

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

Julie Staunton
*Physics Department,
University of Warwick, U. K*



- 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 $\implies$ **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,
 - ...



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

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

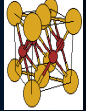
$$F[\mathbf{M}(\mathbf{r})] = A \int ((\nabla M_x)^2 + (\nabla M_y)^2 + (\nabla M_z)^2) d\mathbf{r} \\ - \int \mathbf{H}^{app.} \cdot \mathbf{M}(\mathbf{r}) d\mathbf{r} - \frac{1}{2} \int \mathbf{H}'(\mathbf{r}) \cdot \mathbf{M}(\mathbf{r}) d\mathbf{r} + \int U_{an.}(\mathbf{M}, \hat{n}) d\mathbf{r}$$

- 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.

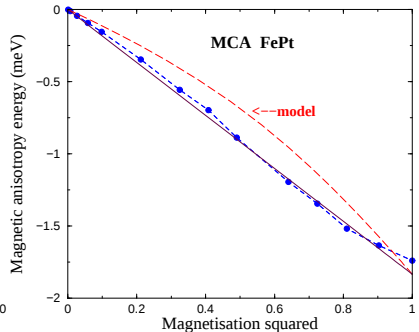
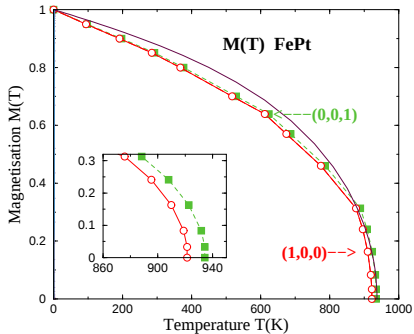
Temperature and magnetocrystalline anisotropy

- 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 , $\frac{k_l(T)}{k_l(0)} \approx \left(\frac{M(T)}{M(0)}\right)^{l(l+1)/2}$, e.g. $k_2 \approx \left(\frac{M(T)}{M(0)}\right)^3$
At higher T , $\frac{k_l(T)}{k_l(0)} \approx \left(\frac{M(T)}{M(0)}\right)^l$, $k_2 \approx \left(\frac{M(T)}{M(0)}\right)^2$



Ab-initio calculations of K for $L1_0$ FePt

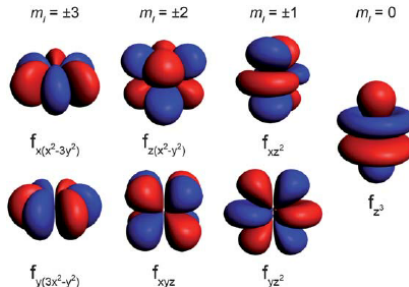


- 'Disordered local moment' (DLM) Theory of magnetism to define 'spins' in itinerant electron system, B. L. Gyorffy et al., J.Phys. F **15**, 1337, (1985); J. B. Staunton et al., Phys. Rev. Lett. **93**, 257204, (2004).
- Shows anisotropy of exchange interactions (more later).

Magnetic anisotropy and crystal field effects

- Rare earth charge density from localised f-electrons, configuration determined by Hunds' first and second Rules.

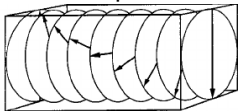
e.g. $\text{Sm } 4f^5 \uparrow\uparrow\uparrow\uparrow\uparrow \circ\circ$, $S = \frac{5}{2}$, $L = 5$



- 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

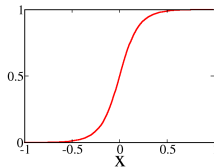


$$\mathbf{M} = (0, \sin \theta(x), \cos \theta(x)),$$

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

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

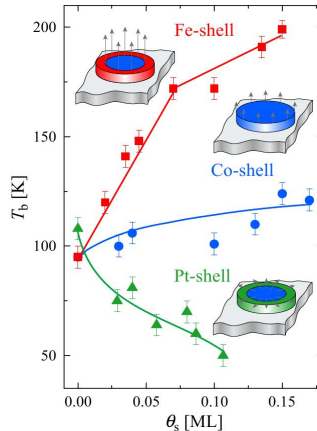
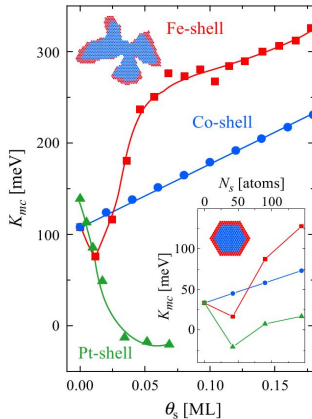
$$\text{i.e. } A \frac{d^2 \theta}{dx^2} + K \sin \theta(x) \cos \theta(x) = 0$$



- Non-linear sine-Gordon equation:

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

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).

$$E[\rho, \vec{M}] =$$

$$T_s[\rho, \mathbf{M}] + E_H[\rho] + E_{xc}[\rho, \mathbf{M}] + \int (V^{\text{ext}}(\mathbf{r})\rho(\mathbf{r}) - \mu_B \mathbf{B}^{\text{ext}}(\mathbf{r}) \cdot \mathbf{M}(\mathbf{r})) d\mathbf{r}$$

- $\rho(\mathbf{r})$ and $\mathbf{M}(\mathbf{r})$ given by solving single electron Kohn-Sham-Dirac equations self-consistently.
- $(-i\hbar\alpha \cdot \nabla + \beta mc^2 + V^{\text{eff}}(\mathbf{r}) - \mu_B \beta \sigma \cdot \mathbf{B}^{\text{eff}}(\mathbf{r}))\psi_\lambda(\mathbf{r}) = \varepsilon_\lambda \psi_\lambda(\mathbf{r})$
 $\rho(\mathbf{r}) = \sum_\lambda^{\text{occ.}} \psi_\lambda^\dagger(\mathbf{r})\psi_\lambda(\mathbf{r}); \mathbf{M}(\mathbf{r}) = \sum_\lambda^{\text{occ.}} \psi_\lambda^\dagger(\mathbf{r})\beta\sigma\psi_\lambda(\mathbf{r})$
 $V^{\text{eff}}(\mathbf{r}) = V^{\text{ext}}(\mathbf{r}) + \frac{e^2}{4\pi\epsilon_0} \int \frac{\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} d\mathbf{r}' + \frac{\delta E_{xc}}{\delta \rho(\mathbf{r})};$
 $\mathbf{B}^{\text{eff}}(\mathbf{r}) = \mathbf{B}^{\text{ext}}(\mathbf{r}) + \frac{\delta E_{xc}}{\delta \mathbf{M}(\mathbf{r})(\mathbf{r})}$

Origin of spin-orbit coupling

- Leading relativistic effects:

$$\left(-\frac{\hbar^2 \nabla^2}{2m} + V(\mathbf{r}) + h_{mv} + h_D + h_{SO} - \mu_B \boldsymbol{\sigma} \cdot \mathbf{B}(\mathbf{r})\right) \phi_\lambda(\mathbf{r}) = \varepsilon_\lambda \phi_\lambda(\mathbf{r})$$

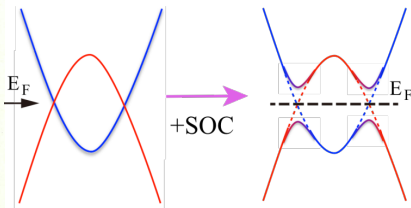
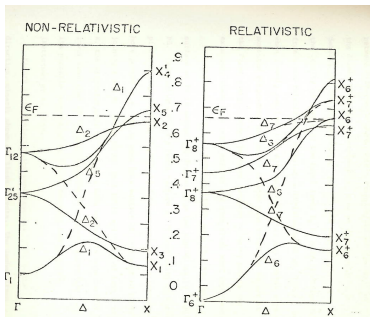
- h_{mv} , mass-velocity term, h_D , Darwin term and

$$h_{SO} = -\frac{\hbar e}{4m^2 c^2} (\mathbf{E}(\mathbf{r}) \times \mathbf{p}) \cdot \boldsymbol{\sigma}, \quad \mathbf{E} = -\nabla V(\mathbf{r}),$$

factor $\frac{1}{2}$ times smaller than term derived semi-classically (Thomas precession).

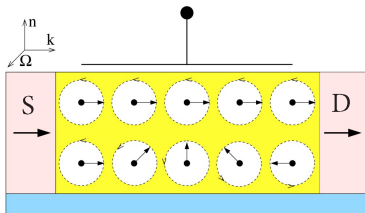
- Platinum bands

Topological Insulator



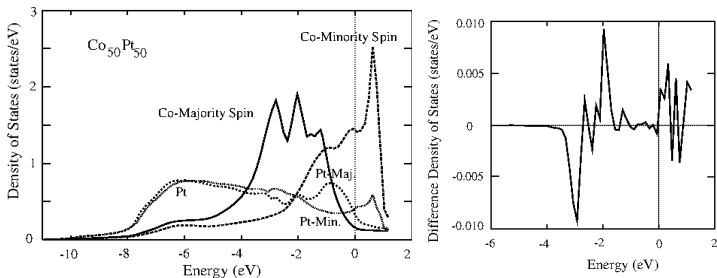
Spin-orbit coupling and broken inversion symmetry

- 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 ($x, y, 0 \leq z \leq d$) with external electric field $\mathbf{E} = (0, 0, E)$ and $h_{SO} = -i \frac{\hbar e}{4m^2 c^2} E (\sigma_x \frac{\partial}{\partial y} - \sigma_y \frac{\partial}{\partial x})$.
- Show that $E_{k,n}^{\uparrow(\downarrow)} = \frac{\hbar^2 k^2}{2m} + \frac{\hbar^2 n^2 \pi^2}{2md^2} + (-) \frac{e\hbar^2 Ek}{2m^2 c^2}$ and $\phi_{k,n}^{\uparrow(\downarrow)}(x, y, z) \propto e^{i(k_x x + k_y y)} \sin(\frac{n\pi z}{d}) \frac{1}{k} \begin{pmatrix} -(+)k_y + ik_x \\ -(+)k_y - ik_x \end{pmatrix}$
- 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. B56,8082, (1997))



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

Anisotropic magnetic interactions

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



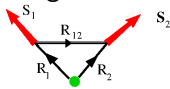
The diagram shows two red arrows representing spins s_1 and s_2 . A black arrow labeled R_{12} points from the tip of s_1 to the tip of s_2 .

$$H = V \mathbf{s}_1 \cdot \mathbf{s}_2 \frac{(2k_F R_{12} \cos(2k_F R_{12}) - \sin(2k_F R_{12}))}{R_{12}^4}.$$

- With spin-orbit coupling, interaction becomes anisotropic (Staunton et al. JPCM, **1**, 5157, (1989)), uniaxial anisotropy.

$$H = H((\mathbf{R}_{12} \cdot \mathbf{s}_1)(\mathbf{R}_{12} \cdot \mathbf{s}_2), (\mathbf{R}_{12} \cdot (\mathbf{s}_1 \times \mathbf{s}_2))^2).$$

- 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

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

Rep. Prog. Phys. 74 (2011) 096501

H Ebert *et al*

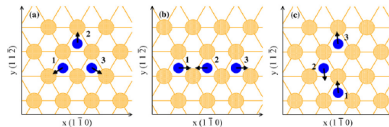


Figure 22. Geometries of the Cr trimers (solid circles) deposited on top of a Au(111) surface (patterned circles): (a) equilateral triangle, (b) linear chain and (c) isosceles triangle. The arrows denote the ground-state orientation of the spin magnetic moments of the Cr atoms [230].

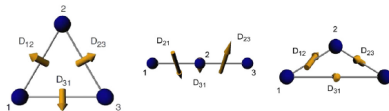
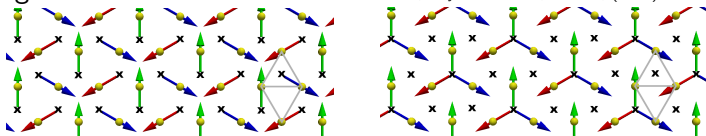


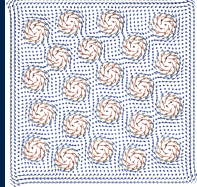
Figure 23. Schematic view of the DM vectors for an equilateral (left), linear (middle) and isosceles (right) Cr trimer on Au(111) [230].

- 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).



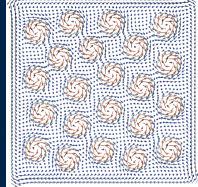
- $F[\mathbf{M}(\mathbf{r})]$, phenomenological free energy - sum of 'exchange' + 'magnetic anisotropy' + 'applied magnetic field interaction' + 'magnetostatic' energies.
- In materials formal underpinning from relativistic quantum electrodynamics $\rightarrow$ R-DFT.
- Magnetic anisotropy - hardness 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
Dzyaloshinskii-Moriya interactions, Skyrmion lattices and data storage (M.Beg et al. Sci. Reports **5**, 17137, (2015))

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.

Relativistic Q.M. and DFT: H. J. F. Jansen, (1999), Phys. Rev. B **59**, 4699; J. Kübler, (2009), 'Theory of itinerant magnetism', (O.U.P); J. B. Staunton, (1994), Reports on Progress in Physics **57**, 1289-1344; P. Strange, (1998), 'Relativistic Quantum Mechanics', (C.U.P.); R. M. Martin, (2008), 'Electronic structure: Basic Theory and Practical Methods', (C.U.P); J. D. Björken and S. D. Drell, (1965), 'Relativistic Quantum Fields', (McGraw-Hill); P. Strange et al., (1984), J. Phys. C **17**, 3355.

Rare earth magnetism and crystal fields: M. Richter, (1998), J. Phys.D **31**, 1017-1048; R. J. Elliott, (1972), 'Magnetic properties of rare earth metals', (Springer); J. Jensen and A. R. Mackintosh, (2011) 'Rare earth magnetism - structure and excitations', (The International Series of Monographs in Physics).

Calculations of magnetic anisotropy and magnetic interactions: H. Ebert *et al.*, (2011), Rep. Prog. Phys. **74**, 096501; J. B. Staunton *et al.*, 2006, Phys. Rev. B **74**, 144411; S. Mankovsky *et al.*, Phys. Rev. B **80**, 014422 (2009); S. Bornemann, *et al.*, 2012, Phys. Rev. B **86**, 104436.