Final Technical Report for DE-SC0006888

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1 Executive Summary

The main goal of this project was to understand novel ground states of spin systems probed by thermal and electrical transport measurements. They are well-suited to characterize the nature of low-energy excitations as unique property of the ground state. More specifically, it was aimed to study the transverse electrical conductivity in the presence of non-collinear and non-coplanar spin ordering and the effects of gauge field as well as novel spin excitations as a coherent heat transport channel in insulating quantum magnets. Most of works done during the grant period focused on these topics. As a natural extension of the project's initial goals, the scope was broadened to include transport studies on the spin systems with strong spin-orbit coupling. One particular focus was an exploration of systems with strong magnetic anisotropy combined with non-trivial spin configuration. Magnetic anisotropy is directly related to implement the non-collinear spin ordering to the existing common geometry of planar devices and thus our investigation poses a significant potential. Work in this direction includes the comparison of the topological Hall signal under hydrostatic pressure and chemical doping, as well as the angular dependence dependence of the non-collinear spin-ordered phase and their evolution up on temperature and field strength. Another focus was centered around the experimental identification of spin-originated heat-carrying excitation in quasi two dimensional honeycomb lattice, where Kitaev type of quantum spin liquid phase is predicted to emerge. In fact, when its long range magnetic order is destroyed by the applied field, we discovered anomalously large enhancement of thermal conductivity, for which proximate Kitaev excitations in field-induced spin liquid state are responsible for. This work, combined with further investigations in materials in the similar class may help establish the experimental characterization of new quantum spin liquid and their unique low energy excitation, e.g. Majorana fermions.

The project ran from July 15, 2011 to December 31, 2016. This period includes one time no-cost extension for 6 months, from July 15, 2016 to December 31, 2016. The approved budget was $766,550 over this period. This grant has been so far acknowledged in 3 publications and 5 manuscripts which have been submitted to peer-reviewed journals and currently under review. The grant supported the PI's summer salary throughout its period and 4 graduate research assistants and 1 research associate for different time periods. Details of the publications and their acknowledgement and the list of people who were supported by this grant are attached in Appendix 1 and 2 respectively at the end of this report.

2 Highlights

The following are the main accomplishments of the Principal Investigator's research group supported by this DOE grant:

- The PI's group collaborated with Thomas Wolf at Karlsruhe Institute of Technology, Germany and studied the electrical conduction metallic helical magnet pure MnSi and Fe-doped samples $Mn_{1-x}Fe_xSi$. Particularly, our investigations were focused on the A-phase, which refers a small pockets in the field (B) – temperature (T) phase diagram where the skyrmion lattice (SkL) emerges. Hexagonal SkL is merely one example of a variety of possible non-collinear or non-coplanar spin structures. The non-coplanarity of these spin structures can give rise to an emergent magnetic field $\boldsymbol{b}_r$ expressed as $b_r^i = \frac{\Phi_0}{8\pi}\epsilon_{ijk}\hat{n}\cdot(\partial_j\hat{n}\times\partial_k\hat{n})$, where ϵ_{ijk} is the Levi-Civita symbol whose indices run over x, y, and z, $\hat{n}(\boldsymbol{r})$ is a unit vector of the magnetization, $\boldsymbol{M}(\boldsymbol{r})$, and $\Phi_0 = h/|e|$ is the single-electron flux quantum. The hexagonal SkL in MnSi is formed on in 2 dimensional plane perpendicular to $\hat{z}$ direction, which gives only the z-component of $\boldsymbol{b_r}$ non-zero, i.e. $\boldsymbol{b_r} = (0, 0, b_r)$. This gauge field arises in the strong Hund coupling limit, where the spin of conduction electrons orients parallel to the local magnetization, twisting to follow it as they move through the material. This results in the acquisition of an extra phase factor in their wave functions, represented by the line integral of a vector potential, analogous to the Aharonov-Bohm effect. The curl of that vector potential corresponds to $\boldsymbol{b_r}$, that acts on conduction electrons in a similar way as the physical magnetic field , and give arise a new type of Hall effect, the topological Hall effect (THE).

In a SkL phase, the area integral of $\boldsymbol{b_r}$ over a magnetic unit cell is quantized to an integer times Φ_0 . For MnSi that integer is -1, and the similarity in SANS data of 8% Fe doped samples suggests this topological quantum number is unchanged with the level of doping considered here:

$$\oint_{\mathcal{A}_{sk}} \boldsymbol{b_r}\cdot\ \mathrm{d}\boldsymbol{\mathcal{A}} = -\Phi_0, \tag{1}$$

where $\mathcal{A}_{sk} = (2/\sqrt{3})\lambda^2$ is the area of the magnetic unit cell with λ a helical pitch length. The minus sign implies that the emergent field on average opposes the normal vector of the unit cell, which is parallel to the applied B. Eq. 1 allows estimation of an *upper bound* for the average topological magnetic field induced by the spin texture: $\bar{b_r} = -\Phi_0/\mathcal{A}_{sk}$, typically tens of Tesla for $Mn_{1-x}Fe_xSi$.

The primary effect of increasing Fe content and application of pressure (P) are a suppression of T_C, the saturated magnetic moment m_s and the helical pitch length . However, Fe doping and P take different function forms in reducing these quantities. Upon increasing Fe, both of T_C and m_S monotonically go to zero, vanishing at the critical doping concentration $x_c \simeq 15-19\%$. However, with pressure, m_S would not goes to zero as P approaches P_C and thus $T_C \to 0$. Similarly, the pitch length λ monotonically decreases as increasing Fe content, while $\lambda(P)$ changes little from the ambient pressure value then begins to decrease when T_C was suppressed more 2/3 of the value at ambient pressure. The Hall effect signals arising from non-trivial spin textures provide a rare opportunity to directly access the gauge fields of purely quantum-mechanical origin. To connect with experiment and compare to $\bar{b_r}$, an effective field B_{eff} may be estimated from the magnitude of THE and the normal Hall coefficient R_H, *viz.* $\rho_{yx}^T = R_H B_{\mathrm{eff}}$.

Motivated to tune the skyrmion size set by λ, and thus control the magnitude of $\bar{b_r}$ via Eq. 1, we studied the Hall effect in Fe-doped MnSi with 6% and 9% Fe content. Previous studies have shown that Fe doping leads to a linear decrease in J while leaving D unchanged, resulting in a decrease in λ observed in scattering experiments. This compresses the spatial extent of the skyrmions, squeezing their single flux quantum through a smaller area, and necessitating a larger $\bar{b_r}$. In our previous work , we compared these two cases extensively. The topological contribution in the sample with 9% Fe is enhanced by a factor of 5 compared to pure MnSi.

We compared the behavior of THE of Fe doped MnSi and MnSi under pressure.Along with this changing magnitude, we find that the sign of the THE changes as well, remaining opposite to the sign of the R_H, in accordance with the minus sign in Eq. 1. The ratio $f = B_{\mathrm{eff}}/\bar{b_r}$ ranges between 0.3 and 0.4 for doped and pressurized samples, except it is much lower, only 0.05 for MnSi at ambient P. Interestingly, despite the larger size of the THE in MnSi under P,

f remains more or less constant across both doping and pressure. We suggest three physical considerations that will be reflected in the value of f: (i) The strength of the coupling between charge carriers and the spin texture. Only in the strong Hund coupling limit, where the spin of conduction electrons tightly follows the spin-texture, can f approach unity. (ii) The ratio of conduction electrons with majority to minority spins. This can be varied by changes in the band structure through addition of Fe. Such alterations change the magnitude of the THE, as electrons with opposite spin feel opposite emergent fields. These induce opposite Hall voltages which cancel with one another. (iii) The presence of strong fluctuations in the spin texture on much shorter time scales than our measurement in millisecond. This is expected to reduce the time-averaged value of the emergent field B_{eff}, and hence diminish f.

This work was published in Physical Review B **88**, 214406 (2013).

- As an extension of investigating the SkL phase in the Fe-doped MnSi, we have studied the magnetic anisotropy of the SkL phase in Fe-doped MnSi using the same series of Fe-doped samples provided by Thomas Wolf from Karlsruhe. MnSi and Fe-doped family are known to have weak magnetic anisotropy, which in turn allows such smoothly varying spin-texture. However little has been known how the A-phase, where the SkL emerges, is affected by the anisotropy. Fe impurities increases the magnitude of gauge field generated by the SkL and consequently, enhances the angular dependence of topological Hall signal and MR in the A phase region. We found distinct angular responses in the THE signal and the magnetoresistance in the A-phase which allows us to categorize different regions of region I,II and III. Our result also revealed that the center of A-phase (region II) shows the spin texture is completely detached from the underlying crystal geometry. Meanwhile, surrounding areas of region I (low T and low H) and III (high T and high H) are attributed to the role of impurity and the crystalline anisotropy (region I) and the polarized Skyrmion state (region III), both of which are fundamentally distinguishable from region II.

  This work was submitted and under review in Physical Review Letter (2016).

- In collaboration with Professor Tomo Uemura at Columbia University, we were involved in a comprehensive study of muon spin relaxation (μSR) on bulk single crystals of MnSi and (Mn,Fe)Si, a thin film of MnSi, and a ceramic specimen of Cu_2OSeO_3. The main results of this study relevant to our current work in $Mn_{1-x}Fe_xSi$ was to discover "fluctuating Skyrmion liquid" (FSL) region near and above T_C. In this regions, critical slowing down of Mn spin fluctuations with a wide range of fluctuation time scales emerges simultaneously to the appearance of a large THE signal and a non-Fermi-liquid exponent of resistivity. For a while, our H dependences of Hall data above T_C in $Mn_{1-x}Fe_xSi$ have shown unusual feature – which cannot be fitted to the usual anomalous Hall effect – just above T_C and we believe that such behavior must have resulted from FSL. However, with the Hall data alone, it was hard to argue the existence of FSL phase above T_C. That is because the H-dependence of anomalous Hall effect from the ordinary magnetic order parameter is not well-defined above T_C thus it is extremely non-trivial to separate out THE signal from other contributions. In this work, the μSR data by Uemura's group, demonstrated that the muon relaxation rate ($1/T_1$) took a form of a stretched exponential above T_C, which immediately implies the wide range of fluctuation time scales. The region of H-T phase diagram showing the stretched exponential coincides with non-zero THE signal above T_C, which is an evidence for FSL phase in $Mn_{1-x}Fe_xSi$.

  This work was submitted and under review in Physical Review X.

- Another collaboration with Uemura's group led to study the restoration of quantum critical behavior by the disorder in Fe-doped MnSi. In this study, the pressure dependence of electrical transport properties (resistivity the Hall signal) and μSR for the volume ratio of the ordered phase in high Fe concentration of $Mn_{1-x}Fe_xSi$. In the presence of disorders (here Fe substitutions), the second order quantum phase transition is found to be restored, which is absent and replaced with first order transition in pure MnSi as the long range order diminishes upon increasing pressure. This work illustrates a novel role of disorder, namely that the presence of disorders favors the behavior closer to the second order transition. This study uniquely showed the ordered volume fractions from μ-SR measurements as a function of temperature and pressure. We contributed to the resistivity characterization and the pressure dependence of the Hall signals in 9% Fe doped sample.

This work was submitted to Nature Physics.

- In collaboration with David Mandrus and his group at University of Tennessee and Oak Ridge National Lab and David Parker at Oak Ridge National Lab, we studied layered-structure helical magnet $Cr_{1/3}NbS_2$. Unlike isotropic B20 structure helimagnets, where MnSi family belong, $Cr_{1/3}NbS_2$ has much larger magnetic anisotropy with the *ab*-plane as easy magnetic plane. We were initially motivated to search for other than B20 crystalline systems that host non trivial spin texture and exhibit strong correlation between the magnetic and the electrical properties. However, our investigation revealed intriguing role of spin-orbit coupling in determining the magnetic anisotropy and in altering electrical transport properties accordingly. This is surprising as $3d$ electrons of Cr^{3+} are expected to have a negligible spin-orbit coupling.

  In order to gain insight into the spin structure, first, we came up with a toy spin model for $Cr_{1/3}NbS_2$. We found that he spin structure of $Cr_{1/3}NbS_2$in a magnetic field B is well-described by a 1-dimensional (1D) Heisenberg spin model with Dyzaloshinkskii-Moriya (DM) interactions a Zeeman interaction, and strong magnetocrystalline anisotropy. Within a single *ab*-plane, the only spin-spin interaction is exchange, which results in a uniformly polarized state with all spins in the plane aligned. This allows the low-temperature magnetic structure of $Cr_{1/3}NbS_2$ to be studied on a 1D lattice, where sites correspond to local moments of Cr^{3+} ions at different position along the *c*-axis. The magnetic moment of the spin on the i^{th} site is represented classically as a 3 dimensional vector of magnitude s: $\boldsymbol{s}_i = s\boldsymbol{n}_i$, $\boldsymbol{n}_i$ a unit vector. The Hamiltonian for the system is

$$\mathcal{H} = \sum_i \Big[- J\boldsymbol{s}_i \cdot \boldsymbol{s}_{i+1} - \boldsymbol{D} \cdot (\boldsymbol{s}_i \times \boldsymbol{s}_{i+1}) - \mu_B \boldsymbol{B} \cdot \boldsymbol{s}_i + A\,(\hat{z} \cdot \boldsymbol{s}_i)^2 \Big],$$

  with J and A both positive and $\hat{z}$ along the *c*-axis. The four terms in the above expression represent the exchange interaction, the DM interaction, a Zeeman interaction, and magnetocrystalline anisotropy, respectively. μ_B is the Bohr magneton.

  We found that the simulation results were consistent with the experimental magnetization data measured with applied field along oblique angles. They also successfully captured the field induced transition from the helical state to the soliton lattice (when $H \parallel ab$) or the conical state ($H \parallel c$). consequently, we found the relatively strong anisotropy of $Cr_{1/3}NbS_2$ prevents from having more complex spin texture (e.g. SkL) and concluded that anomalous H dependence of the Hall signal is not originated from topological spin texture but from the modification of band structure depending on the magnetization orientation. Yet, it is clear that unusually angle dependence of MR must have different origin other than usual enhanced spin scattering along the hard-axis.

Subsequent magnetoresistance measurement upon rotating field, we found that the MR variation as the field was rotated out of plane is 23 times larger than that of in-plane. Although the magnitude of spin orbit coupling (SOC) in $3d$ magnetic materials is considered much smaller than other energy scale such as U or the band width W, we conclude that it was crucial to determine major magnetotransport properties, especially related to the magnetic anisotropy.

Such dramatic change of the field orientation dependent MR indicates a significant modification in the electronics structure depending on the magnetization direction. Estimated energy scale calculated from the first principle calculations is found consistent with the temperature such anisotropy disappears. We found that unusual H profile of ρ_{yx} and the larger amplitude of out-of-plane AMR occur in the same temperature regions. Thus, two different phenomena are likely to share the same origin.

This work is published in Applied Physics Letter **105**, 072405 (2014) and Physical Review B **91**, 184401 (2015).

- We investigated thermal conductivity and magnetic torque response as a function of T and H in the honeycomb structured quasi 2D magnet α-$RuCl_3$. This work was done in a collaboration with Kwang-Young Choi and his group in South Korea, who provided single crystalline samples and Bruce Normand from Paul Scherer Institute in Switzerland as well as David Graf at the National High Magnetic Field Lab.

  $RuCl_3$ is currently a leading candidate to realize the Kitaev state. Very little is known the Kitaev model under a magnetic field. Experimentally, little significant results (transport, NMR, inelastic neutron scattering) are yet available, either on iridate materials or on α-$RuCl_3$ that afford any insight into the nature of the quasiparticle excitations under applied magnetic field.

  At low field we find only weak and conventional contributions, which in the Heisenberg-ordered state is no surprise. However, we find an unambiguous field-induced phase transition to a disordered state – the field-induced spin liquid (FISL) – at $H = 7$ T. The FISL has a large and linearly field-dependent thermal conductivity at low temperatures that can only be explained by a coherent spin excitation at zero energy. The presence of a gapless excitation in the FISL, which from its linear growth has a Dirac-type spectrum, is truly remarkable, because it is a property of the exact Kitaev solution. Thus we name this feature a massless proximate-Kitaev spin excitation. Its discovery is a genuine first in the field and poses a clear challenge to theory. Of less immediate impact, our results also allow us to deduce the presence of a high-density continuum of massive (and likely fractional) spin excitation – also a feature of the pure Kitaev system – which do not constitute coherent spin modes but act to cause a strong and field-enhanced suppression of the phonon contribution to heat conduction.

  Furthermore, in the course of trying to estimate the phonon- spin scattering contribution in $RuCl_3$, we quickly realized there has been very little theoretical or experimental understanding on how the spin-orbit coupling (SOC) modifies the phonon contribution to the thermal conductivity. It is natural to assume that spin-lattice interaction is expected to be stronger in the strong SOC limit, which in turn, makes a large impact on pure phonon heat conduction, that can be hardly captured by the Debye theory.

  This provides a natural extension of this work into the future project: The system with unusual spin excitation arising from coexisting strong electronic correlation and the strong SOC will provide unique opportunity to investigate the modification of phonon heat conduction due to the enhanced spin-lattice interaction mediated by the SOC. Even in the system of a simple Heisenberg exchange coupling (J), the spin-lattice coupling is magnetoelastic such that the strength of J can be modified by the bond length between magnetic ions and this can leads stronger scattering of phonons by spin excitation. Such magnon-phonon scattering can also emerge the material with DM interaction, even in the limit of very weak SOC. Consequently, overall much reduced thermal conductivity is expected but detailed T and applied H dependence is likely specific to types of interactions. For example, thermal conductivity of Sr_2IrO_4 is found much lower there than in the worst quality La_2CuO_4, the $3d$- isostructured counterpart for Sr_2IrO_4. It is likely because of the enhanced magnon-phonon scattering.

This work is submitted to Physics Review Letters and currently under review.

- Lastly, we have worked on developing a novel thermometry using the shot noise from metallic tunnel junction sandwiched by insulator, $Al/AlO_x/Al$, which was one of the initially proposed goals of this project. We performed statistical error analysis of our measurement as a shot noise thermometer (SNT). Using a minimal parameter noise model, we calculate the resolution limits for shot noise tunnel junction readings of temperature, noise temperature and gain bandwidth. Our semiquantitative study of SNT resolution provides a useful guide for its applicability as a thermometer and they are in good agreement with experimental data. However, we found that in the low temperature limit $T \to 0$, the resolution of temperature T and the circuit noise temperature T_N and the gain G is increasingly limited by a finite T_N and verify the difficulty of operating SNT in the regime of $T \ll T_N$. This problem can be mitigated by the fast read-out circuit and processing real time, which will be resolved in further effort.

Appendix 1. List of papers acknowledged this award

1. "Large enhancement of emergent magnetic fields in MnSi with impurities and pressure" by Benjamin J. Chapman, Maxwell J. Grossnickle, Thomas Wolf and Minhyea Lee. *Published in* Physical Review B **88**, 214406 (2013).

   Excerpt of the acknowledgement: This work is supported by the US Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award No. ER 46797.

2. "Spin structure of the anisotropic helimagnet $Cr_{1/3}NbS_2$ in a magnetic field" by Benjamin J. Chapman, Alexander C. Bornstein, Nirmal J. Ghimire,David Mandrus, and Minhyea Lee. *Published in* Applied Physics Letters **105**, 072405 (2014).

   Excerpt of the acknowledgement: This work was supported by the US DOE, Basic Energy Sciences, Materials Sciences and Engineering Division (ORNL) and at CU under Award No. DE-SC0006888.

3. "Out-of-Plane Spin-Orientation Dependent Magnetotransport Properties in the Anisotropic Helimagnet $Cr_{1/3}NbS_2$" by Alexander C. Bornstein, Benjamin J. Chapman, Nirmal Ghimire, David. G. Mandrus, David S. Parker and Minhyea Lee. *Published in* Physical Review B **91**, 184401 (2015).

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4. "Anomalous thermal conductivity and magnetic torque response in magnetic torque response in the honeycomb magnet α-$RuCl_3$. arXiv:1612.03881 (2016) and *Submitted to* Physical Review Letters (2016).

   Excerpt of the acknowledgement: This work was supported by the US DOE, Basic Energy Sciences, Materials Sciences and Engineering Division under Award Number DE-SC0006888. Torque magnetometry was performed at the National High Magnetic Field Laboratory, which is supported by National Science Foundation Cooperative Agreement No. DMR-1157490 and the State of Florida. KYC acknowledges financial support from Korea NRF Grant No. 2016-911392.

5. "Multiple Magnetic States Within The A-Phase: Angular Dependence Study Of $Mn_{0.9}Fe_{0.1}Si$. arXiv:1612.03881 (2016) and *Submitted to* Physical Review Letters (2016).

   Excerpt of the acknowledgement: This work is supported by the US Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award No. DE-SC0006888

6. "Restoration of quantum critical behavior by disorder in pressure-tuned (Mn,Fe)Si. *Submitted to* Nature Physics (2016).

   Excerpt of the acknowledgement: Work at Columbia was supported by the US National Science Foundation grants DMR-1105961, DMR-1436095 (DMREF grant), DMR-1610633, OISE-0968226 (PIRE), the Japan Atomic Energy Agency Reimei project during 2011-16,

the Friends of University of Tokyo Inc. M.L. was supported by the US DOE, Basic Energy Sciences, Materials Sciences and Engineering Division under Award Number DE-SC0006888.

7. “Dynamic Spin Fluctuations and Static Order Parameter of Skyrmions”, by L. Liu, C. Arguello, S. Cheung, B. Frandsen, T. Goko, G. Pohl, H. Luetkens, T. Prokscha, E. Morenzoni, S.R. Dunsiger, P. Böni, A. Hallas, T. Medina,T.J.S. Munsie, M. Wilson, G.M. Luke, C. Ding, H. Man, F.L. Ning, B.J. Chen, D. Zheng, C.Q. Jin, G. Shibata, A. Fujimori, T. Ito, W. Higemoto, E. Svanidze, N. Kanazawa, N. Nagaosa, S. Seki, Y. Tokura, A. Bornstein, Minhyea Lee, D. Reznik, and Y.J. Uemura. *Sumitted to* Physical Review X.

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Appendix 2. List of people supported by the award

- Minhyea Lee (PI) supported for the summer salary from 2011 -2016.
- Benjamin Chapman (Graduate Research Assistant) from June 2012 to June 2014.
- Alexander Bornstein (Temporary Research Associate) from July 2014 to February 2015.
- Ian Leahy (Graduate Research Assistant) from June 2015 to October 2016.
- Peter Siegfried (Graduate Research Assistant) from June 2015 to October 2016
- Christopher Pocs (Graduate Research Assistant) from June 2016 to October 2016.