Searching for a Kitaev spin liquid in α-$Ru_{1-x}Ir_xCl_3$

The ruthenium trichloride α-$RuCl_3$ has recently garnered tremendous research interest as a possible realization of the celebrated s=1/2 Kitaev honeycomb magnet that harbors quantum and topological spin liquids. During my research stay at the IMR, we have conducted specific heat measurements of α-$Ru_{1-x}Ir_xCl_3$ (x~0.2) with a view to examining the robustness of Kitaev physics under spin vacancy. Our detailed temperature- and field-dependent specific heat data unveil a quantum-critical-like behavior for H//plane and a field-induced unknown phase above 5 T for H⊥plane.

The search for Kitaev quantum spin liquids and concomitantly occurring Majorana fermions is one of the most intensively pursued goals in contemporary condensed matter physics. In particular, Majorana fermions have been sought after due to their applicability to fault-free quantum computation.

In the quest for quantum spin liquids, the s=1/2 Kitaev model on a honeycomb lattice was proposed by Alexei Kitaev in 2006 as a new platform that hosts a quantum spin-liquid ground state and two types of Majorana fermions: itinerant Majorana fermions and Z_2 static fluxes.

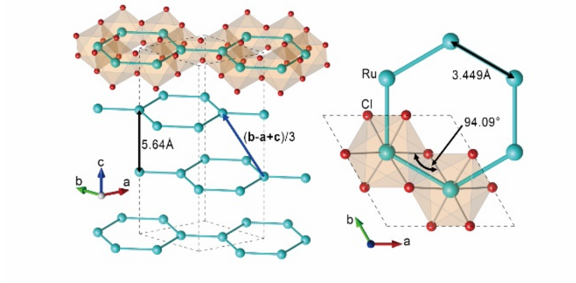
Since then, an enormous amount of theoretical and experimental work has been devoted to verifying these exotic quasiparticles in Kitaev candidate materials. Because the Kitaev model is analytically solvable, it has a significant advantage in identifying thermodynamic and spectroscopic features of a spin liquid over other spin-liquid systems. Nonetheless, people have soon found that real materials contain unavoidably non-Kitaev interactions that stabilize magnetic order at low temperature while preempting an expected Kitaev spin-liquid ground state.

The detrimental effects of the non-Kitaev terms can be partly nullified by the application of an external magnetic field as well as by the introduction of spin vacancies. During my research visit to the IMR, we have explored the possibility of achieving a quantum spin liquid in the diluted α-$Ru_{1-x}Ir_xCl_3$.

α-$RuCl_3$ is closest to the Kitaev candidate materials reported so far and constitutes honeycomb lattices of octahedrally coordinated Ru^{3+} (J_{eff}=1/2) ions as shown in Fig. 1. α-$RuCl_3$ shows the zigzag antiferromagnetic order T_N~7 K, thereby obscuring the theoretically predicted Kitaev quantum spin liquid.

One route to suppress the zigzag magnetic order is by introducing nonmagnetic impurities to α-$Ru_{1-x}Ir_xCl_3$ [3]. Earlier studies of α-$Ru_{1-x}Ir_xCl_3$ have suggested the presence of a spin-liquid-like state in the wide range of x. Especially, α-$Ru_{0.8}Ir_{0.2}Cl_3$ is located in the vicinity of a quantum critical point. Considering the intriguing role of nonmagnetic impurities, a careful study of the ground state is urgently requested.

Specific heat is a powerful tool for probing low-energy magnetic excitations in quantum magnets. The high-field lab, IMR provides extreme conditions for specific heat experiments.



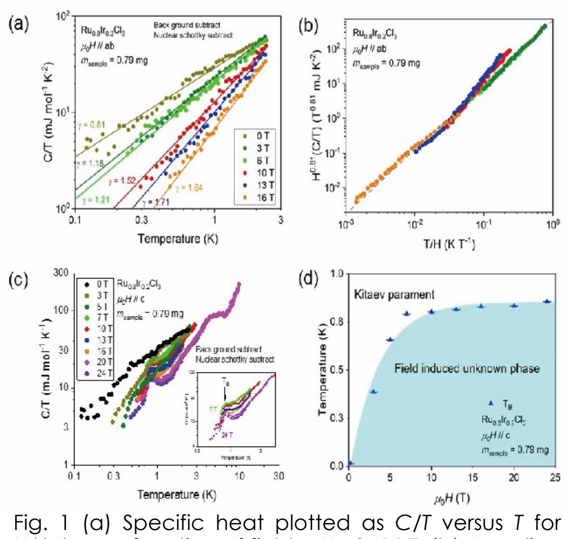
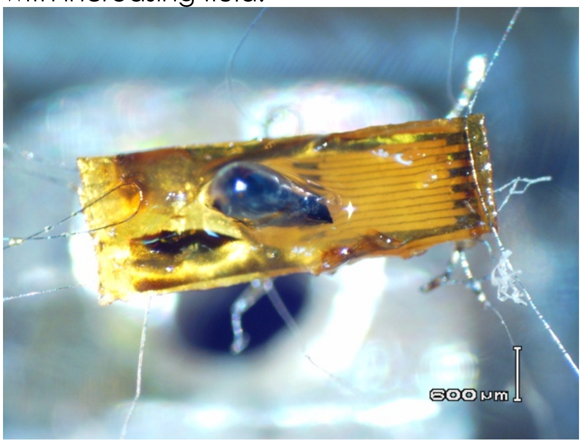


Fig. 1 (a) Specific heat plotted as C/T versus T for $H//ab$ as a function of field μ_0H= 0- 16 T. (b) A scaling plot of $H^{0.81}(C/T)$ versus T/H. (c) C/T versus T for $H//c$ (d) T-H phase diagram of α-$Ru_{0.8}Ir_{0.2}Cl_3$.

We measured low-temperature specific heat using the 20 T superconducting and the 25 T Cryogen-free superconducting magnet equipped with dilution fridge and ^{3}He cryostats. The data were taken in the temperature range of T=0.12-2.5 K and the field range of μ_0H=0-24 T for the external field parallel ($H//ab$) and perpendicular ($H//c$) to the honeycomb plane.

In Fig. 1(a), the low-T specific heat $C(T)$ of α-$Ru_{0.8}Ir_{0.2}Cl_3$ is plotted on the log-log scale in the in-plane fields of μ_0H= 0 - 16 T. Upon cooling in zero fields, C/T shows a power-law decrease of $C/T \propto T^{0.81}$, being close to the T dependence of what is expected for a quantum spin liquid with a Dirac node. With increasing field, the exponent gradually increases towards $C/T \propto T^{1.84}$. This suggests the presence of highly degenerate low-lying excitations around energy E = 0. The low-energy density of states is gradually depleted with increasing field.



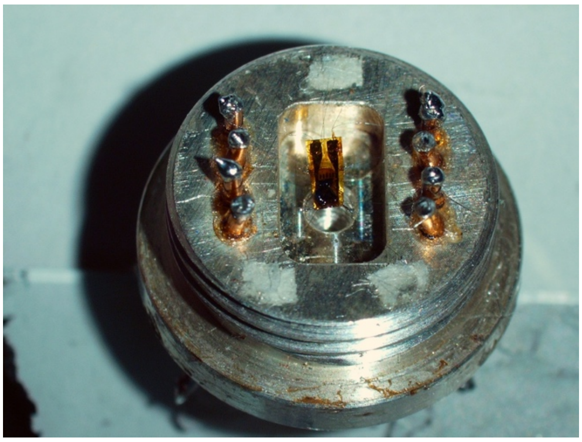


Fig. 2 An exxample of the sample stage for heat capacity measurement in very low temperature.

In the examination of the scaling relation, we plot $H^{\alpha}C/T$ vs T/H in Fig. 1(b). We find the data collapse of C/T onto a single curve over three orders of magnitude with the scaling exponent α=0.81. The power-law scaling is consistent with quantum-critical-like behavior. Indeed, it has been observed in a class of quantum spin liquids with bond randomness [4, 5]. At the moment, dc magnetic susceptibility and magnetization measurements are underway to crosscheck the universal quantum scaling behavior in thermodynamic quantities.

Fig. 1(c) shows the out-of-field dependence of the low-T specific heat plotted on a log-log scale of C/T vs T. With the application of the magnetic field above 5 T, the weak magnetic anomaly starts to appear at 0.7 K and slightly increases with increasing field up to 24 T. For $H//c$ the T-H phase diagram of α-$Ru_{0.8}Ir_{0.2}Cl_3$ is summarized in Fig. 1(d).

It is quite striking that the magnetic behavior relies strongly on the field-orientation direction. Possibly, this is linked to a large anisotropy of the pristine α-$RuCl_3$. We have a plan for torque magnetometer experiments to draw a complete phase diagram.

In summary, our temperature- and field-dependence of the specific heat unravels a highly anisotropic magnetism of the diluted α-$Ru_{0.8}Ir_{0.2}Cl_3$. Our results suggest that α-$Ru_{0.8}Ir_{0.2}Cl_3$ is proximate to a quantum critical point. In future work, we will elucidate whether the spin-liquid-like state in α-$Ru_{0.8}Ir_{0.2}Cl_3$ has a Kitaev spin-liquid flavor.

I am grateful to Prof. Hiroyuki Nojiri and Nojiri group members for the generous support of this research project and their hospitality during my stay at the IMR. This work was supported by ICC-IMR, Tohoku University.

References

[1] A. Kitaev, Ann. Phys. (Amsterdam) **321**, 2 (2006).
[2] S.-H. Do *et al.*, Nat. Phys. **13**, 1079 (2017).
[3] S.-H. Do *et al.*, Phys. Rev. B **98**, 014407 (2018).
[4] K. Kitagawa *et al.*, Nature (London) **554**, 341 (2018).
[5] Y.S. Choi *et al.*, Phys. Rev. Lett. **122**, 167202 (2019).

Keywords: Kitaev honeycomb lattice, Spin vacancy, quantum criticality
Prof. Kwang-Yong Choi (Department of Physics, Chung-Ang University, Korea)
E-mail:kchoi@cau.ac.kr
http://physics.cau.ac.kr