Dzyaloshinkii-Moriya interactions and chiral magnetism (in B20 epilayers and) at ferromagnet/heavy metal interfaces

Christopher Marrows

c.h.marrow@leeds.ac.uk
@ChrisMarrows
Acknowledgements


M. D. Robertson – Acadia University, Canada

A. Dobrynin – Diamond Light Source, UK

S. McVitie, M.-J. Benitez & D. McGrouther – University of Glasgow, UK

C. J. Kinane, T. R. Charlton, and S. Langridge – STFC Rutherford Appleton Laboratory, UK

Funding gratefully received from:
Interesting phenomena in magnetism

Magnetic Properties
- Magnetocrystalline anisotropy
- Dzyaloshinkii-Moriya interaction

Magneto-Optical Properties
- Magnetooptic Kerr effect
- X-ray magnetic circular dichroism

Magnetotransport Properties
- Anisotropic magnetoresistance
- Anomalous Hall effect
- Spin Hall effect
- Tunnelling anisotropic magnetoresistance
Dzyaloshinskii-Moriya Interaction requires structural inversion asymmetry + SOC

\[ E_{DM} = D \cdot S_1 \times S_2 \]

monolayers of metal on heavy element substrate

Asymmetric layers (with different SOC) around FM break inversion symmetry

W(110)/Fe(2 ML)
Meckler PRL 103, 157201 (2009)

Cu(100)/Fe/Ni
Chen PRL 110, 177204 (2013)

Crystal lacks inversion symmetry e.g. B20 unit cell


Bulk DMI: B20 systems
B20 bulk crystal phases

Mühlbauer et al., Science 323, 915 (2009)
Wilson, Thesis (2013)
Chiral Magnetic Skyrmions

Topologically stable vector field object
“Combed hedgehog”

Emergent electrodynamics arising from Berry phase
Each skyrmion = \( \varphi_0 \) of fictitious magnetic flux
Moving skyrmions => effective electric field

Tony Skyrme FRS

Fe_{0.5}Co_{0.5}Si - Yu Nature (2010)
B20 alloys

transitional metal monosilicides

FeSi - paramagnetic narrow gap semiconductor

Fe$_{1-x}$Co$_x$Si - helimagnetic doped semiconductor

MnSi - helimagnetic metal

CoSi - diamagnetic metal

transition metal monogermanides

FeGe - 'high' temp. helimagnetic metal

MnGe - short period helimagnetic metal

Wilhelm et al., PRL 107, 127203 (2011)
Kanazawa et al., PRL 106, 156603 (2011)
Sputtered FeGe XRD and magnetometry

FeGe co-sputtered at ~470 °C in Ar:H₂(4%) at 3 mTorr textured films in (111) orientation

High ordering temperature: 276 K

B20 helimagnets under field

**Helimagnet in an applied field**

- For a magnetic field applied parallel to $Q$, a conical phase forms.
- In a bulk crystal if H is not parallel to Q, Q will align to the field.
- In a thin film there is a uniaxial anisotropy that fixes the direction of the helix.
- An *in-plane* applied field distorts the helix shape into a helicoid.

---

Interference fringes produced from reflections and reflections within film

Shift due to different polarisations of neutrons experiencing a different scattering potential

Structure parameters determined above $T_c$ and kept constant (e.g. scattering length density, film thickness, etc.)

A helicoid profile is used to generate the magnetic scattering length density and a fit is performed.

Helicoid structure:
- $M$ is the magnetic moment
- $y$ – depth of film
- $\lambda$ is the helix wavelength
- $\phi_0$ is a fitting parameter

Specular reflection

$$ q_z = \frac{4\pi \sin \theta}{\lambda} $$


FeGe Results

In-plane magnetic field

1. $T = 50$ K

2. $T = 50$ K

3. $T = 50$ K

$t = 69 \pm 1$ nm

Bulk $\lambda \sim 70$ nm, from fit $\lambda = 70 \pm 5$ nm
FeGe and FeGe/Fe Results

FeGe

FeGe/Fe

T = 50 K

arXiv:1506.01575
Transition Metal Substitution: Previous Doping Studies

- Ordering temperature – Blue
- Ratio of ordering temperature to helix wavelength - Red

Previous study in Mn$_{1-x}$Fe$_x$Ge

B20 helimagnetic material

Change in skyrmion chirality

Divergence in helical wavelength found $x \sim 0.8$

What happens when you add Co?

MnGe $\rightarrow$ FeGe $\rightarrow$ CoGe

adding electrons

Fe\(_{1-x}\)Co\(_x\)Ge Characteristics

- Samples grown by molecular beam epitaxy
- Si(111) wafer for lattice matching
- Growth along the (111) zone axis
- Lattice matched with a 30° in-plane rotation

Low-energy electron diffraction patterns

100 eV
x = 0

Si (7 x 7)

FeGe (111)

\(\alpha \sim 0.1-0.03\% < \text{Bulk}\)
x = 0 to 1

~10% larger than bulk

a) Lattice constant

b) \(M_{\text{sat}}\)

c) Ordering temperature
Fe\textsubscript{1-x}Co\textsubscript{x}Ge PNR Results

- Helix wavelength $\lambda$ increases with Co content
- Both samples less than one period
- Only a lower bound of 170 nm could be set for $x = 0.54$

<table>
<thead>
<tr>
<th>$x$</th>
<th>$\lambda$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>70 ± 5</td>
</tr>
<tr>
<td>0.36</td>
<td>115 ± 15</td>
</tr>
<tr>
<td>0.54</td>
<td>&gt;170</td>
</tr>
</tbody>
</table>
**Fe$_{1-x}$Co$_x$Ge Results**

**Graphs and Data**

- **Graph a)**: Comparison of $T_c$ (K) vs. $\lambda$ (nm) for different compositions.
- **Graph b)**: Helical wavelength ($\lambda$) vs. concentration $x$ for MnGe, FeGe, and CoGe, with symbols indicating different compositions.
- **Graph c)**: Plot of $T_c$ (K) vs. $\lambda$ (nm) for FeGe and CoGe.
- **Graph d)**: Plot of $\lambda$ vs. $x$ for MnGe, FeGe, and CoGe.

**Tables**

<table>
<thead>
<tr>
<th>$x$</th>
<th>$\lambda$ (nm)</th>
<th>$T_c \lambda^{-1}$ (K nm$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>70 ± 5</td>
<td>-3.9</td>
</tr>
<tr>
<td>0.36</td>
<td>115 ± 15</td>
<td>-2.0</td>
</tr>
<tr>
<td>0.54</td>
<td>&gt; 170</td>
<td>&gt; -0.8</td>
</tr>
</tbody>
</table>

**Equations**

$\lambda \propto J/D$  
$T_c \propto J$

**Text**

- Keeping negative ratio in-line with previous study
- Possible divergence again in helical wavelength

---

FeGe - magnetoresistance

Metallic $\rho(T)$ behaviour, with no sign of cusp at magnetic ordering temperature.

In out-of-plane field see negative magnetoresistance (MR) up to $H_c$.

Low-field MR is indicative of saturation of the conical phase.

GMR-like mechanism: generalised Levy-Zhang model for DW resistance.

FeGe – Hall effect

- OHE linear up to about 200 K
- Can be fitted by 2-band model
- AHE peaks at about 200 K

\[ \rho_{xy}^a = \left[ \alpha \rho_{xx} + \beta \rho_{xx}^2 + b \rho_{xx}^2 \right] \mu_0 M(H) \]

- Quadratic scaling with \( \rho_{xx} \)
  => Intrinsic or side-jump?
FeGe – anomalous Hall effect

Topological Hall effect

- as an electron moves adiabatically through spatial varying spin topography its spin orientation maps the magnetisation:
  \[ \hat{n}(\mathbf{r}, t) = \mathbf{M}/|\mathbf{M}| \]
- electrons gain Berry phase as they traverse skyrmion.

- Berry phase \( \propto \sin \theta (\partial_x \theta \partial_y \phi - \partial_y \theta \partial_x \phi) \)
- Considering Berry phase as an AB phase in an emergent magnetic field, we expect a Hall effect
- one quantum \( \Phi_0 \) of emergent flux per skyrmion.

T. Schultz et al., Nature Physics, 8, 301 (2012)
Total Hall effect

\[ q_{yx}(H) = q_{yx}^o(H) \]

ordinary

\[ Q_{yx}^o = \frac{1}{ne} \]

Hall bars patterned by photolithography, hard mask, ion milling.

Our devices
Total Hall effect

\[ q_{yx}(H) = q_{yx}^o(H) + q_{yx}^e(H) \]

- Ordinary: \( q_{yx}^o \)
- Extraordinary/Anomalous: \( q_{yx}^e \)

\( q_{yx}^o \)simply \( \frac{1}{ne} \)

• Deflection of carriers due to magnetic material:
  - EHE often a larger than the OHE
  - Arising from spin orbit coupling
Total Hall effect

\[ \rho_{yx}(H) = \rho_{yx}^o(H) + \rho_{yx}^e(H) + \rho_{yx}^T(H) \]

- ordinary
- extraordinary/anomalous
- topological
FeGe – topological Hall effect

our 80 nm film: THE over full temperature range

Huang et al. - Johns Hopkins


Huang et al. PRL, 108, 267201 (2012)
Giant THE in Fe$_{0.7}$Co$_{0.3}$Si

Combined two techniques:
1. 4 wire Hall measurements
2. SQUID-VSM magnetometry

\[ \eta_y(H) - \eta_y^T(H) = aM(H) + bH \]

by scaling the magnetisation to fit the Hall data one can extract the THE as the difference.

Largest THE to date: 820 n\(\Omega\)cm.

Useful for electrical detection of skyrmions?

\(J = 4 \times 10^8\) A/m$^2$ (results insensitive to J down to $2 \times 10^4$ A/m$^2$)
Prior THE measurements

Discovered in MnSi – few nΩcm

~200 nΩcm in MnGe: 2 nm skyrmions


Recent comprehensive study by Ritz et al. in MnSi – up to 50 nΩcm


Giant generic topological Hall resistivity of MnSi under pressure

R. Ritz, M. Halder, C. Franz, A. Bauer, M. Wagner, R. Bamler, A. Rosch, and C. Pfleiderer
Technische Universität München, Physik-Department E21, D-85748 Garching, Germany
Institute of Theoretical Physics, Universität zu Köln, D-50937 Köln, Germany
(Received 13 November 2012; published 29 April 2013)
THE isotherms =>
Skyrmion phase diagram

THE shows two contributions:
• Broad, weakly hysteretic background (few 100 mT)
• Sharp, hysteretic extremum (~50 mT)
why is the effect so large?

\[
\rho_{xy}^T = nPR_0B_{\text{eff}}
\]

\[
B_{\text{eff}} \sim \frac{4\phi_0}{\sqrt{3}a^2}
\]

Huang et al. PRL, 108, 267201 (2012)
Giant THE

high spin polarization $\rightarrow$ 0.77

spin polarization of the current

ordinary Hall coefficient

relative skyrmion density ($\sim$1 if dense)

emergent gauge (one flux quanta)

skyrmion separation

$$\rho_{xy}^T = nPR_0B_{\text{eff}}$$

$$B_{\text{eff}} \sim \frac{4\phi_0}{\sqrt{3}a^2}$$

Huang et al. PRL, 108, 267201 (2012)
Giant THE

high spin polarization $\rightarrow 0.77$

spin polarization of the current

Emergent gauge
(one flux quanta)

ordinary Hall coefficient

doped semiconductor

$x = 0.30$, $n \sim 1 \times 10^{22} \text{ cm}^{-3}$

relative skyrmion density
(~1 if dense)

$doped semiconductor$

file, $x = 0.3$

$Q_{xy}^T = nPR_0 B_{\text{eff}}$

$B_{\text{eff}} \sim \frac{4\phi_0}{\sqrt{3}a^2}$

$\rho_{xy}^T (\mu\Omega \cdot \text{cm})$

$\mu_0 H (\text{mT})$

$\rho_{xy}^T (\mu\Omega \cdot \text{cm})$

$\rho_{xy}^T (\mu\Omega \cdot \text{cm})$

$\rho_{xy}^T (\mu\Omega \cdot \text{cm})$

$\rho_{xy}^T (\mu\Omega \cdot \text{cm})$

Huang et al. PRL, 108, 267201 (2012)
Giant THE

high spin polarization → 0.77

relative skyrmion density (~1 if dense)

ordinary Hall coefficient

low carrier concentration (high Hall coefficient)

doped semiconductor
x = 0.30,
n ~ 1 \times 10^{22} \text{ cm}^{-3}

emergent gauge (one flux quanta)

2λ / \sqrt{3} = 11 \pm 1 \text{ nm}
strain reduces separation relative to bulk (48 nm*)

\rho_{xy}^T = nPR_0B_{eff}

Huang et al. PRL, 108, 267201 (2012)
Giant THE

Skyrmion separation

Ordinary Hall coefficient

Relative skyrmion density (~1 if dense)

calculate: n \sim 0.2

High spin polarization \rightarrow 0.77

Spin polarization of the current

Low carrier concentration (high Hall coefficient)

Doped semiconductor

x = 0.30,
n \sim 1 \times 10^{22} \text{ cm}^{-3}

Emergent gauge

(one flux quanta)

2\phi_0 / \sqrt{3} = 11 \pm 1 \text{ nm}

Strain reduces separation relative to bulk (48 nm*)

Skyrmion separation

Effective magnetic field

B_{\text{eff}} \sim \frac{4 \phi_0}{\sqrt{3} a^2}

Huang et al. PRL, 108, 267201 (2012)


Topological contribution to $\rho_{xx}$?

Hysteretic dips in $\rho_{xx}$ coincide with THE peaks (after linear and WL background subtraction)

Only present below $\sim 20$ K

Peaks in $\rho_{xx}$ predicted by Monte Carlo simulations — Yi et al. Phys. Rev. B 80, 054416 (2009).
Interface DMI: multilayers
4 DW configurations in PMA films

- 2 possible magnetization re-orientation
  - perpendicular to the magnetization plane (Bloch)
  - in the magnetization plane (Néel)

- 2 possible chiralities for each

- Bloch wall state has lower magnetostatic energy

\[ \Delta = \sqrt{\frac{A}{K}} \]
Imaging of chiral DWs

Spin polarized STM

Kubetzka et al., PRB (2003)

Lorentz-TEM

Yu et al., Nature (2010)

SPLEEM

Chen et al., Nature Comm. (2013)

NV center

Tetienne et al., Science (2014)
• Investigated Ta(3.2)\Pt(3)\Co(0.8)\AlO_\text{x}(5.3) films

• JEOL TEM
• Fresnel mode at 100kV
• Defocus microscope by $\Delta z$ to reveal contrast

$$\beta_L(x) = \frac{e\lambda}{h} \int_{-\infty}^{\infty} B_y(x, z)dz$$
Calculated Contrast

- No contrast observed for Néel wall at normal incidence.
- Tilting the sample reveals contrast.
- For a Bloch wall contrast variation is always observed.

Bloch wall

Néel wall
Experimental contrast

- $B = 0 \, T; \, \theta = 0^\circ$

- No contrast variation is observed at $\theta=0^\circ$
- DWs are observable at $\theta=30^\circ$
- Symmetric linetraces

✓ Néel walls
• L-TEM

• Domain growth until DW meet each other and form a 360° structure

• Néel type DW confirmed in both directions

• In case of Néel wall magnetostatic charges are created on either side of DW
• Rigidity of the Néel wall is locked by the DMI
• To annihilate the two walls this topological barrier must be overcome
• Annihilation is a measure of the DMI
DW annihilation: polar Kerr microscopy
- Measurement of annihilation field
- Problem reproduced by micromagnetic simulations
- $D = 0.35 \pm 0.05 \text{ mJ/m}^2$
- Value is artificially low (no thermal activation in model, assume perfect material)
Materials

- Epitaxially grown materials, studied in-situ

Kubetzka et al, PRB (2003)  W\Fe

- Pt(111)\Ni  →  Ir(111)\Ni  shows opposite DMI

- Do Pt\Co\Ir layers have larger DMI? Can we enhance the effective DMI?
Film characterization

- Films grown by DC sputtering
- Kerr microscopy used to measure hysteresis loops
- Out-of-plane anisotropy measured by SQUID/VSM → no significant change with inserted Ir layer
Experimental setup

- DW displacement measured by Kerr microscopy
- Differential mode employed
- Displacement radially symmetric in case of out-of-plane field
- Strong asymmetry with presence of in-plane field
• Huge asymmetry in Pt\Co\Pt
• 2.3Å of Ir lifts the asymmetry
• 4.6Å of Ir reverses the asymmetry
• Different contribution from Co\Pt and Co\Ir!
Creep regime

- Thermally activated creep regime in general
  \[ \nu = \nu_0 \exp \left( -\zeta H_z^{-\mu} \right) \quad \mu = 1/4 \]
  - Where the scaling \( \zeta \) parameter is field-dependent
    \[ \zeta = \zeta_0 \sqrt[4]{\frac{\sigma_{DW}(H_x)}{\sigma_{DW}(0)}} \]

- DW energy density

Transition from Bloch wall to Néel wall

\[ \sigma_{DW}(H_x) = \sigma_0 - \frac{\pi^2 \Delta M_s^2}{4K_D} (H_x + H_{DMI})^2 \]

Néel wall

\[ \sigma_{DW}(H_x) = \sigma_0 + 2K_D\Delta - \pi \Delta M_s |H_x + H_{DMI}| \]

Simulations

Modelling:
creep law including DMI fields

- Model well reproduces experimental data
- D-M constant obtained by using $D = \mu_0 H_{\text{DMI}} M_s \Delta$
- Bloch–Néel wall transition $\frac{4K_D}{\pi M_s} \simeq 18 \text{ mT}$
- DMI changes sign around Pt\Co\Ir(2.5Å)

Why do we observe DMI in symmetric stack?

- Epitaxial sample grown by sputtering @ >150°C on Al₂O₃

Ta\Pt\Co\Pt stack must not be symmetric!
Crystallographic order is extremely important!

Mihai et al, APL (2013)
Towards exotic textures

Non-homogeneous ground state

- DW energy \( E = 4\sqrt{AK} \mp \pi D \)

\[ D_{\text{crit}} \sim 3 \text{ mJ/m}^2 \]

Isolated skyrmions

\[ D < D_{\text{crit}} \]

Pt/Co/Ta

Woo et al. arXiv:1502.07376

Ta/CoFeB/TaOx


\{Pt/Co/Ir\} \times N

Conclusions

• examined scattering mechanisms and observed conical phase MR and THE in textured FeGe

• evidence for chiral magnetic structure in PNR and a giant THE in Fe$_{1-x}$Co$_x$Si.
  Porter et al., arXiv:1312.1722 [cond-mat.mes-hall]

• topological protection of homochiral walls in Pt/Co/AlO$_x$
  Benitez et al., arXiv:1503.07668 [cond-mat.mtrl-sci]

• interface engineering of DMI in Pt/Co/Ir/Pt