Spin-orbit coupling effects on electrons, magnetic anisotropy, crystal field effects.

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Introduction

- Magnetism in condensed matter: 'exchange interactions' between many interacting electrons. Electrons' wavefunction, including their spins, must be *antisymmetric*, consistent with the Pauli Exclusion Principle.
- Generates spontaneous magnetisation in some materials.
  - Insulators: $H = -\frac{1}{2}\sum_{i,j} J_{i,j}\mathbf{s}_i \cdot \mathbf{s}_j$.
  - Spin-polarised electronic bands in metals.
- Nothing to link electron spins to any spatial direction -$\Longrightarrow$ Spin-orbit coupling does this, $H_{SO} \propto \mathbf{L} \cdot \mathbf{s}$.
- Role in
  - magnetic easy axis and magnetic hardness,
  - domain wall structure,
  - transport properties for spintronics,
    e.g. Anomalous Hall Effect, Spin-Orbit Torque,
  - topological materials,
  - magnetic nanostructures,e.g. skyrmion structures,
  - $\cdots$

Spin-Orbit Coupling and Magnetic Anisotropy, $U_{an.}$

Free energy of magnet, large length scale, $\mathbf{M} = M\,\hat{n}$:

$$F[\,\mathbf{M}(\mathbf{r})\,] = A\int((\nabla M_x)^2 + (\nabla M_y)^2 + (\nabla M_z)^2)\ d\mathbf{r}$$
$$-\int \mathbf{H}^{app.}\cdot\mathbf{M}(\mathbf{r})d\mathbf{r}\ -\frac{1}{2}\int \mathbf{H}'(\mathbf{r})\cdot\mathbf{M}(\mathbf{r})\ d\mathbf{r} + \int U_{an.}(\mathbf{M},\hat{n})\,d\mathbf{r}$$

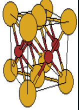
- Fundamental property $U_{an.}$ links magnetisation direction to structure via *spin-orbit coupling* of electrons.
- Large $U_{an.}$ $\rightsquigarrow$ magnetic hardness, permanent magnets.
  Small $U_{an.}$ $\rightsquigarrow$ magnetic softness, high permeability.
- Large $U_{an.}$ $\rightsquigarrow$ stability of magnetic information, $\rightsquigarrow$ smaller magnetic particles, higher blocking temperatures T_b's
- $U_{an.}$ is big for high Z materials. Has strong T, compositional and structural dependence.

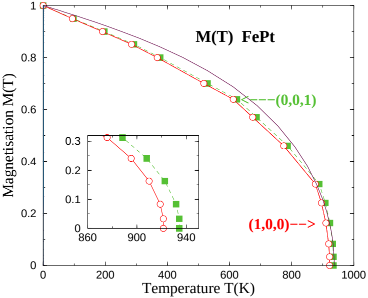
Temperature and magnetocrystalline anisotropy - spin model illustration

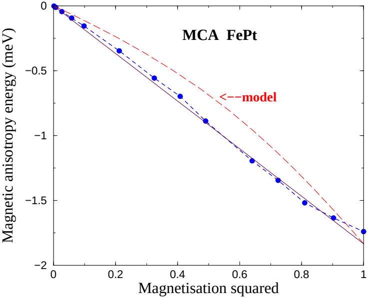
- $U_{an.} = \sum_l k_l g_l(\hat{n})$, where k_l are the magnetic anisotropy constants, and g_l's are polynomials consistent with crystal point group symmetry.
- As T rises, the k_l's decrease rapidly.
- Localised spin model for a uniaxial magnet, with $H = -\frac{1}{2}\sum_{i,j} J_{ij}\mathbf{s}_i \cdot \mathbf{s}_j - K\sum_i (\hat{n}\cdot\mathbf{s}_i)^2$.
- k_l's T dependence given by Zener, Akulov, Callen and Callen and others (H.B.Callen and E.Callen, J.Phys.Chem.Solids, **27**, 1271, (1966)).
- At low T, $\frac{k_l(T)}{k_l(0)} \approx (\frac{M(T)}{M(0)})^{l(l+1)/2}$, e.g. $k_2 \approx (\frac{M(T)}{M(0)})^3$

  At higher T, $\frac{k_l(T)}{k_l(0)} \approx (\frac{M(T)}{M(0)})^l$, $k_2 \approx (\frac{M(T)}{M(0)})^2$

Ab-initio calculations of K for $L1_0$ FePt



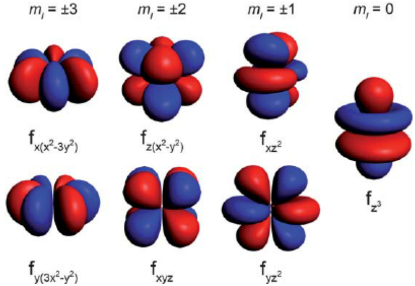




- 'Disordered local moment' (DLM) Theory of magnetism to define 'spins' in itinerant electron system, B. L. Gyorffy et al., J.Phys. F **15**, 1337, (1985); J. B. Staunton et al., Phys. Rev. Lett. **93**, 257204, (2004).
- Shows anisotropy of exchange interactions (more later).

Magnetic anisotropy and crystal field effects

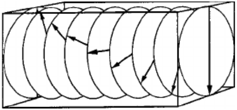
- Rare earth charge density from localised f-electrons, configuration determined by Hunds' first and second Rules. e.g. Sm $4f^5$ $\uparrow\uparrow\uparrow\uparrow\uparrow \circ\circ$, $S = \frac{5}{2}$, $L = 5$



- Spin-orbit coupling causes the f-electron charge distribution to be dragged around with spin as magnetisation direction is altered.
- There is an electrostatic energy cost from the surrounding charges, the crystal field, that generates an on-site magnetic anisotropy.

Magnetic anisotropy and domain wall widths

- Simple model of a Bloch domain wall

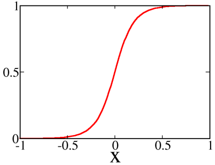


$$\mathbf{M} = (0, \sin\theta(x), \cos\theta(x)),$$

Free energy: $F[\theta(x)] = \int_{-L}^{L} (A(\frac{\partial\theta}{\partial x})^2 - K\cos^2(\theta(x))dx$

Solution $\frac{\delta F}{\delta\theta(x)} = 0$ for $\theta(-L) = 0$, $\theta(-L) = \pi$, $(L \to \infty)$.

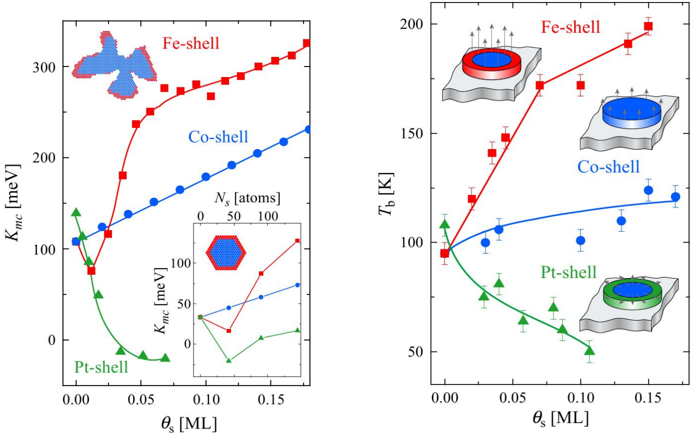
i.e. $A\frac{d^2\theta}{dx^2} + K\sin\theta(x)\cos\theta(x) = 0$



- Non-linear sine-Gordon equation:

$\tan(\theta(x) - \frac{\pi}{2}) = \sinh(x\sqrt{\frac{K}{A}})$ and width of wall $\sim \sqrt{\frac{A}{K}}$.

K and blocking temperature in magnetic nanostructures



S. Ouazi et al., Nature Commun.,12/2012; 3:1313

- Blocking temperature, $T_b = \frac{K\,V}{k_B}$

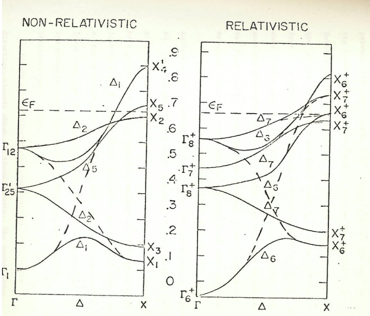
- Computational materials modelling must describe $\approx 10^{24}$ interacting electrons.
- Density functional theory (DFT) makes this problem tractable. It focusses on dependence of the energy of a material on electronic charge, ρ and magnetisation, $\mathbf{M}$, densities, $E[\rho, \mathbf{M}]$.
- Many interacting electrons described in terms of non-interacting electrons in effective fields (Kohn Sham).
- The effective fields have subtle exchange and correlation effects for nearly bound d- and f-electrons.
- Relativistic effects - spin-orbit coupling- lead to magnetic anisotropy, $E[\rho, M\,\mathbf{n}_1] - E[\rho, M\,\mathbf{n}_2]$ or torque $\frac{\delta E}{\delta \mathbf{n}}$.

Fundamental origins - interacting electrons and relativistic effects

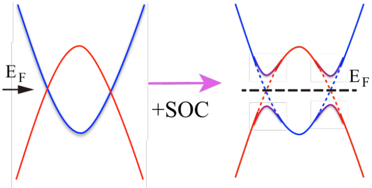
- Official starting point - Quantum Electrodynamics. Leads to Relativistic Density Functional Theory (A.K.Rajagopal (1980),Adv.Chem.Phys.**41**,59).
  $E[\rho, \vec{M}] =$
  $T_s[\rho, \mathbf{M}] + E_H[\rho] + E_{xc}[\rho, \mathbf{M}] + \int (V^{ext}(\mathbf{r})\rho(\mathbf{r}) - \mu_B \mathbf{B}^{ext}(\mathbf{r}) \cdot \mathbf{M}(\mathbf{r}))d\mathbf{r}$
- $\rho(\mathbf{r})$ and $\mathbf{M}(\mathbf{r})$ given by solving single electron Kohn-Sham-Dirac equations self-consistently.
- $(-i\hbar\alpha \cdot \nabla + \beta mc^2 + V^{eff}(\mathbf{r}) - \mu_B \beta\sigma \cdot \mathbf{B}^{eff}(\mathbf{r}))\psi_\lambda(\mathbf{r}) = \varepsilon_\lambda \psi_\lambda(\mathbf{r})$
  $\rho(\mathbf{r}) = \sum_\lambda^{occ.} \psi_\lambda^\dagger(\mathbf{r})\psi_\lambda(\mathbf{r})$; $\mathbf{M}(\mathbf{r}) = \sum_\lambda^{occ.} \psi_\lambda^\dagger(\mathbf{r})\beta\sigma\psi_\lambda(\mathbf{r})$
  $V^{eff}(\mathbf{r}) = V^{ext}(\mathbf{r}) + \frac{e^2}{4\pi\epsilon_0} \int \frac{\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} d\mathbf{r}' + \frac{\delta E_{xc}}{\delta\rho(\mathbf{r})}$;
  $\mathbf{B}^{eff}(\mathbf{r}) = \mathbf{B}^{ext}(\mathbf{r}) + \frac{\delta E_{xc}}{\delta \mathbf{M}(\mathbf{r})(\mathbf{r})}$

Origin of spin-orbit coupling

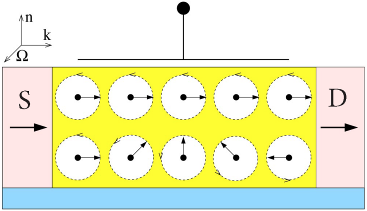
- Leading relativistic effects:
  $(-\frac{\hbar^2\nabla^2}{2m} + V(\mathbf{r}) + h_{mv} + h_D + h_{SO} - \mu_B\sigma\cdot\mathbf{B}(\mathbf{r}))\phi_\lambda(\mathbf{r}) = \varepsilon_\lambda\phi_\lambda(\mathbf{r})$
- h_{mv}, mass-velocity term, h_D, Darwin term and
  $h_{SO} = -\frac{\hbar e}{4m^2c^2}(\mathbf{E}(\mathbf{r}) \times \mathbf{p}) \cdot \sigma, \quad \mathbf{E} = -\nabla V(\mathbf{r})$,
  factor $\frac{1}{2}$ times smaller than term derived semi-classically (Thomas precession).
- Platinum bands



Topological Insulator

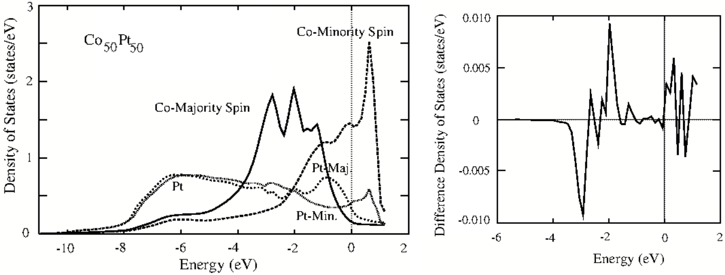


- When an inversion symmetry is broken there is a spin polarisation of the electronic states by SO coupling.
- Rashba E.I.Rashba and Y.A.Bychov,J.Phys.C **17**, 6039, (1984) and Dresselhaus G.Dresselhaus, Phys.Rev,**100**, 580, (1955) Effects.
- Electron confined in 2D $(x, y, 0 \leq z \geq d)$ with external electric field $\mathbf{E} = (0, 0, E)$ and $h_{SO} = -i\frac{\hbar e}{4m^2c^2}E(\sigma_x\frac{\partial}{\partial y} - \sigma_y\frac{\partial}{\partial x})$.
- Show that $E^{\uparrow(\downarrow)}_{k,n} = \frac{\hbar^2 k^2}{2m} + \frac{\hbar^2 n^2 \pi^2}{2md^2} + (-)\frac{e\hbar^2 Ek}{2m^2c^2}$ and $\phi^{\uparrow(\downarrow)}_{k,n}(x, y, z) \propto e^{i(k_x x + k_y y)}\sin(\frac{n\pi z}{d})\frac{1}{k}\begin{pmatrix} (-(+)k_y + ik_x) \\ (-(+)k_y - ik_x) \end{pmatrix}$
- Spin Field Effect Transistor (SFET) (S.Datta and B.Das, App.Phys.Lett, **56**,665,(1990); I.Zutic et al. Rev.Mod.Phys. **76**,323,(2004)).



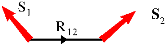
Magnetic anisotropy again

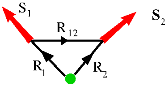
- Time reversal invariance is broken when a magnetic term $-\mu_B \sigma \cdot \mathbf{B}$ is added.
- With spin-orbit coupling and magnetic terms, electronic structure varies with direction of magnetisation, e.g. CoPt (S.S.A.Razee et al., Phys.Rev. B**56**,8082, (1997))



- Origin of anisotropy: $\Delta E_{MAE} = \sum_{\lambda}^{occ.} \int_{BZ} (\varepsilon_{\lambda}^{\mathbf{n}_1}(\mathbf{k}) - \varepsilon_{\lambda}^{\mathbf{n}_2}) d\mathbf{k}$

Anisotropic magnetic interactions

- RKKY interaction between 2 magnetic impurities in free electron gas, 

$$H = V \mathbf{s}_1 \cdot \mathbf{s}_2 \frac{(2k_F R_{12} \cos(2k_F R_{12}) - \sin(2k_F R_{12}))}{R_{12}^4}.$$
- With spin-orbit coupling, interaction becomes anisotropic (Staunton et al. JPCM, **1**, 5157, (1989)), uniaxial anisotropy.
$$H = H((\mathbf{R}_{12} \cdot \mathbf{s}_1)(\mathbf{R}_{12} \cdot \mathbf{s}_2), (\mathbf{R}_{12}.(\mathbf{s}_1 \times \mathbf{s}_2))^2).$$
- Break inversion symmetry by including third site (A.Fert and A.M.Levy, Phys.Rev.Lett. **44**,1538, (1980)) 

and find unidirectional anisotropy, $H = H((\mathbf{R}_1 \times \mathbf{R}_2) \cdot (\mathbf{s}_1 \times \mathbf{s}_2))$ - Dzyaloshinskii-Moriya-type (I.Dzyaloshinskii, J.Phys.Chem.Solids, **4**, 241, (1958); T.Moriya, Phys.Rev.Lett. **4**, 5 ,(1960))

Modelling magnetic nanostructures

- $H = -\frac{1}{2}\sum_{i,j}(J_{ij}\mathbf{s}_i \cdot \mathbf{s}_j + \mathbf{s}_i \mathcal{J}^s_{ij}\mathbf{s}_j + \mathbf{D}_{ij}\cdot(\mathbf{s}_1 \times \mathbf{s}_2)) + \sum_i K_i(\mathbf{s}_i)$

Rep. Prog. Phys. **74** (2011) 096501 H Ebert *et al*

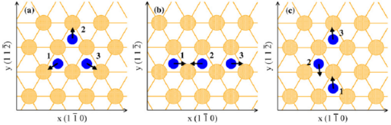


Figure 22. Geometries of the Cr trimers (solid circles) deposited on top of a Au(1 1 1) surface (patterned circles): (*a*) equilateral triangle, (*b*) linear chain and (*c*) isosceles triangle. The arrows denote the ground-state orientation of the spin magnetic moments of the Cr atoms [230].

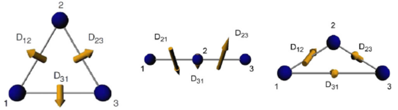
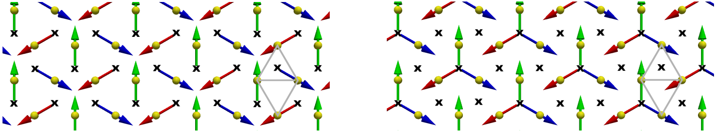


Figure 23. Schematic view of the DM vectors for an equilateral (left), linear (middle) and isosceles (right) Cr trimer on Au(1 1 1) [230].

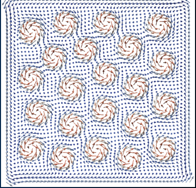
- Magnetic monolayer on f.c.c. $(1,1,1)$ substrate, chiral magnetic structures. M. dos Santos Dias et al., Phys.Rev. B **83**, 054435, (2011).



Summary

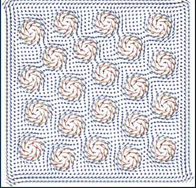
- $F[\mathbf{M}(\mathbf{r})]$, phenomenological free energy - sum of 'exchange' + 'magnetic anisotropy' + 'applied magnetic field interaction' + 'magnetostatic' energies.
- In materials formal underpinning from relativistic quantum electrodynamics $\rightarrow$ R-DFT.
- Magnetic anisotropy - hardness of permanent magnets, domain wall thicknesses, blocking temperatures.
- Crystal field origin of on-site (single ion) anisotropy.
- Spin-orbit coupling and spin polarisation of electrons as relativistic effects. Rashba, Dresselhaus effects.
- Broken spatial inversion symmetry. Disruption of time reversal invariance.
- Ab-initio DFT calculations used to explain/provide A, K parameters for modelling magnetic properties.

Summary



- Anisotropic magnetic interactions
  Dzyaloshinskii-Moriya interactions, Skyrmion lattices and data storage (M.Beg et al. Sci. Reports **5**, 17137, (2015))

Summary



- Anisotropic magnetic interactions
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More references:
General, domains, domain walls etc.: S. Blundell, (2000), ‘Magnetism in Condensed Matter’, (O.U.P); R. O’Handley, R., (2000), ‘Modern magnetic materials: principles and applications’, (Wiley); W. F. Brown Jr., W.F., (1962), ‘Magnetostatic Principles in Ferromagnetism’, (North Holland, Amsterdam); D. J. Craik and R. S. Tebbell, (1965), ‘Ferromagnetism and Ferromagnetic Domains’, (North Holland, Amsterdam); M. E. Schabes, (1991), J.Mag.Magn.Mat. **95**, 249-288.
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