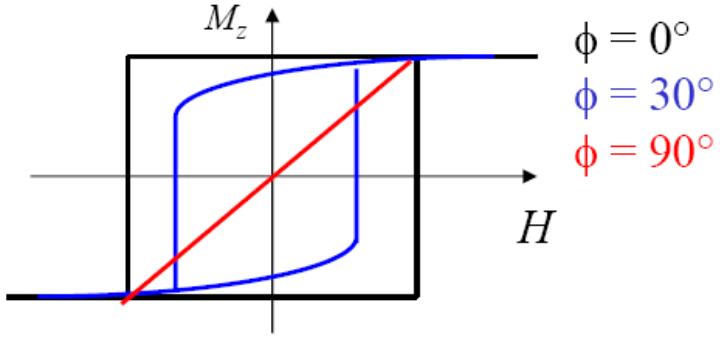
The magnetiic moment $\mu = MV$ will point along a direction that makes E a minimum:

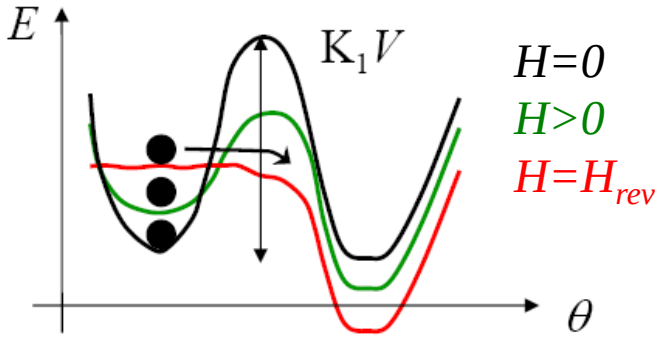
$$\frac{\partial E}{\partial \vartheta} = \mu H \sin\vartheta + K_1 V \sin(2(\vartheta - \varphi)) = 0 \quad (2)$$

Eq. 2 can be solved for θ and we can plot $M_z = M\cos\theta$ (this is what one usually measures) as a function of H.

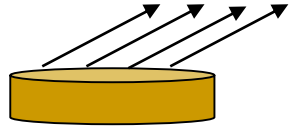
The reversal field is the field at which the energy minimum in eq. (1) vanishes ($\partial^2 E/\partial\vartheta^2 = 0$)







During the magnetization reversal all the atom spins in the particle stay aligned



M(H) curve for *H* along easy axis

H_{rev} is the field required to reverse the magnetization i.e. to write a bit

$$H_{rev} = 2E_{rev} / (M_{atom} V)$$

N.B.: K = 1.2 meV,
M_{atom} = 2 μ_B=2 0.06 meV/T ➡ H_{rev}= 20 T

Particle ensemble

Ensemble of noninteracting monodomain particles (macrospin *m*) and with uniaxial anisotropy K

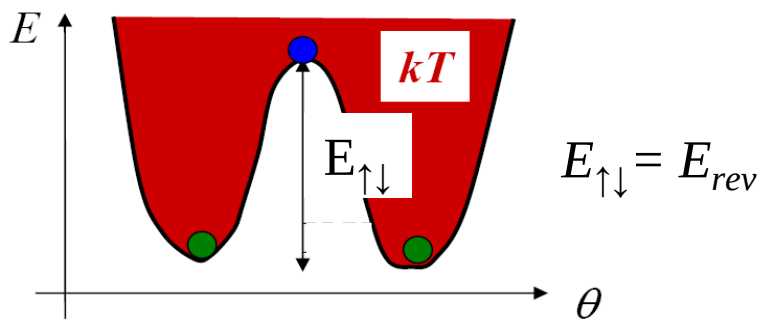
The magnetization curve represents the asymmetry in the number of particles pointing up ($n\uparrow$) or down ($n\downarrow$) changing over time with the applied field

$$\frac{dn_\uparrow}{dt} = -\kappa_{\uparrow\downarrow} n_\uparrow + \kappa_{\downarrow\uparrow} n_\downarrow$$

Transition rates

$$\kappa_{\uparrow\downarrow} = \nu_0 e^{-E_{\uparrow\downarrow}/k_B T}$$

$$E_{\uparrow\downarrow} = K \sin^2 \vartheta - m H \cos(\vartheta - \varphi)$$



$$E_{\uparrow\downarrow} = E_{rev}$$

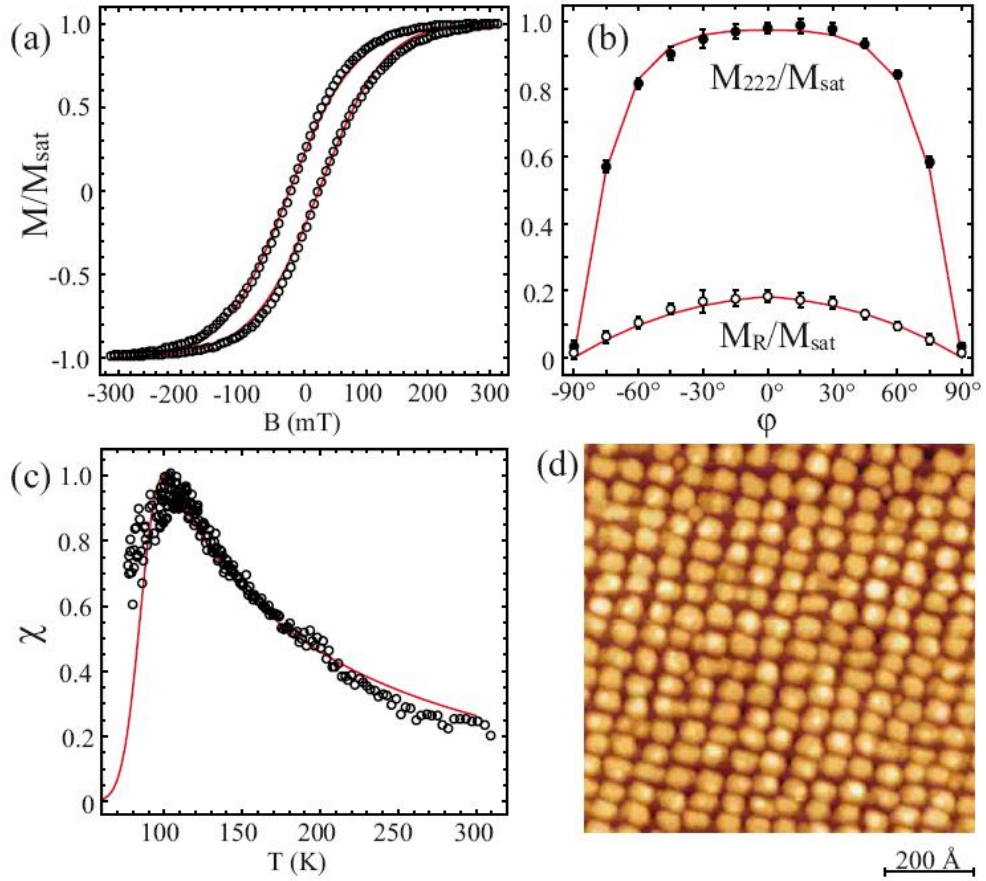
J. J. Lu *et al.*, J. Appl. Phys. **76**, 1726 (1994)

Distribution of writing fields

(experimentally we have a distribution in K and *m*)

1.1 ML Co/Au(11,12,12)

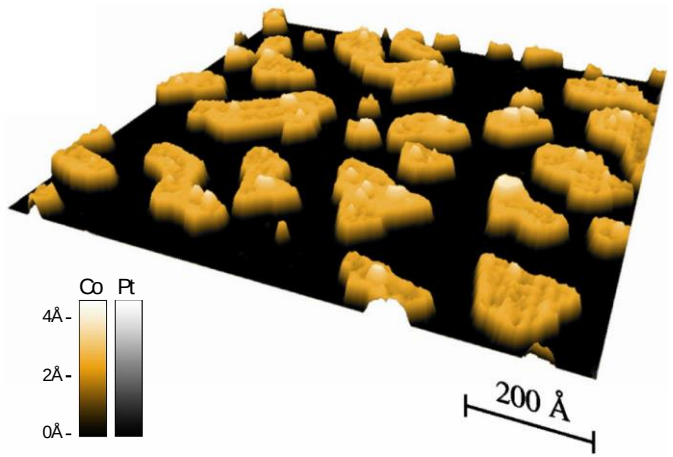
Two atomic layer high particles; mean size = 600 atoms



A. Lehnert *et al.*, Rev. Sci. Inst. **80**, 023902 (2009)

The edge does the job

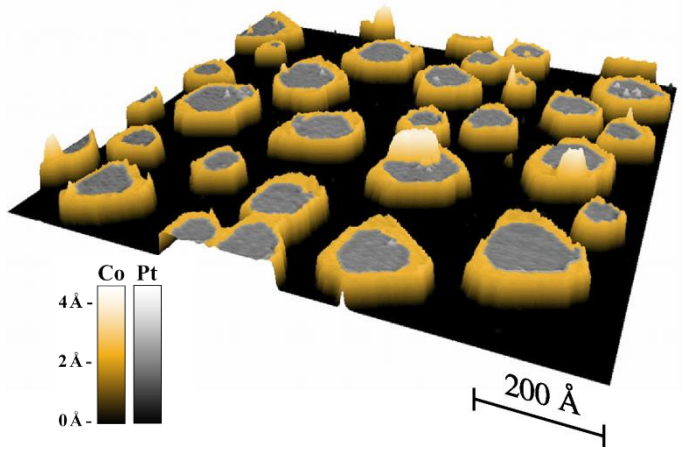
Co islands

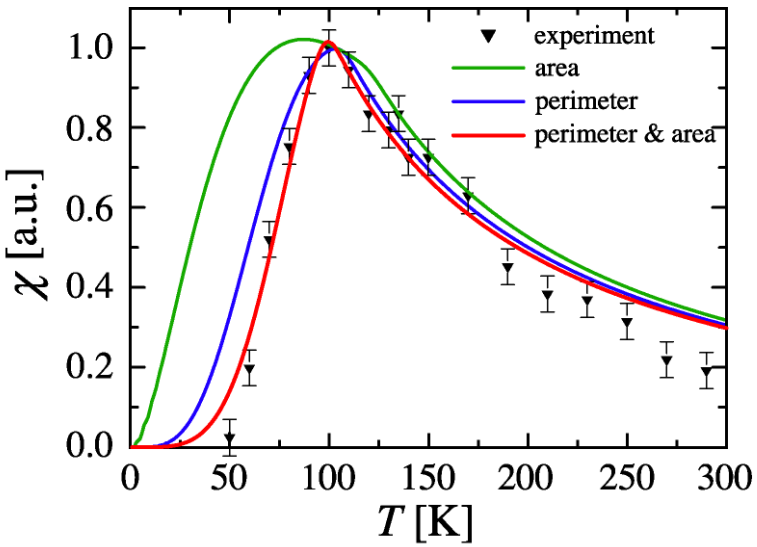


1 atomic layer high islands
Size about 1000 atoms

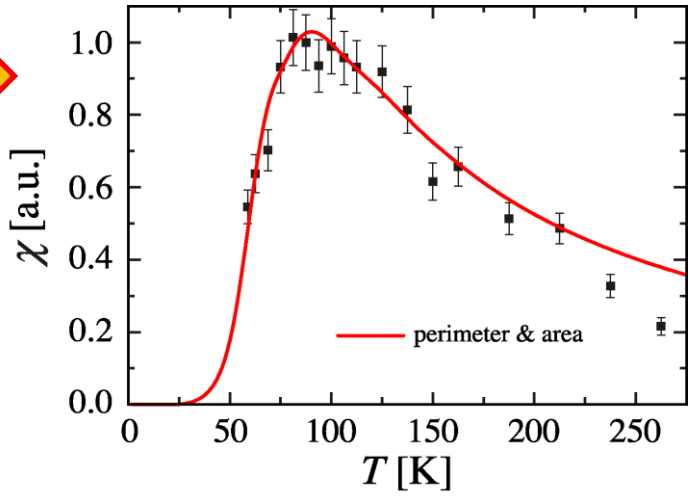
Out-of-plane easy axis

Pt core and Co shell





Same blocking temperature
T_b = 100 K



MAE originates from the edge

S. Rusponi *et al.*, Nature Mat. **2**, 546 (2003).

Atomic-scale engineering of the MAE

nature COMMUNICATIONS

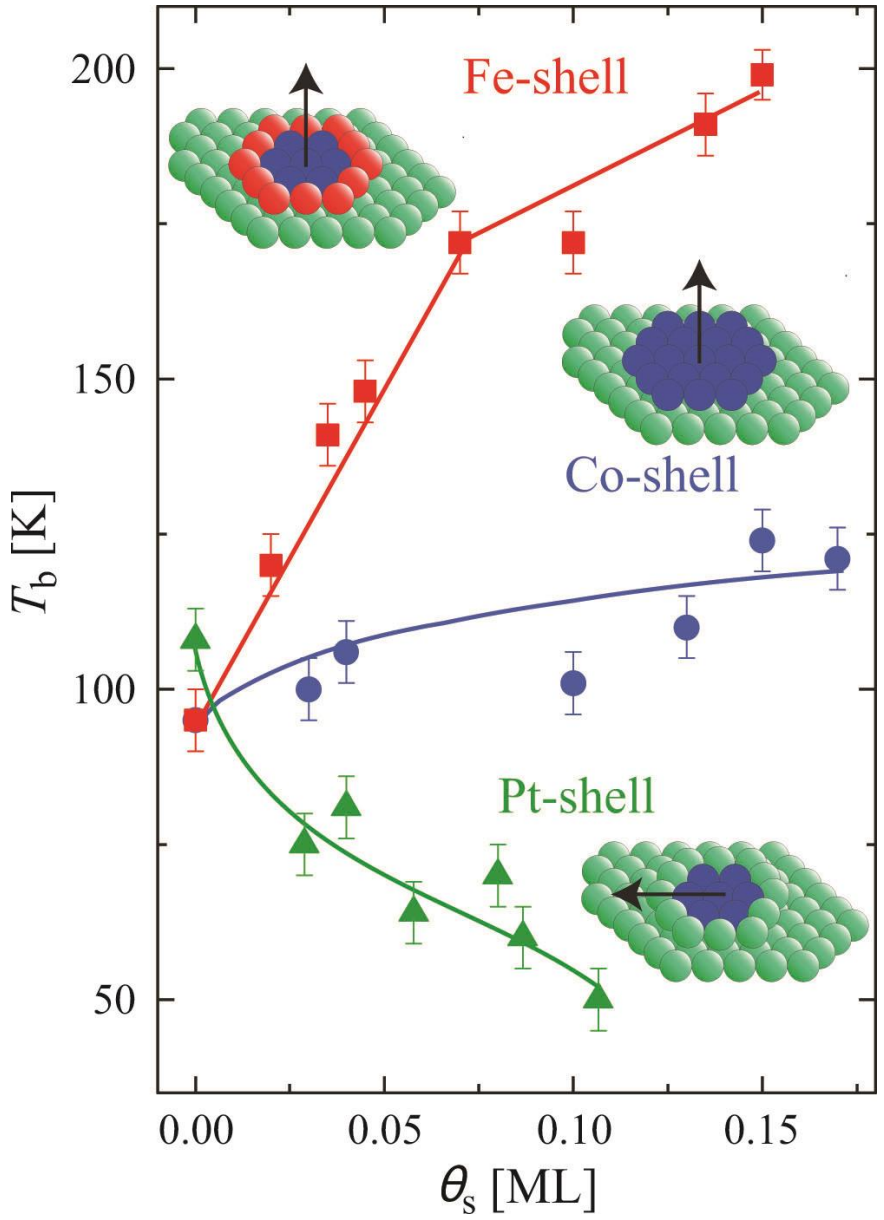
ARTICLE

Received 10 Sep 2012 | Accepted 22 Nov 2012 | Published 27 Dec 2012

DOI: 10.1038/ncomms2316 OPEN

Atomic-scale engineering of magnetic anisotropy of nanostructures through interfaces and interlines

S. Ouazi[1,*,†], S. Vlaic[1,*], S. Rusponi[1], G. Moulas[1], P. Buluschek[1], K. Halleux[1], S. Bornemann[2], S. Mankovsky[2], J. Minár[2], J.B. Staunton[3], H. Ebert[2] & H. Brune[1]

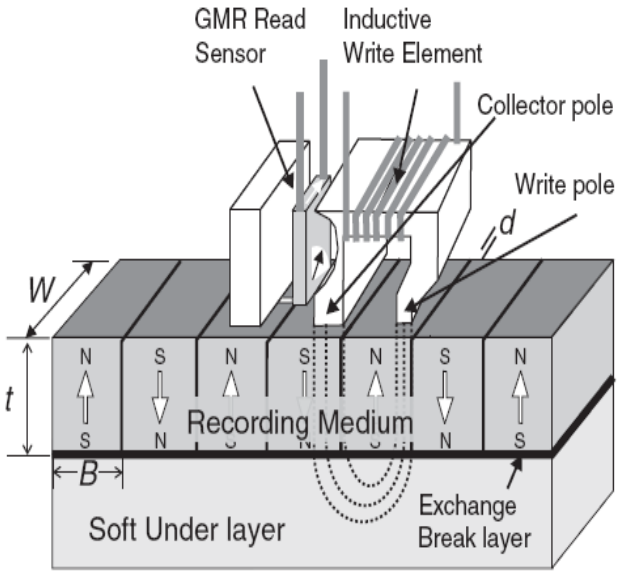


Island size
about 1000 atoms

FePt $L1_0$ phase for perpendicular recording

Perpendicular recording

HDD media: FePt in the $L1_0$ phase



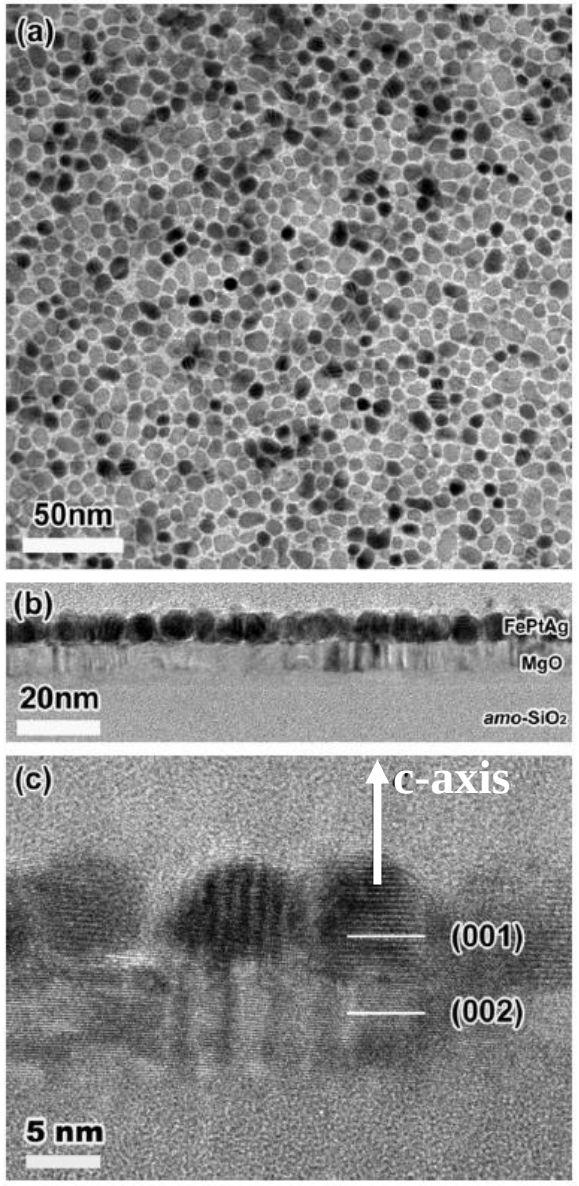


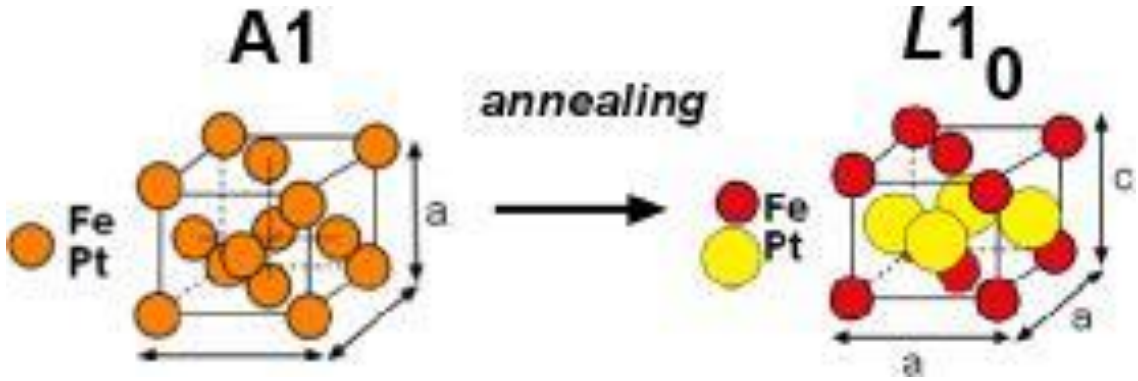
Fig. 8. (a) Bright field plane view image, (b) cross-sectional bright field image, and (c) cross-sectional high-resolution TEM image of the $(FePt)_{0.9}Ag_{0.1}$ –50vol%C film.

L. Zhang *et al.*, JMMM **322**, 2658 (2010)

Effect of crystal structure

Random distribution of Fe and Pt atoms

Ordered distribution of Fe and Pt atoms

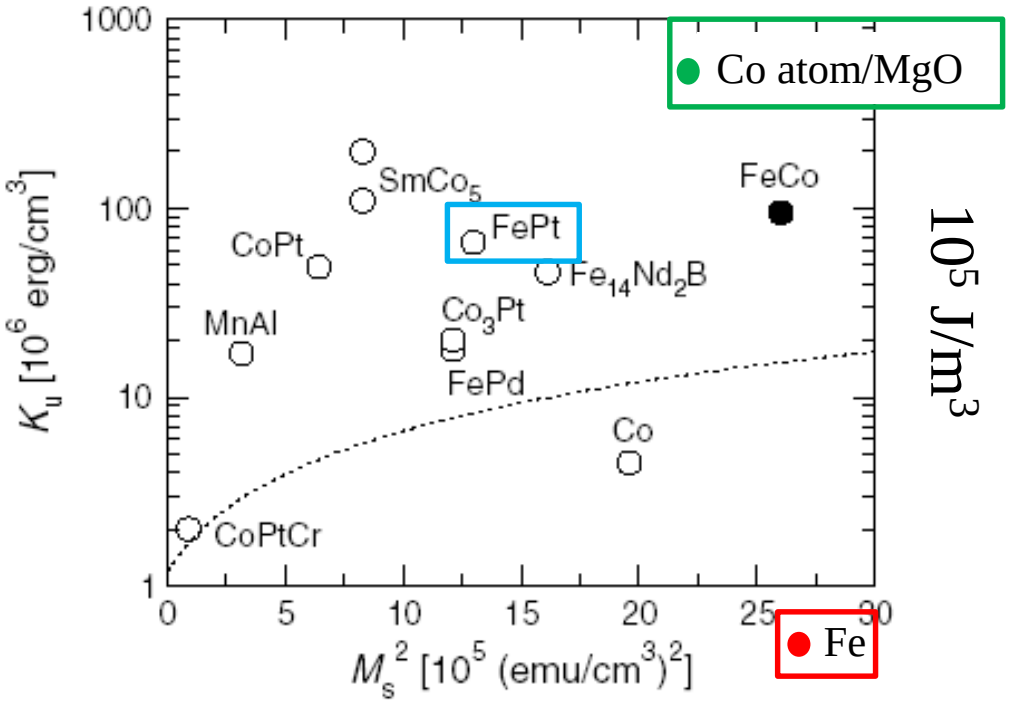


Basically isotropic

Strong easy axis along c-axis

In the alloy, every atom counts the same for the MAE (volume property)

Why FePt in $L1_0$ phase and not simply Fe bcc?



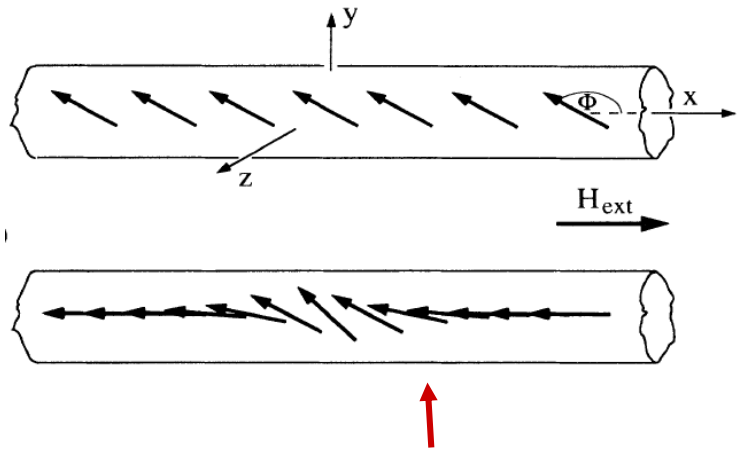
Storage media require materials with both:

- high K (to keep the magnetization orientation against thermal disorder)
- high M_s (to keep $H_{rev} = 2K/M_s$ within technological limit)

T. Burkert et al., *Phys. Rev. Lett.* 93, 027203 (2004).

Shape affects magnetization reversal mechanism: coherent *vs.* incoherent reversal

Wire with length L, section W, lattice parameter a.
Each atom contributes with a MAE = K



Coherent reversal: E_{rev} = K L W

Reversal by domain wall creation and displacement: $E_{rev} = 4\,W\sqrt{AK}$

$A = 2J_{ex}S^2/a$ is the stiffness (a is the atomic pitch)

This solution is true in the limit of H_{ext} ->0

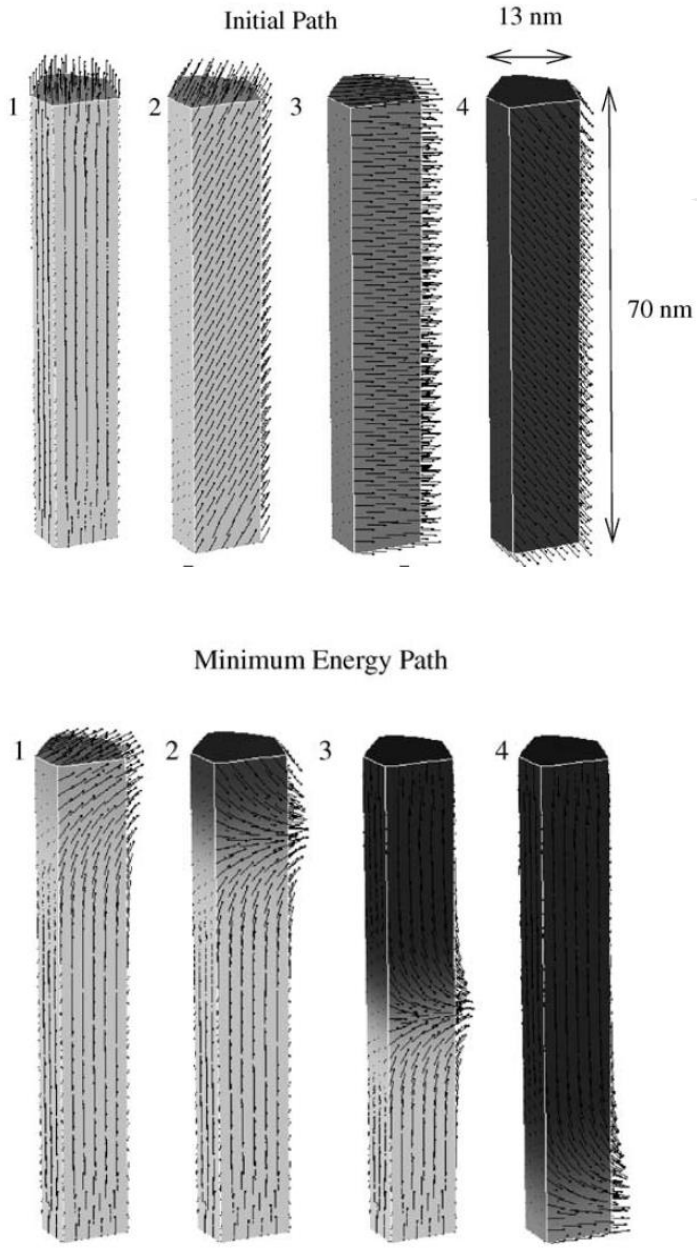
Domain wall displacement is favored if:

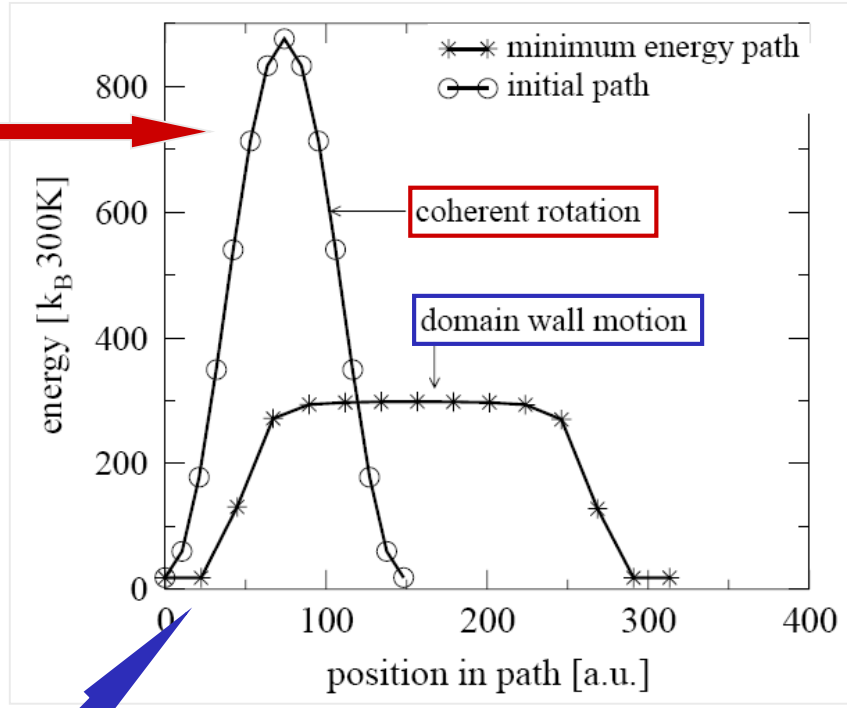
$$4\,W\sqrt{AK} < K\,L\,W$$

$$L > 4\sqrt{A/K}$$

H.-B. Braun Phys. Rev B **50**, 16502 (1994); H.-B. Braun Phys. Rev B **50**, 16485 (1994);
H.-B. Braun Phys. Rev Lett. **71**, 3557 (1993); H.-B. Braun J. Appl. Phys. **85**, 6172 (1999);

Particle shape affect magnetization reversal





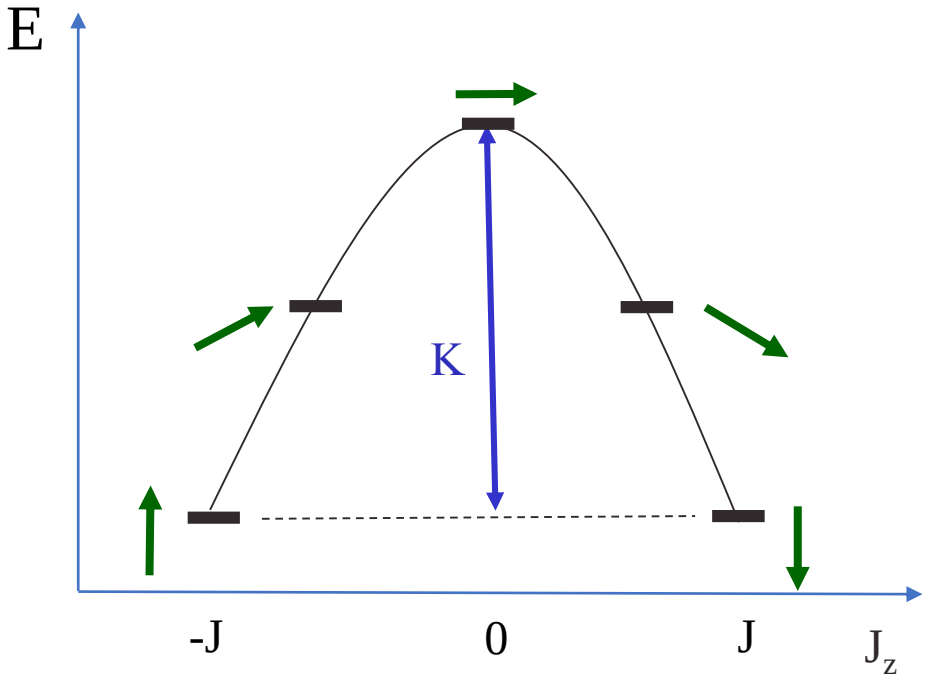
Domain wall motion costs less energy than coherent rotation

R. Dittrich *et al.* J. Magn. Magn. Matter. **250**, L12 (2002)

MAE: classic *vs.* quantum object

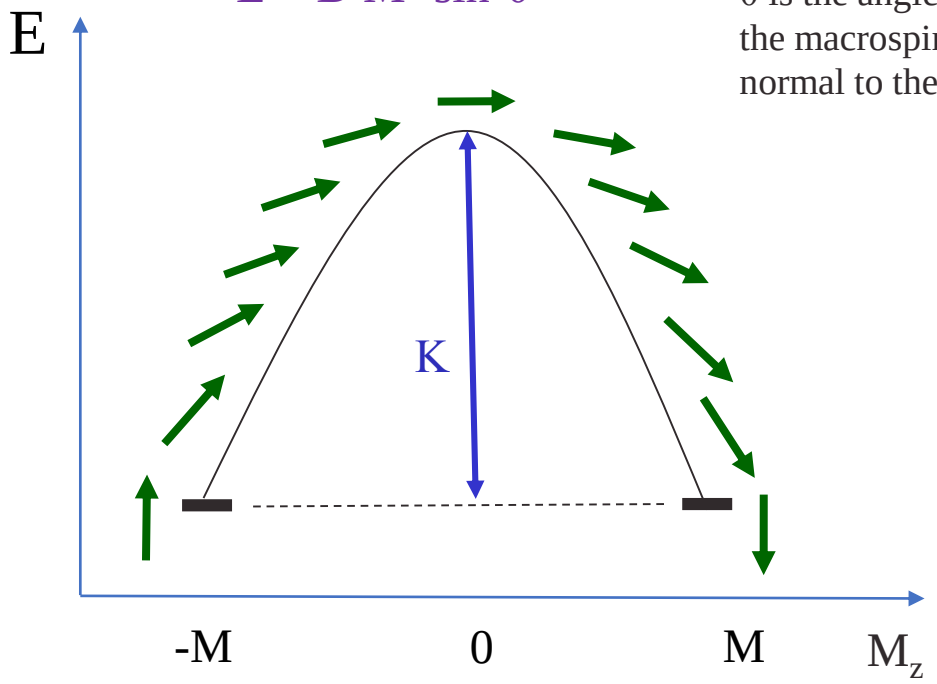
Ideal uniaxial case: crystal field with C_∞ symmetry around z direction

Quantum:
$E = D J_z^2$



Only a discrete number (2J+1)
of states are available

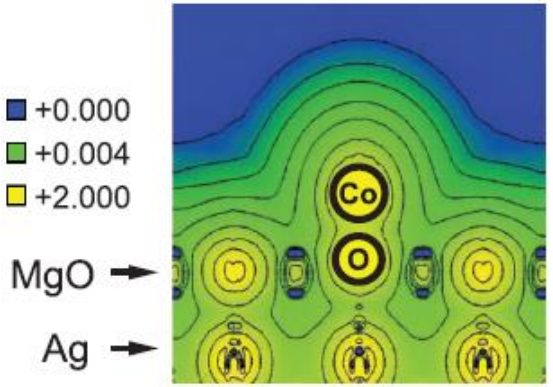
Classic:
$E = D M^2 \sin^2\theta$

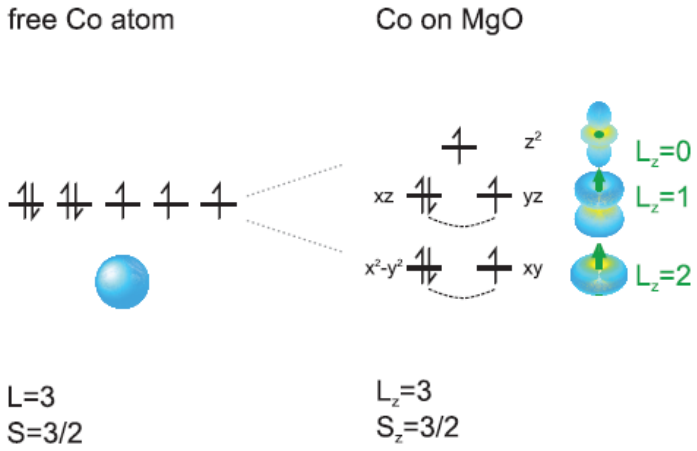


θ is the angle between the macrospin M and the normal to the surface

Continuous magnetization rotation

Reaching the MAE limit in 3d metal atoms

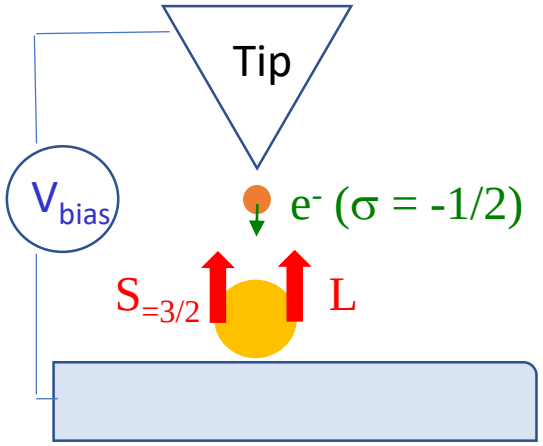


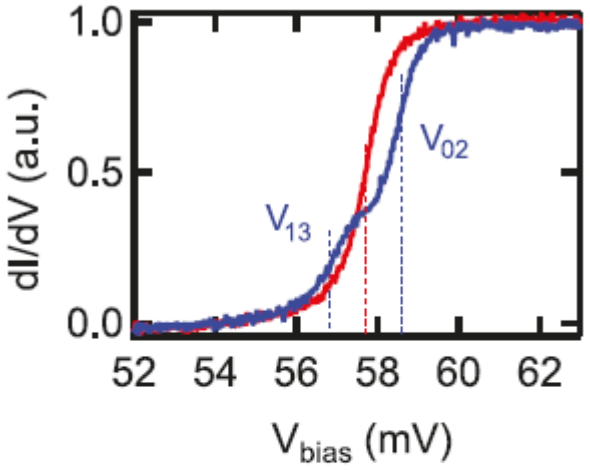


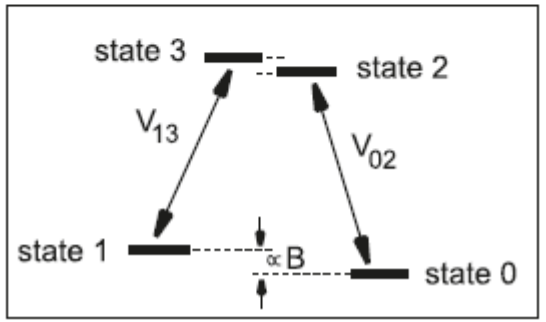
Co atom on MgO adsorbs on top of oxygen and forms a strong uniaxial bond preserving atomic values of L and S

From XMCD:

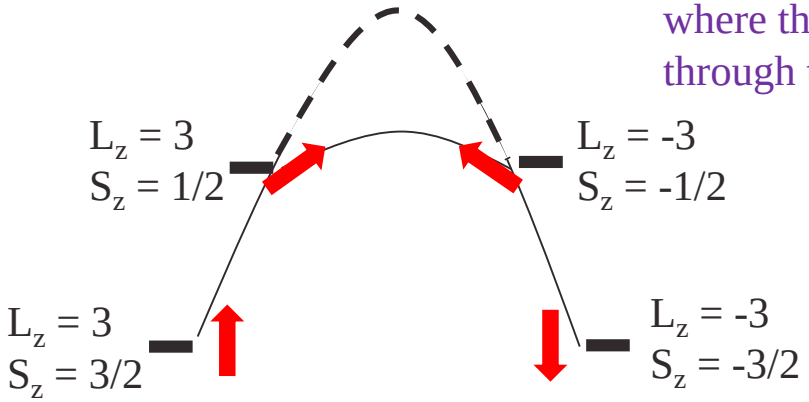
- $L_z = 2.9$; $S_z = 3/2$
- The uniaxial bond generates a strong out-of-plane anisotropy

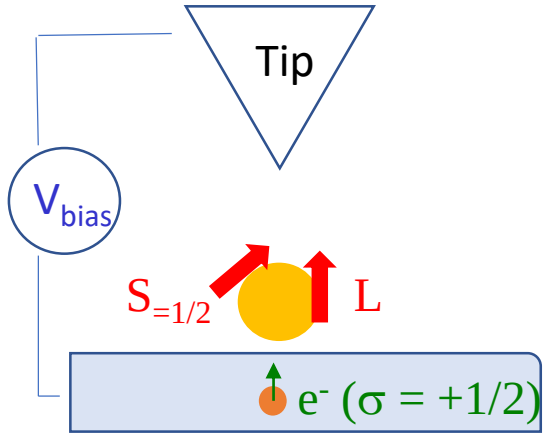






- To reverse we need $\Delta S_z = 1$.
- Different from the classical case where the magnetization passes through the $S_z=0$ state ($\Delta S_z = 3/2$)





$V_{bias} = \lambda\, \Delta S \cdot L_z$

$\Delta\sigma = 1 \rightarrow \Delta S = -1$

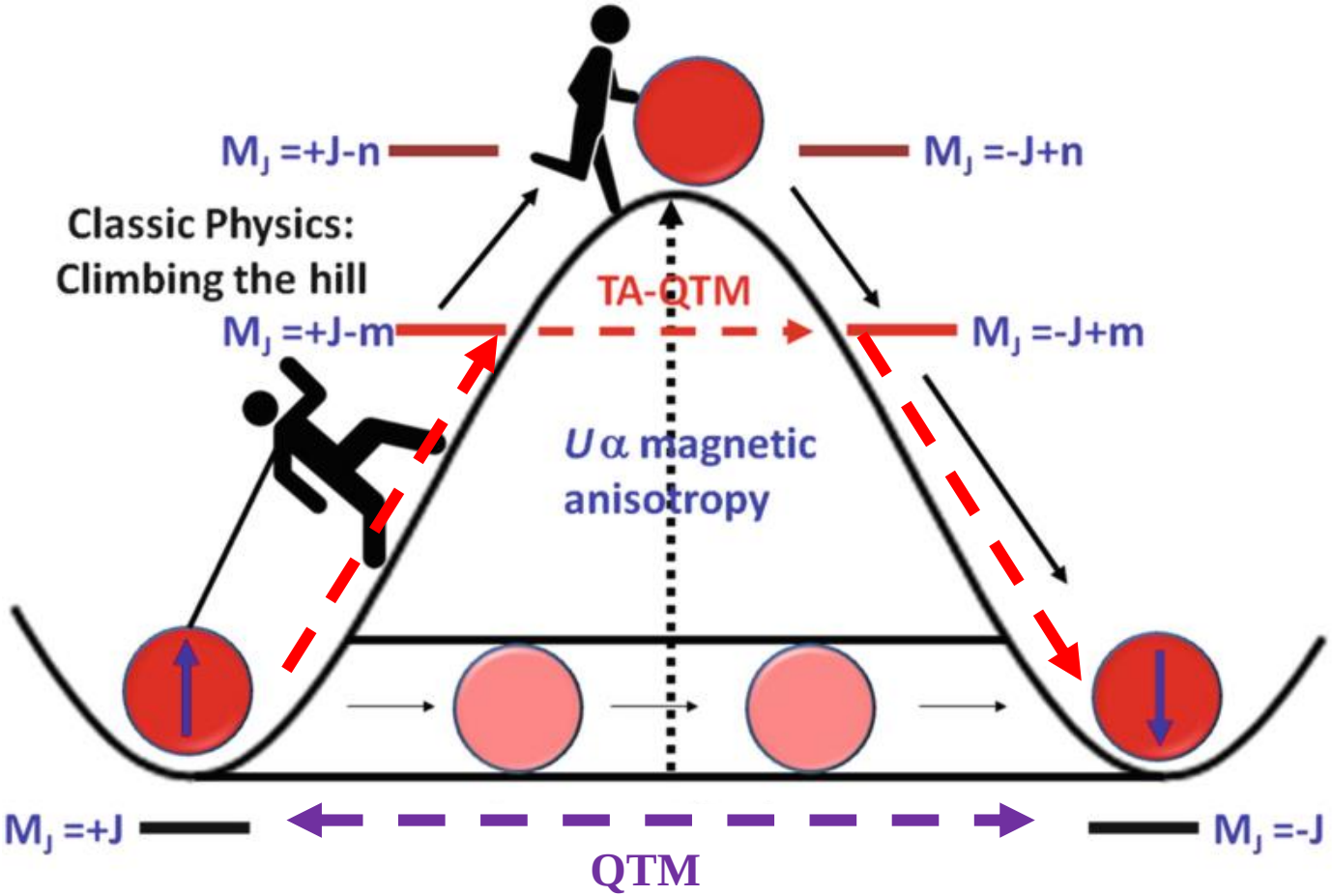
I. Rau *et al.*, Science **344**, 988 (2014)

Quantum tunneling of magnetization (QTM)

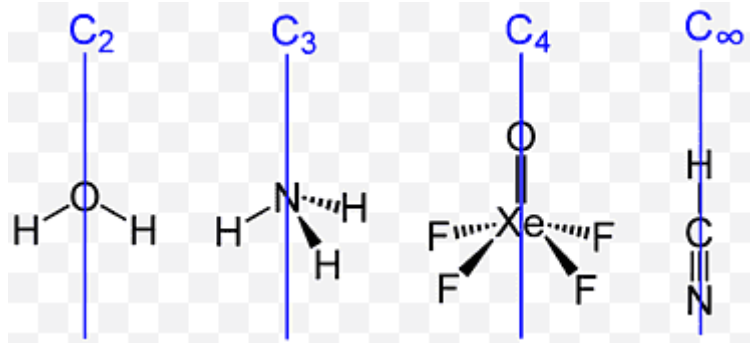
Real case: crystal field with C_n symmetry

Reversal via: - QTM

- TA-QTM



Ex.: crystal field with C_n symmetry



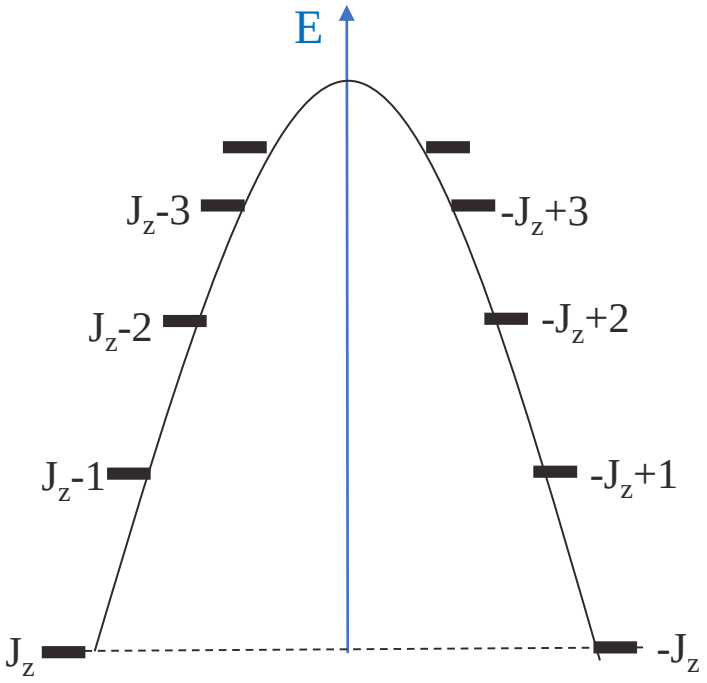
TA-QTM: thermal assisted QTM

W. Wernsdorfer, C. R. Chimie **11,** 1086 (2008)

Level splitting induced by crystal field

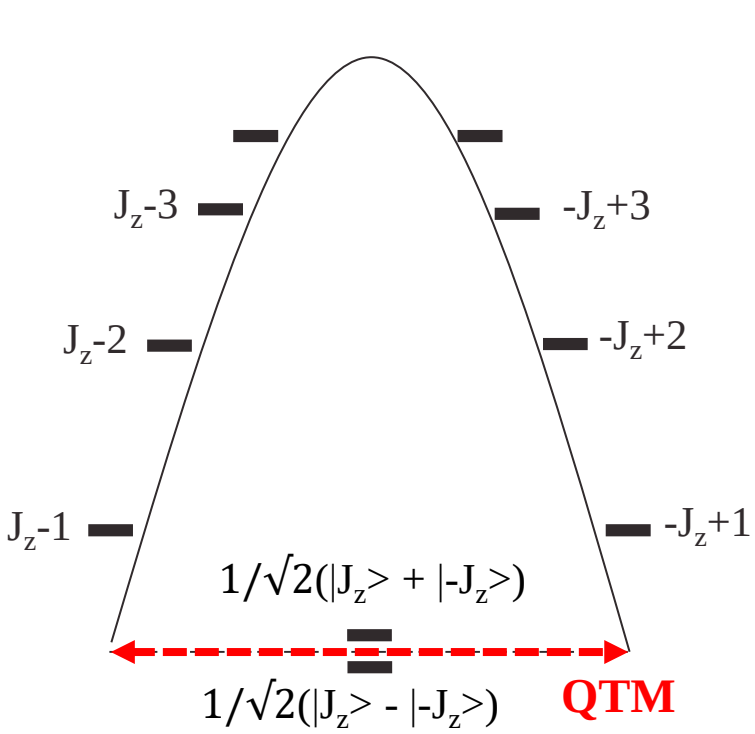
C_∞ symmetry: $E = D J_z^2$

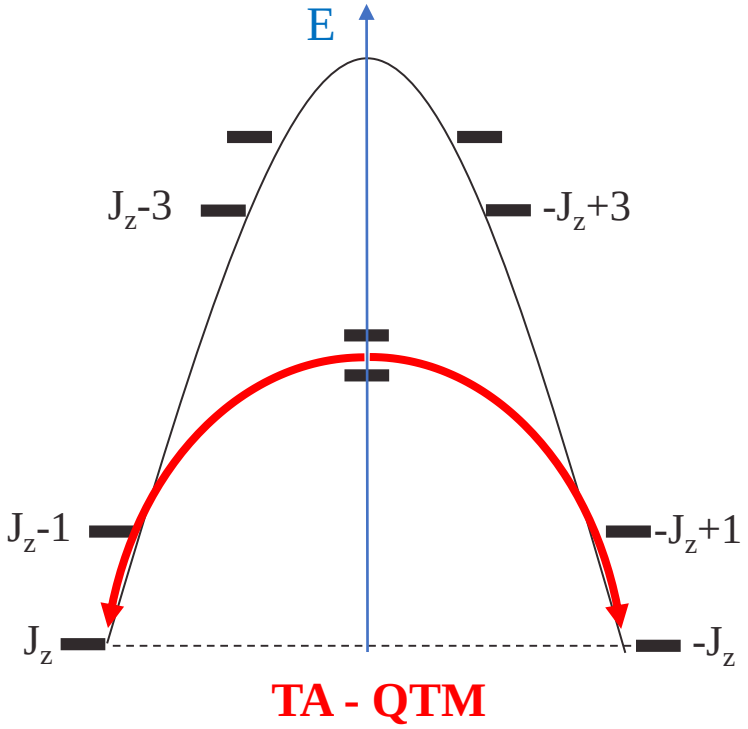
Pure J_z states -> No QTM



C_{nv} symmetry: $E = D J_z^2 + a (J_+^n + J_-^n)$

$J_\pm^n$ operators mix states satisfying J_z-J_z, = n k (n, k integers) -> QTM or TA-QTM





In case of QTM a net magnetization can not be stabilized in the ground state (the ground state is a superposition of spin-up and spin-down states) -> the particle can not be a bit

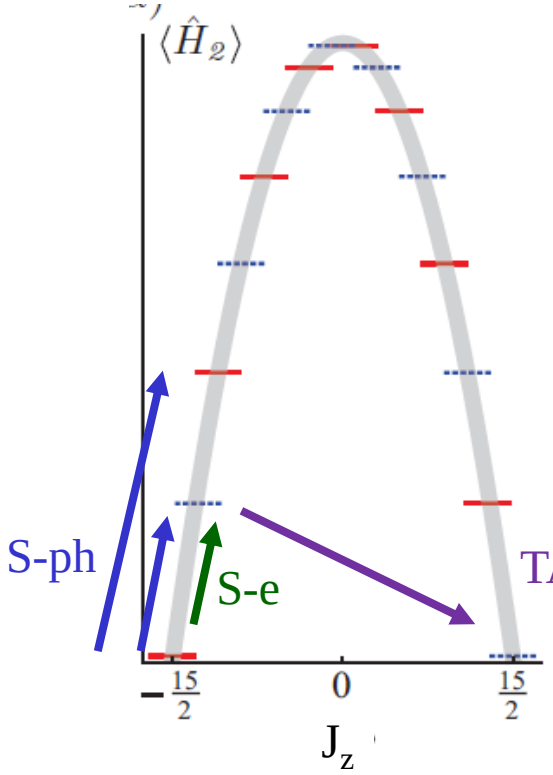
Transitions induced by spin-phonon or spin-electron scattering

Phonons are distortions of the crystal field -> $H_{spin\text{-}phonon} = \alpha (J_+, J_-) + \beta (J_+^2, J_-^2)$
Spin-phonon scattering induces transitions between states differing by $\Delta J_z = \pm 0,1, 2$

A. Fort, et al. Phys. Rev. Lett. **80**, 612 (1998)

Electrons have spin $\sigma = 1/2$ -> $H_{spin\text{-}electron} = J_0 J_z \sigma_z + 1/2\ J_0 (J_+ \sigma_- + J_- \sigma_+)$
Spin-electron scattering induces transitions between states differing by $\Delta J_z = \pm\ 0, 1$

C. Hubner *et al.*, Phys. Rev.B **90**, 155134 (2014)



Example:

CF with C_{2v} symmetry + interaction with conduction electrons + interaction with phonons

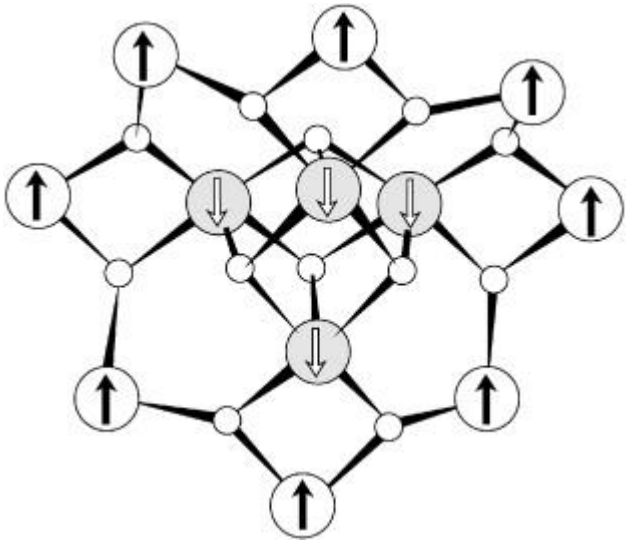
$$H_2 = D_2 J_z^2 + E_2 (J_+^2 + J_-^2) + J_0 J_z \sigma_z + 1/2\ J_0 (J_+ \sigma_- + J_- \sigma_+) + \alpha (J_+, J_-) + \beta (J_+^2, J_-^2)$$

TA-QTM: spin reversal via first excited state and not via the top of the barrier

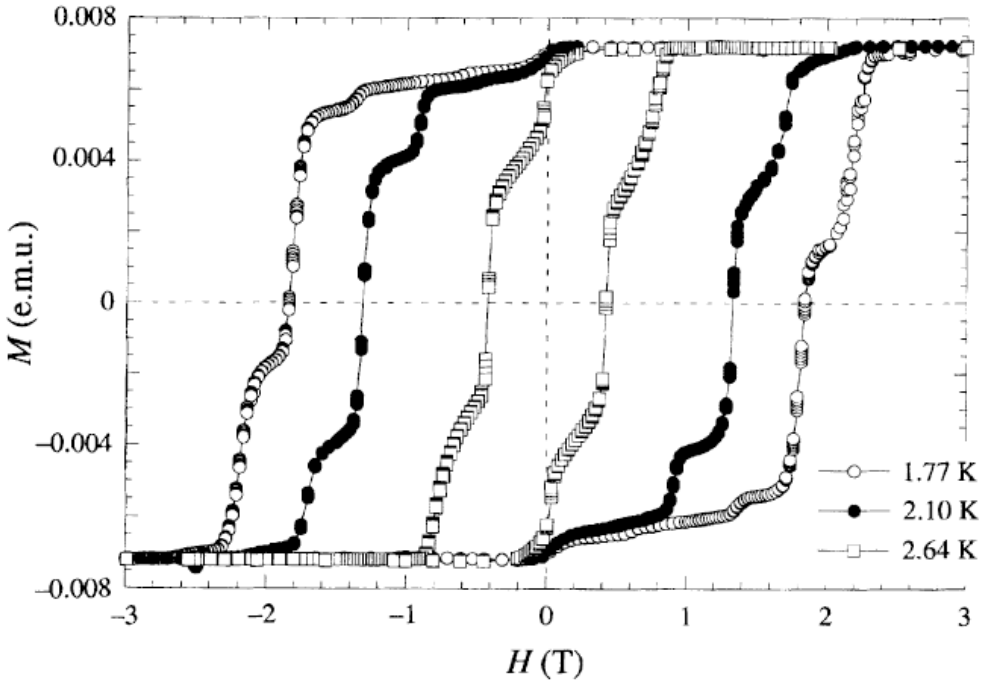
Magnetization is stable only if QTM, electron and phonon transitions are forbidden

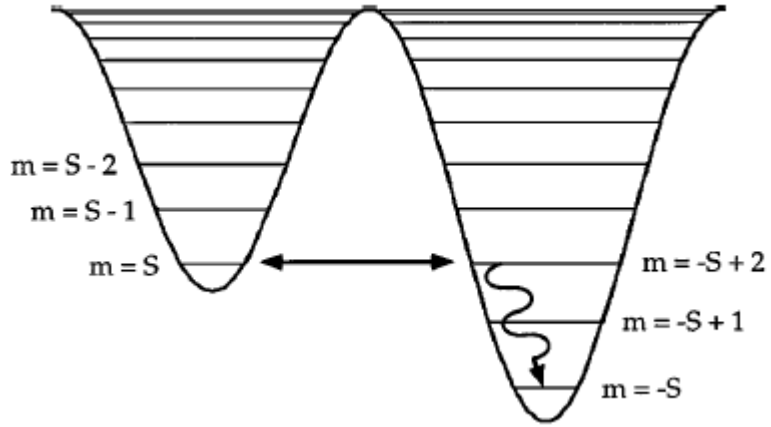
Quantum tunneling of magnetization in Mn_{12}-acetate

Single molecule magnet:
Mn_{12}-acetate



Mn atoms have S = 5/2, L = 0
8 Mn pointing up; 4 Mn pointing down
Total J = S = 10



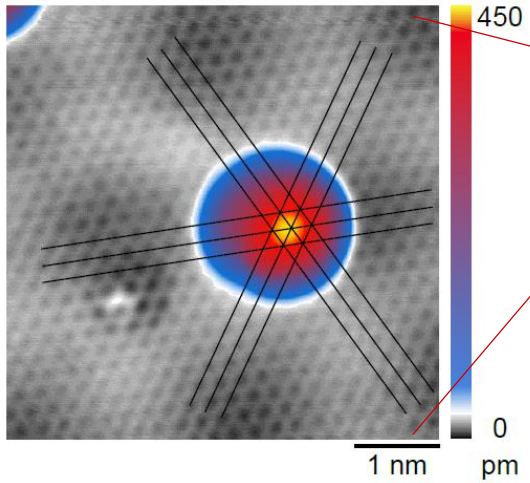


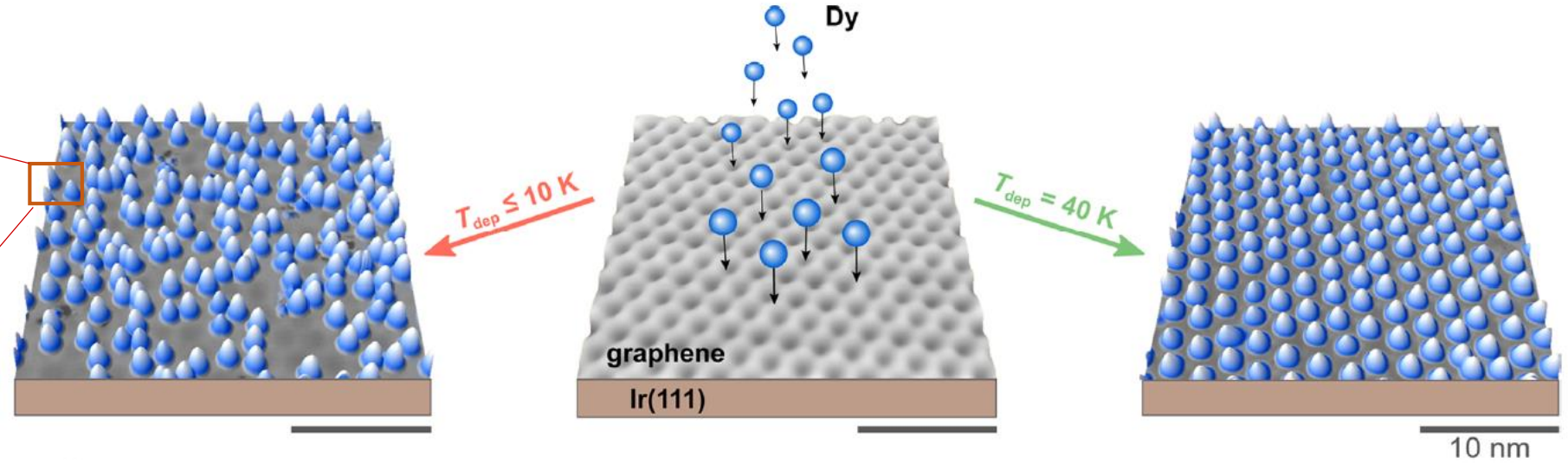
QTM when states with opposite J_z are degenerate

L. Thomas *et al.* Nature **383**, 145 (1996); J. R. Friedman *et al.* Phys. Rev. Lett. **76**, 3830 (1996)

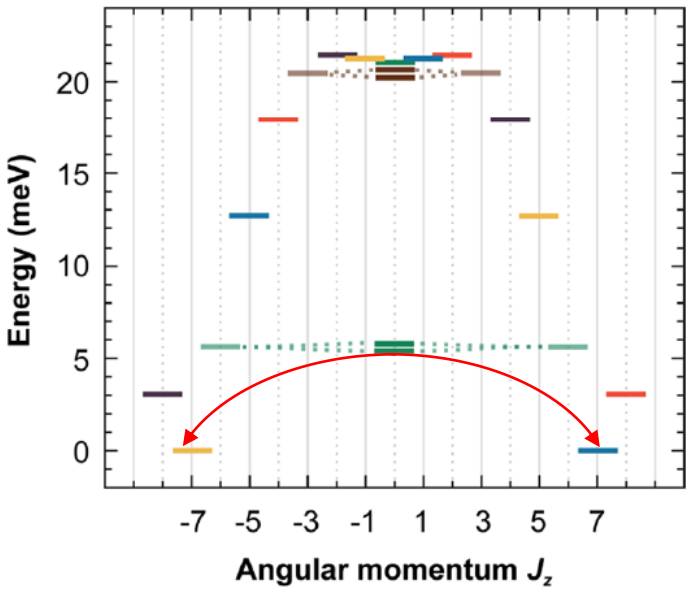
Superlattice of single atom magnets

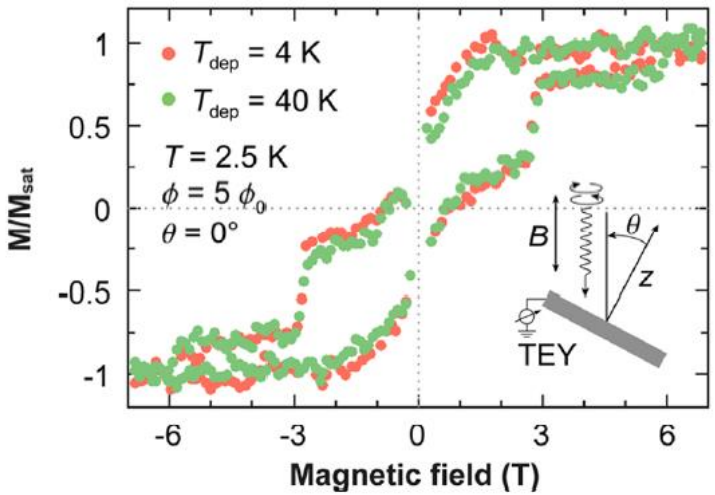
Dy adsorbs in the center of a graphene hexagon (hollow site)

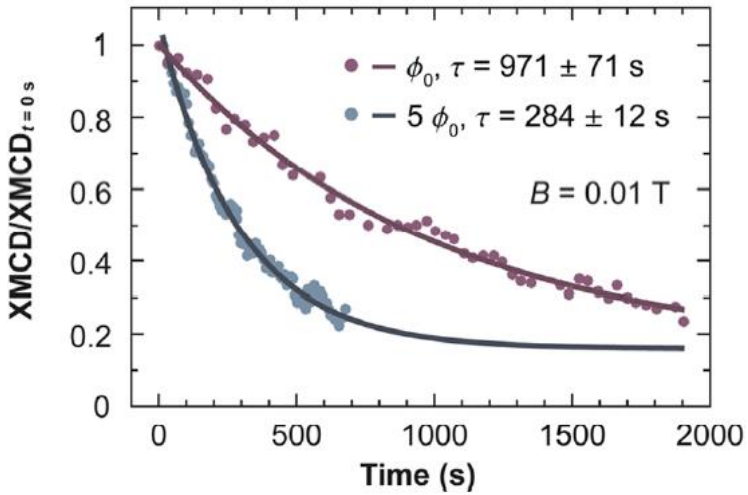




CF with C_{6v} symmetry







TA-QTM -> M ≠ 0 at B = 0 T, and T low enough (< 10 K)

R. Baltic *et al.*, Nano Letters **16**, 7610 (2016)