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Understanding the magnetic ground states for improper multiferroic materials

Jason T. Haraldsen

Theoretical Division and Center for Integrated Nanotechnologies
Los Alamos National Laboratory
Los Alamos, New Mexico 87544, USA

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Associated Publications

- J.T. Haraldsen** *et al.*, *Phys. Rev. Lett.* **102**, 237204 (2009)
M. Swanson, **J.T. Haraldsen**, and R.S. Fishman, *Phys. Rev. B* **79**, 184413 (2009)
J.T. Haraldsen and R.S. Fishman, *J. Phys.: Condens. Matter* **21**, 216001 (2009)
J.T. Haraldsen *et al.*, *Phys. Rev. B* **82**, 020404(R) (2010)
J.T. Haraldsen and R.S. Fishman, *Phys. Rev. B* **82**, 144441 (2010)
R.S. Fishman and **J.T. Haraldsen**, *J. Appl. Phys.* **109**, 07E117 (2011)
M. Frontzek, **J.T. Haraldsen** *et al.*, *Phys. Rev. B* **84**, 094448 (2011)
R.S. Fishman, G. Brown, and **J.T. Haraldsen**, *Phys. Rev. B* **85**, 020405(R) (2012)
T. Nakajima, S. Mitsuda, **J.T. Haraldsen** *et al.*, *Phys. Rev. B* **85**, 144405 (2012)
J.T. Haraldsen, R.S. Fishman, and G. Brown, *Phys. Rev. B* **86**, 024412 (2012)
F. Ye, R.S. Fishman, **J.T. Haraldsen** *et al.*, *J. Appl. Phys.* **111**, 07E137 (2012)
R.S. Fishman, N. Furukawa, **J.T. Haraldsen** *et al.*, *Phys. Rev. B* **86**, 220402(R) (2012)
J.T. Haraldsen and A.V. Balatsky, *Mater. Res. Lett.* **1**, 39 (2013)
R.S. Fishman, **J.T. Haraldsen** *et al.*, Accepted *Phys. Rev. B* (2013)

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ENERGY

A.V. Balatsky – LANL/T-4/CINT

R.S. Fishman – ORNL/MSTD

G. Alvarez – ORNL/CNMS

G. Brown – Florida State University

S.A. Trugman – LANL/T-4/CINT

M. Graf – LANL/T-4

J.-X. Zhu – LANL/T-4/CINT

P. Wölfle - Karlsruhe Institute of Technology – Germany

R.M. Fernandes – University of Minnesota

M. Klintenberg – Uppsala University – Sweden

Experimental

F. Ye – ORNL/NSSD

J.A. Fernandez-Baca – ORNL/University of Tennessee

M.R. Fitzsimmons – LANL

R. Remesh – University of California Berkeley/LBNL

J. Musfeldt – University of Tennessee

M. Frontzek – ORNL/NSSD

A. Podlesnyak – ORNL/NSSD

T. Nakajima – University of Tokyo – Japan

K. Kimura – Osaka University – Japan

T. Kimura – Osaka University – Japan

N.J. Curro – University of California, Davis

Q.X. Jia – LANL/CINT

D. Yarotski – LANL/CINT

Outline

- Introduction to multiferroics
 - Why Multiferroics?
 - Ferromagnetism/Ferroelectricity
 - Types of multiferroicity
- CuFeO_2 – Improper multiferroic
 - Magnetic phases and doping effects
 - Inelastic neutron scattering data
- Rotational algorithm for magnetic systems
 - Non-collinear rotation
 - Calculation of Spin Dynamics
- Modeling the dynamics of Ga-doped CuFeO_2
 - 3-D frustrated antiferromagnet
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Why Multiferroics?

Advancement of technology through the combination of magnetic ordering and electric polarization

Magnetoelectric Coupling

Bulk Multiferroics

- Magnetic Field Sensors
- Tunable microwave devices
- Electric field tunability

Composite Multiferroics

- Spintronic devices
- Tunneling magnetoresistance
- Solid state memory devices

Ferromagnets

A ferromagnet develops a spontaneous magnetization of the atomic spins in the absence of an external magnetic field.

Image from <http://www.seagate.com>

Antiferromagnets

An antiferromagnet develops a distinct magnetic order with no net magnetization.

Collinear spins

Noncollinear spins

Ferroelectrics

Electric polarization: $\mathbf{P} = \sum_i \mu_i$

A ferroelectric develops a spontaneous electric polarization in the absence of an external electric field.

Applications:

Ferroelectric RAM
Ferroelectric capacitors
Medical ultrasound sensors
Infrared cameras
Camera flashes

“Proper” ferroelectrics like BaTiO_3 were first discovered in 1920.

Ginzburg-Landau

$$F_{GL} = \alpha P^2 + \beta P^4$$

Multiferroicity

A material that has both ferroelectric and ferromagnetic properties is called multiferroic. These materials are important because charge can be controlled by a magnetic field (**H**) or magnetism can be controlled by an electric field (**E**). Potentially, multiferroic materials have wide-ranging technological applications.

In “**proper**” multiferroics, the coupling between magnetism and ferroelectricity is **weak**.

Multiferroicity

A material that has both ferroelectric and ferromagnetic properties is called multiferroic. These materials are important because charge can be controlled by a magnetic field (**H**) or magnetism can be controlled by an electric field (**E**). Potentially, multiferroic materials have wide-ranging technological applications.

In “**improper**” multiferroics, the electric polarization is associated with a non-collinear spin state (the individual moments do not all lie along the same axis). The coupling between magnetism and ferroelectricity is **strong**.

Coupling Mechanisms for Improper Multiferroics

1. Inverse Dzyaloshinskii-Moriya interaction (Balatsky *et al.*, Mostovoy *et al.*):

ordering vector perpendicular to polarization

Spiral ordering produces lattice asymmetry that creates an electric polarization.

2. Atomic symmetry breaking involving the non-collinear spins of neighboring transition-metal ions and a central oxygen atom (Jia *et al.*, Arima *et al.*):

ordering vector parallel to polarization

Spiral ordering produces a hybridization of orbitals.

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CuFeO₂ – A Frustrated Antiferromagnet

CuFeO₂ is hexagonal structure that consists of $S = 5/2$ Fe³⁺ ions.

Multiple Competing interactions.

Kimura *et al.* PRB 73, 220401 (2006)

Magnetic field induces electric polarization

Associated with non-collinear spin configuration

"Proper" Spiral

Nakajima *et al.*, JPSJ 76, 043709 (2007)

Phase Diagram - $\text{CuFe}_{1-x}\text{Al}_x\text{O}_2$ and $\text{CuFe}_{1-y}\text{Ga}_y\text{O}_2$

Kimura *et al.* PRB 73, 220401 (2006)

CuFeO_2 exhibits multiferroic at zero field with Al or Ga doping.

Terada *et al.* J. Phys.: Conf. Series 145, 012071 (2009)

Correlation between doping and anisotropy

Terada et al., J. Phys.: Conf. Series 145, 012071 (2009)

Terada et al., J. Phys. Soc. Jpn. 74, 2604 (2005)

Increasing doping concentration lowers anisotropy by introducing disorder.
This softens spin-wave modes.

Materials and Experiments

- **Samples:**

A series of well-characterized samples of 3.5% Ga doped CuFeO_2 were provided by the T. Kimura group from Osaka University.

- **Inelastic neutron scattering: SNS and HFIR (Oak Ridge National Laboratory)
JRR-3 (Japan Atomic Energy Agency)**

CNCS time-of-flight spectrometer at SNS

CG4C triple-axis spectrometer at HFIR

3.5% Ga-doped CuFeO_2 – INS Data

Inelastic neutron scattering data from the HFIR and the SNS and ORNL.

Excitations at $H \sim 0.2(Q)$ and $0.3 (2\pi - Q)$.
Symmetric around $H = 0.25 (\pi)$.
Spiral ordering along the x-direction

Dispersion along $[0,0,L]$ indicates interlayer interactions in the z-direction.

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Variational Method for the Spin Ground State

Modification of the S_z and S_y components to account for anisotropy and lattice distortions.

$$S_z(R) = A \cos[Q \cdot x] + \sum_{l=1} C_{2l+1} \cos[Q(2l+1) \cdot x] \quad \text{Anisotropy}$$

$$+ \sum_{l=0} B_{2l+1} \cos[(2\pi - Q)(2l+1) \cdot x] \quad \text{Lattice Distortion}$$

$$S_y(R) = \sqrt{S^2 - S_z(R)^2} \operatorname{sgn}(Q \cdot x)$$

Spin Dynamics

General Technique for Spin Rotation

Goals

- Generate a rotation matrix.
- Examine the spin moment through its local reference frame.

θ - Rotation in xz -plane

ϕ - Rotation in xy -plane

$$\underline{U}_i = \begin{vmatrix} \cos(\theta_i)\cos(\phi_i) & \cos(\theta_i)\sin(\phi_i) & -\sin(\theta_i) \\ -\sin(\phi_i) & \cos(\phi_i) & 0 \\ \sin(\theta_i)\cos(\phi_i) & \sin(\theta_i)\sin(\phi_i) & \cos(\theta_i) \end{vmatrix}$$

Rotation of the Spin Hamiltonian

$$H = -\frac{1}{2} \sum_{i \neq j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - D \sum_i S_{zi}^2 - g\mu_B H \sum_i S_{zi}$$

$$= -\frac{1}{2} \sum_{i \neq j} J_{ij} \bar{\mathbf{S}}_i \cdot \underline{U}_i \underline{U}_j^{-1} \mathbf{S}_j - D \sum_i \left(\underline{U}_i^{-1} \bar{S}_{zi} \right)^2 - g\mu_B H \sum_i \underline{U}_i^{-1} \bar{S}_{zi}$$

Using a Holstein-Primakoff expansion ($1/S$), the Hamiltonian becomes

$$H = E_0 + H_1 + H_2 + \text{Higher Order Terms}$$

E_0 = classical energy

H_1 = vacuum contribution to the spin-waves (must vanish)

H_2 = spin dynamics (spin-wave frequencies and intensities)

Higher-order terms can be ignored since quantum fluctuations are negligible with large S .

$$i \frac{dv_q}{dt} = -[H_2, v_q] = \underline{M}(q) v_q$$

$$\text{Det}[\underline{M}(q) - \omega_q \underline{I}] = 0$$

The spin-wave frequencies can be determined by through a diagonalization of the equations-of-motion.

Spin-Wave Neutron Scattering Intensity

$$I(q, \omega) = S(q, \omega) F_q^2 e^{\frac{-(\omega - \omega_q)^2}{2\delta^2}}$$

Spin-Spin Correlation Function

$$S(q, \omega) = \sum_{\alpha\beta} (\partial_{\alpha\beta} - q_\alpha q_\beta) S^{\alpha\beta}(q, \omega)$$

$$S(q, \omega) = \frac{1}{N} \int e^{-i\omega t} dt \sum_{ij} e^{iq \cdot (R_j - R_i)} \langle S_i^\alpha(0) S_j^\beta(t) \rangle$$

$$= -\frac{4}{\pi} \lim_{\delta \rightarrow 0} \Im \left[Tr \left(\underline{G}(q, \omega + i\delta) \underline{C}_{\alpha\beta} \right) \right]$$

$$\underline{G}_{nm}(q, \omega + i\delta) = \left[\frac{-1}{(\omega + i\delta) \underline{I} - \underline{M}(q)} \underline{N} \right]_{nm}$$

a and b = x, y, z

Modeling the intensities helps “lock down” the magnetic structure by providing another dimension to the problem.

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The Magnetic Interactions for the 3-D Model

$$H = -\frac{1}{2} \sum_{i \neq j} J_{ij} S_i \cdot S_j - D \sum S_{zi}^2 - g\mu_B H \sum S_{zi}$$

J_{ij} – super-exchange interaction between magnetic ions

D – single ion anisotropy

H – external magnetic field

$$J_1^{(1)} = J_1^{(2)} = J_1 - K_1/2$$

$$J_1^{(3)} = J_1 + K_1$$

Predicted Spin-Wave Dynamics

Simple Spiral

$C_1(Q)$

With large anisotropy and no lattice distortion

$B_1(2\pi - Q)$

CNC Spiral

$\Delta\Theta_1 \approx 22^\circ$ and $\Delta\Theta_2 \approx 134^\circ$

With small anisotropy and large lattice distortion

“Proper” spiral is not a stable ground state!

Spin-Wave Dynamics Comparison

Multi
Domain
(in meV)

$J_1 = -0.19$
 $J_2 = -0.10$
 $J_3 = -0.13$
 $J_{z1} = -0.05$
 $J_{z2} = 0.02$
 $J_{z3} = -0.01$
 $D = 0.01$
 $K_1 = 0.07$

Single
Domain
(in meV)

$J_1 = -0.23$
 $J_2 = -0.07$
 $J_3 = -0.10$
 $J_{z1} = -0.07$
 $J_{z2} = 0.01$
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 $D = 0.01$
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Field vs. Anisotropy: Competing Phases

Prediction of the CuFeO_2 trajectory

Experiments currently being planned to examine these phases!

R.S. Fishman, G. Brown, and J.T. Haraldsen, Phys. Rev. B. 85, 020405(R) (2012).
J.T. Haraldsen, R.S. Fishman, and G. Brown, Phys. Rev. B. 86, 024412 (2012).

Summary

- Working towards understanding the multiferroic mechanism.
- Examined the underlying magnetic ground states for strong magneto-electric coupling.
- Developed a rotational technique to examine the spin dynamics of non-collinear magnetic systems.
- Modeled the magnetic structure for the multiferroic phase in CuFeO_2 .
- Predicted the high-magnetic-field phases.
- Started working towards the coupling of magnetism to electric polarization through interfacial symmetry breaking in multiferroic heterostructures.

Modulations due to magnetoelectric coupling.

Explicit $z \rightarrow -z$ symmetry is broken

$$F_I = \alpha_I M^2 + \frac{\beta_I}{2} M^4 + \gamma_I P^2 + \frac{\beta_I}{2} P^4 + \lambda_I PM^2$$

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Research Interests: Magnetic and Complex Oxide Materials

Quantum Spin Nanostuctures: Understanding intricate low-dimensional magnetism through an examination of the various interactions within clusters and molecular magnets.

Magnetic Heterostructures: Investigating the magnetic chirality produced through stacking mechanisms and interactions in heterostructures.

Multiferroic Materials: Exploring the nature of magnetoelectric coupling and various order parameters in bulk and composite multiferroics.

Complex Oxide Interfaces: Examining the complex interplay between multiple degrees of freedom (spin, charge, orbital, etc.) through the evolution from interfacial phenomena to bulk properties.

J.T. Haraldsen *et al.*, *Phys. Rev. B* 86, 024412 (2012)

J.T. Haraldsen *et al.*, *J. Condens. Matter* 22, 186002 (2010)

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Advanced Data Analysis: Simulations and modeling of complex data sets through computation and visualization techniques.

J.T. Haraldsen *et al.*, Phys. Rev. B 86, 024412 (2012)

Funding Opportunities

U.S. DEPARTMENT OF DEFENSE

- Multiscale multidisciplinary modeling of electronic materials
- Young faculty award

U.S. DEPARTMENT OF ENERGY

- Predictive theory and modeling
- Early career research award

National Science Foundation WHERE DISCOVERIES BEGIN

- Computational and data driven materials research
- Materials analysis
- Research experiences for undergraduates
- Faculty early career program

McHUMOR.com by T. McCracken

“Reading this grant proposal, I conclude that you must have gotten an A in creative writing.”

Program managers are interested in supporting predictive theory and modeling, especially in support of current experimental facilities.

Projects Overview

➤ Magnetic Structure Investigations

- ✧ Na_2RuO_4 (cluster to long-range order)
- ✧ Honeycomb lattice (co-existence of 2-D and 3-D order)

➤ Multiferroics

- ✧ Expand the magnetic Hamiltonian and couple to electric polarization through orbital parameters.
- ✧ Investigate the relevance of three dimensional magnetic ordering on the multiferroic state.
- ✧ Examine induced polarization from magnetic states in the bulk and at heterostructure interfaces.

➤ Development of magnetic structure software

- ✧ Fast and efficient tool for the determination and understanding of magnetic materials.
- ✧ Dynamic analysis for theory and experiment.

➤ Interfacial Phenomena

- ✧ Determine the affects of defects and impurities on interfacial phenomena.
- ✧ Incorporate spin, lattice, orbital, and electronic degrees of freedom.

➤ Departmental Collaborations

- ✧ Given the detailed crossover within the department. I am very much open to collaborative projects.

“I think you should be more explicit here in step two!”

Teaching and Mentoring

Teaching and mentoring experience:

- University of Tennessee – Teaching Assistant (General – Honors Physics)
- Oak Ridge National Laboratory – Mentored undergraduate students in research resulting in publications in PRL and PRB.
- Los Alamos National Laboratory – Mentored graduate students in research resulting in a NanoLetter.

Teaching philosophy:

- Motivate students through critical thinking and education enrichment.
- Work directly with students using examples, homework, and experiments.
- Prepare them for writing papers, proposals, and presentations.
- Helping them achieve their goals.

Courses:

- I can teach a number of courses in the undergraduate and graduate curriculum: General Physics, Quantum Mechanics, Condensed Matter Theory, Electricity and Magnetism, Statistical Mechanics, etc.
- I would also look to offer special topics courses: Nanomagnetism, mathematical/computational condensed matter physics, and the physics of materials.

“Well, a pop-up Master’s thesis is certainly an original idea...”

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- Los Alamos National Laboratory
- Center for Integrated Nanotechnologies
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- Osaka University
- Florida State University
- University of Tokyo