Nanomagnetism — Part III
Atomic-scale properties

Olivier Fruchart
Institut Néel (CNRS-UJF-INPG)
Grenoble - France

http://neel.cnrs.fr
SKETCH OF THE LECTURES

Part I – Magnetization reversal

Part II – Techniques

Part III – Atomic-scale properties
MOTIVATING THE LECTURE – The need for nanomagnetism (reminder)

Fundamental issues for nanomagnetism

- Is a small grain (ferro)magnetic?
  - Count number of surface atoms

- Is a small grain stable against thermal fluctuations?
  - $k_B T (300 \, \text{K}) \approx 4 \times 10^{-21} \, \text{J} \approx 25 \, \text{meV}$
  - $100 k_B T (300 \, \text{K}) \approx 2.5 \, \text{eV}$
  - Derive from macroscopic arguments

Magnetic grain media of current hard disks

- Decades-old (yet still modern) topic
1. Ferromagnetic order in low dimensions
   - Structure and magnetic order
   - Magnetic moments (surfaces etc.)
   - Magnetic ordering (thermal effects)

2. Magnetic energy anisotropy

3. Interfacial effects
Properties of bulk Fe

(P,T) ambiant conditions

Body-Centered Cubic (bcc)

Ferromagnetic
\[ \approx 2.2 \mu_B \text{ atom}^{-1} \]

\[ T_c = 1043 \text{ K} \]

T>1185 K

Face-Centered Cubic (fcc, \( \gamma \)-Fe)

No magnetic order
1. FERROMAGNETIC ORDER — Metastable phases (fcc Fe)

Effect of strain on the crystalline structure

Fe/Cu(001)

300K growth with MBE: fcc>bcc

(a) 3.5 ML  (b) 5.3 ML
60 nm  60 nm

300K growth with PLD: fcc

(c) 3.5 ML  (d) 5.3 ML
60 nm  60 nm

Magnetism of fcc Fe

High-spin and low-spin fcc phases?

P. Ohresser et al., PRB59, 3696 (1999)
1. FERROMAGNETIC ORDER – Metastable phases (fcc Fe)

Spin-density wave antiferromagnetism

D. Qian et al., PRL87, 227204(2001)

FIG. 4. Magnetic structures proposed for 6, 7, 8, and 9 ML Fe on Cu(100); the inset gives the layer dependent magnetic moments for fcc Fe along the $z$ direction, $z(d) = 0$ corresponding to the first AFM layer. (Note: all the moments drawn here are lying in the planes parallel to the front plane of the structure section.)

1. FERROMAGNETIC ORDER — Surface magnetism — Naive views

Probing surface magnetization

**Surface techniques at OK**
- Mossbauer with probe layers

**Plot \( m(t) \) at 0K:**
- Magnetometry
- XMCD

**Some results**
- Fe/W(110) : 0.14ml(+0.35\( \mu_B \))
- UHV/Fe(110); Ag/Fe(110): 0.26ml(+0.65\( \mu_B \))
- Cu/Ni(111): -0.5ml
- Overlayers: Pd/Ni(111)/Re(0001)

(Too) simple picture: band narrowing at surfaces

**Enhanced moment at surfaces**

Bulk picture

Surface picture
Conclusions

- Bulk: $m_L = 0.14 \mu_B$/at.
- Surface: $m_L = 0.31 \mu_B$/at.
- Bi-atomic wire: $m_L = 0.37 \mu_B$/at.
- Mono-atomic wire: $m_L = 0.68 \mu_B$/at.
- Bi-atom: $m_L = 0.78 \mu_B$/at.
- Atom: $m_L = 1.13 \mu_B$/at.


Conclusions

- From bulk to atoms: considerable increase of orbital moment
- 2 atoms closer to wire than 1 atom
- Bi-atomic wire closer to surface than wire


1. FERROMAGNETIC ORDER — Surface magnetism — Polarizability and Stoner criterium

**Exchange polarization at interfaces**

<table>
<thead>
<tr>
<th>Pd(D)/Ni(111)/Re(0001)</th>
</tr>
</thead>
</table>

**TOM**

- U. Gradmann, Handbook...

**Fe/Pd multilayers**

- J. Vogel et al., PRB55, 3663 (1997)

**XMCD**

**Conclusion:** Pd significantly polarized over several layers

**Spontaneous polarization — Stoner criterium**

**Small Rh(4d) clusters studied in flight**

- (Stern-Gerlach experiment)


**Handwavy explanation based on Stoner criterium**

\[ I \rho_{\uparrow,\downarrow}(\epsilon_F) > 1 \]

**Conclusion:** reduced bandwidth may even drive ferromagnetism

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1. FERROMAGNETIC ORDER — Magnetic ordering

Elements of theory

- Ising (1925). No magnetic order at $T>0K$ in 1D Ising chain.
- Bloch (1930). No magnetic order at $T>0K$ in 2D Heisenberg. (spin-waves; isotropic Heisenberg)
- $\rightarrow$ N. D. Mermin, H. Wagner, PRL17, 1133 (1966)
  2D Ising model: $T_c>0K$

Magnetic anisotropy stabilizes ordering

Experiments

Ni(111)/Re(0001)


Tc interpreted with molecular field
1. FERROMAGNETIC ORDER — Magnetic ordering

Naïve model

\[ T_C = \frac{\mu_0 z_n W_{1,n} g_J^2 \mu_B^2 J (J+1)}{3 k_B} \]

Less naïve...

\[ \langle z \rangle = z_b - \frac{2(z_b - z_s)}{N} \]

\[ \Delta T_C(t) \sim t^{-\lambda} \]

\[ \lambda = 1 \]

G.A.T. Allan, PRB1, 352 (1970)

Conclusion:
Naïve views are roughly correct

Experiments


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1. FERROMAGNETIC ORDER — Magnetic ordering

**Effect of lateral size**

![Graph showing the effect of lateral size on the magnetic ordering temperature \( T_c \).]

- Data from H.J. Elmers et al., Phys. Rev. Lett. 73, 898 (94)

**Conclusion**

\( T_c \) also depends on size of islands (lateral dimensions)
3. Magnetic anisotropy

1. Ferromagnetic order in low dimensions

2. Magnetic energy anisotropy
   - Microscopic origins of Magnetic Anisotropy Energy (MAE)
   - Surface versus magneto-elastic anisotropy
   - From surfaces (2D) to atoms (0D)

3. Interfacial effects
2. Magnetic anisotropy — Basics

Dipolar energy

Mutual energy of two magnetic dipoles:

\[
E_{1,2} = \frac{\mu_0}{4\pi r^3} \left[ \vec{\mu}_1 \cdot \vec{\mu}_2 - \frac{3}{r^2} (\vec{\mu}_1 \cdot \hat{r}) (\vec{\mu}_2 \cdot \hat{r}) \right]
\]

Let us assume two magnetic dipoles with vertical direction, either ‘up’ or ‘down’:

\[
E_{1,2}(\theta) = \frac{\mu_0}{4\pi r^3} \mu_1 \mu_2 \left[ 1 - 3 \cos^2 \theta \right]
\]

\[\cos^2(\theta_C) = \frac{1}{3}\]

Parallel alignment is favored for \( \theta < \theta_C \approx 54.74^\circ \)

Antiparallel alignment is favored for \( \theta > \theta_C \approx 54.74^\circ \)

‘Cone’ of alignment

Conclusions

- Nanostructures: long axis favored
- Films: in-plane favored

\[ e_d^Z = \frac{1}{2} \mu_0 M_Z^2 \]
2. Magnetic anisotropy — Basics

Magnetocrystalline anisotropy energy

- Electronic cloud
- Atom nucleus (crystal structure)

Spin-orbit coupling ⇒ the energy of both spin and orbital moment depends on orientation

Series development on an angular basis:

**Anisotropy energy**

\[
E_{mc} = K_1 m_z^2 + K_2 m_z^4 + ... 
\]

Uniaxial

\[
E_{mc} = K_4 (m_x^2 m_y^2 + m_y^2 m_z^2 + m_z^2 m_x^2) + ... 
\]

Cubic

**Normalized magnetization components**

- Alignment of magnetization is favored along given axes of the crystal

(Derived from slide of A. Thiaville – CNRS/Orsay)

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2. Magnetic anisotropy — Basics

**Magneto-elastic anisotropy**

\[
E_{\text{mel}} = K_{\text{mel,1}} \cos^2 (\theta) + \ldots
\]

**Origin**
Distortion of orbitals & crystal field

**Result**
Correction to the magneto-crystalline energy

\[
K_{\text{mel,1}} \sim B_i \varepsilon
\]
Surface anisotropy


« Superficial magnetic anisotropy and orientational superstructures »

Overview

Breaking of symmetry for surface/interface atoms

Correction to the magneto-crystalline energy

\[ E_s = K_{S,1} \cos^2(\theta) + K_{S,2} \cos^4(\theta) + \ldots \]

« This surface energy, of the order of 0.1 to 1 erg/cm², is liable to play a significant role in the properties of ferromagnetic materials spread in elements of dimensions smaller than 100Å »

Pair model of Néel:

- \( K_s \) estimated from magneto-elastic constants
- Does not depend on interface material
- Yields order of magnitude only: correct value from experiments or calculations (precision !)
2. Magnetic anisotropy — Surface anisotropy

Magnetic Anisotropy Energy (MAE): Link with anisotropy of orbital moment

**Theory**

Perturbation theory for 3d metals:

\[
\text{MAE} = \alpha \frac{\xi}{4\mu_B} \Delta \mu_L
\]


\[\mu_L \text{ does not rotate in 3d metals} \Rightarrow \text{MAE reflects cost in } \xi\]

Covers magnetocrystalline, magnetoelastic and surface anisotropy

**Ab initio calculations**

High precision needed: \(1 \mu \text{eV} \ll 10 \text{eV}\)

**Experiments**

Bulk (Fe, Ni, ...)

\[\Delta \mu_L \approx 10^{-4} \mu_B / \text{atom} \Rightarrow \text{MAE} \leq 1 \mu \text{eV}\]

O. Hjortstam et al., PRB55, 15026 (1997)

**Conclusions**

- Origin of MAE = anisotropy of orbital moment
- No strict linearity
- \(\alpha\) may also depend on thickness in thin films (band structure) → Direct measurement of MAE preferable
2. Magnetic anisotropy — Surface anisotropy

**History of surface anisotropy: STEP 1 (1/t plot)**

\[ E_{\text{tot}}(t) = k_V t + 2k_S \]

\[ e(t) = k_V + \frac{2k_S}{t} \]

**First example of perpendicular anisotropy**


Bulk

\[ \mu_0 H_S \]

\[ 1/D \]

\[ T=2\Lambda L \]
2. Magnetic anisotropy — Surface anisotropy

### Structural relaxation

- Pseudomorphic range
- Relaxation range (introduction of dislocation)

\[
e(t) \sim (a_{\text{substrate}} - a_{\text{bulk}}) \frac{t_c}{t}
\]


### Effect on anisotropy

Magneto-elastic anisotropy:

\[
k_{\text{mel}} \sim B_{\text{mel}} \epsilon
\]

Strain relaxation regime:

\[
k(t) = k_{\text{bulk}} + \alpha \frac{B_{\text{mel}}}{t}
\]

**Conclusion:**

Mixing of surface and magneto-elastic contributions

\[
e(t) = k_V + \frac{2k_S}{t}
\]

C. Chappert and P. Bruno., JAP64, 5736 (1988)
2. Magnetic anisotropy — Interface anisotropy — What use?

Main use in applications: perpendicular magnetic anisotropy

**Materials and geometry**

- Interfacial elements with large spin-orbit: Pt, Au, Pd
- Often: multilayers
- Co/Au film

**Magneto-optical recording**

- Why: large magneto-optical response
- Material: 3D-Rare-Earth based

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Main use in applications: perpendicular magnetic anisotropy

Decreased dipolar coupling in HDD media

Longitudinal recording (1956 - )

Perpendicular recording (2005 - )


⇒ High anisotropy with low spread angle
⇒ Reduced intra- and inter-grain dipolar coupling

See lecture: Laurent RANNO

Enhanced anisotropy for solid-state memories

Concerns MRAM: Magnetic Random Access Memories

C. Chappert et al., The emergence of spin electronics in data storage, Nat. Mater. 6, 813 (2007)

Reminder for the thermal stability for small flat elements:

\[ H_c = \frac{2K}{\mu_0 M_S} \left( 1 - \sqrt{\frac{25k_B T}{KV}} \right) \]

⇒ In-plane magnetization

\[ K = N \times \frac{1}{2} \mu_0 M_S^2 \]

Issue: N is small with flat elements

⇒ Perpendicular magnetization

\[ K \approx \frac{1}{2} \mu_0 M_S^2 \]
2. Magnetic anisotropy — Interface anisotropy — from 2D to atoms

From surfaces (2D) to wires (1D) and atoms (0D)

Method
- XMCD
- Fit magnetization curves

Magnetic Anisotropy Energy
- Bulk Co: 40μeV/atom
- Co ML: 140μeV/atom
- Co bi-wire: 0.34meV/atom
- Co wire: 2meV/atom
- Co bi-atom: 3.4meV/atom
- Co atom: 9.2meV/atom

Conclusions:
- Model systems to highlight trends in applied materials
- Anisotropy per atom increases in low dimensions
- The TOTAL anisotropy decreases → Not thermally stable

STM, 8.5nm, 5.5K

1. Ferromagnetic order in low dimensions

2. Magnetic energy anisotropy

3. Interfacial effects
   - Exchange bias
   - RKKY coupling
   - Dipolar effects
3. INTERFACIAL EFFECTS – Exchange-bias

Seminal studies

Oxidized Co nanoparticles

\[ \mu_0 H_E \approx 0.2 \, \text{T} \]

Field-cooled hysteresis loops:
- Increased coercivity
- Shifted in field

**Exchange bias**
J. Nogués and Ivan K. Schuller

**Exchange anisotropy—a review**
A E Berkowitz and K Takano

Meiklejohn and Bean,
Phys. Rev. 102, 1413 (1956),
Phys. Rev. 105, 904, (1957)
3. INTERFACIAL EFFECTS — Exchange-bias: what use?

Increase coercivity of layers

AF

F2

Crude approximation for thin layers:

\[ H_{F-\text{AF}} \approx H_F \left(1 + \frac{K_{\text{AF}} t_{\text{AF}}}{K_F t_F}\right) \]

Application

Concept of spin-valve in magneto-resistive elements


► Sensors

► Memory cells

► Etc.
3. INTERFACIAL EFFECTS — RKKY interlayer coupling

The physics

Spin-dependent quantum confinement in the spacer layer

\[ J(t) = \frac{A}{t^2} \sin(q \alpha t + \Psi) \]

Coupling strength:

\[ E_S = J(t) \cos \theta \]

in \( J/m^2 \)

\[ \theta = \langle m_1, m_2 \rangle \]

with:

\[ J(t) = \frac{A}{t^2} \sin(q \alpha t + \Psi) \]

\[ q^\alpha = k^+ - k^- \]

Spin-independent

\[ \Delta \phi = q t + \phi_A + \phi_B \]

Forth & back phase shift

Spin-dependent

\[ r_A, \phi_A, r_B, \phi_B \]

Constructive and destructive interferences

\[ \Rightarrow \text{Maxima and minima of } n(\epsilon) \]

Figures

### Illustration of coupling strength

**Figure 3.** Dependence of the normalized exchange coupling constant on the 3$d$, 4$d$ and 5$d$ transition metals in (a) Co/TM and (b) Fe/TM multilayers.

Note: $J(t)$ extrapolated for $t=3\text{Å}$


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<table>
<thead>
<tr>
<th>Element</th>
<th>(A_1)</th>
<th>(\Delta A_1)</th>
<th>(J)</th>
<th>(P)</th>
<th>(\Psi)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe/TM multilayers 300 K</td>
<td>0.1</td>
<td>0.2</td>
<td>0.3</td>
<td>0.4</td>
<td>0.5</td>
</tr>
<tr>
<td>Co/TM multilayers 300 K</td>
<td>0.6</td>
<td>0.7</td>
<td>0.8</td>
<td>0.9</td>
<td>1.0</td>
</tr>
</tbody>
</table>

\[ J(t) = \frac{A}{t^2} \sin \left( \frac{2\pi t}{P} + \Psi \right) \]
3. INTERFACIAL EFFECTS — RKKY interlayer coupling — What use? What constraints?

**Synthetic Ferrimagnets (SyF) — Crude description**

**F2**

**F1**

Hypothesis:

- Two layers rigidly coupled
- Reversal modes unchanged
- Neglect dipolar coupling

\[ M = \left| \frac{e_1 M_1 - e_2 M_2}{e_1 + e_2} \right| \]

\[ K = \frac{e_1 K_1 + e_2 K_2}{e_1 + e_2} \]

\[ H_c = \frac{e_1 M_1 H_{c,1} + e_2 M_2 H_{c,2}}{\left| e_1 M_1 - e_2 M_2 \right|} \]

**What use?**

- Increase coercivity of pinned layers
- Decrease intra- and inter- dot dipolar coupling

**Practical aspects**

- Ru spacer layer (largest effect)
- Control thickness within a few Angströms!
3. INTERFACIAL EFFECTS – Collective effects: bilayers

**Stacked dots: dipolar**

**In-plane magnetization**

![In-plane magnetization diagram]

**Out-of-plane magnetization**

![Out-of-plane magnetization diagram]

**Hint:**

An upper bound for the dipolar coupling is the self demagnetizing field

**Notice:** similar situation as for RKKY coupling

**Stacked dots: orange-peel coupling**

**In-plane magnetization**

![In-plane magnetization diagram]

**Out-of-plane magnetization**

![Out-of-plane magnetization diagram]

**Always parallel coupling**


(valid only for thick films)


(valid for any films)

**Out-of-plane magnetization**

May be parallel or antiparallel

**J. Moritz et al., Europhys. Lett. 65, 123 (2004)**
3. INTERFACIAL EFFECTS — Collective effects: range of interaction

Upper bound for dipolar fields in 2D

Estimation of an upper range of dipolar field in a 2D system

\[ \| H_d(R) \| \leq \int_0^R \frac{2\pi r dr}{r^3} \]

Integration

Local dipole: \( 1/r^3 \)

\[ \| H_d(R) \| \leq \text{Cte} + 1/R \]

Convergence with finite radius (typically thickness)

Non-homogeneity of dipolar fields in 2D

Example: flat stripe with thickness/height = 0.0125

\[ \text{Real} \]

\[ \text{Average} \]

Dipolar fields are weak and short-ranged in 2D or even lower-dimensionality systems

Dipolar fields can be highly non-homogeneous in anisotropic systems like 2D

Consequences on dot’s non-homogenous state, magnetization reversal, collective effects etc.
Some literature

Moment and anisotropy of ultrathin films


O. Fruchart et al., Magnetism in reduced dimensions, C. R. Phys. 6, 921 (2005)

Perpendicular anisotropy


Exchange-bias


Magneto-elasticity in thin films


Theory (misc)


F. E. Gabaly et al., Noble metal capping effects on the spin-reorientation transitions of Co/Ru(0001), N. J. Phys. 10, 073024 (2008)
The lectures of all ESM schools since 2003 are ordered here in terms of topics. Those pertaining to several topics are listed several times. The topics are:

**Magnetic field and moments**
- [2005] Basis and magnetic materials: [K.H. Müller](#) [Abstract](#) | [Slides](#) (2.2MB)  

**Exchange, magnetic ordering, magnetic anisotropy**

**Exchange and magnetic ordering**
- [2009] Magnetism at finite temperature: molecular field, phase transitions: [Claudine Lacroix, Institut Néel - Grenoble, France](#) [Slides](#).

**Magnetic anisotropy**
- [2009] Magnetic anisotropy and how it can be controlled: [Dirk Sander, MPI-Halle, Germany.](#) [Abstract](#) | [Slides](#)  

**Low-dimensional and surface magnetism**
- [2003] Low dimensional magnetism - experiments: [O. Fruchart](#) [Abstract](#) | [Slides](#)  
- [2003] Low dimensional magnetism: the role of the electronic structure: [H. Dreyssé](#) [Content](#) | [Abstract](#)  
- [2003] From the atom to magnetic nanoparticles: [E. Bonet](#) [Abstract](#) | [Slides](#)
1. Overview of self-organization processes

1. Introduction
2. Self-assembled epitaxial growth
3. Self-organized epitaxial growth
4. Engineered and 3D self-organization
5. Perspectives of self-organization
6. X-ray investigation of SO systems