Magnon-magnon/phonon coupling in two-dimensional triangular lattice antiferromagnets





Center for Correlated Electron Systems, Institute for Basic Science

Je-Geun Park

Dept. Physics & Astronomy

Seoul National University

IBSPCS—KIAS International Workshop Frustrated Magnetism









Why magnon-magnon/phonon coupling in 2D TAF?

◆ A Tale of Two Cities: magnon-magnon/phonon couplings in YMnO₃

- Magnons measured by inelastic neutron scattering
- Phonons measured by inelastic X-ray scattering





Magnetic excitations in non-collinear metallic antiferromagnet CrB₂, Pyeongjae Park

Spin texture and its dynamics of Al-doped triangular lattice

antiferromagnet h-YMnO₃, Kisoo Park



Triangular Antiferromagnets





Triangular Antiferromagnets at J-G Park's Group







Magnon-Phonon Coupling



PHYSICAL REVIEW

VOLUME 110, NUMBER 4

MAY 15, 1958

Interaction of Spin Waves and Ultrasonic Waves in Ferromagnetic Crystals*





Textbook Example: FeF₂





FeF₂





S. Lovesey, "Theory of Neutron Scattering from Condensed Matter systems," sect. 9.8

T. Chatterji et al., JPCM **22**, 316001 (2010)



Magnon-Phonon for Spintronics



Spintronics: spins as data storage

Writing process by photon & heat



nature **LETTERS PUBLISHED ONLINE: 21 AUGUST 2011** | DOI: 10.1038/NMAT3099

Long-range spin Seebeck effect and acoustic spin pumping

K. Uchida^{1,2}, H. Adachi^{2,3}, T. An^{1,2}, T. Ota^{1,2}, M. Toda⁴, B. Hillebrands⁵, S. Maekawa^{2,3} and E. Saitoh^{1,2,3,6}*

Photodrive of magnetic bubbles via magnetoelastic waves

Naoki Ogawa^{a,1}, Wataru Koshibae^a, Aron Jonathan Beekman^a, Naoto Nagaosa^{a,b}, Masashi Kubota^{a,c,2}, Masashi Kawasaki^{a,b}, and Yoshinori Tokura^{a,b}

^aRIKEN Center for Emergent Matter Science, Wako, Saitama 351-0198, Japan; ^bDepartment of Applied Physics and Quantum Phase Electronics Center, University of Tokyo, Tokyo 113-8656, Japan; and ^cResearch and Development Headquarters, ROHM Company, Ltd., Kyoto 615-8585, Japan

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- Magnon-phonon coupling is important for spin conversion.
- Conversion factor \propto coupling strength



Magnon-Phonon in YIG



Spintronics: spins as data storage

Writing process by photons & heat



Photo-induced magnetic domain in YIG



N. Ogawa et al., PNAS 112, 8977 (2015)

Magnon-Phonon coupling: exchange-striction



$$H = H_{Heis} + H_{lattice} = \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + H_{lattice}$$

$$J_{ij} \left(\mathbf{R}_i, \mathbf{R}_j, \mathbf{R}_{O_{ij}} \right) = J_0 + (\mathbf{u}_1 \cdot \nabla_1 + \mathbf{u}_2 \cdot \nabla_2 + \mathbf{u}_3 \cdot \nabla_3) J_{ij} + \cdots$$

$$(\mathbf{u}_1 \cdot \nabla_1 + \mathbf{u}_2 \cdot \nabla_2 + \mathbf{u}_3 \cdot \nabla_3) J_{ij} = \left(\mathbf{u}_{O_{ij}} - \mathbf{u}_i \right) \cdot \nabla_1 (J_{ij}) + \left(\mathbf{u}_{O_{ij}} - \mathbf{u}_j \right) \cdot \nabla_2 (J_{ij}) + \left(\mathbf{u}_j - \mathbf{u}_i \right) \cdot \nabla_3 (J_{ij})$$

Superexchange-striction Direct exchange-striction

Magnon-Phonon coupling: exchange-striction

- Exchange energy is dominant in most 3d TM based magnets
- No linear coupling in collinear spin structure



Longitudinal spin fluctuation $S_i^z = S - b_i^{\dagger} b_i$ Transverse spin fluctuation $S_i^x, S_i^y = b_i^{\dagger}, b_i$

$$H = J \sum_{i,j} S_i \cdot S_j$$
$$= J \sum_{i,j} \left(S_i^x S_j^x + S_i^y S_j^y + S_i^z S_j^z \right)$$

$$H_{mp} = 0 SJS \sum_{i,j} \left(S_i^x S_j^x + S_i^y S_j^y + S_i^z S_j^z \right)$$

1 phonon 2 magnon

Magnon-Phonon coupling via exchange-striction

Noncollinear magnetic structure: Transverse-longitudinal coupling
 → Direct mixing of magnons and phonons



Longitudinal spin fluctuation $S_i^z = S - b_i^{\dagger} b_i$ Transverse spin fluctuation $S_i^x, S_i^y = b_i^{\dagger}, b_i$





Joosung Oh PhD Thesis (2017)



magnon

1 phonon

Calculation of Hybrid ME Excitations

- Lattice & spin modulation \rightarrow phonon & magnon operators
- Numerical diagonalization of Hamiltonian
- Calculation of dynamical spin structure factor

$$H = H_{spin} + \hbar \sum_{i=1}^{90} \omega_i b_k^{\dagger} b_k + \frac{\alpha J}{2d} \sum_{ij} \left(\boldsymbol{e}_{0_{ij}i} \cdot \boldsymbol{U}_i + \boldsymbol{e}_{0_{ij}j} \cdot \boldsymbol{U}_j \right) \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

Lattice modulation:
$$U_{j,l} = \sqrt{\frac{\hbar}{2Nm_j\omega_{k\lambda}}} \sum_{k,\lambda} V_{j,k\lambda} e^{ik \cdot (R_l + r_j)} (b_{k\lambda} + b_{-k\lambda}^{\dagger})$$

Spin modulation: $S_i^- \simeq \sqrt{2S}a_i^\dagger$

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Matrix from:
$$H = S \sum_{k} X_{tot}^{\dagger} \underbrace{\bigcap_{iN} \bigcap_{iN} -iN^{\dagger}}_{iN} \underbrace{\bigcap_{iN} \bigcap_{iN} -iN^{\dagger}}_{iN} \underbrace{O}_{iN} \underbrace{O}_{i$$



Joosung Oh PhD Thesis (2017)



Estimated α For YMnO₃ $\alpha = \frac{d(P_0)}{T_N(P_0)} \frac{\partial T_N / \partial P}{(\partial d / \partial P)} = 14$ • T. Lancaster et al., PRL 98, 197203 (2007) • D. P. Kozlenko, JGP et al., JETP 82, 193 (2005)

> La₂CuO₄: $\alpha = 6 \sim 7$ CuCrO₂: $\alpha = 30$

- M. C. Aronson et al., PRB 44, 4657 (1991)
- K. Park, JGP et al., PRB 94, 104421 (2016)



Two Experimental Tools



Inelastic Neutron Scattering

Inelastic X-ray Scattering









- Wavelengths of neutrons and x-ray are si milar to atomic spacing!
- Energy of neutrons and x-ray are similar to elementary excitations in solids!

Advantage of Inelastic neutron scattering

- Probes both spin & lattice excitation
- Good energy resolution
- Sensitive to light atoms

Advantage of Inelastic x-ray scattering

- Probes lattice vibration
- Good momentum resolution
- Low background
- Small sample down to $10 \mu m$ and 100 nm film

Magnon-Magnon/Phonon Coupling in hexagonal Manganites

Inelastic Neutron and X-ray Scattering

2D Triangular Antiferromagnet (Y,Lu)MnO₃



- Structural transition from paraelectric ($P6_3$ /mmc) to ferroelectric ($P6_3$ cm) below T_c~1250 K
- Mn atoms form triangular layers

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• 120° noncollinear magnetic order below T_{N} due to geometrical frustration



H. Sim, J-G. Park et al., Acta Cryst. B72, 1 (2016)

- YMnO₃: T_N=75 K, θ_{CW}=705 K
- LuMnO₃: T_N =90 K, θ_{CW} =887 K

4 possible magnetic structure of P6₃cm space group





Ferroelectric-Magnetic domain locking





Several papers from S-W Cheong's group Y. Geng et al., Nano Lett., 12, 6055 (2012) H. Das et al., Nat. Commun. 5, 2998 (2014)





Spin-Lattice Coupling in h-RMnO₃



- Atomic displacement below T_N
- Large displacement of Mn x position induces
 Mn trimerization.









S. Lee, JGP et al., PRB (2005) S. Lee, JGP et al., Nature (2008)



Spin-Lattice Coupling in h-RMnO₃



• Numerous evidences of spin-lattice(phonon) coupling in YMnO₃



M. N. Iliev et al., PRB 56, 2488 (1997)
P. A. Sharma et al., PRL 93, 177202 (2004)
M. Poirier et al., PRB 76, 174426 (2007)



Magnon dispersion relation



- Three magnon modes
- J=2.5 meV, D=0.28 meV



$$H_{ex} = J \sum_{r} S_r \cdot S_{r+\delta} + D \sum_{r} (S_r^z)^2$$

Antiferro Easy plane
1 > 0 D1 > 0



O. P. Vajk et al., PRL 94, 087601 (2005)

M. A. van der Varte et al., arXiv:0907.3055



Magnon-Magnon coupling in LuMnO₃







Linewidth Broadening in LuMnO₃







Magneto-Elastic Excitation in (Y,Lu)MnO₃





J. Oh, J-G. Park et al., Nat. Commun. 7, 13146 (2016)



Exchange-Striction Model



Mn-O Bond-length change is dominant for exchange-striction in $YMnO_3$

$$H = H_{spin} + \hbar \sum_{i=1}^{90} \omega_i b_k^{\dagger} b_k + \frac{\alpha J}{2d} \sum_{ij} \left(\boldsymbol{e}_{O_{ij}i} \cdot \boldsymbol{U}_i + \boldsymbol{e}_{O_{ij}j} \cdot \boldsymbol{U}_j \right) \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$





DFT phonon result of YMnO₃



INS Dynamical Structure Factor



Dynamical Structure Factor: HoMnO₃









Overall features are all similar for all three different models although the ESP model is slightly better.

Inelastic X-ray scattering (IXS) at Spring-8

16 CEES Center for Correlated Electron Systems

BL 43XU at Spring-8, Japan

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- Simultaneous measurement of 24 different q points
- q & E resolution: 0.05 Å $^{-1}$ & 1.5 meV

Main Operating Conditions at BL43LXU				
Spectrometer	High Resolution (Atomic Dynamics)			
X-ray Energy (<u>keV</u>)	25.70	21.75	17.79	15.82
Energy Resolution FWHM (meV)	>0.75	>1.25	>2.8	~6
Analyzer Reflection Si (nnn)	(13 <u>13 13</u>)	(11 <u>11 11</u>)	(9 <u>9 9</u>)	(8 <u>8 8</u>)
Maximum Momentum Transfer (Å ⁻¹)	12	10	8.3	7.4
Photons at the Sample (Relative: 1 unit ~25 GHz)	~0.2	~1	~3	~10
Analyzer Solid Angle	~ 0.2 x 0.2 mrad ² (Min.) to 9.4 x 8.9 mrad ² (HxV, Max.)			
Beam Size at Sample Diameter, FWHM	~50 µm (Default), ~15 µm (Compound Focus)			













IXS result and DFT calculations for YMnO₃





Key Observations

- We have succeeded in measuring several key phonons by using inelastic Xray scattering technique.
- We have also found a significant shift induced by cooling.



(0 k 0)

Magneto-Elastic Excitations in YMnO₃



K. Park, J.-G. Park et al., in preparation





Kisoo Park



Red: HT (120 K) Blue: LT (14 K)

How to quantify magnon-phonon hybridization?

Each magneto-elastic modes can be characterized by the ratio between magnon/phonon components $(p_i^{mag}(q) + p_i^{ph}(q) = 1, \quad p_i^{mag/ph}(q): magnon/phonon character of$ *i*-th band at**q**point).

Mangeto-elastic modes colored with the magnon character $p_i^{mag}(\pmb{q})$

Matrix from:
$$H = S \sum_{k} X_{tot}^{\dagger} \begin{pmatrix} L & -iN^{\dagger} & M & -iN^{\dagger} \\ iN & \Omega & -iN & 0 \\ M & iN^{\dagger} & L & -iN^{\dagger} \\ iN & 0 & iN & \Omega \end{pmatrix} X_{tot}$$







Magnon-Phonon Mixing Entropy



- Each magneto-elastic modes can be characterized by the ratio between magnon/phonon components $(p_i^{mag}(q) + p_i^{ph}(q) = 1, \quad p_i^{mag/ph}(q): magnon/phonon character of$ *i*-th band at**q**point).
- We can define the 'magnon-phonon mixing entropy' $S(q) = -\sum_i p_i^{mag}(q) log(p_i^{mag})(q)$ to quantify the mode-integrated momentum dependence of magnon-phonon coupling.



Magnon-Phonon Mixing Entropy





Detailed Phonon Mode Analysis

We can diagonalize pure magnon Hamiltonian in MP-coupled Hamiltonian.

$$\mathcal{H}_{mag}(\mathbf{k}) = \begin{pmatrix} A_{\mathbf{k}} & B_{\mathbf{k}} \\ B_{\mathbf{k}} & A_{\mathbf{k}} \end{pmatrix}, \qquad \mathcal{H}_{mp}(\mathbf{k}) = \begin{pmatrix} A_{\mathbf{k}} & N_{\mathbf{k},\lambda}^{\dagger} & B_{\mathbf{k}} & N_{\mathbf{k},\lambda} \\ N_{\mathbf{k},\lambda} & E_{ph}(\mathbf{k},\lambda) & N_{\mathbf{k},\lambda}^{\dagger} & 0 \\ B_{\mathbf{k}} & N_{\mathbf{k},\lambda} & A_{\mathbf{k}} & N_{\mathbf{k},\lambda}^{\dagger} \\ N_{\mathbf{k},\lambda}^{\dagger} & 0 & N_{\mathbf{k},\lambda} & E_{ph}(\mathbf{k},\lambda) \end{pmatrix}$$
where $V_{mag}^{\dagger} \cdot \mathcal{H}_{mag}(\mathbf{k}) \cdot V_{mag} = E_{mag}, V_{mag} = \begin{pmatrix} \alpha_{\mathbf{k}} & \beta_{\mathbf{k}} \\ \gamma_{\mathbf{k}} & \delta_{\mathbf{k}} \end{pmatrix}$

$$\rightarrow \begin{pmatrix} \alpha_{k} & 0 & \beta_{k} & 0 \\ 0 & I & 0 & 0 \\ \gamma_{k} & 0 & \delta_{k} & 0 \\ 0 & 0 & 0 & I \end{pmatrix}^{\dagger} \cdot \begin{pmatrix} A_{k} & N_{k,\lambda}^{\dagger} & B_{k} & N_{k,\lambda} \\ N_{k,\lambda} & E_{ph}(\mathbf{k},\lambda) & N_{k,\lambda}^{\dagger} & 0 \\ B_{k} & N_{k,\lambda} & A_{k} & N_{k,\lambda}^{\dagger} \\ N_{k,\lambda}^{\dagger} & 0 & N_{k,\lambda} & E_{ph}(\mathbf{k},\lambda) \end{pmatrix} \cdot \begin{pmatrix} \alpha_{k} & 0 & \beta_{k} & 0 \\ 0 & I & 0 & 0 \\ \gamma_{k} & 0 & \delta_{k} & 0 \\ 0 & 0 & 0 & I \end{pmatrix}$$
$$= \begin{pmatrix} E_{mag}(\mathbf{k}) & g_{k,\lambda}^{\dagger} & 0 & g_{k,\lambda} \\ g_{k,\lambda} & E_{ph}(\mathbf{k},\lambda) & g_{k,\lambda}^{\dagger} & 0 \\ 0 & g_{k,\lambda} & E_{mag}(\mathbf{k}) & g_{k,\lambda}^{\dagger} \\ g_{k,\lambda}^{\dagger} & 0 & g_{k,\lambda} & E_{ph}(\mathbf{k},\lambda) \end{pmatrix}$$
Mode-dependent magnon-phonon coupling strength

 $\frac{1}{\left|g_{k,\lambda}\right|^{2}}$ 0.4









K point (1/3 1/3 0), 8th phonon mode (16.17 meV)

 \rightarrow Mn-trimer breathing phonon mode is most strongly coupled with the magnons at K point.









Magnetic Graphene & Model Magnetism





Jae-Ung Lee, JGP, Hyeonsik Cheong et al., Nano Lett. 16, 7433 (2016)

CrGeTe₃

C Gong et al., Nature, 546, 265 (2017)

$\mathbf{CrI}_{\mathbf{3}}$

B. Huang et al., Nature, 54 270 (2017)



- S. Kim, JGP et al., Phys. Rev. Lett. 120, 136402 (2018)
- Kangwon Kim, JGP, Hyeonsik Cheong et al., Nat. Comm. 10, 345 (2019)

E (a.u.)

Carina Belvin, JGP, Nuh Gedik, (in preparation)

T.,~155 K

Temperature (K)

Bulk
 10 m

+ DL + 41,

4 SL

÷ 21.



- MnPS₃
- K. Kim, JGP, Hyeonsik Cheong et al., 2D Materials (2019)
- H. Chu, JGP, D. Hsieh et al., PRL (submitted)



μ₀Η (T)





Frustration on real 2D limits



Honeycomb Lattice

- Most of magnetic van der Waals materials have honeycomb lattice
- Ising: $FePS_3$, CrI_3 , VI_3 , $Fe_3Ge_2Te_6$...
- XY: NiPS₃
- Heisenberg: MnPS₃

cf: RuCl₃

Triangular Lattice

- 1T TaS₂: possible quantum spin liquid state
 - M. Kratochvilova, NPG
 Quantum Materials 2,
 42 (2017)
 - Y. J. Yu, Phys. Rev. B96, 081111(R) (2017)

cf: Y Matsuda's talk on Tuesday

Kagome Lattice

- $Pd_3P_2S_8$
- Nb₃I₈
- Nb₃(Br,Cl)₈



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• Key Players at my group

Kisoo Park, Joosung Oh, Ki Hoon Lee, Manh Duc Le, Jaehong Jeong, J. C. Leiner, Je-Geun Park (CCES, SNU)

• Sample preparations

Hasung Sim (CCES, SNU) Hiroshi Eisaki & Yoshiyuki Yoshida (AIST, Japan) S. W. Cheong (Rutgers, USA)

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Je-Geun Park's Group



http://magnetism.snu.ac.kr



We have several positions open at the moment. If interested, please contact us.



Summary



- Strong magnon-phonon coupling produces a new hybrid magneto-elastic mode.
- We developed a quantitative formalism/procedure how to combine magnon and phonon.
- We provide a detailed momentum- and mode- analysis on the effect of magnon-phonon coupling.
- <u>More general remark</u>: In a noncollinear magnet, magnon mixes with other magnon and phonon, resulting in a creation of magneto-elastic excitation.

