Final DOE Grant Progress Report

1. **DOE Award number:** DE-SC0008110

   Recipient: University of Michigan

   Project Title: Probing High Temperature Superconductors with Magnetometry in Ultrahigh Magnetic Fields

   Principal Investigator: Lu Li

   Team Member: None

2. There are no limitations for authorized distributions.

3. **Executive Summary**

The objective of this research is to investigate the high-field magnetic properties of high temperature superconductors, materials that conduct electricity without loss. A technique known as high-resolution torque magnetometry that was developed to directly measure the magnetization of high temperature superconductors. This technique was implemented using the 65 Tesla pulsed magnetic field facility that is part of the National High Magnetic Field Laboratory at Los Alamos National Laboratory. This research addressed unanswered questions about the interplay between magnetism and superconductivity, determine the electronic structure of high temperature superconductors, and shed light on the mechanism of high temperature superconductivity and on potential applications of these materials in areas such as energy generation and power transmission. Further applications of the technology resolve the novel physical phenomena such as correlated topological insulators, and spin liquid state in quantum magnets.

4. **Comparison with the project goals and accomplishments**

The project has accomplished the major goals of torque magnetometry in pulsed magnetic fields. The major objective of high temperature superconductivity is achieved and the result is published in *Proceedings of National Academy of Sciences*. We further applied the high magnetic field torque magnetometry techniques to study topological materials and quantum magnets. The results are published in *Science, Physical Review X,* and *Physical Review B*.

5. **Project activities**.

(1). Revealing a charge-order-fluctuation transition and the vortex liquid state in underdoped cuprate high temperature superconductors (*Proceedings of National Academy of Sciences, 2016,* DOI:10.1073/pnas.1612591113).

The magnetic torque of single crystalline high temperature superconductors $YBa_2Cu_3O_{6.55}$ (YBCO) samples were also measured using the 60 T pulsed-field solenoid at the Los Alamos National Laboratory. To eliminate torques induced by eddy currents, we fabricated insulating cantilevers with the bottom faces metalized for capacitive detection of the deflection. To make measurements within the pulse window of 100 msec, we built a capacitance bridge operating at a frequency of 20 kHz.

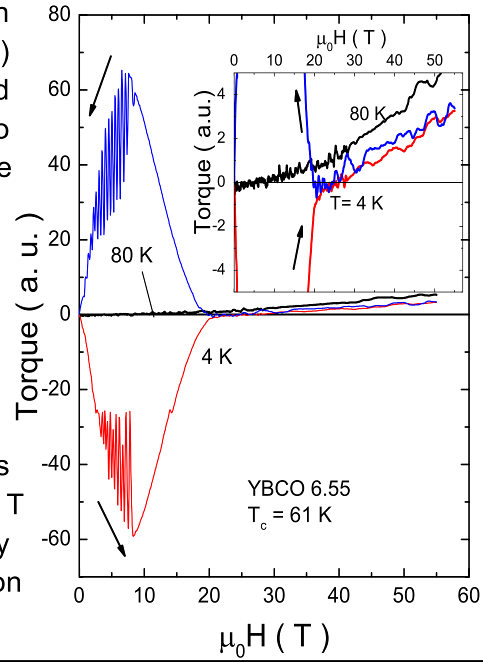

FIG. 1: Plots of the torque signal τ at 80 K and 4 K measured in ortho-II YBCO in a pulsed field up to 55 T, measured with $\theta \sim$ 10 degree. The curves of τ vs. H at 4 K is shown in red (H increasing) and blue (decreasing). Rapid oscillations below 10 T likely arises from collective flux motion. The trace at 80 K is shown in black. The insert displays the same data set in an expanded scale.

An example of the magnetization curve taken on ortho-II YBCO is shown in Fig. 1. The torque curve at 4 K is plotted in comparison with that at 80 K, well above T_c. At T = 80 K the torque τ vs. magnetic field H curve is perfectly parabolic, reflecting the weak superconducting fluctuation diamagnetism at 80 K. By contrast, at T = 4 K, the sample is deep in the superconducting state. Below 20 T, we observed hysteresis caused by flux pinning. As shown in the insert in Fig. 1, the hysteresis ends at the melting field H_m ~20 T. At $H > H_m$, the $\tau - H$ curve at T = 4 K is smaller than that at 80 K.

The difference between the torque curves at the two temperatures is the diamagnetic signal in vortex-liquid state. Furthermore, we observed that the torque signal shows a series of prominent spikes for H <10 T. These spikes may be caused by collective flux-bundle entry or exit (flux avalanches) within the vortex-solid state.

A detailed analysis shows that the measured *M-H* curves have a kink at H at ~ 22 T. We further carried detailed measurements of the diamagnetic signal M_d using the 35 T DC magnetic field in National High Magnetic Field Laboratory (NHMFL). At low T, the kink signature of H_K can be resolved in the differential susceptibility $\chi_d = dM_d/dB$ - as shown in Fig. 2. The curve of χ_d is initially featureless at 40 K. Below 40 K, a broad anomaly appears with a peak that grows with decreasing T. The peak occurs when the component of the field H equals 22 T. Such a kink feature in the magnetization, a thermodynamic measurement, suggests a magnetic-field-driven transition at a finite magnetic field around 22 T.

(2). Demonstrating the Dirac electronic surface states using quantum oscillations in topological Kondo insulator SmB_6. (*Science 2014* DOI: 10.1126/science.1250366, *Physics Review B* 2015 DOI: 10.1103/PhysRevB.92.115110).

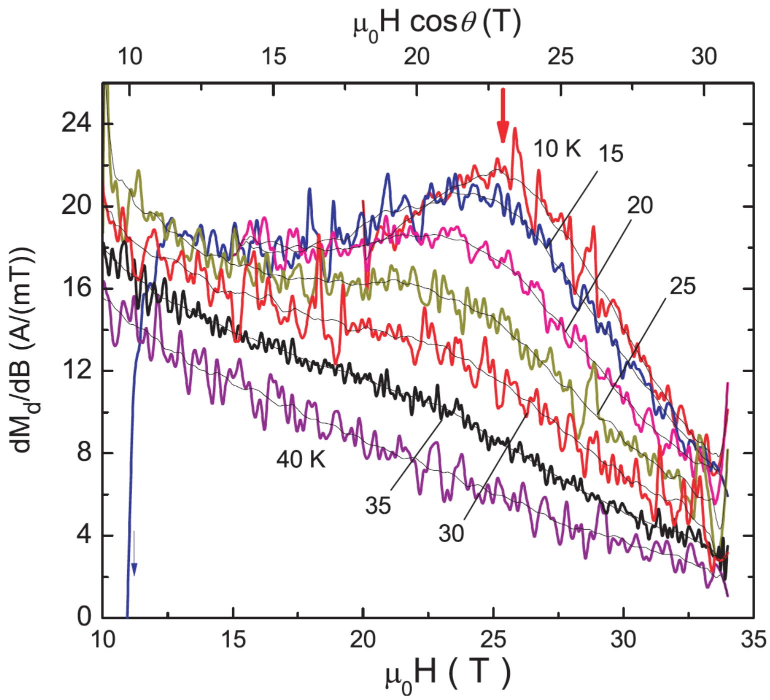

Fig. 2. The differential susceptibility $\chi_d = \partial M/\partial B$ plotted vs. $H_Z = H\cos\theta$ in YBCO (solid curves are digitally smoothed averages). The top axis is the magnetic field along the crystal c-axis. At 40 K, χ_d is a smoothly decreasing function of H. Below 40 K, a conical anomaly rises above this background, growing in height as T → 10 K. The peak occurs at H_K = 23 T (red arrow). The vertical drop in the curve at 15 K near 12 T signals the abrupt onset of hysteresis as H decreases below the melting field Hm.

The discovery of topological insulators and possible topological superconductors opens a new area of study and brings hope to more exciting applications such as topological quantum computation. Strong electronic interaction usually prevents simple surface states from forming. However, on the surface of Kondo insulator Samarium Hexaboride SmB_6, topologically protected surface states are predicted to exist as a result of a gap forming through the hybridization of the local orbital and the mobile electrons. The surface conductance has been demonstrated with non-local experiments in SmB_6. Our group has recently observed the dHvA effect in SmB_6. The angular dependence of the oscillation period shows that the observed Fermi surfaces are two-dimensional, and exists on both the (101) surface and the (100) surface.

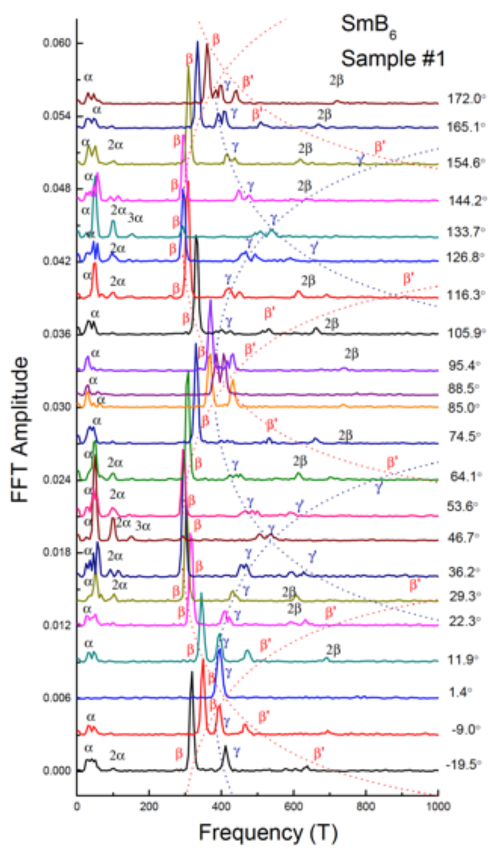

Fig. 3. The Fast Fourier Transformation (FFT) amplitude curves of the torque signal of SmB6 sample 1 at tilt angles expanding over 180 degrees. All the FFTs were taken on the torque curves measured in magnetic field up to 18 T at T = 300 mK.

High-resolution torque magnetometry was used to measure the magnetic moment of the samples. Torque is measured by the change of the capacitance between a metal cantilever and a nearby metal plate. Using a rotator probe we were able to map the dHvA oscillation frequencies of SmB_6 as a function of angle. Our torque magnetometry is able to resolve the magnetic oscillation (de Haas – van Alphen effect). Using the 45 T hybrid magnet and a regular superconducting magnet, we mapped the angular dependence of the quantum oscillation frequencies. Fig.3 shows the Fast Fournier Transformation FFT spectra of the magnetic torque vs. 1/H curves of a single crystalline SmB_6. Three set of Fermi surfaces and their higher harmonics are observed, and a strong angular dependence of oscillation frequencies indicates a strong anisotropy electronic state. Further analysis of the angular dependence shows that the frequencies are proportional to the inverse of sinusoidal functions, confirming the two-dimensional electronic state in SmB_6.

Our result of resolving the Fermi surface is the first step to verify the topological nature of the Kondo insulator SmB_6. We need to perform further tests to show the topological nature of the surface state. We have pushed the torque magnetometry measurement to 45 T to resolve the Landau level indexing plot. Fig.4 shows the Landau Level index plot for 3 observed Fermi pockets. Like those in graphene and Bi_2Te_3, the interval of the index is about -1/2, which results from the Dirac nature of the electronic state.

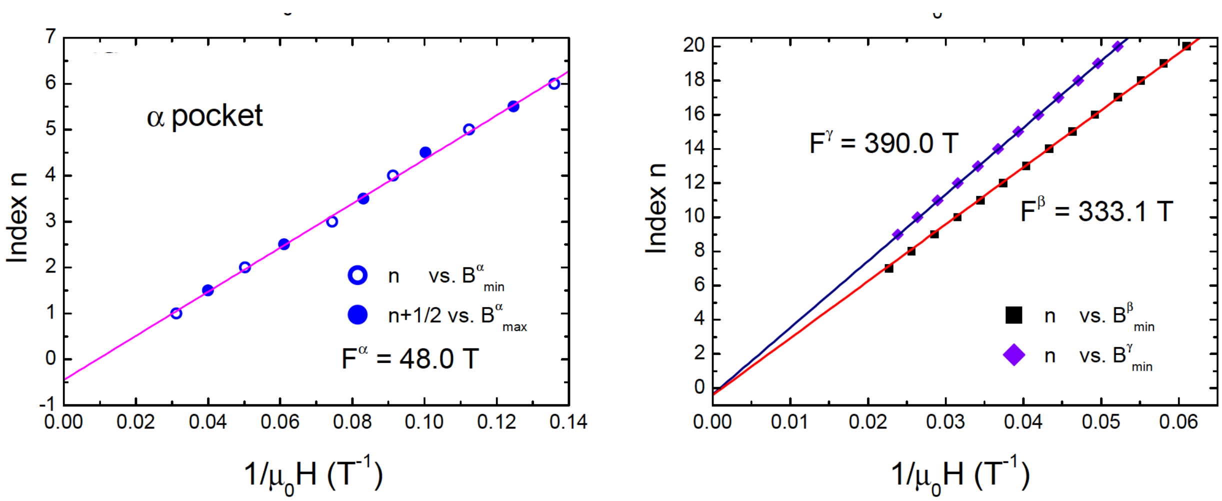

Fig. 4. (Left) The Landau Level number n is plotted against $1/B_n$ from Fermi pocket α (open circles). The maximum points of the susceptibility are also plotted against n + 1/2 for pocket α (solid blue circles). The infinite field limit of the index plot intercept is around -1/2. (right) The Landau Level index plot for Fermi pocket β and γ. Extrapolating the linear dependence of the index n vs. $1/B_n$ for frequencies also yields the intercept -1/2 at $1/B \rightarrow 0$.

An important aspect of these results is that we directly measured the magnetization of the single atomically layered surface states. Traditional experimental tools for studying surface states are photoemission, tunneling, and electrical transport. Thanks to our advanced magnetometry, we determined the magnetization of the surface state via torque for the first time. Our results open a new avenue for the detection of surface electronic states in novel materials.

(3). High Field Magnetic Ground State of Kagome Lattice Spin Liquid Herbertsmithite (*Physical Review B, 2014,* DOI:10.1073/pnas.1612591113).

An exciting field of modern condensed matter physics is the quantum spin liquid. The strong frustration leads to the lack of magnetic ordering and the emergence of novel physical phenomena in the ground state. Herbertsmithite $ZnCu_3(OH)_6Cl_2$ is a kagome lattice antiferromagnet with 1/2 spin and has been demonstrated to be a likely candidate of spin liquid by recent neutron scattering measurements. However, the direct determination of the magnetic ground state at extremely low temperature has always been challenging because of two reasons: (a) The direct magnetization measurements are quite challenging to perform at extremely low temperatures; and (b) In the Zn plane, there are extra free Cu moments whose Curie-like paramagnetic signal dominates the magnetic response at low temperature. To solve the challenge, we applied torque magnetometry on single crystalline Herbertsmithite using high-resolution cantilever setups. Using high field torque magnetometry, we measured the angular dependence of the magnetic torque and the magnetization of Herbertsmithite at 20 mK < T < 40 K and in intense magnetic field. Shown in Fig. 5, the intrinsic magnetization from the kagome lattice turns out to be linear with magnetic field, and the magnetic susceptibility is independent of temperature at 20 mK < T < 5 K.

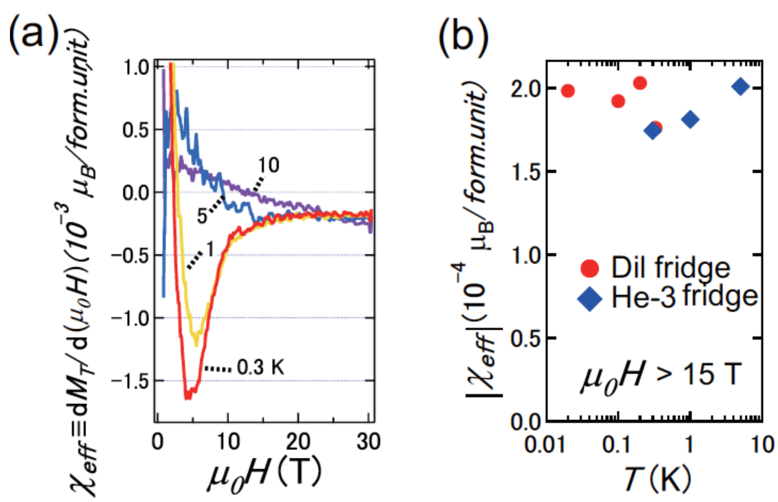

Fig.5 (a) Effective magnetic susceptibility χ_{eff} is plotted as function of field H up to 31 T for single crystal Herbersmithite. (b) The temperature T dependence of the absolute value of the high-field χ_{eff} of the kagome lattices.

Our results show that the high magnetic field phase of Herbertsmithite exists as a gapless spin liquid ground state. Such a state is not affected by the field-driven anomalies at 7 T – 15 T, which possibly originate from the interlayered Cu spins. Far from the gapped spin liquid state expected by theorists, our result is the first demonstration of the gapless spin liquid ground in Kagome-lattice quantum magnets.

(4). Observation of breaking of the rotational symmetry in a topological superconductor candidate Nb-doped Bi_2Se_3 (*Physical Review X, 2017* DOI: 10.1103/PhysRevX.7.011009, *Physical Review B, 2016* DOI: 10.1103/PhysRevB.94.041114, *Physical Review B, 2014* DOI: 10.1103/PhysRevB.90.195141).

Unconventional superconductors are characterized by superconducting order parameters that are non-invariant under crystal symmetry operations. When the order parameter is single-component, this non-invariance is manifested solely in the phase of the superconducting wavefunction, and can only be detected by phase-sensitive measurements. On the other hand, when the order parameter is multi-component, the magnitude of the superconducting gap can be different along symmetry-related crystallographic directions. The gap anisotropy directly leads to thermodynamic property of the superconducting state that spontaneously breaks the crystal rotational symmetry of the normal state. However, direct thermodynamic signature of rotational symmetry breaking due to superconductivity has not been found in any crystals.

In this work, we applied torque magnetometry to measure the magnetic anisotropy in the ab crystalline plane in the superconducting state of niobium (Nb)- doped bismuth selenide (Bi_2Se_3). We observed direct evidence that the superconducting magnetic response couples strongly to the underlying 3-fold crystal symmetry in the recently discovered superconductor with trigonal crystal structure, Nb-doped Bi_2Se_3, shown in Fig. 6(A).

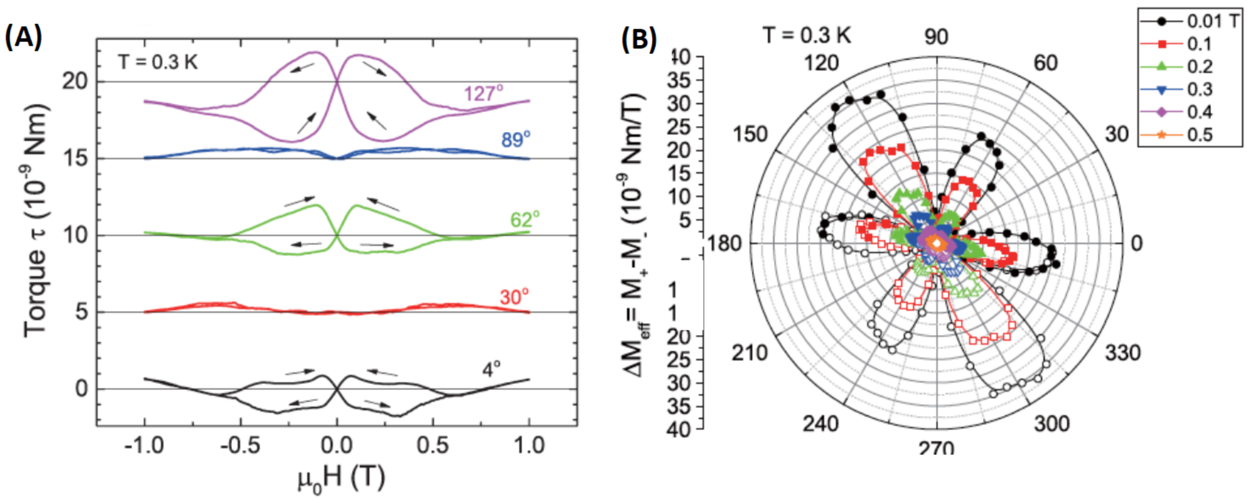

Fig. 6. (A) Selected torque curves at 0.3 K of Nb-doped Bi_2Se_3 with external magnetic field between -1 and 1 T swept in the crystalline a-b planes. The magnitude of the hysteresis loop is maximum at around 120° and is nearly zero at 30° and 90°. (B) Polar plot of spontaneous magnetization $\Delta M_{eff} = M_+ - M_-$, where M_+ is the magnetization signal from the up-sweep of the magnetic field, and M_- is the magnetization signal from the down-sweep of the magnetic field. Angle ϕ is defined as the tilt angle between the crystalline a- axis and the magnetic field direction.

More importantly, we observed that the magnetic response is greatly enhanced along one preferred direction spontaneously breaking the rotational symmetry, shown in Fig. 6(B). Instead of a simple 3-fold crystalline symmetry, the superconducting hysteresis loop shows dominating 2-fold and 4-fold symmetry. This observation confirms the breaking of the rotational symmetry and indicates the presence of nematic order in the superconducting ground state of Nb-doped Bi_2Se_3. Our further heat capacity measurements display an exponential decay in superconducting state and suggest that there is no line node in the superconducting gap. These observations provide strong evidence of odd-parity topological superconductivity.

6. **Product**

Publications:

(1). G. Li, Z. Xiang, F. Yu, T. Asaba, B. Lawson, P. Cai, C. Tinsman, A. Berkley, S. Wolgast, Y. S. Eo, Dae-Jeong Kim, C. Kurdak, J. W. Allen, K. Sun, X. H. Chen, Y. Y. Wang, Z. Fisk and Lu Li "Two-dimensional Fermi surfaces in Kondo Insulator". Science 346, 1208 (2014).

(2). Tomoya Asaba, Tian-Heng Han, B. J. Lawson, F Yu, C Tinsman, Z Xiang, G Li, Young S Lee, and Lu Li "High-field magnetic ground state in S=1/2 kagome lattice antiferromagnet $ZnCu_3(OH)_6Cl_2$". Physical Review B 90, 064417 (2014).

(3). BJ Lawson, G Li, F Yu, T Asaba, C Tinsman, T Gao, W Wang, YS Hor, and Lu Li "Quantum oscillations in CuxBi2Se3 in high magnetic fields". Physical Review B 90, 195141 (2014).

(4). Steven Wolgast, Yun Suk Eo, T. Ozturk, G. Li, Z. Xiang, C. Tinsman, T. Asaba, B. Lawson, F. Yu, J. W. Allen, K. Sun, Lu Li, C. Kurdak, D-J Kim, Z Fisk "Magnetotransport measurements of the surface states of samarium hexaboride using Corbino structures". Physical Review B 92, 115110 (2015).

(5). B. J. Lawson, Paul Corbae, G. Li, F. Yu, Tomoya Asaba, Colin Tinsman, Y. S. Qiu, J. E. Medvedeva, Y. Hor, and Lu Li, "Multiple Fermi surfaces in superconducting Nb-doped Bi_2Se_3", Physical Review B, 94, 041114(R) (2016).

(6). Colin Tinsman, G. Li, Caroline Su, Tomoya Asaba, B. J. Lawson, F. Yu, and Lu Li, "Probing the thermal Hall effect using miniature capacitive strontium titanate thermometry", Applied Physics Letters, 108, 261905 (2016).

(7). Fan Yu, Max Hirschberger, Toshinao Loew, Gang Li, Benjamin J. Lawson, Tomoya Asaba, J. B. Kemper, Tian Liang, Juan Porras, G. S. Boebinger, J. Singleton, B. Keimer, Lu Li, and N. P. Ong, "Magnetic phase diagram of underdoped YBa2Cu3O6.6 inferred from torque magnetization and thermal conductivity", Proceedings of the National Academy of Sciences, 113, 12667(2016).

(8).Tomoya Asaba, B.J. Lawson, Colin Tinsman, Lu Chen, Paul Corbae, Gang Li, Y. Qiu, Y.S. Hor, Liang Fu, Lu Li. "Rotational Symmetry Breaking in a Trigonal Superconductor Nb-doped Bi_2Se_3". Physical Review X 7, 011009 (2017).