

Exploiting polarised neutron and resonant x-ray reflectivity techniques to study nanoscale magnetism



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Outline

- Motivation
 - The importance of interfaces
- Reflectivity
 - Introduction to the basic Ideas
 - Information contained
 - ◆ Specular/Off-specular
 - Practical Considerations
- Examples
- Outlook
 - Bright!



Nanoscale Electronic Phenomena Research

Heusler, DMS

Surface Magnetism

Exchange Bias

- Conventional EXB
- Synthetic EXB
- Fruzen magnetism

Domain Structures

Frustration

Singlet-Triplet Superconductivity

Helimagnetism-Skyrmions

Organic Spintronics

Interfacial Magnetism

To understand and control spin and interfacial phenomena
 Probe length scale (<1nm to >1000nm)
 Vector Magnetometry ~<0.1μ_B per f.u.

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The Importance of interfaces

I. N. Krivorotov et al., Science 307, 228 (2005).

■ S.S.P.Parkin et al. Science 220 5873 (2008)

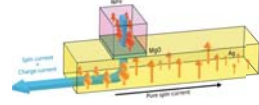
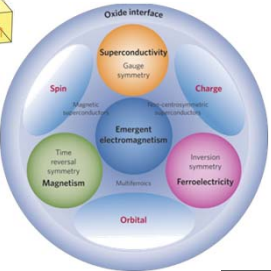
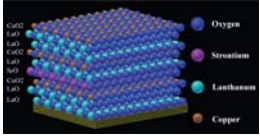
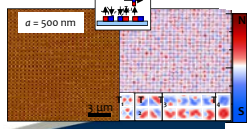
■ Ramesh and Spaldin Nat. Mat. 6, 21 (2007)

■ Maccherozzi et al. Phys. Rev. Lett. 101, 267201 (2008)

■ Awschalom and Flatté Nat. Phys. 3 153 (2007)

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Complex physics: advanced characterisation

◆ x-section
◆ Polarisation
◆ Interfacial sensitivity


Nat. Phys. 7, 75–79 (2010)

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Nature Materials 11, 103–113 (2012)

Complementarity



- Bulk measurements
 - Magnetometry
 - Transport...






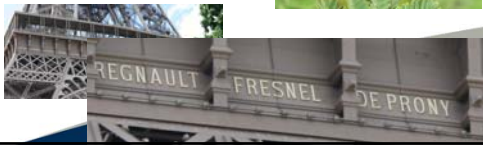
- X-rays
 - Element specific
 - Shell Selective
 - Good Q-resolution
 - Weak interaction
 - High incident flux
 - Large resonant enhancements
 - Strong absorption (resonant)
 - Polarisation dependence
 - Small coherence length (<10μm)
- Imaging
 - XPEEM
 - Lorentz
- Neutrons
 - Strong magnetic scattering
 - Penetrative probe
 - Good energy resolution
 - Absolute measurement
 - Polarisation analysis
 - Long coherence length (~100μm)

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Interference effects

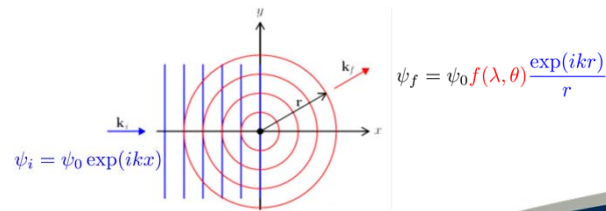







■ Fresnel reflection 1815

Neutrons & Magnetism

Scattering from a single (fixed) atom

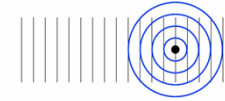
- atomic nuclei via the short-range (fm) strong force;
- unpaired orbital electrons via a magnetic dipole interaction



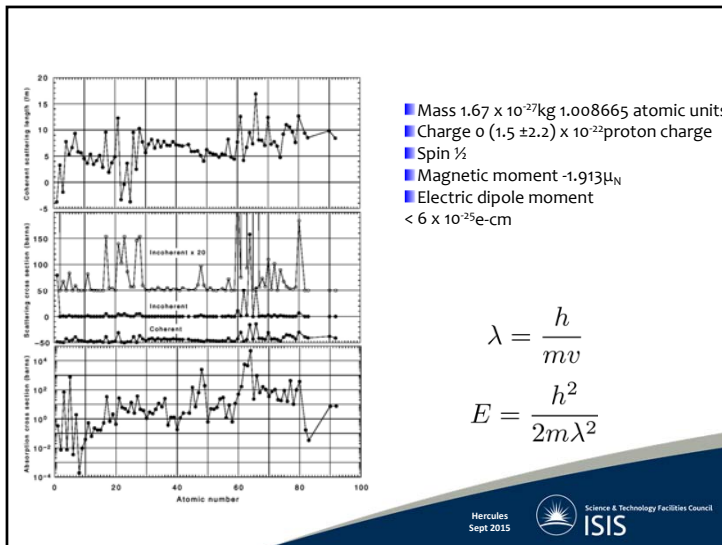
Scattering length

$$\sigma_{tot} = 4\pi b^2$$

Where b is the scattering length



- The sign of b is arbitrary
- A negative sign implies a change in the phase of the scattered wave
- b is sometimes complex and wavelength dependent due to resonant absorption
- b depends on the isotope
- b depends on the spin states of the neutron and nucleus



Isotope Dependence

Nickel Isotope	Scattering length b (fm)	Hydrogen Isotope	Scattering length b (fm)
^{58}Ni	15.0(5)	1H	-3.7409(11)
^{60}Ni	2.8(1)	2D	6.674(6)
^{61}Ni	7.60(6)	3T	4.792(27)
^{62}Ni	-8.7(2)	O	5.803
^{64}Ni	-0.38(7)		

$$|11\rangle = \uparrow\uparrow$$

$$|10\rangle = (\uparrow\downarrow + \downarrow\uparrow) / \sqrt{2}$$

$$|1-1\rangle = \downarrow\downarrow$$

$$|00\rangle = (\uparrow\downarrow - \downarrow\uparrow) / \sqrt{2}$$

- Isotopic substitution for contrast
- Isotopic substitution to move peak positions in spectroscopy
- Incoherent scattering

SLD

<http://www.ncnr.nist.gov/resources/sldcalc.html>

Neutron activation and scattering calculator

This calculator uses neutron cross sections to compute activation on the sample given the mass in the sample and the time in the beam, or to perform scattering calculations for the neutrons which are not absorbed by the sample.

1. Enter the sample formula in the material panel.
2. To perform activation calculations, fill in the thermal flux, the mass, the time on and off the beam, then press the calculate button in the neutron activation panel.
3. To perform scattering calculations, fill in the wavelength of the neutrons and/or neutrons, the thickness and the density of the sample in the formulae, then press the calculate button in the absorption and scattering panel.

Co at 8.90 g/cm³

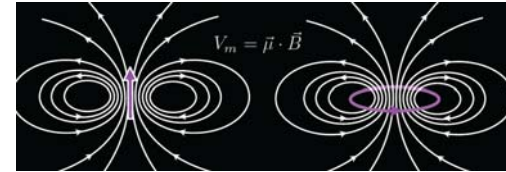
Source neutrons: 1.000 Å = 11.80 meV = 3956 m/s
Source X-rays: 1.542 Å = 8.042 keV

1/e penetration depth (cm)	Scattering length density (10 ²⁴ Å ⁻³)	Scattering cross section (E=meV)	X-ray SLD (10 ¹⁰ Å ⁻²)
abs	real	real	real
abs-incoh	imag	abs	imag
abs-incoh-coh	incoh	incoh	

Neutron transmission is 9.86% for 3 cm of sample (after absorption and incoherent scattering).
Terminated flux is 9.85e+6 n/cm²s for a 1e11 n/cm²s beam.

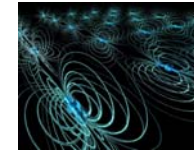
Neutrons and Magnetism

B-field due to both orbit and spin



The neutron has a magnetic moment

- Can solve magnetic structures
- Can study magnetic excitations



Field due to spin and orbital moments is

$$\vec{B} = \vec{B}_s + \vec{B}_L = \frac{\mu_0}{4\pi} \left\{ \vec{\nabla} \times \left(\frac{\vec{\mu}_e \times \hat{R}}{R^2} \right) - \frac{2\mu_B \vec{p} \times \hat{R}}{\hbar R^2} \right\}$$

Magnetic vector potential \vec{A} , of a dipolar field due to electron spin moment

Biot-Savart Law for a single electron of linear momentum, \vec{p}

Evaluating the spatial part of the transition matrix element

$$\langle \vec{k}_f | V | \vec{k}_i \rangle \propto \exp(i\vec{Q} \cdot \vec{r}_k) \left\{ \vec{Q} \times (\vec{s}_j \times \vec{Q}) + \frac{i}{\hbar Q} (\vec{p} \times \vec{Q}) \right\}$$

Basic Ideas

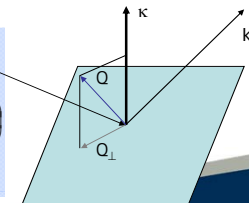
- Magnetic dipole moment:
- Differential cross-section

$$\vec{\mu}_n = -\gamma \frac{e\hbar}{2m} \vec{\sigma}$$

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k'}{k} \left(\frac{m}{2\pi\hbar^2} \right)^2 \left| \langle k' \sigma' \lambda' | V_m | k \sigma \lambda \rangle \right|^2 \delta(E_\lambda - E_{\lambda'} + \hbar\omega)$$

(See e.g. Squires eqn 7.15)

$$\begin{aligned} V_m &= -\vec{\mu}_n \cdot \mathbf{B} \\ \langle k' | V_m | k \rangle &= -r_0 \vec{\sigma} \cdot \hat{Q}_\perp \\ \hat{Q}_\perp &= \sum_j \exp(i\vec{\kappa} \cdot \vec{r}_j) \left(\vec{\kappa} \times (\vec{s}_j \times \vec{\kappa}) - \frac{i}{\hbar\kappa} (\vec{\kappa}_j \times \vec{p}_j) \right) \\ Q &= -\frac{1}{2\mu_B} \mathbf{M}(\vec{\kappa}) \end{aligned}$$



$$Q = Q_s + Q_l = -\frac{1}{2\mu_B} \mathbf{M}(\vec{\kappa})$$

- Neutrons only see the components of the magnetisation perpendicular to the scattering vector

- 1972 first non-resonant magnetic x-ray scattering experiment by Bergevin and Brunel
- 1985 first prediction of magnetic resonance scattering by Blume
- 1985 first experimental observation of resonance enhancement at the K-shell of Ni by Namikawa et al.
- 1987 discovery of resonance absorption of circular polarized x-rays in Fe by G. Schütz et al.
- 1988 seminal paper by Gibbs et al. on magnetic scattering at Ho spin spiral
- 1990 first demonstration of resonance magnetic reflectivity at Fe -film by Kao et al.

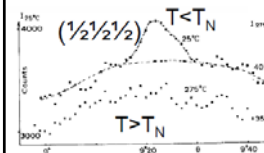
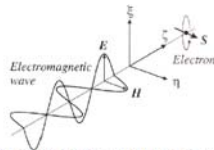


Fig. 1. Intensity $I_s(\theta)$ near the $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ position at $T = 25^\circ \text{C}$ and 275°C in counts/225 min. The hump which cover the interval could be due to some impurity.

Non-resonant Spin term



E-field interacts with the charge, but H-field interacts with the spin of the electron. This gives rise to a weak spin scattering amplitude with the following characteristics:

- Spin scattering amplitude is weaker than charge scattering by a factor: $\tau = h\nu/m_e c^2 = E_{\text{photon}}/E_{\text{electron}} \approx 10\text{keV}/511\text{keV} = 2 \times 10^{-2}$
- For a single electron the intensity is reduced by $\tau^2 \approx 10^{-4}$, such that only with high energy x-rays non-resonant spin scattering makes sense.
- Charge and spin scattering for a plane polarized incident beam are 90° out of phase
- Spin scattering causes a partial rotation of the plane of polarization
- The combined charge and spin scattering amplitude becomes:

$$f = -r_e [(e_f \cdot e_i) F_c - i\tau F_s \cdot B]$$

Non-resonant orbital term

Scattering of EM-waves at atoms slightly changes the orbit of electrons, giving rise to orbital magnetic scattering. The combined charge and spin and orbital scattering amplitude becomes:

$$f = -r_e [(e_f \cdot e_i) F_c - i\tau (F_s \cdot B_s + F_l \cdot B_l)]$$

$F_{c,s,l}$ are unit cell structure factors for the charge, spin, and orbital distribution, respectively.

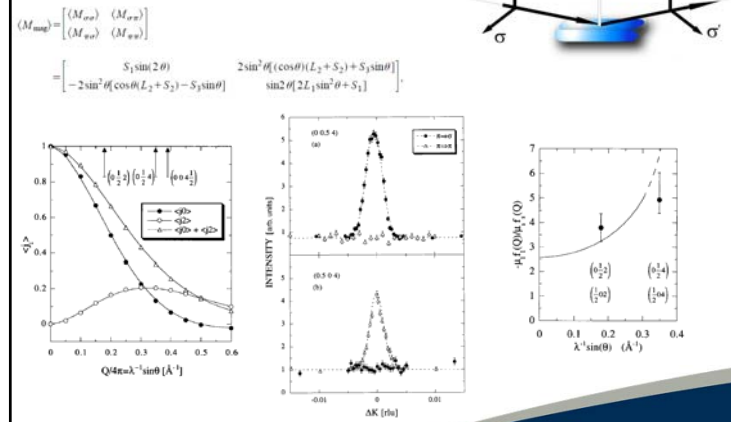
$B_{s,l}$ express selection rules, similar to magnetic neutron scattering, such that the cross section are only sensitive to magnetic moments, which are perpendicular to the scattering vector.

However, B_s and B_l describe different rotations of the plane of incident polarization, which allows a separation of spin and orbital contributions, unlike neutron scattering.

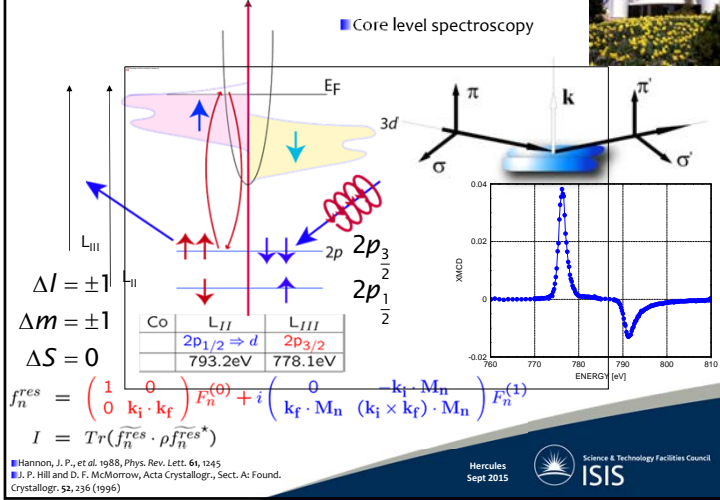
Non-Resonant Summary

- Magnetic scattering of spin and orbital moment rotates the plane of polarization, but differently, allowing separation of spin and orbital contributions; for forward scattering, orbital sensitivity is lost. Spin scattering is therefore best achieved by using high energy x-rays at small scattering angles.
- For a linearly polarized incident wave, the scattering amplitudes of charge and magnetic scattering (spin and orbital) are 90° out of phase. Because of the $\pi/2$ -shift, charge and magnetic scattering can not interfere (interference term can not be used for scattering enhancement);
- Magnetic scattering is very weak compared to charge scattering. The magnetic cross section scales with v^2 . (In the case of the Brunel & de Bergevin experiment the intensity ratio was about 10^{12}) (e^2/mc^2)²
- Magnetic and charge scattering can either be separated in case of an antiferromagnetic reflection (NiO), or by σ - π scattering.

Example UAs



Resonance Enhancement



Transmission

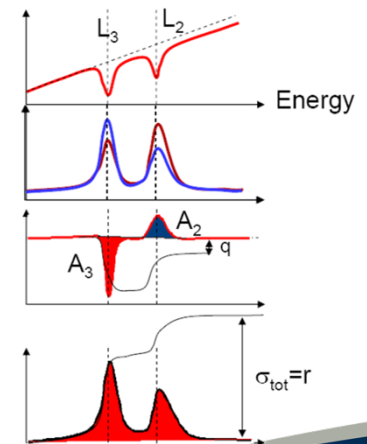
Difference signal

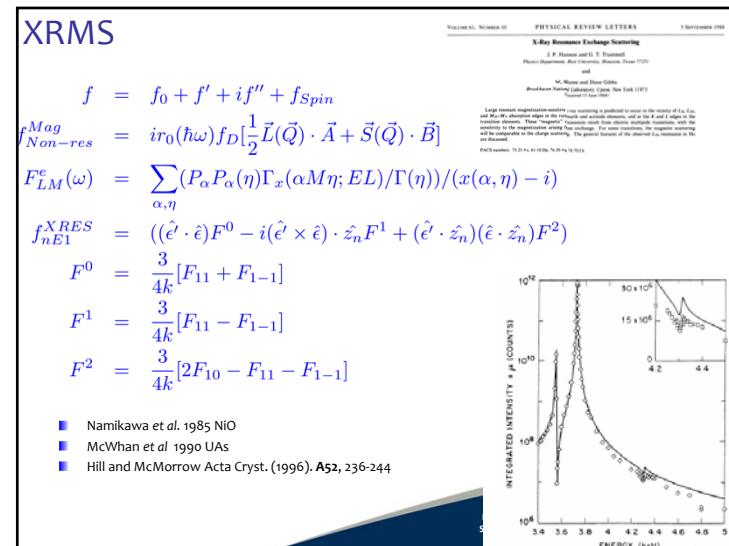
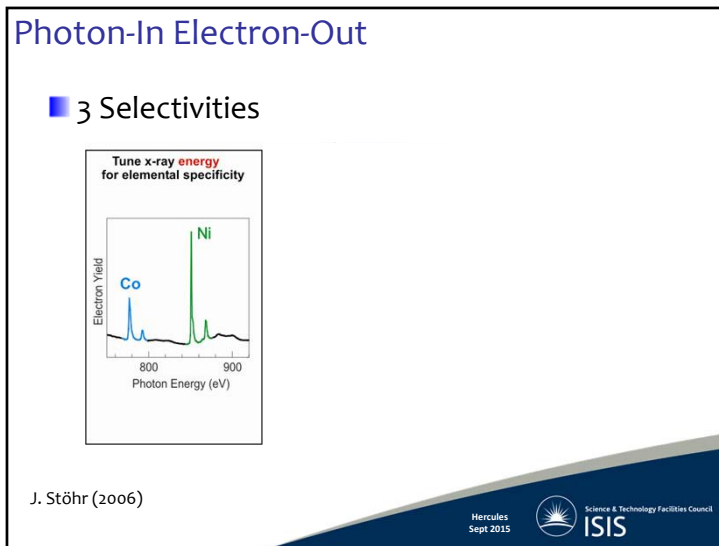
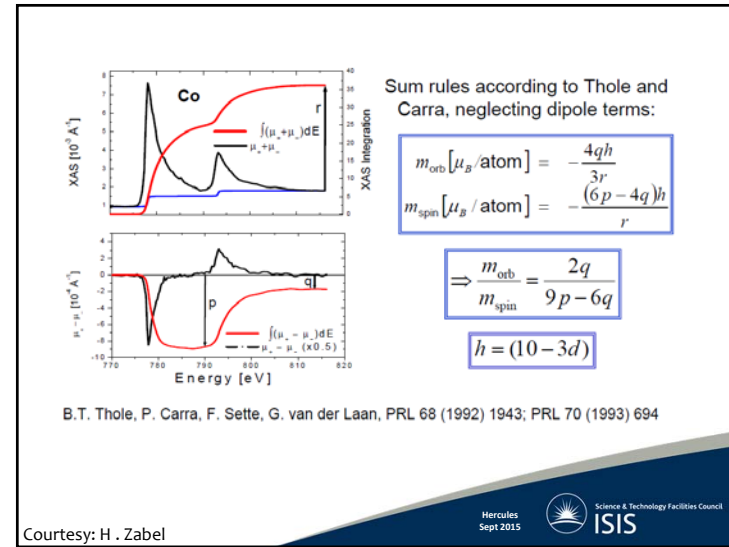
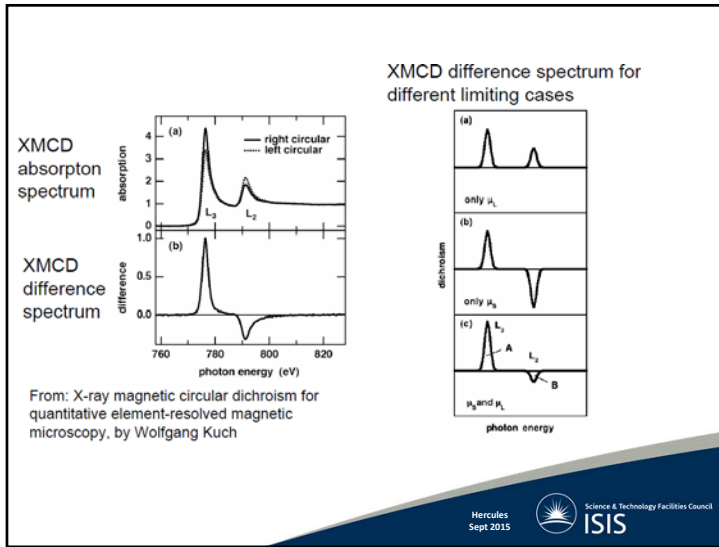
$$q = \int (\mu_{\perp} - \mu_{\parallel}) dE$$

\propto orbital moment

Total absorption

$$r = \int (\mu_{\perp} + \mu_{\parallel}) dE$$



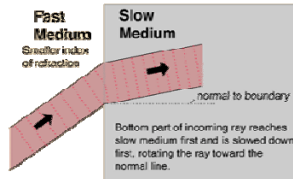
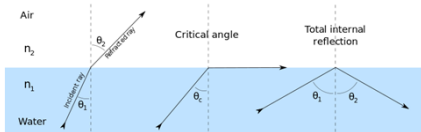


Refractive Index

$$n = \frac{c}{v}$$

- n varies with wavelength: dispersion

$$n_1 \sin \theta_1 = n_2 \sin \theta_2$$



<http://hyperphysics.phy-astr.gsu.edu>
en.wikipedia.org

$$n = 1 - \lambda^2 A - i\lambda B \tag{1}$$

$$A = \frac{Nb}{2\pi} \tag{2}$$

$$B = \frac{N(\sigma_a + \sigma_i)}{4\pi} \tag{3}$$

$$n = 1 - \alpha - i\beta \tag{1}$$

$$\alpha = \frac{N\lambda^2 |r_e|}{2\pi} \tag{2}$$

$$\beta = \frac{\lambda\mu}{4\pi} \tag{3}$$

n < 1 Total External reflection

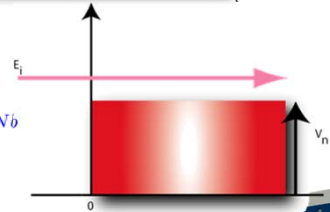
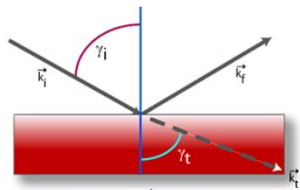


Index of Refraction: Neutrons

$$n = \frac{\sin \gamma_i}{\sin \gamma_t} = \frac{|\vec{k}_t|}{|\vec{k}_i|}$$

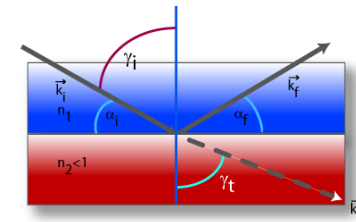
$$n^2 = \frac{|\vec{k}_t|^2}{|\vec{k}_i|^2} = \frac{E_t}{E_i} = \frac{E_i - V_n}{E_i} = 1 - \frac{4\pi}{k_i^2} N b$$

$$V_n = \frac{2\pi\hbar^2}{m_n} N b$$



Critical Reflection

- $\frac{\cos \alpha_i}{\cos \alpha_f} = \frac{n_2}{n_1}$
- At the critical angle $\frac{\cos \alpha_i}{\cos 0} = n$

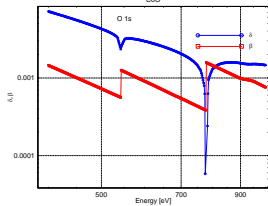
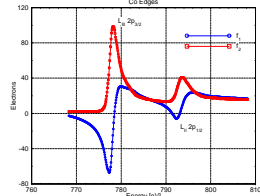


$$Q_c = \frac{4\pi}{\lambda} 2k \sin \alpha_c = 2k \sqrt{1 - \cos^2 \alpha_c} = \sqrt{4k^2 (1 - n^2)} \cong \sqrt{4k^2 \cdot 2\delta} = \sqrt{16\pi N b}$$

Material	$\theta_c / \text{\AA}$
Ni	0.1
Cu	0.083
Al	0.047
Si	0.047
D ₂ O	0.082

- Q_c only depends on the material!

Index of Refraction: Photons



http://www-cxro.lbl.gov/optical_constants/

$$n = 1 - \delta - i\beta$$

$$= 1 - \frac{r_e}{2\pi} \lambda^2 \sum_i n_i f_i(0)$$

$$f(0) = f_1 + i f_2 \frac{\sigma_a}{2r_e \lambda}$$

$$= Z^* + \frac{1}{\pi r_e h c} \int_0^\infty \frac{\epsilon^2 \sigma_a(\epsilon)}{E^2 - \epsilon^2} d\epsilon$$

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Specular Scattering

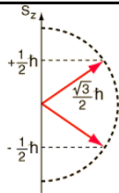
Neutron and x-ray reflectivity

How do we connect the scattering length profile with the reflectivity

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Using the neutron's spin

- The neutron is a spin $\frac{1}{2}$ particle
- The neutron possesses an intrinsic magnetic moment: spin
- Caution...



$$V = V_n + V_m \quad (1)$$

$$V = \frac{\hbar}{2\pi m} N b - \mu \cdot \mathbf{B} \quad (2)$$

$$= -4\pi \mu_n \cdot \mathbf{M} \quad (3)$$

$$= -\frac{2\hbar\pi^2}{m_n} \rho(z) \sin(\theta) \quad (4)$$

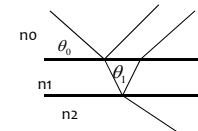
$$= \frac{2\hbar\pi^2}{m_n} \frac{M_p}{M_s} N \frac{m_n \mu_n \mu_0}{2\pi \hbar^2} \mu_s \quad (5)$$

$$\vec{\mu}_n = \gamma \mu_N \vec{\sigma}$$

$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$

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Fresnel's law for a surface and thin film



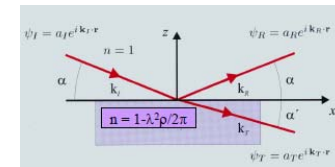
$$r_{ij} = \frac{a_r}{a_i} = \frac{k_i - k_j}{k_i + k_j}$$

$$t_{ij} = \frac{a_j}{a_i} = \frac{2k_i}{k_i + k_j}$$

$$k_i = n_i \sin \theta$$

$$\beta_i = \frac{2\pi}{\lambda} n_i d_i \sin \theta_i$$

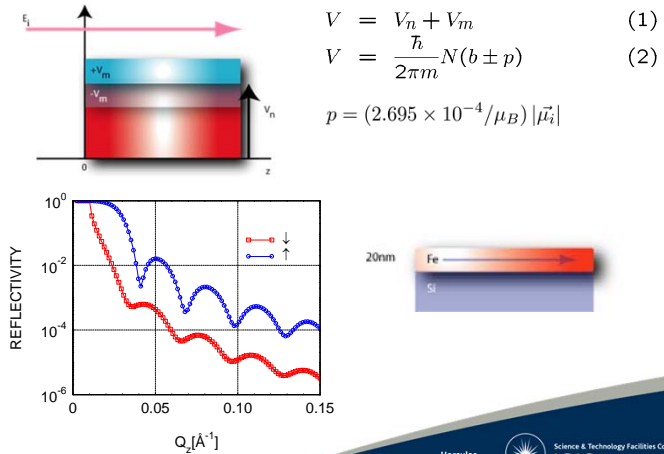
$$R(Q) = \left| \frac{r_{01} + r_{12} \exp(-2i\beta_1)}{1 + r_{01} r_{12} \exp(-2i\beta_1)} \right|^2$$



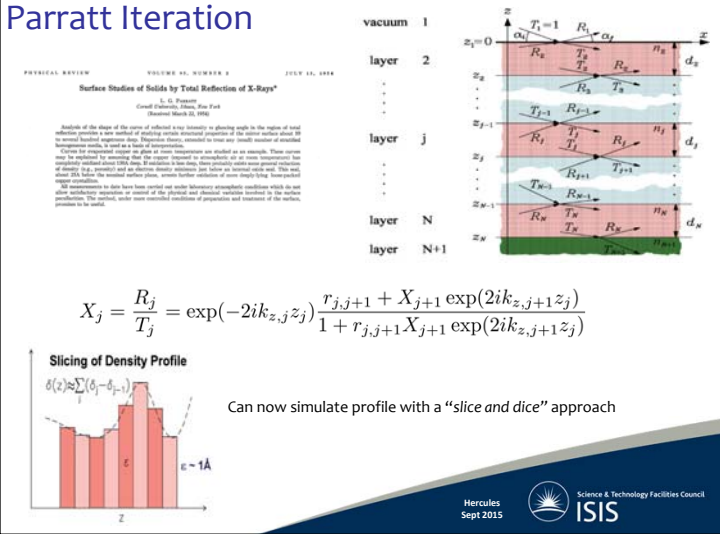
<http://www.che.udel.edu/cns/pdf/Reflectometry.pdf>

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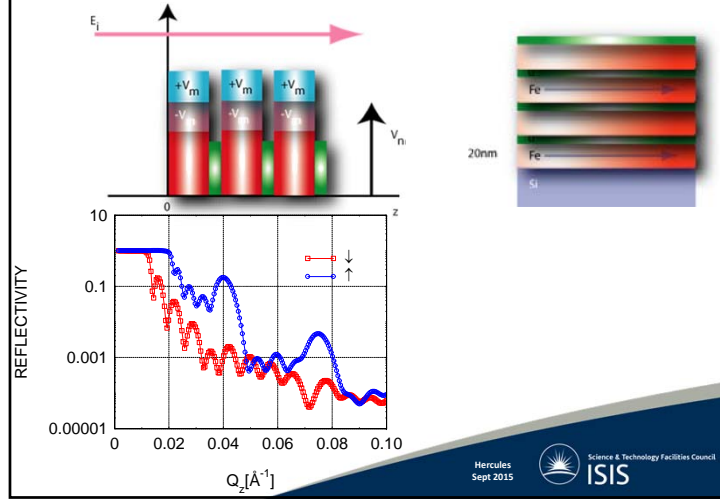
PNR from a single layer



Parratt Iteration



PNR from a multiple layers



Spin dependent cross-section

- In-plane orientation of magnetisation obtainable from 4 spin dependent cross-sections
- Components of the magnetisation, m give rise to
 - $m \parallel H$: Non Spin Flip Scattering (NSF)
 - $m \perp H$: Spin Flip Scattering (SF)
- Dynamical analysis gives absolute depth dependence profile

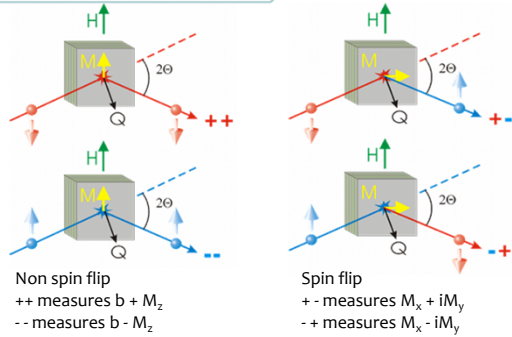
$$b = b + p \sin \phi$$

$$pm \cos \phi = px$$

$$\left[\frac{-\hbar^2 \nabla^2}{2m_n} + V(r) \right] \psi^{\uparrow, \downarrow} = E \psi^{\uparrow, \downarrow}$$

Polarisation

Polarised neutron reflection

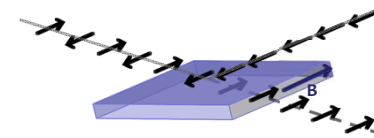


By fitting all components the direction and strength of the magnetic moment can be measured as a function of depth

Polarisation

Polarising supermirrors

Magnetic materials have a spin dependent term in their refractive index



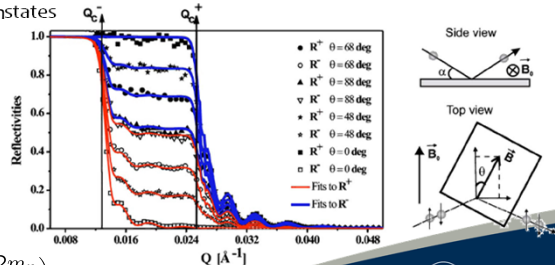
An ~60%/40% Fe/Co mirror works well at saturation

Caveat to Classical Description

- CD predicts a continuous variation of critical edge

$$\frac{4\pi \sin(\alpha_c^\pm)}{\lambda} = Q_c^\pm = \sqrt{\frac{2m_n}{\hbar^2} (V_n \pm |\mu| |B_s| \cos(\theta))}$$

- Stern-Gerlach effect? Only 2 eigenstates



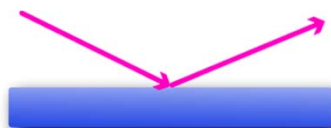
$$k_{\pm}^2 = \left(\frac{2m_n}{\hbar^2}\right) (V_n \pm |\mu| |B|)$$

PHYSICAL REVIEW B 71, 214423 (2005)
 Quantum states of neutrons in magnetic thin films
 F. Radu,^{1,*} V. Leiner,² M. Woith,³ V. K. Ignatovich,⁴ and H. Zabel¹
¹Department of Physics, Ruhr-University Bochum, D-44780 Bochum, Germany
²Institut für Werkstoffforschung WFN, GRSO Forschungszentrum Geesthacht, 21502 Geesthacht, Germany
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 (Received 12 April 2005; published 23 June 2005)

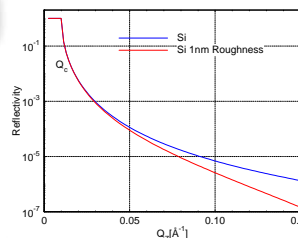
Roughness

Structural and magnetic interfacial phenomena

Structural Roughness



$$R(Q) = R_F \exp(-Q^2 \sigma^2)$$

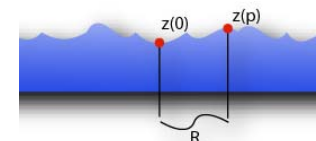


X-ray and neutron scattering from rough surfaces

S. K. Sinha, E. B. Sirota, and S. Garoff*
Corporate Research Science Laboratory, Exxon Research and Engineering Company, Clinton Township, Route 22 East, Annandale, New Jersey 08801

H. B. Stanley†
University of Maryland, College Park, Maryland 20742
(Received 30 November 1987)

$$C(R) = \langle [z(x, y) - z(x', y')]^2 \rangle = \sigma^2 \exp(-R/\xi)^{2h}$$



- σ = roughness
- ξ = cut-off length:
 - for $R > \xi$, interface appears smooth,
 - for $R < \xi$, interface appears rough, fractal behaviour
- $h=3$ -D Hurst parameter for jaggedness($0 < h < 1$)
 - smooth: $D=2, h=1$
 - very rough $D=3, h=0$

Simulation Packages

Simulation Packages: neutron

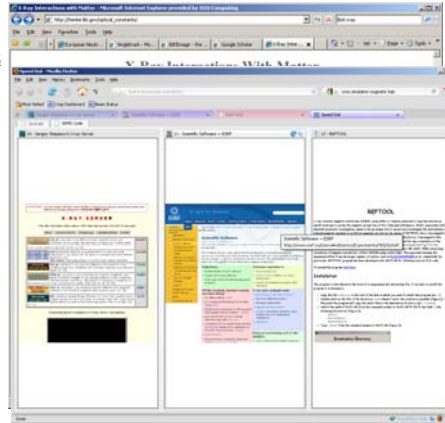
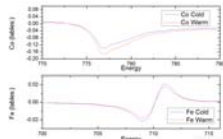
<http://neutronreflectivity.neutron-eu.net/main/SimulationPrograms>



Simulation Packages: photon

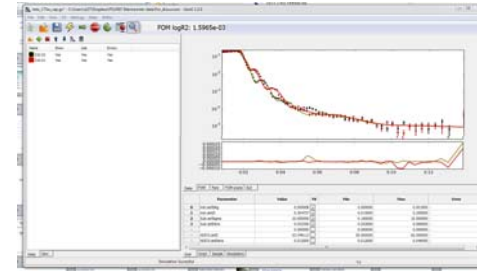
- X-ray

- <http://sergey.gmca.aps.anl.gov/>
- ESRF
- RefTool
- GenX



GenX (neutron and x-ray reflectivity)

- Sourceforge

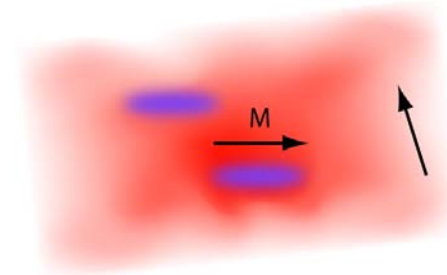


Off-specular scattering

Probing in-plane lengthscales

$L_{\text{Coh}} < \text{Domain size}$

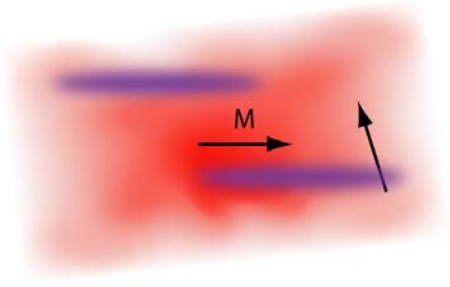
- $I \propto \sum_i (A_i^2)$
- Only specular scattering



$L_{\text{Coh}} > \text{Domain size}$

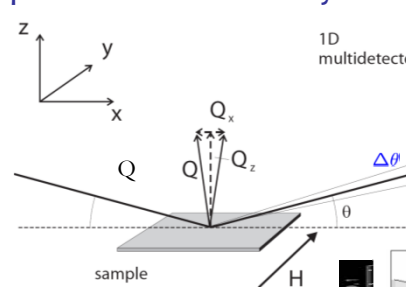
$I \propto (\sum_i A_i)^2$

Only diffuse scattering



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Experimental Geometry

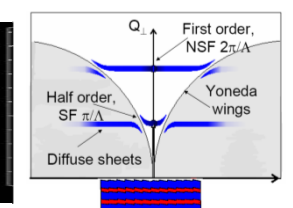


1D multidetector

$Q_x = |Q| \sin \theta \times \Delta \theta$

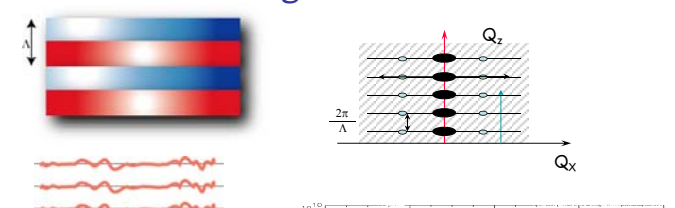
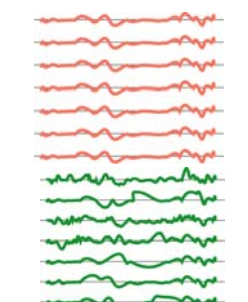
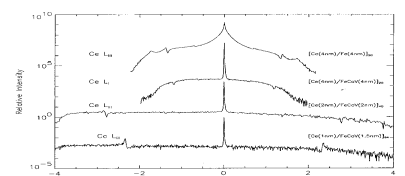
$Q_y = |Q| \times \Delta \phi$

- λ range 0.5-6.5 Å
- 1-d Detector
- Typical acquisition ~2 hours/field
- Measured coherence length ~30 μm



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Diffuse scattering

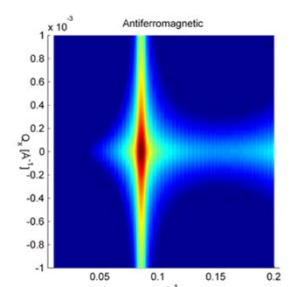




(Ce/Fe) Tixier, Mannix et al.

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Holy and Baumbach, prb, 49, 10668, (1994)

Kinematic calculation



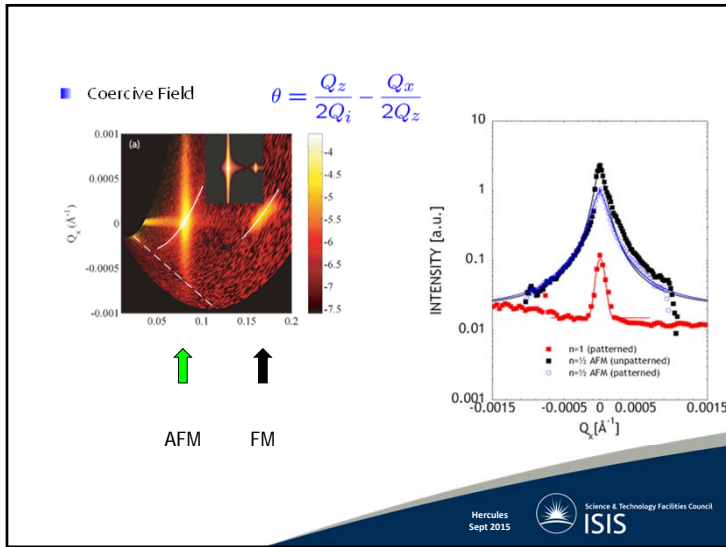
Antiferromagnetic

$I(\mathbf{Q})/I_0$

$Q_z [\text{Å}^{-1}]$

- Langridge et al. Phys. Rev. Lett. (2000) 85, 4964

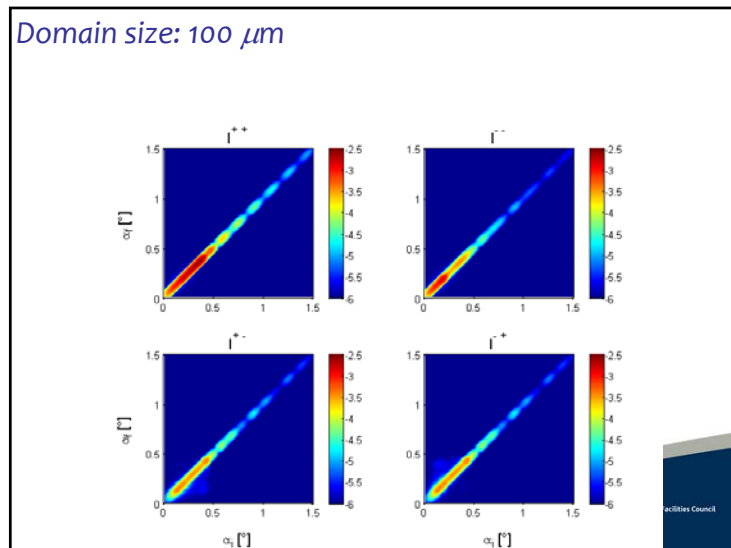
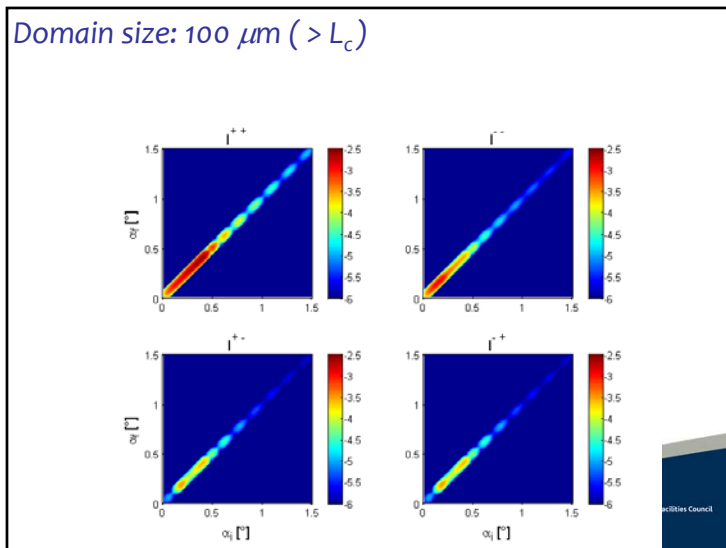
Hercules Sept 2015 Science & Technology Facilities Council ISIS



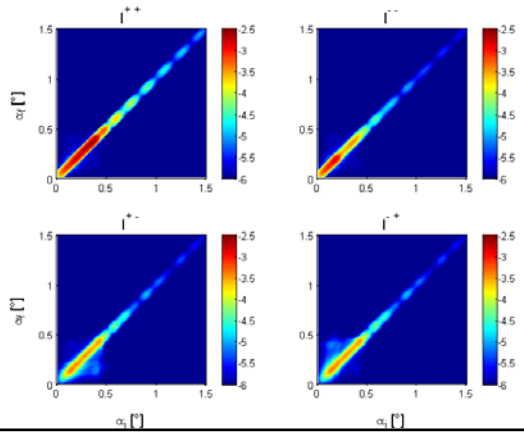
Domains and the coherence volume

Many thanks to H. Zabel for slides

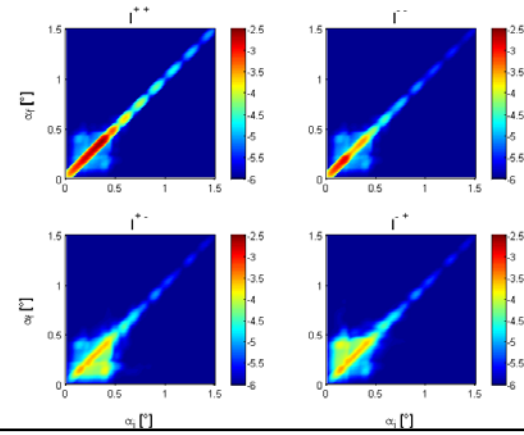
Science & Technology Facilities Council ISIS



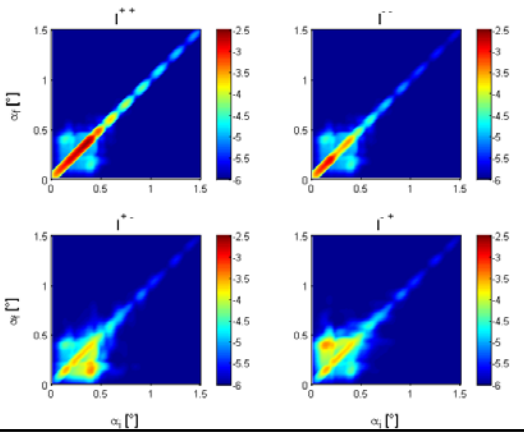
Domain size: 50 μm



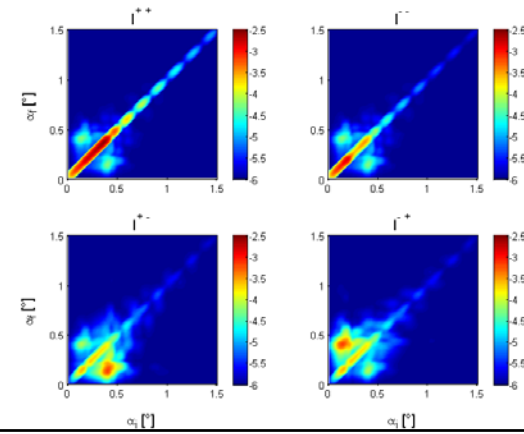
Domain size: 10 μm



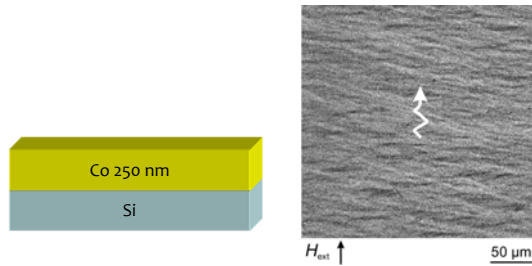
Domain size: 5 μm



Domain size: 1 μm

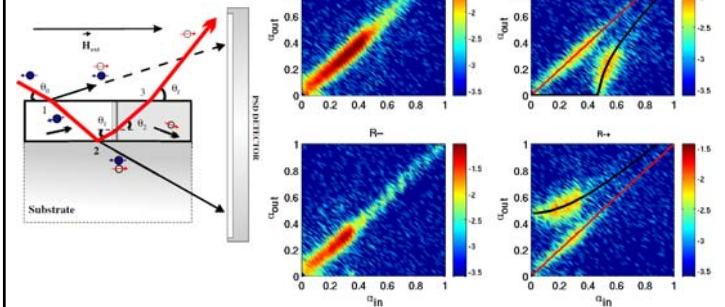


Single ferromagnetic film in the domain state

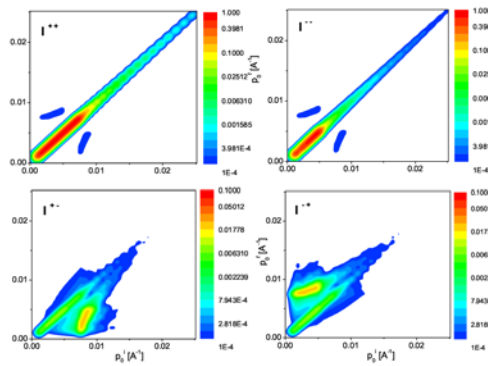


F. Radu et al, J. Phys.: Condens. Matter 17 (2005) 1711-1718

Banana shape off-specular scattering from domain state



Simulation of domain state



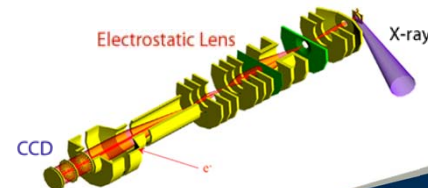
B.P. Toperverg Physica B 297 (2001) 160-168

XPEEM: X-ray Photoelectron Emission Microscopy

- Spatial resolution ~ 30 nm
- Short acquisition times
- Element sensitive technique (XMC(L)D)
- Surface sensitive technique
- Real Space Technique

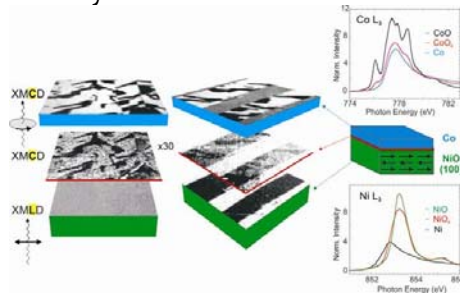


<http://www.diamond.ac.uk>



Imaging Interfacial Magnetism

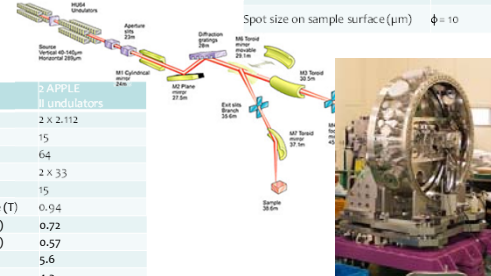
Mixed layer



H. Ohldag, et al Phys. Rev. Lett. 87, 7201 (2001)
<http://www-ssrl.slac.stanford.edu/stohr/magneticexchange.htm>

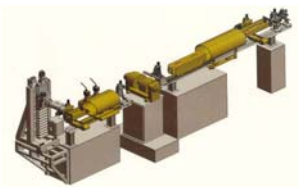
Soft x-ray beamline i06/i10

06	diamond
Energy range (first harmonic circular) (eV)	106 - 1300
Energy range (linear horizontal) (eV)	80 - 2100
Energy range (linear vertical) (eV)	130 - 1500
Resolving power ($\Delta E/E$)	10,000 @ 400eV
Spot size at PEEM (μm) (FWHM)	10 (H) x 3 (V)
Spot size on sample surface (μm)	$\phi = 10$

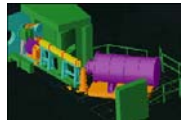


Insertion device	3 APPLE II undulators
Nominal magnet length (m)	2 x 2.112
Nominal magnet gap (mm)	15
Magnet period (mm)	64
Number of periods	2 x 33
Nominal magnet gap (mm)	15
Peak field in horizontal mode (T)	0.94
Peak field in vertical mode (T)	0.72
Peak field in circular mode (T)	0.57
K_{MAX} in horizontal mode	5.6
K_{MAX} in vertical mode	4.3
K_{MAX} in circular mode	3.4

Reflectometers



- The CRISP reflectometer at ISIS is a white beam time of flight (tof) polarised neutron reflectometer viewing a 20K hydrogen moderator.



<http://www.llf.fr/YellowBook/D17/>



<http://www.llf.fr/YellowBook/ADAM/>

- Continuous or tof mode of operation
- 5Å Monochromator (6Å Polarised)
- White beam flux 9.6×10^9 n/s/cm²

PolRef



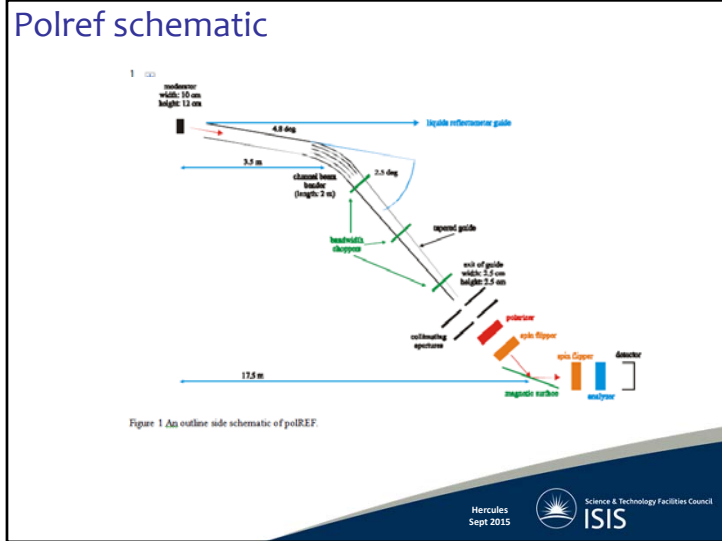
- Wavler
- Unpolar
- Polarize

- Source t
- Beam in

- Well shi
- 640 channel linear gas detector with 0.5mm pixel

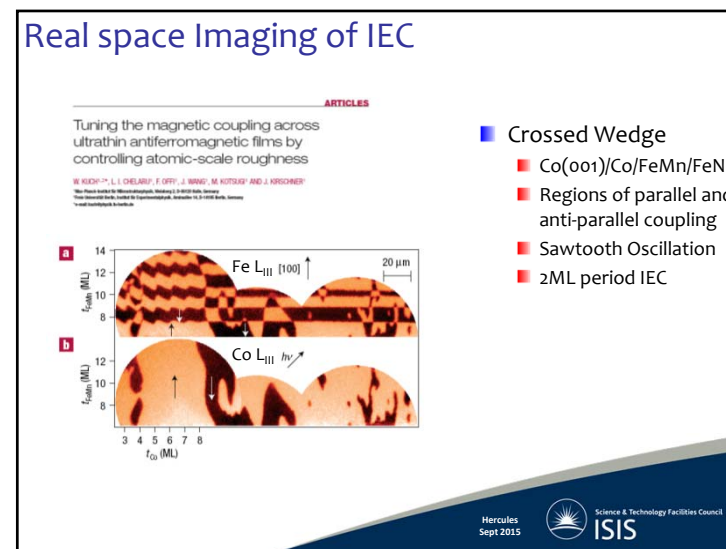
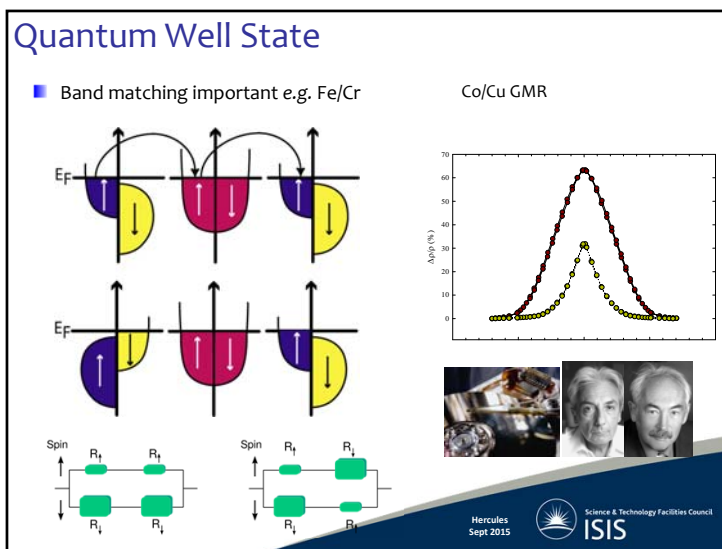
- Vertical 2θ 7.5°
- Horizontal 2θ 22°



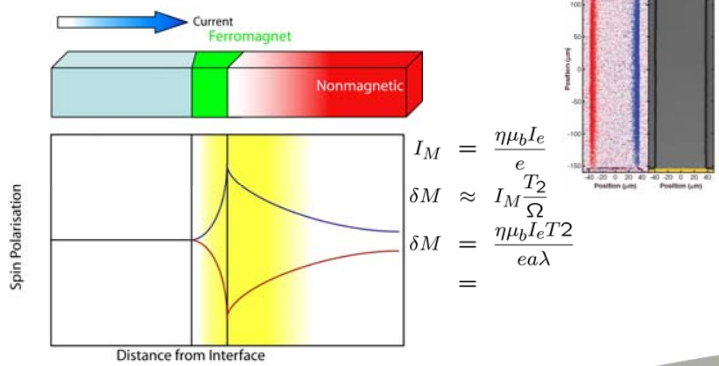


Examples of reflectivity

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Spin Accumulation



■ A. Fert et al., J. Phys. D: Appl. Phys. 35 (2002) 2443-2447
 ■ M. Johnson and R.H. Silsbee Phys. Rev. Lett. 55 (1985) 1790
 ■ Kato et al. Science 306 1910 (2004)

Structural Characterisation

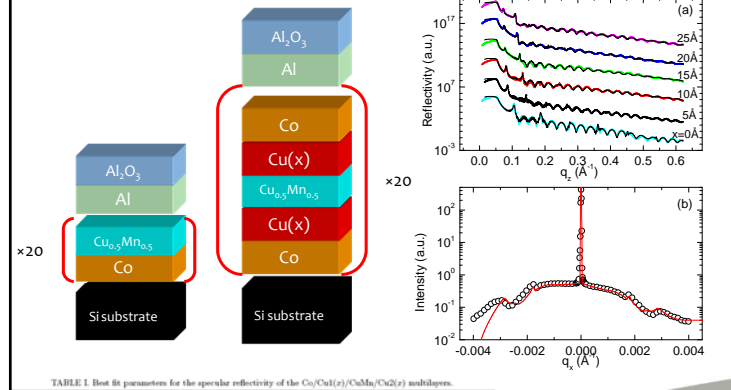
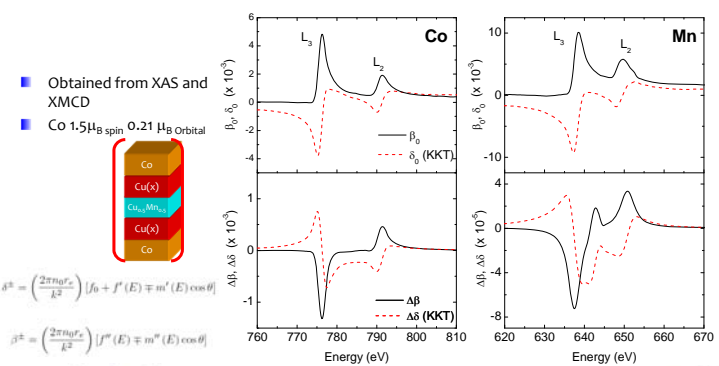


TABLE 1. Best fit parameters for the specular reflectivity of the Cu/Cu(x)/CuMn(x)/Cu₂(x) multilayers.

p	Thickness				Interface Morphology			
	Co ±1Å	Cu1 ±0.5Å	CuMn ±0.5Å	Cu2 ±0.5Å	σ _{Co/Cu1} ±0.5Å	σ _{Cu1/CuMn} ±0.5Å	σ _{CuMn/Cu2} ±0.5Å	σ _{Cu2/Si} ±0.1
0	42	-	23.5	-	5.5	1.0	180	0.5
5	41	6.5	22.5	5.5	4.0	0.5	170	0.5
10	41	12.0	19.5	11.5	5.0	1.0	220	0.5
15	43	16.5	23.0	17.5	5.5	0.5	180	0.5
20	39	22.5	20	21.0	5.5	0.5	180	0.5
25	43	27.0	22.5	26.5	6.0	1.0	250	0.5

Optical constants



$$\beta^\pm = \left(\frac{2\pi n_0 r_e}{k^2} \right) [f_0 + f''(E) \mp m''(E) \cos \theta]$$

$$\delta^\pm = \left(\frac{2\pi n_0 r_e}{k^2} \right) [f''(E) \mp m''(E) \cos \theta]$$

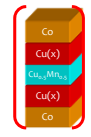
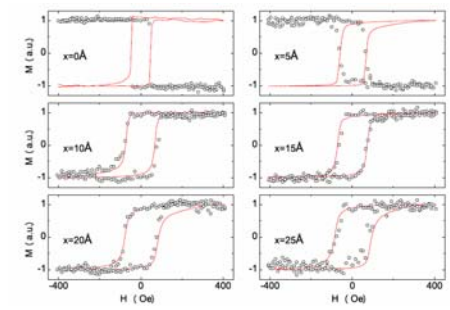
$$\delta^\pm = \delta_0 \mp \Delta \delta$$

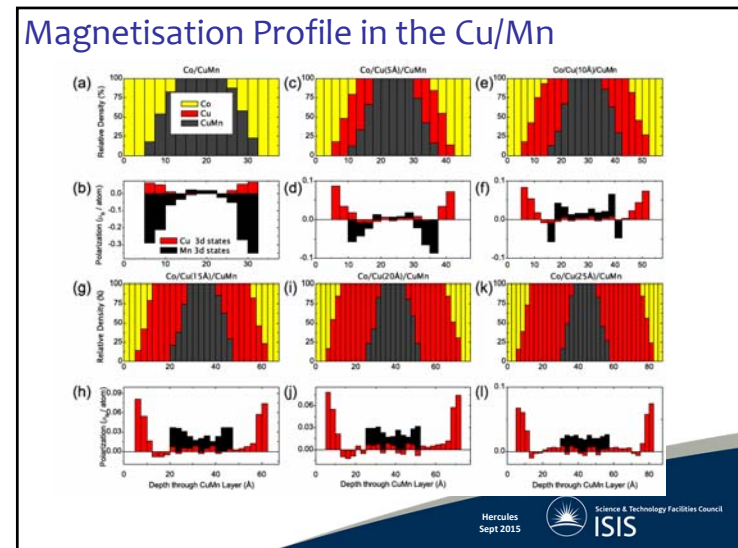
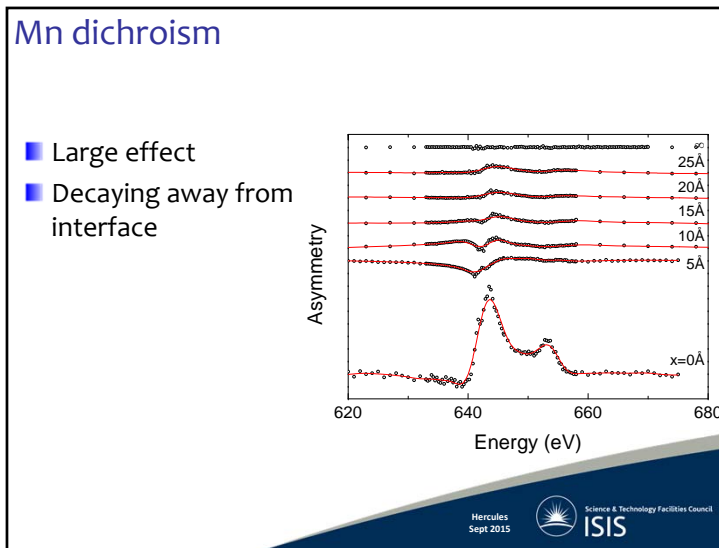
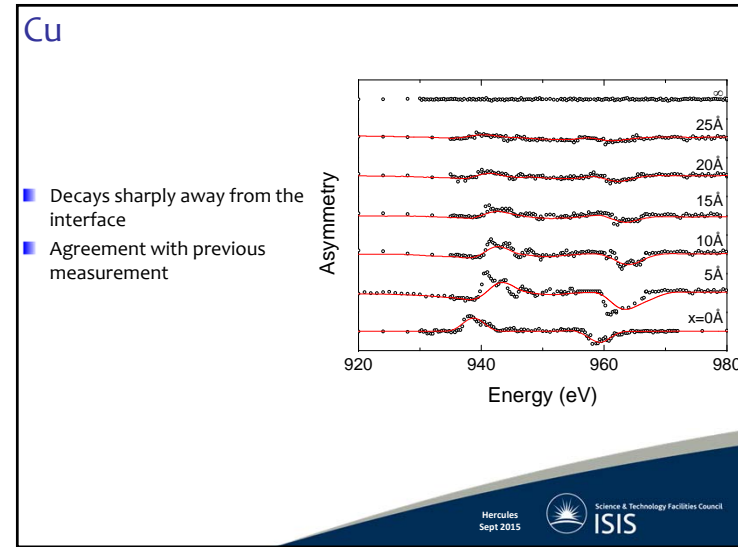
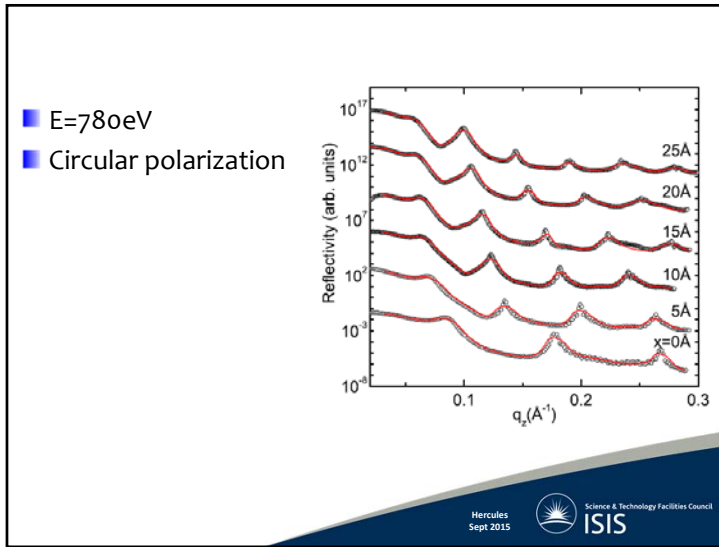
$$n_j^\pm = 1 - \delta_j^\pm + i \beta_j^\pm$$

$$\beta^\pm = \beta_0 \mp \Delta \beta$$

Hysteresis Loops

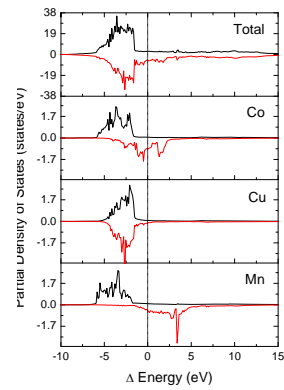
- Comparison of Co and Mn
- Clear transition from ↓↓ to ↑↑
- Mn rotates with the Co



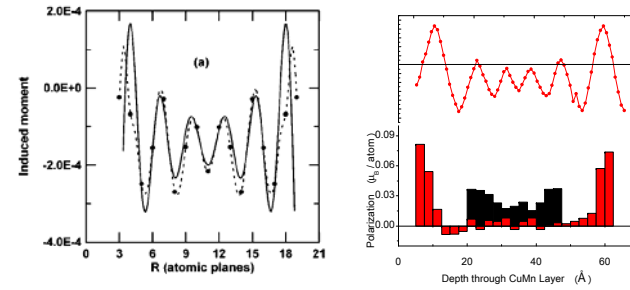


DFT-DOS

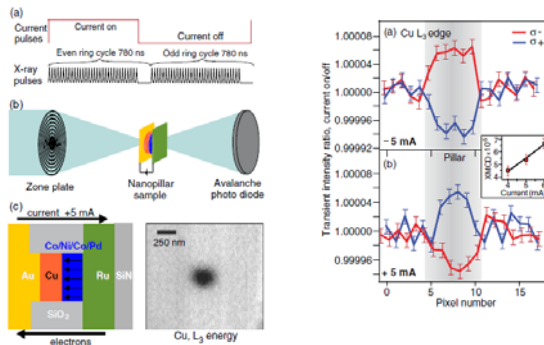
- Supercell 4 atoms of Cu and Mn
- $0.05\mu_B$ on Cu d orbital
- Decays rapidly from interface



Quantum Oscillations



■ Mathon et al. PRB 59 6344 (1999)



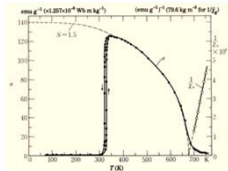
Kukreja R et al. X-ray Detection of Transient Magnetic Moments Induced by a Spin Current in Cu Phys. Rev. Lett. 115 096601 (2015)

A Controllable Magnetic Interface

Motivation

Tuneable Magnetostructural phase transition

- J.S. Kouvel, C.C. Hartelius, *J. Appl. Phys.*, **33**, 1343 (1962)
- L. Zsoldos, *Phys. Stat. Sol.*, **20**, K25 (1967)
- Anomalously strong electron-lattice coupling
- Fundamental interest experimentally and theoretically
 - Nature of the stability of the FM phase
 - Kudrnovsky, *J. et al. Phys. Rev. B* **91**, 014435 (2015)



HAMR, magnetic refrigeration

- FePt/FeRh system
 - Thiele et al. *Appl. Phys. Lett.* **82** 2859 (2003)
- Control MTJ cell coercivity

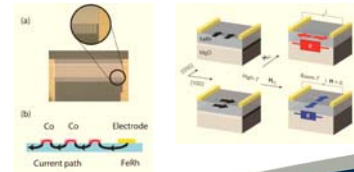
Ultra-fast switching

- Ju et al. *Phys. Rev. Lett.* **93**, 197403 (2004)
- Radu et al. *prb* **81**, 104415 (2010)
- Quirin et al. *prb* **85**, 020103(R) (2012)

Ability to control through heterostructure design

Spintronic Applications

- Marti, X. et al. *Nat. Mater.* **13**, 367 (2014)
- Naito, T. et al. *J Appl Phys* **109**, 07C911 (2011)



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- CsCl structure
 - $a=2.99\text{\AA}$
 - α' phase
- 300K: Type G AF
- Fe: $\sim 3.3 \mu_B$
- Rh: no moment
- FM alignment within $\langle 111 \rangle$ planes
- AF alignment between $\langle 111 \rangle$ planes
- 350 K: AF \rightarrow FM
- Fe: $\sim 3.1 \mu_B$
- Rh: $\sim 1 \mu_B$

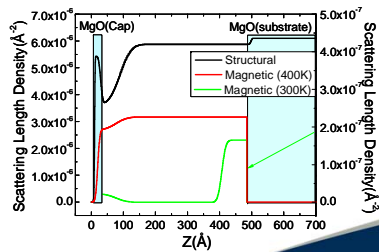
- J. S. Kouvel and C. C. Hartelius, *JAP*, **33**, 1343 (1962).
- G. Shirane et al. *Phys. Rev.* **134**, A1547 (1964)

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Sept 2015

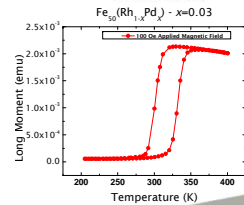
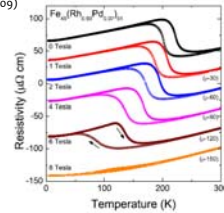
Strain and Pd doping

Transition temperature is tuneable via doping and field

- T_{AF-F} can be increased by doping with Ir and Pt and decreased by doping with Pd and Ni.
- Shift to lower temperatures observed by application of applied field.



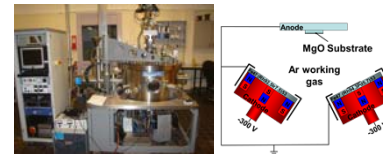
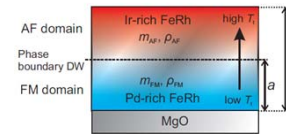
P. Kushwaha, A. Lakhani, R. Rawat, and P. Chaddah, *Physical Review B* **80**, (2009)



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Sample Growth



- Base pressure: $\sim 5 \cdot 10^{-8}$ torr.
- Sputtered using two angled DC magnetrons.
- Alloy targets used to make $FeRh_{1-x}Pd_xIr_y$:
 - $Fe47(Rh92.5Ir7.5)53$
 - $Fe47(Rh94.3Pd5.7)53$
- Substrate: MgO(001)
- Nominal FeRh(Pd/Ir) thickness 50nm.
- Post-growth anneal at 600°C for 60 min.
- Al (3nm) Cap deposited at 100°C.
- Aim is for anneal to diffuse layers for smooth doping gradient.

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- Transition controllable by:
 - Temperature
 - Field (H)
 - Doping – Pd, Pt (decrease T_c) or Ir, Ni (increase T_c)
 - Strain - R. Fan, et. al. Phys Rev B 82, 184418 (2010)
 - Pressure – S. Yuasa et. al. Phys. Soc. Jap. 63, 3, 855-858(1994).
 - Ion Beam – Koide, T. et al. J. Appl. Phys. 117, 17E503 (2015).
- Possible to create a controllable magnetic interface via a doping gradient

- Basis for a field or temperature sensor

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Magnetisation measurements

- Phenomenological characterisation of the FM phase

$$a(T) = t \left(\frac{m(T) - m_{AF}(T)}{m_{FM}(T) - m_{AF}(T)} \right)$$

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Resistance measurements

- Large change in carrier density across the transition

$$a(T) = t \left(\frac{1 - \frac{R(T)}{R_{AF}(T)}}{\frac{R(T)}{R_{FM}(T)} - \frac{R(T)}{R_{AF}(T)}} \right)$$

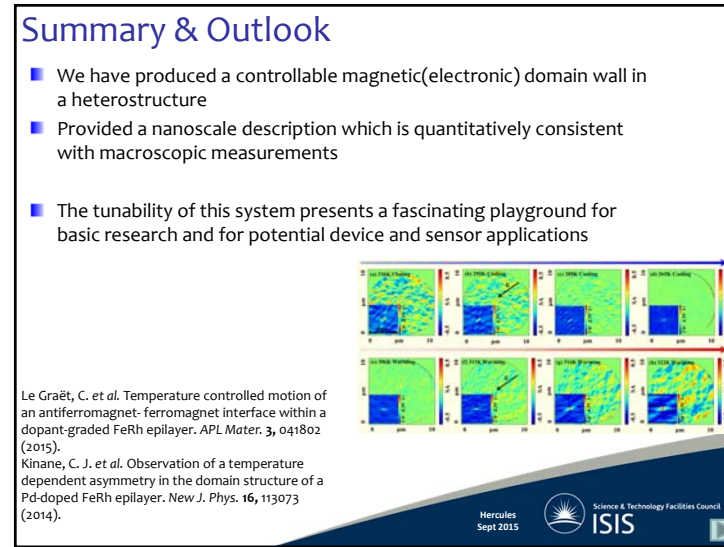
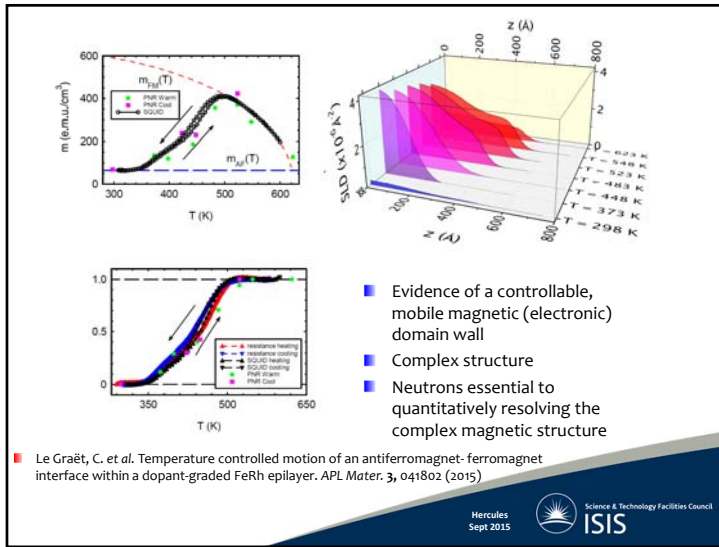
de Vries, M. A. et al. Hall-effect characterization of the metamagnetic transition in FeRh. New J. Phys. 15, 13008 (2013).

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Neutron Measurements

- Saturating Field
- Constrained two layer model
- The minimum to describe the data

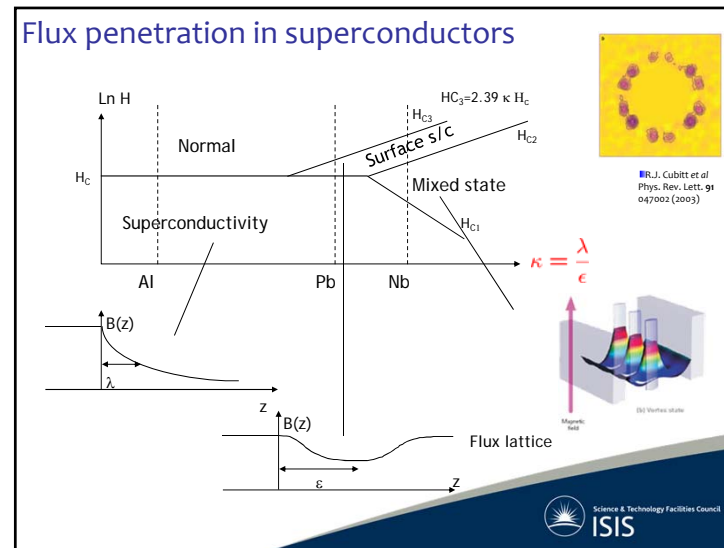
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Superconductivity and reflectivity

Towards superconducting spintronics

ISIS Science & Technology Facilities Council



Flux penetration in superconductors

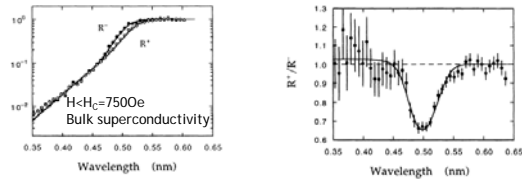


FIG. 2. The spin-dependent reflectivities R^+ and R^- measured in an applied magnetic field of 8.0×10^4 A/m (750 Oe). The continuous lines are the reflectivities calculated for the two polarization states, with the same instrumental and surface parameters as in Fig. 1, and an exponential decay of magnetic induction with a penetration depth of 39 nm.

PHYSICAL REVIEW B VOLUME 49, NUMBER 22 1 JUNE 1994-01

Magnetic-induction profile in a type-I superconductor by polarized-neutron reflectometry

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Department of Physics, Clarendon Laboratory, University of Oxford, Parks Road, Oxford, OX1 3PU, United Kingdom

D. M.K. Paul

Department of Physics, University of Warwick, Coventry, CV9 7AL, United Kingdom

J. Profide

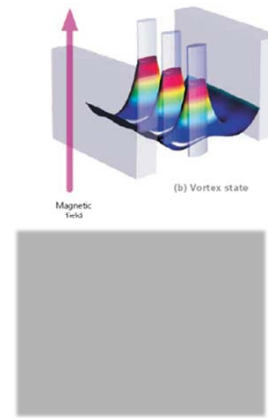
ISIS Science Division, Rutherford Appleton Laboratory, Chilton, Didcot, OX11 0QX, United Kingdom

(Received 17 February 1994)

- 1 μm Pb film
- PbO surface layer
- $B(z) = \mu_0 H \exp(-z/\lambda)$
- $\lambda = 39 \pm 1 \text{ nm}$
- No field dependence

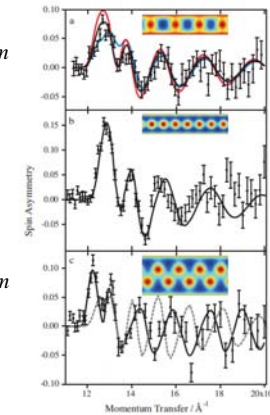


Visualising the Flux distribution



$d = 195 \text{ nm}$

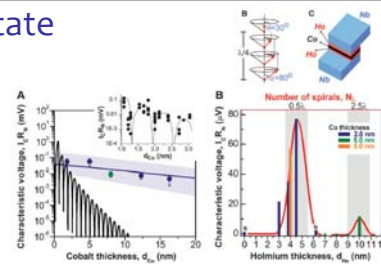
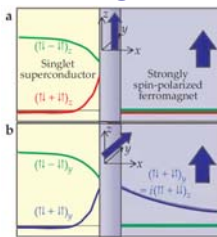
$d = 250 \text{ nm}$



Drew et al Phys. Rev. B. 80 134510 (2009)



Generating a triplet state

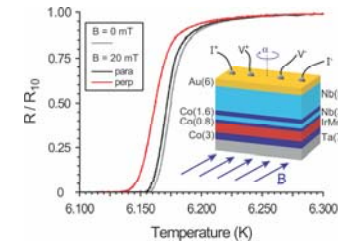
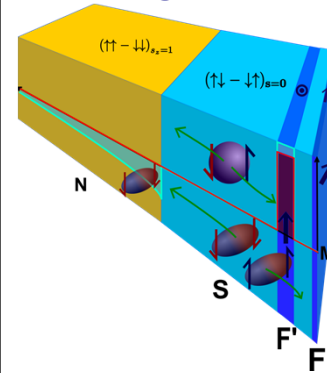


- first found in heavy fermion materials such as UGe_2 or URhGe
- orbital and spin components of the wavefunction are both even with respect to electron exchange, thus they must be an odd-frequency; odd with respect to time reversal
- non-collinear magnetism within a distance of the coherence length required

- Robinson et al. Science 329, 59 (2010)
- Banerjee et al. Nat. Commun. 5, 1 (2014).
- M. Eschrig Physics Today (2011)

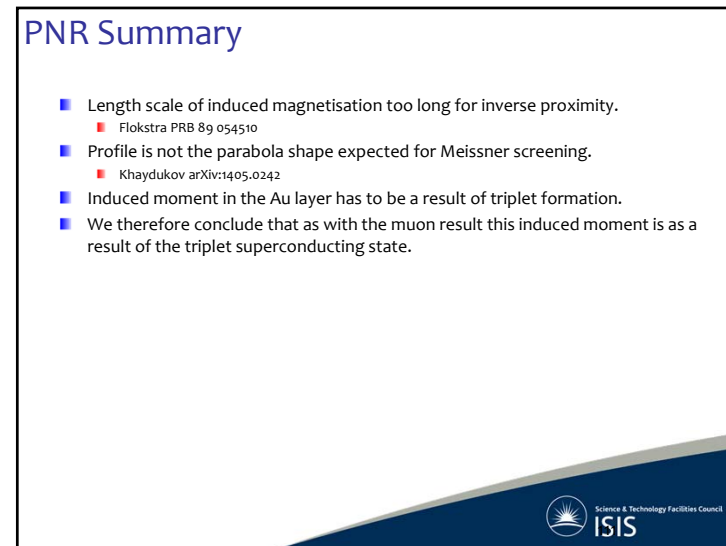
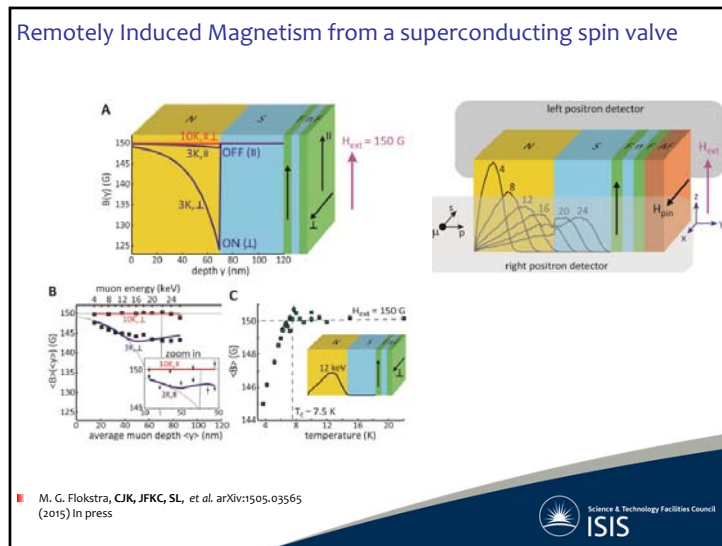
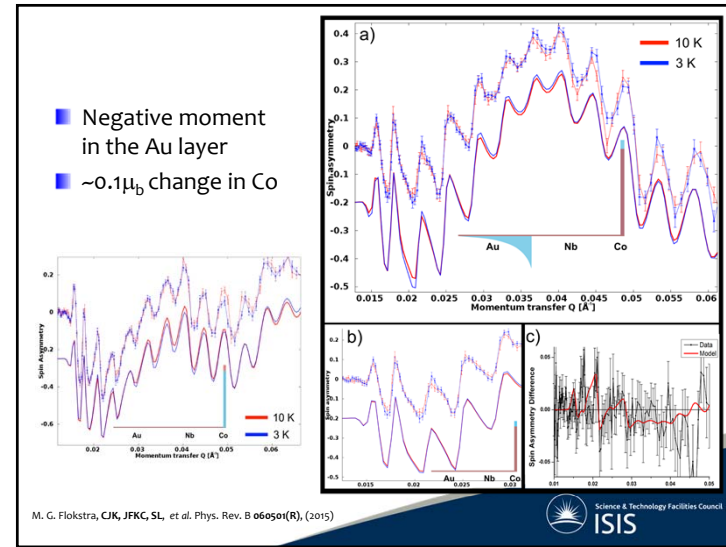
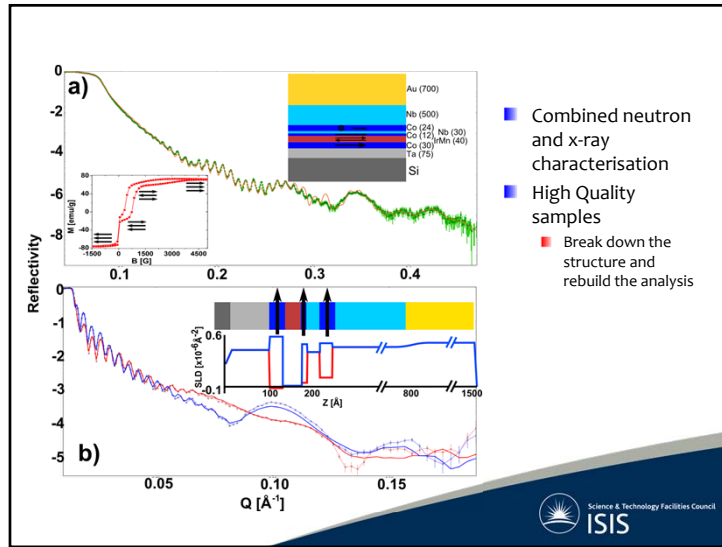


Generating a triplet state



- F. S. Bergeret, A. F. Volkov, and K. B. Efetov, Rev. Mod. Phys. 77, 1321 (2005)
- M. G. Flokstra, CJK, JFKC, SL, et al. Phys. Rev. B 060501(R), (2015)

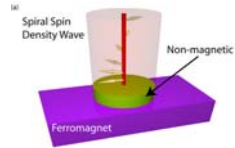




Motivation

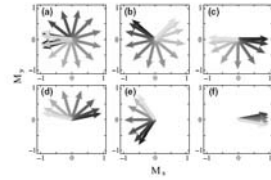
Bulk Spin Transfer Torque

- PRL 96, 256601 (2006)
- PRB 79, 104433 (2009)



Exotic Phase

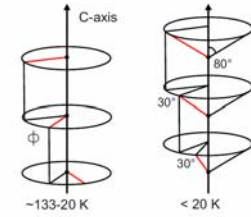
- PRB 78, 020402(R) (2008)
- PRB 79, 134420 (2009)



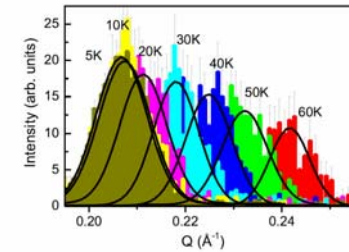
$$|f| = (1/2\pi Jh) \sqrt{(C_{23}j_3)^2 - (6K_0^6)^2}$$

- Spin-triplet SC Science, 329, 59 (2010)
- RMP 77 1321 (2005)

Magnetic Spirals in Holmium thin films

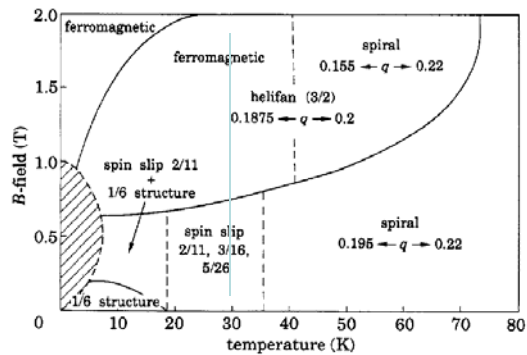


The representative magnetic structure of bulk Ho below its Néel temperature (left) and below its Curie temperature (right).



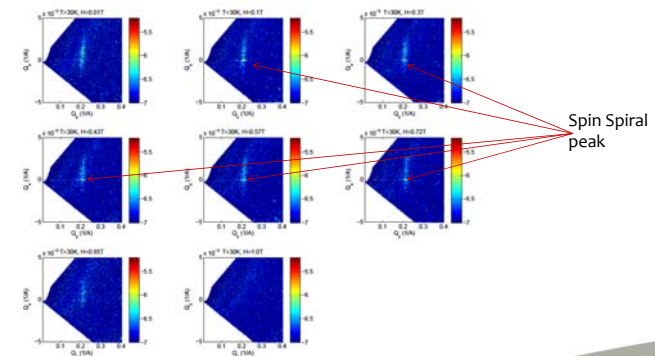
The momentum transfer versus the intensity of the magnetic diffraction peak for the 50 nm thick Ho sample as a function of temperature.

Bulk Ho Field Phase diagram



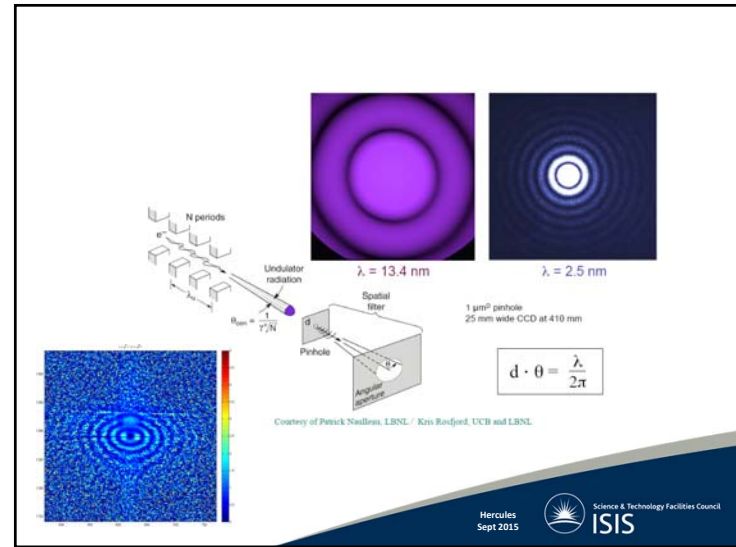
D. A Jehan et al., Europhys. Letts. 17 553 (1992)

Polref: Diffuse scattering from a holmium spin spiral: 7.5T



J. D. S. Witt et al. J Phys: Condens Matter 23, 416006 (2011).

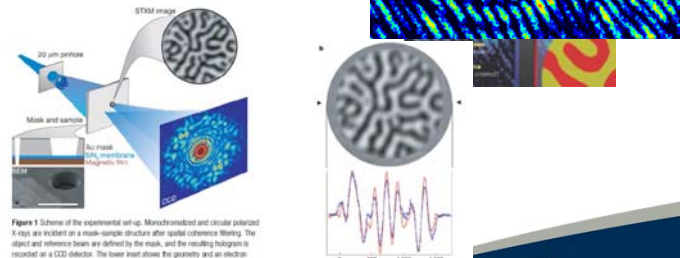
COHERENCE



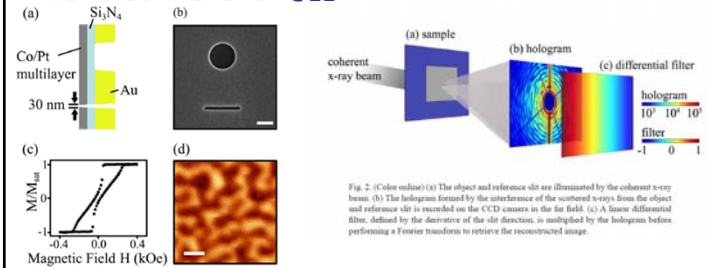
Lensless imaging of magnetic nanostructures by X-ray spectro-holography

S. Eisele¹, J. Lüthy², W. F. Schottler³, M. Lürsen⁴, O. Hellmuth⁵, W. Eberhardt¹ & J. Stöhr⁶

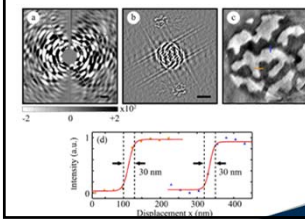
¹HEIST, Albert-Ludwigs-Strasse 15, D-7800 Berlin, Germany
²SLAC, Stanford Linear Accelerator Center, 2575 Sand Hill Road, Menlo Park, California 94025, USA
³Department of Applied Physics, 110 Via Puelio Strad, Stanford University, Stanford, California 94305, USA
⁴San Jose Research Center, Hitachi Global Storage Technologies, 650 Harry Road, San Jose, California 95128, USA

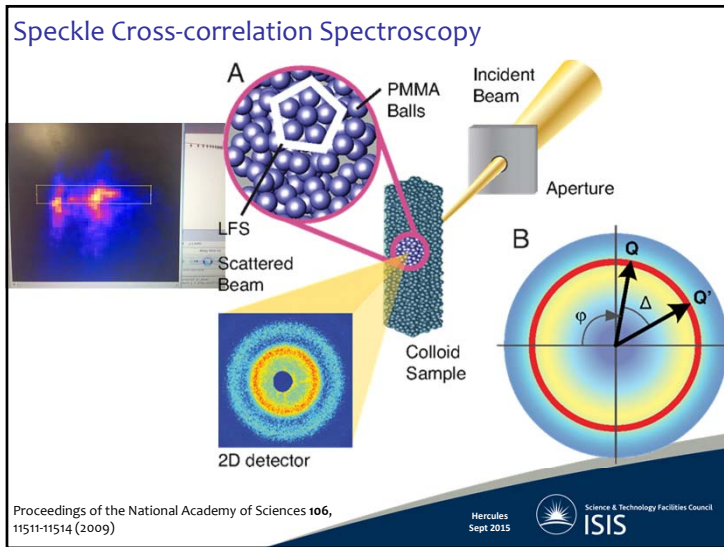


Extended References



- Duckworth, SL et al. Vol. 19, Optics Express 16223 (2011)
- Duckworth, SL et al. New J. Phys. 15 23045 (2013)



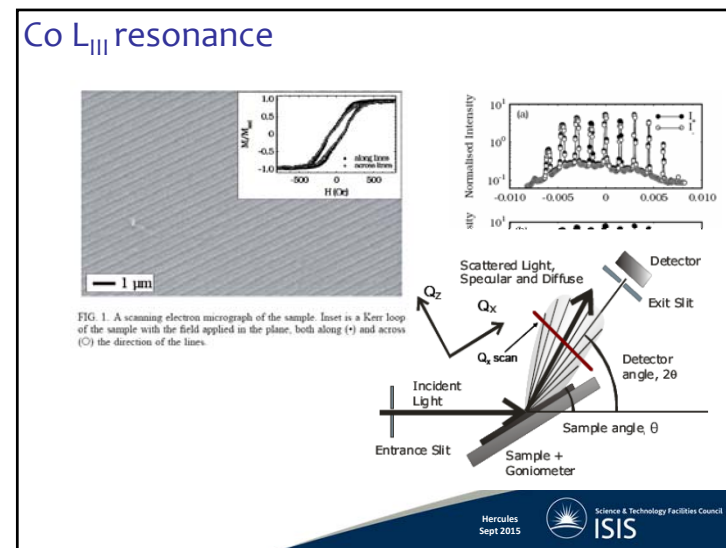
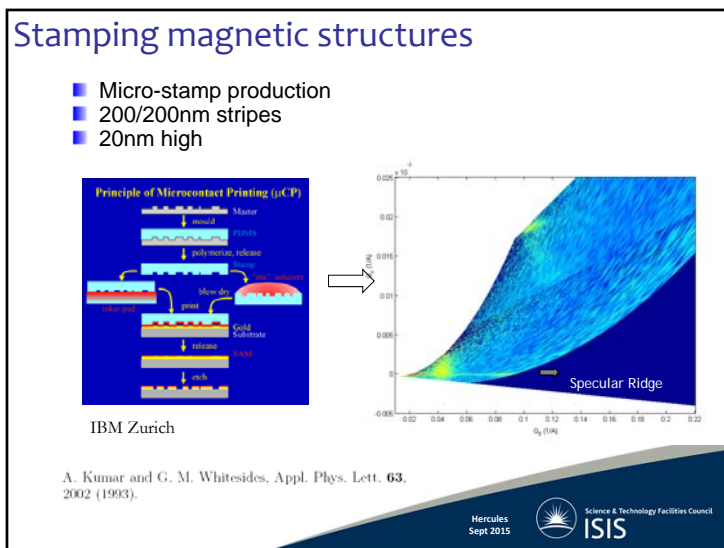


OxfordMaterials

Stamping magnetic structures:
micro-contact printing

NLSL

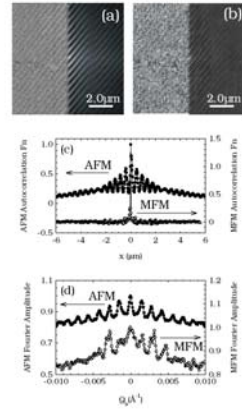
Hercules Sept 2015 Science & Technology Facilities Council **ISIS**



AFM/MFM Measurements

- MFM weak
 - Measured @ remanence
- Xrms measures structure-charge correlation

$$\propto \text{Re}(\bar{f}_m \cdot \bar{f}_s)$$
- MFM sensitive to force gradients
- s-xrms magnetisation
- Magnetism tracks gross features

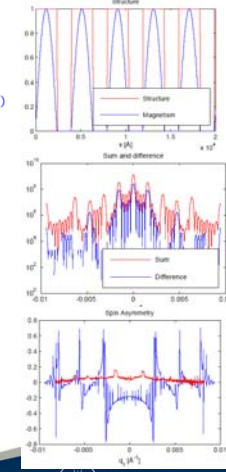
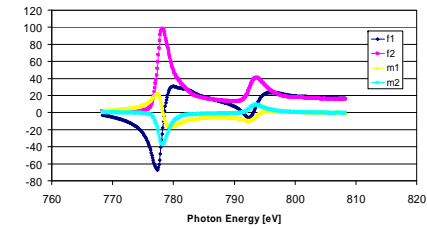


L-A Michez et al. APL **86** 112502 (2005)

Spin Asymmetry

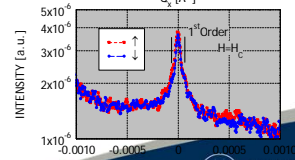
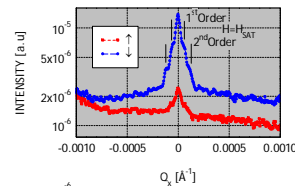
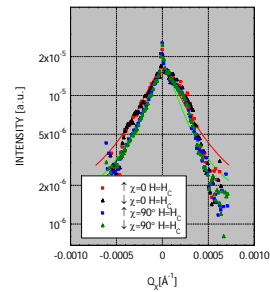
$$f_n^{res} = \begin{pmatrix} 1 & 0 \\ 0 & k_i \cdot k_f \end{pmatrix} F_n^{(0)} + i \begin{pmatrix} 0 & -k_i \cdot M_n \\ k_f \cdot M_n & (k_i \times k_f) \cdot M_n \end{pmatrix} F_n^{(1)}$$

$$I = \text{Tr}(f_n^{res} \cdot \rho f_n^{res*})$$



5 – 10 μm Stripes

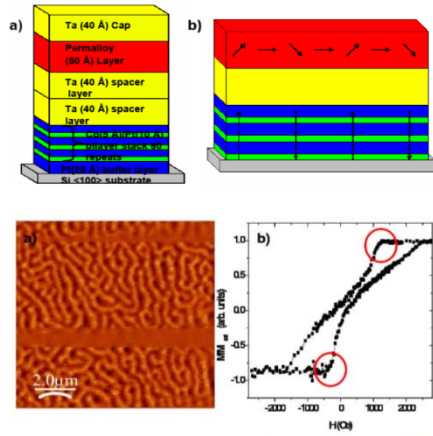
- H = H_c
- Domain size
 - χ = 0° (||) 3.5 μm
 - χ = 90° (⊥) 4.4 μm
- Domain Distribution
 - ~3rad
- No Magnetic Roughness
- H = H_{Sat}
- H || Stripes
- Integer order satellites
- Magnetic Roughness



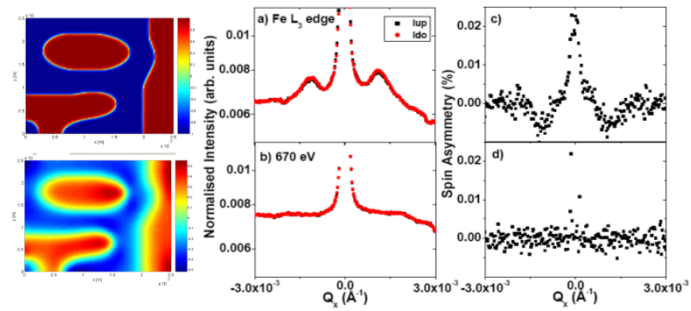
Structurally smooth- Magnetically Rough
out of plane anisotropy



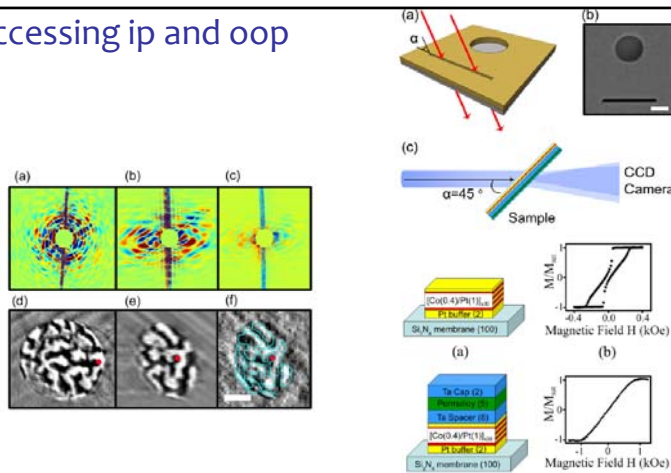
Out of plane Anisotropy



S-XRMS



Accessing ip and oop

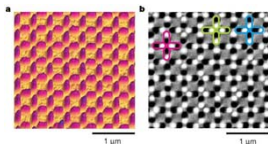
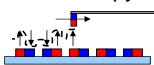


Conclusions

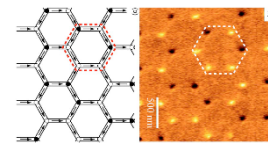
- **Diffuse Measurements**
 - Direct (simple) scattering cross-section allowing the quantitative analysis of the magnetic disorder
 - Statistical analysis of magnetic surface morphology not possible with other techniques
- **Dynospheres**
 - Increased coercivity
 - Patterning geometry does not affect AFM domain size
 - Magnetic roughness driven by the structural periodicity
 - ◆ cf. Smooth structural surfaces where the magnetic roughness is on a longer lengthscale than the structural in-plane correlation length
 - ◆ Free interfacial magnetisation
 - ◆ Fluctuation of surface moments
- **Stripes**
 - Domain size and disorder relatively independent of field direction
 - Magnetic roughness driven by the PMMA roughness
- **Proximity patterning**
 - Good qualitative agreement
 - Requires the neutron measurement
- **Ongoing/future Measurements**
 - Micromagnetic simulation of sphere/stripe structure
 - Dwba analysis

Artificial Spin Ice

- 2D array of single domain, ferromagnetic nanobars
- Ising axes defined by shape anisotropy
- Taylor lattice geometry to enforce frustration
- Square and kagome vertex systems analogous to crystalline "spin ice" materials
 - Frustrated bulk magnet
 - Coulombic magnetic "monopole" excitations
- Athermal ($T_{active} \sim 10^5 K$), therefore, field-ordering currently main focus
- Magnetic microscopy – vertex statistics



Wang, R. F. et al. Nature 439, 303–306 (2006).



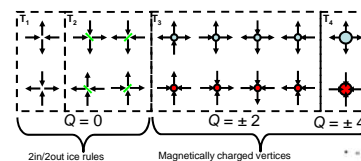
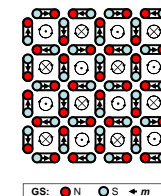
Tanaka et al. PRL 73, 052411 (2006).



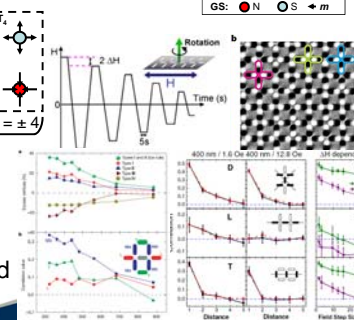
Jennel, T. et al. Physical Review B 70, 134408 (2004)
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Magnetic Square Ice

- $2^4 = 16$ possible vertex configurations
 - 4 vertex types
- 2-fold degenerate ground-state (GS)
- Studied statistically following applied fields

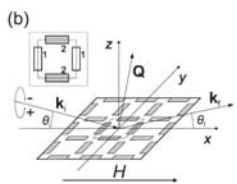
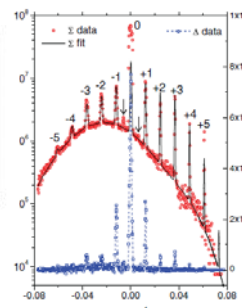
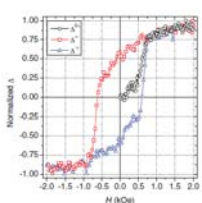
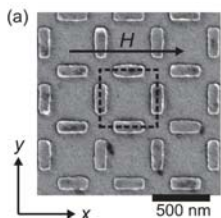


- ac demagnetization
- Energy minimisation
- Only short range correlated states accessed



Wang, R. F. et al. (2006) Nature 439, 303–306
Ke et al. (2008) PRL 101, 037205

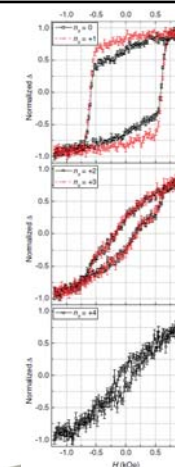
Asi



$$\sum(Q_x) = C \sum_{q_x, q_y} (|F(q_x, q_y)|^2 R(Q_x; q_x, q_y))$$

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- Superposition of loops
- Advantages over Bragg-MOKE
- Opportunities to look at thermal systems



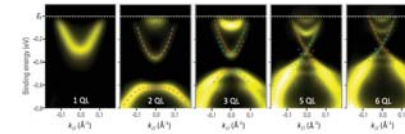
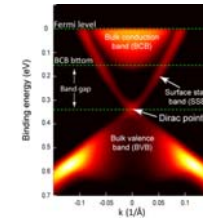
AIP Advances 2, 022163 (2012)

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QUANTUM MATTER

QAHE & Topological Insulators (TIs)

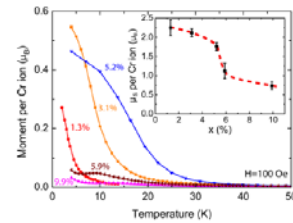
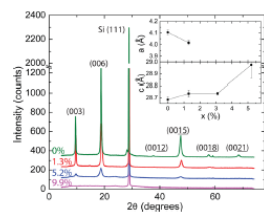
- Quantum Hall effect observed in 2DEG
 - $\sigma = \nu \frac{e^2}{h}$
- Anomalous Quantum Hall effect (QAHE)
 - Band inversion
 - Ferromagnetic insulator breaking TRS
- TIs possess insulating bulk gap and gapless edge states
 - Magnetic dopants



Yu et al. *Science* **329** (2010) 61
Chang et al. *Science* **3340** (2013) 167
He, K., Wang et al. *Natl. Sci. Rev.* **1**, 38–48 (2013)

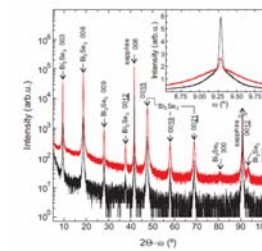
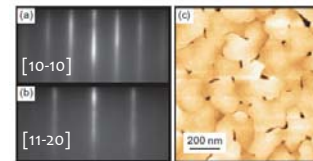
Cr doped Bi_2Se_3

- $T_c \sim 20\text{K}$ for 5%
- Good crystalline quality $\sim <5\%$

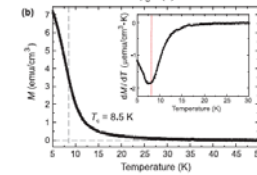
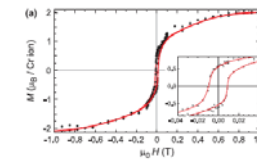


Haazen et al. *APL* **100** (2012) 082404

$(\text{Cr}_{0.12}\text{Bi}_{0.88})_2\text{Se}_3$



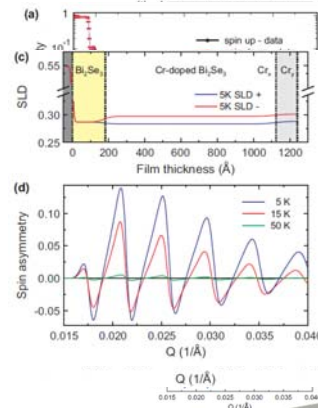
$X = 0.12$



- Cr concentration fixed by RBS
- $1.5\mu_B/\text{Cr}$ @ 0.7T
- No enhancement observed in the near surface region
- Moment less than $3.78\mu_B$ of Cr^{3+} substituted on Bi site

■ **Summary**

- Cr doping up to 12% without a loss of crystallinity (substitutional doping)
- Moment lower (2.1) than expected for Cr^{3+}
- Homogeneous ferromagnetism
- Precursor to observing the QAHE (lower temperatures)



Collins-McIntyre, L. J. et al. Magnetic ordering in Cr-doped Bi_2Se_3 thin films. *Europhys. Lett.* **107**, 57009 (2014).



Summary

- Can take advantage of (i.e. control) the refractive index (polarised neutrons, deuteration, resonant enhancement and isotopic substitution)
- Can extract magnetic structures
- Realistic sample environments
 - Time resolution
 - Speckle
- Sub nm resolution for systems
- Lengthscales (out of plane) monolayer to $\sim 100\text{nm}$



Magnetism Summary

■ **Neutrons**

- see the B-field
- Strong scattering
- See component perpendicular to Q
- Penetrating
- Polarisation dependence
- Simple cross-section

■ **Photons**

- Weak scattering (non-resonant)
- Separation of orbital and spin
- Large resonant enhancements
- Polarisation dependence
- Depth selectivity
- Complex (rich) cross-section

} Highly complementary probe for nanoscale magnetism



References

- Polarized Neutrons, W.G. Williams, Oxford (1988)
- Theory of Magnetic neutron and photon scattering, E. Balcar & S.W. Lovesey, Oxford (1988)
- Introduction to Thermal Neutron Scattering, G.L. Squires, Cambridge (1978)
- Elements of Modern X-Ray Physics, Als-Nielsen and McMorrow, Wiley & Sons (2001)
- Magnetism: from fundamentals to nanoscale dynamics, Stohr and Siegmann, Springer (2006)
- www.ill.eu
- www.isis.stfc.ac.uk
- www.esrf.eu
- www.diamond.ac.uk



Review Articles

Report on Progress

X-rays and magnetism

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