MERIT Long-Term Overseas Dispatch

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Topic : Low-temperature physics of organic spin liquid candidates

Abstract

I have been working on my research project since June last year at the Max Planck Institute of Solid State Physics in Stuttgart, Germany. This report presents the research results up to June 30, 2024. My project is ongoing in the Department of Quantum Materials under the supervision of Prof. Takagi, my supervisor at the University of Tokyo. I have measured the magnetic susceptibility of an organic triangular lattice compound that is known as a quantum spin liquid (QSL) candidate.

Research Details

QSL states were first suggested in triangular-lattice antiferromagnetic systems with spin-1/2[1]. κ-(BEDT-TTF)$_2X$ (X: anions) compounds have been investigated as organic QSL candidate materials. Forming a dimer on each BEDT-TTF molecule in conducting layers, the system changes into a Mott insulator with localized electrons at low temperatures and exhibits antiferromagnetic interactions between spins.

The precise, ultra-low temperature susceptibility measurements are established in MPI. Besides, microscopic electron-spin resonance (ESR) measurements have been done on the same single crystals by collaborating with the Dressel Group at Stuttgart University. We have aimed to measure the low-temperature states in κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ and κ-(BEDT-TTF)$_2$Ag$_2$(CN)$_3$ through the measurements above.

The susceptibility measurements by SQUID confirmed the reproducibility of temperature-dependence of χ_{spin} in high temperatures under 1 T of magnetic field among six single crystals. The derivative of susceptibility, $d\chi_{\mathrm{spin}}/dT$, exhibits a sharp peak at around 6 K, which is consistent with previous research. A finite susceptibility remains down to 50 mK revealed by the Faraday magnetometer with dilution refridge, indicating low-energy excitations in spins (Fig. 1 (a)). We succeeded in measuring the magnetization for the first time, although it shows different field/temperature dependence from previous torque magnetization at very low temperatures. The external field raises the magnetization linearly and the saturation of the magnetization, as generally observed for impurity spins, was not detected under the field up to 7 T (Fig. 1 (b), (c)).

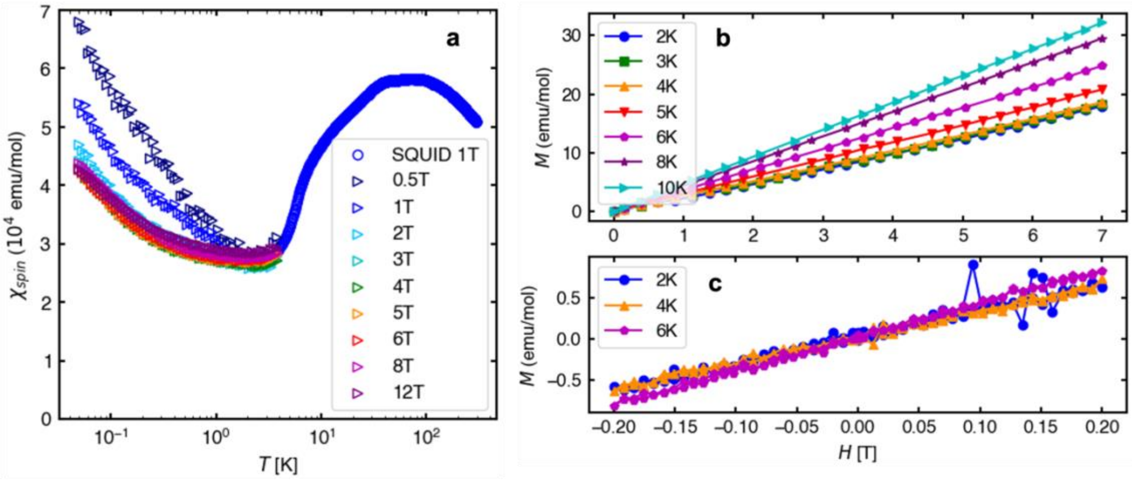

Figure 1 (a) Temperature dependence of magnetization by Faraday magnetometer measurement. (b), (c) Change in magnetization by applying the field.

On the other hand, the anisotropic behavior of the spin g-factor increases below 3 K, as figured out by ESR measurements from the collaborator at the University of Stuttgart. The internal magnetic field detected by nuclear magnetic resonance (NMR) measurements shows nontrivial broadenings below 3 K[3], indicating that the spin states differ from normal paramagnetic states. The non-linear responses of the internal field to the applied field have also been detected in previous μSR measurements.

Based on the results above, it is believed that the field-induced inhomogeneous and magnetic-order-like internal field appears in κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$. The phase transition has not yet been confirmed so the possibility of a QSL remains. Specific heat or ESR at ultra-low temperatures are planned for further measurements.

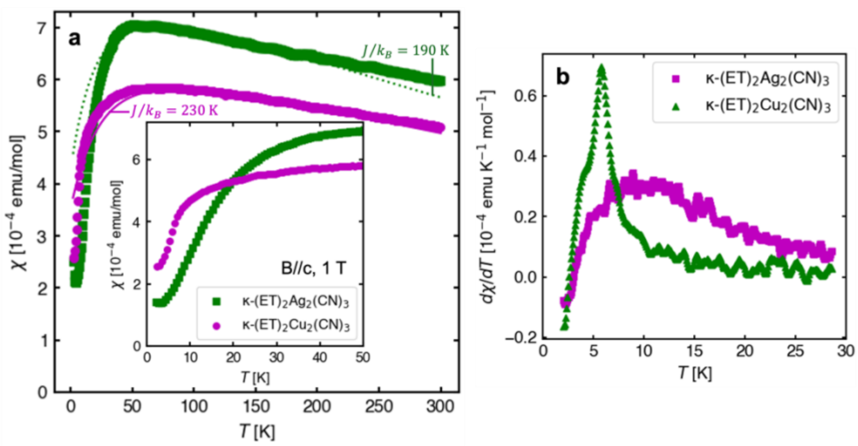

Figure 2 Susceptibility (a) and its temperature derivative (b) obtained from SQUID measurements.

The compound κ-(BEDT-TTF)$_2$Ag$_2$(CN)$_3$, Ag instead of Cu, is recognized as a QSL candidate with a larger electron correlation compared to the Cu compound. The susceptibility measurement by SQUID has reproduced the finite susceptibility, although it is smaller than that of the Cu compound, down to 2 K in several samples. The temperature derivative of susceptibility shows no anomalies, which differs from the behavior observed in the Cu compound. For the next step, ESR measurements are planned to investigate the field dependence of the internal field on κ-$(BEDT\text{-}TTF)_2Ag_2(CN)_3$.

[1] P.W. Anderson, Mater. Res. Bull. **8**, 153 (1973).
[2] Y. Shimizu *et. al.*, PRL**91**, 107001(2003).
[3] K. Kanoda, private communication.

Figure 3 German cuisine.

Life in Stuttgart

The institute is located about 10 km from the city center in Stuttgart, a lush and relaxed place surrounded by forest. I often go to the university, 20 minutes away from the institute on foot. The path to the university runs through the forest and walking along the road makes me feel refreshed. In the research process, it is sometimes necessary to ask for help from the technicians. Students need to clearly specify the types of parts they want—shapes, sizes, etc.—and negotiate directly with the technicians (sometimes some people only speak German).

For the first two months of my stay, I lived in the guest house next to the institute building, which was just a 10-second walk away. Since then, I have been renting a room that is 30 minutes away from the institute. Although the room doesn't have air conditioning systems, summer in Germany is cool and comfortable. Simultaneously, it is warm enough with a heater in winter because the room is well-insulated. Regarding food, German cuisine tends to feature a lot of meat and potatoes, so I mainly cook vegetables at home to give my stomach a break.

Acknowledgments

During my stay, I received guidance and encouragement from many individuals. I appreciate Prof. Takagi for accepting my stay and for your advice. I thank the collaborators, Sudip from the University of Stuttgart for the ESR measurements, Matsumoto-san for the Faraday magnetometer measurements, and Prof. Kanoda and Sekine-san for the discussions. κ-$(BEDT\text{-}TTF)_2$ $Cu_2(CN)_3$ single crystals are given by Prof. Sasaki from Tohoku University. In addition, I would like to express my gratitude to the co-supervisor Prof. Seki involved in this MERIT long-term overseas dispatch, as well as to the staff of the MERIT office. This stay was made possible thanks to the financial support from the Max Planck Institute. I sincerely appreciate it.