μ^+SR Knight shift of the Mott insulator κ-(ET)$_4$Hg$_{2.78}$Cl$_8$

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The hole-doped organic superconductor κ-(ET)$_4$Hg$_{3\text{-}\delta}$Br$_8$, (κ-HgBr), where δ = 11% and ET = bis (ethylenedithio) tetrathiafulvalene, has been the key to bridge the knowledge gap between half-filled organics and doped cuprate systems, as the well-known Mott-Hubbard model in the laboratory. Usually, organic superconductivity appears under pressure when the ratio of Hubbard interaction and bandwidth, U/W, is larger than 1. Nonetheless, the triangular lattice of organics, unlike the square lattice in cuprates, provides extensive geometrically control through nearest, t, and next-nearest, t', transfer integrals between sites. In the case of geometrically triangular frustration ($t \sim t'$), the Mott insulating state cannot be magnetically ordered down to the miliKelvin (mK) order, becoming a Mott quantum spin liquid.[1)] Specifically, both hole-doped superconductors have a region corresponding to a strange metallic state at which resistivity exhibits a linear temperature dependence, $\rho \propto T$, which is not a Fermi-liquid (FL) behavior.[1–3)]

In 2015, an important model to study strange metal was developed by Sachdev-Ye-Kitaev (SYK).[4,5)] It consists of Majorana fermions with random all-to-all interactions, leading to quantum information science and quantum many-body physics. Although the realization of the SYK-strange metal remains complex, Tsuji-Werner theoretically found an SYK-strange metal region after an out-of-time-order correlation treatment of a multi-band Hubbard model.[6)] In the strange metal region, the electron behaves as a system having a non-FL electronic scattering rate with $\rho \propto T$. In this region, the spin fluctuates strongly while adjusting the competition between the localized spin in the Mott insulating state and the itinerant one in the FL state, and U/W should be close to unity, which is known as a quantum critical point (QCP).[6)] Cha *et al.* further showed that when the itinerant spin-1/2 fermions interact via onsite U and random infinite-ranged spin-spin interaction, the QCP appears with quantum spin liquid dynamics, identical to that of SYK-local spin dynamics.[7)] On the other hand, the growth of antiferromagnetic spin fluctuations (AFSFs) towards low temperature without any long-range order can lead to the QCP at which a non-FL behavior is observed, like in the heavy fermion system.[1)]

Our experiment[8)] showed that the time-reversal symmetry is preserved in the superconducting state of κ-HgBr down to 0.3 K, narrowing the similarity down to that of cuprates despite the triangular lattice. To seek evidence of strong AFSFs towards the Mott spin-liquid ground state, we also study the sister insulating compound, which has a different doping content yet slightly higher U/W than that of κ-HgBr, κ-(ET)$_4$Hg$_{3\text{-}\delta}$Cl$_8$ (κ-HgCl), with δ = 22%. It shows a metal-insulator transition at $T_{\mathrm{MI}} = 20$ K and ambient pressure.

The signature of strong AFSFs has been observed in κ-HgBr, in which the temperature dependence of μ^+ Knight shift, $K(T)$, is not proportional to that of susceptibility, $\chi(T)$ in the low-temperature region below 50 K, unlike in other κ-type organic superconductors; this signature is often found in heavy-fermion system.[1)] One possible reason is that μ^+ probes a hyperfine coupling constant different from itinerant electrons in the high-T region and localized electrons. From the linear part of the K-χ plot from 50 to 300 K, the hyperfine coupling constant, A_{hf}, in κ-HgBr was reported to be 166 Oe/μ_{B}. If this deviation is a signature of such a different hyperfine coupling constant, we naively expect a larger A_{hf} in κ-HgCl.

We have followed the same experimental condition to measure the μ^+ Knight shift in κ-HgCl with that of κ-HgBr.[9)] The measurement was performed using DC muon beam on the NuTime spectrometer in M15 beamline, TRIUMF Canada, in a field of 6 T. Figure 1 shows the measured μ^+SR time spectra after Fourier transforming. From 300 down to 2 K, the central peak is gradually shifted to the positive side, and a prominent shift was detected below T_{MI}. $K(T)$ follows Curie-

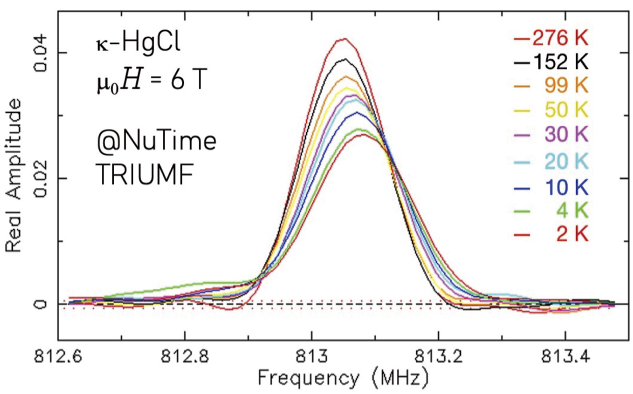

Fig. 1. Fourier transform of μ^+SR time spectra in the field of 6 T in κ-(ET)$_4$Hg$_{2.78}$Cl$_8$ measured at several temperatures, represented by colorful lines, from 276–2 K.

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Weiss paramagnetic behavior well while $\chi(T)$ has a broad peak at 70 K. From the linear region of the K-χ plot above 100 K, our preliminary analysis obtained $A_{\mathrm{hf}} = 230(20)$ Oe/μ_{B}. The value is about 40% larger than that of κ-HgBr. Next, we are going to study the spin dynamics of κ-HgCl down to the mK order.

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