μSR Study of Hole-Doped Organic Metals κ-$(ET)_4Hg_{3-\delta}X_8$; X = Br, Cl
"Superconductivity Nearby Quantum Spin Liquid States"

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Motivations

Strange Metal is abundant in the strongly correlated electron systems, as the normal state of the optimum T_c regime of the superconducting, e. g., cuprates and organics [1].

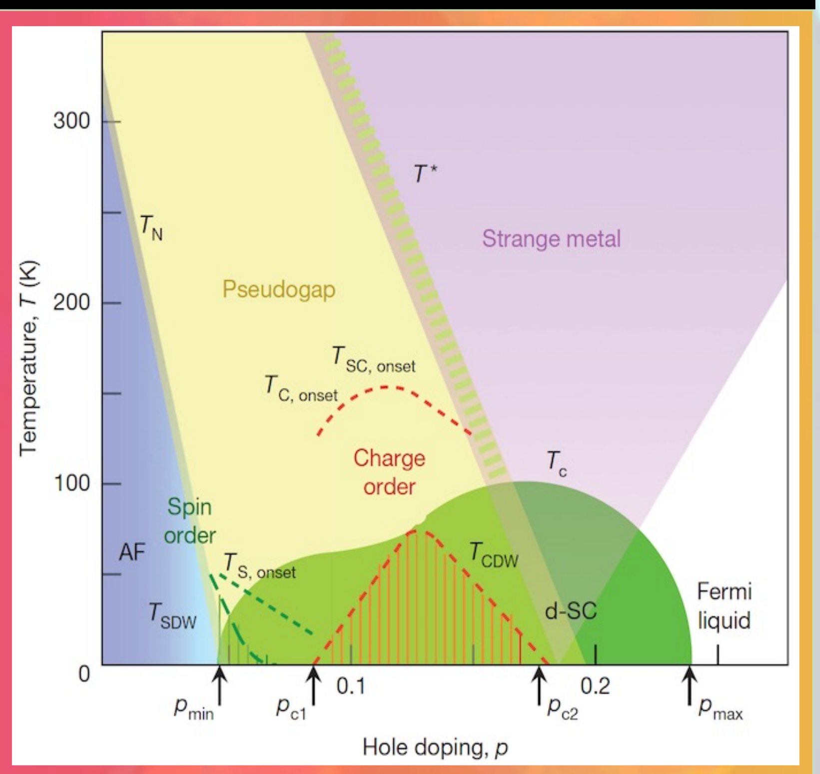
Non-Fermi Liquidity of the incoherent metal is phenomenologically characterized by:

- frequency dependence of electronic scattering rate: $Im\sum(\omega)\sim\sqrt{\omega}$ [2]
- temp. dependence of resistivity: $\rho\sim T$ [3; Ref. therein]
- collision time in resistivity ~ $\hbar/(k_BT)$ [3]
- current flow without quasiparticle [3]
- no long-range order, towards QCP [2]

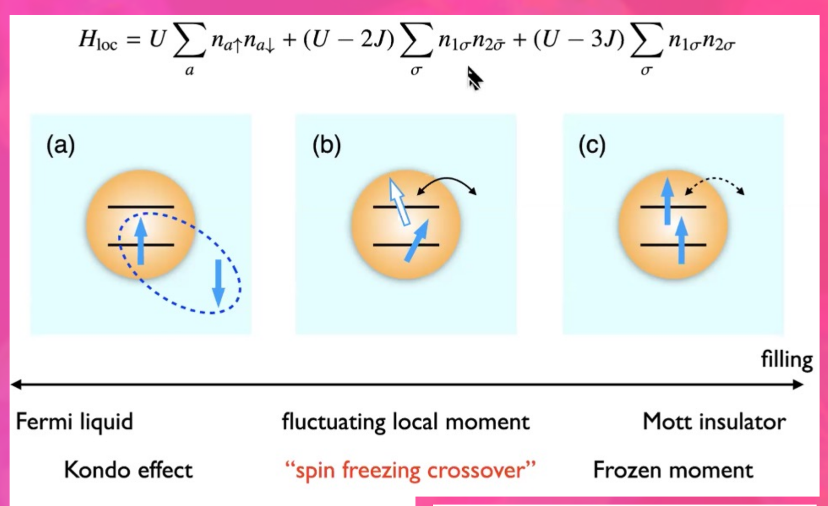
Sachdev-Ye-Kitaev (SYK) model [4–6] describes strange metal (0+1 dimension) dual to black hole in the Anti-de-Sitter (0+2 dimension) AdS_2 horizon → Holography [7]

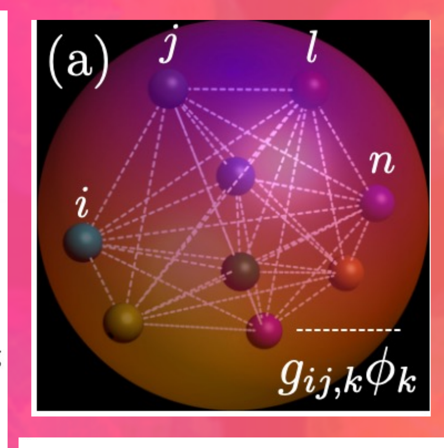
Tsuji-Warner Phase diagram: 4-point out-of-time-order-correlator of Multi-orbital Hubbard model. Strongly fluctuating spins in the spin freezing crossover has the SYK strange metal characteristic [8].

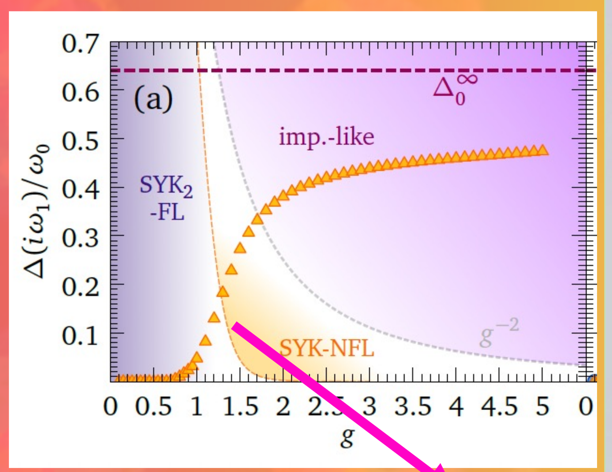
Kim-group showed phase diagram for the ½-filled system [ ].

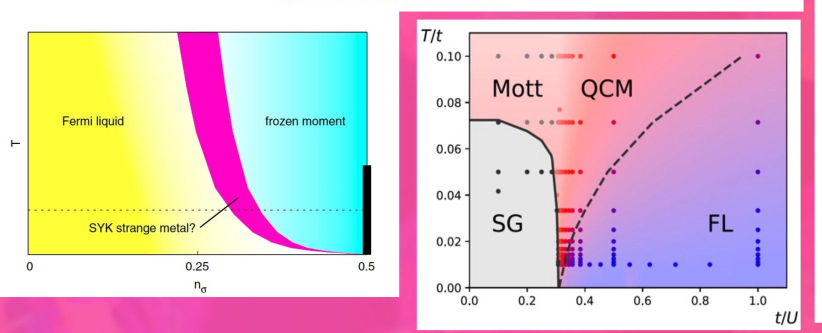


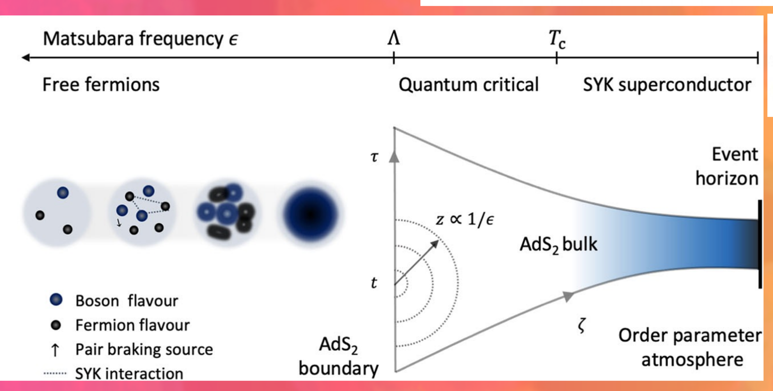
Schmalian-group demonstrated that the Yukawa-SYK superconductor is characterized by coherent quasiparticle excitations and higher-order bound states thereof [9]. Furthermore, it is holographic and quantum critical *conventional* Eliashberg theory [10,].







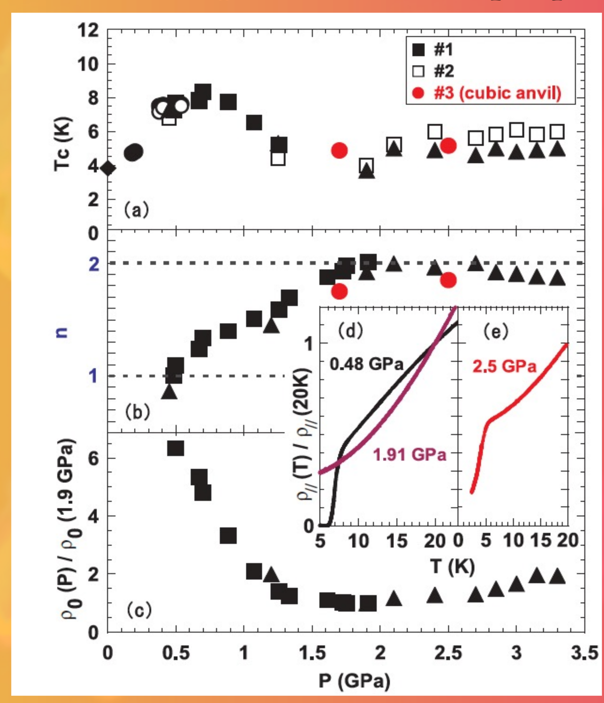




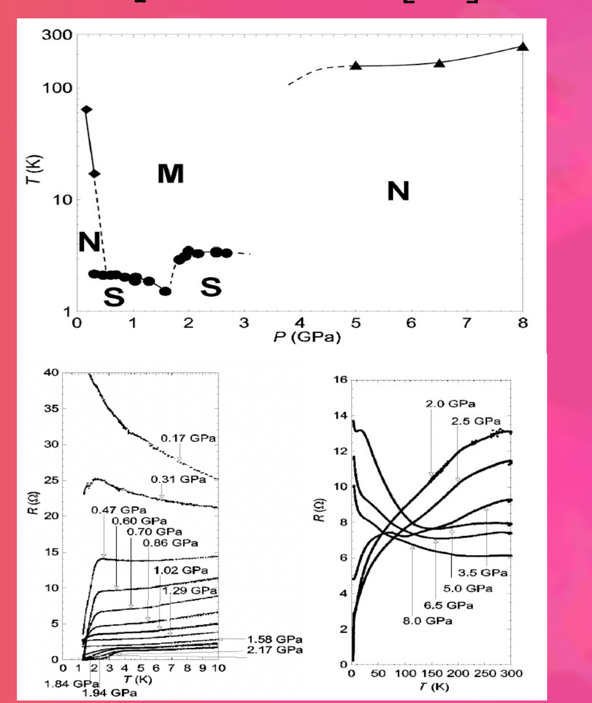
Questions: Can we experimentally observe and characterize the order parameter of a holographic superconductor in actual Non-Fermi Liquid material?? We focus on an organic strange metals which undergoes superconducting or Mott insulating.

Doped Spin Liquid Candidate

κ-$(ET)_4Hg_{3-\delta}Br_8$ (δ=0.11), κ-HgBr
Superconductor; T_c ~ 4.3 K
Pressure Induced Non-Fermi Liquid – Fermi Liquid Crossover [12]

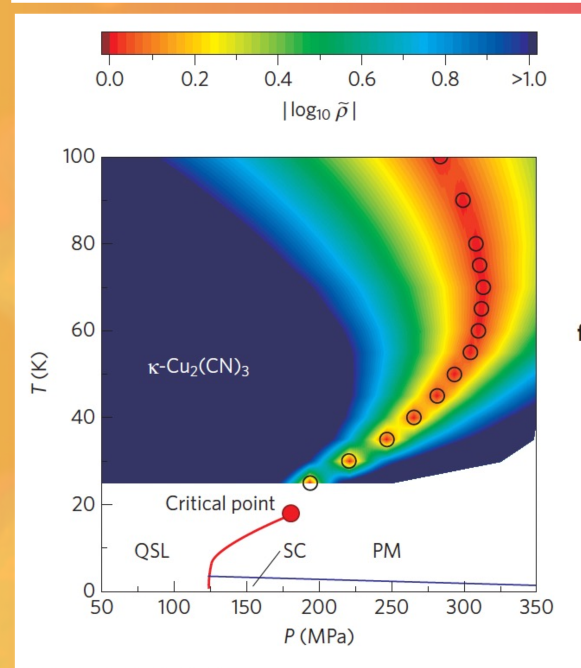


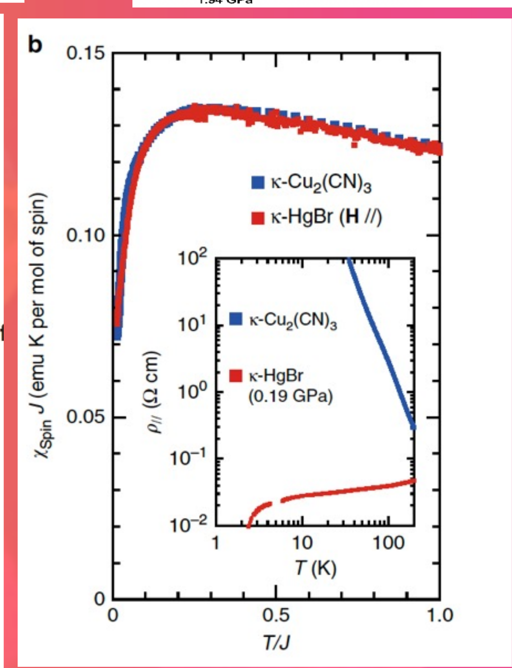
κ-$(ET)_4Hg_{3-\delta}Cl_8$ (δ=0.22), κ-HgCl
Metal – Insulator; T_{MI} ~ 20 K
Mott Insulator, Pressure Induced Superconductor [12]

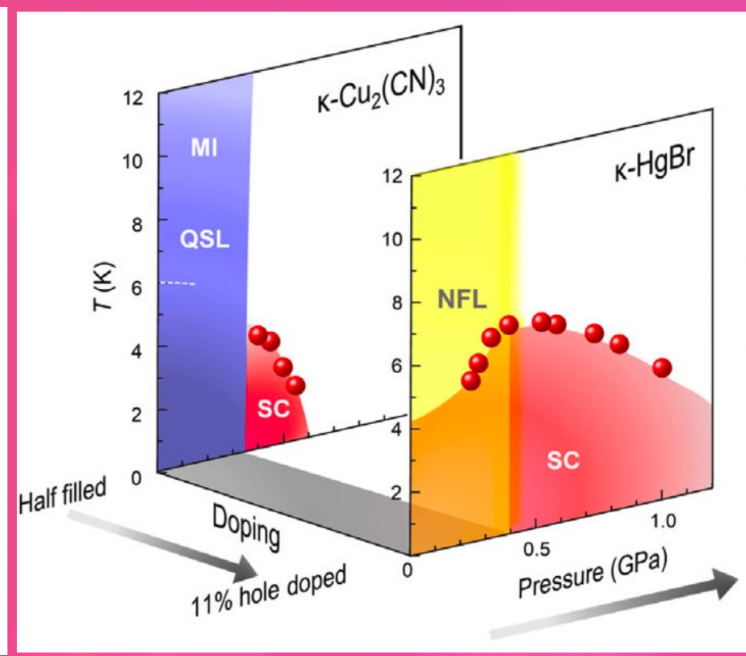


Previous studies:

a. κ-HgBr is a doped spin liquid candidate due to triangular lattice [13].
b. κ-HgCl; NMR showed inhomogeneous spin state below 2 K [19]
c. From μ^+SR, ^{13}C-NMR, and heat capacity → Non-Fermi liquidity is originated by strong AF spin fluctuation → both systems are close to the QCP. [14, 15, 16].

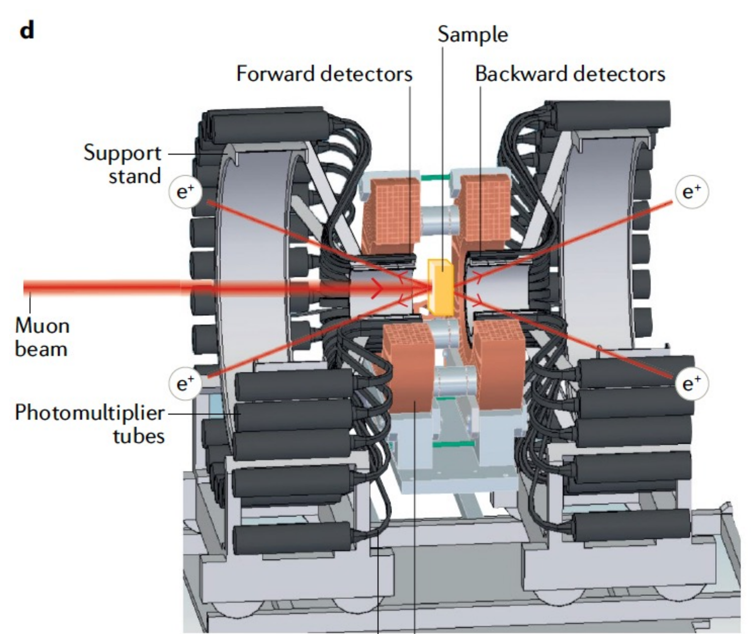


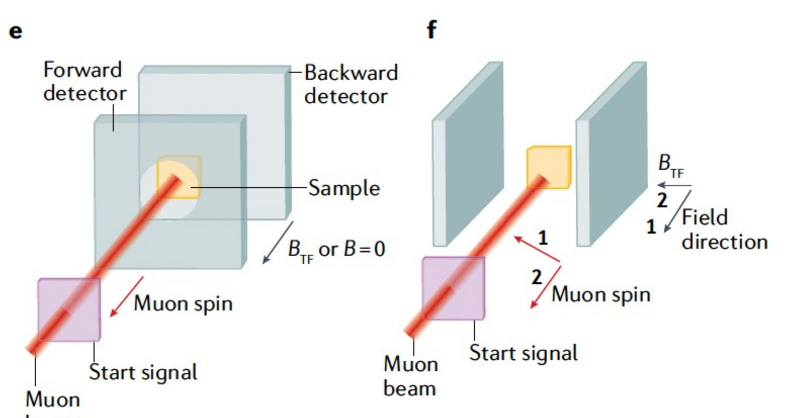




Experimental Details

Muon Spin Rotation and Relaxation (μ^+SR)





μ^+SR spectrometer at ISIS and schematic μ^+SR experiment [18]

Superconducting κ-HgBr

Time Reversal Symmetry and muon Relaxation rate Measurements

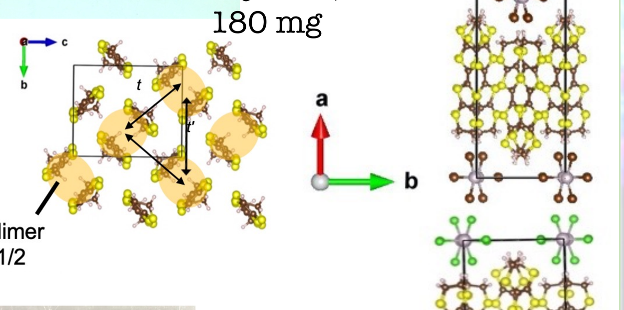
- Zero field (ZF) μ^+SR using pulsed muon at ISIS, UK and JPARC, Japan
- 0.3 K – 300 K, ^{3}He cryostat and ^{4}He gas flow

London penetration depth and Pairing symmetry Determination

- Transverse field (TF) μ^+SR using DC muon at TRIUMF, Canada
- 0.025 K – 6 K, ^{4}He+^{3}He Dilution Refrigerator

Aligned κ-HgBr crystals, 180 mg

a = 38.661(2) Å
b = 8.7148(5) Å
c = 11.2774(7) Å

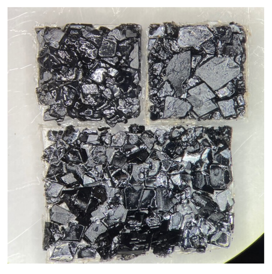


ET dimer
S = 1/2

Mott Insulating κ-HgCl

muon Relaxation rate Measurements and Spin Dynamics

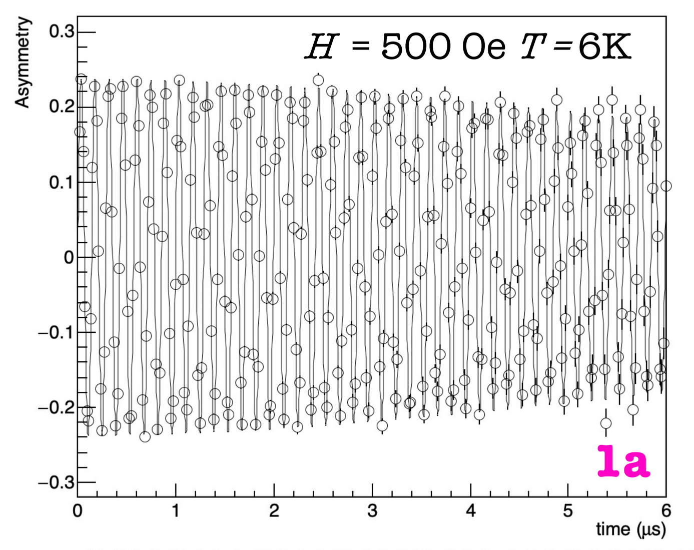
- ZF and Longitudinal Field (LF) μ^+SR using DC muon at TRIUMF, Canada, pulsed muon at ISIS, UK and JPARC, Japan, respectively.
- 0.025 K – 300 K, ^{4}He+^{3}He Dilution Refrigerator, ^{3}He cryostat and ^{4}He gas flow

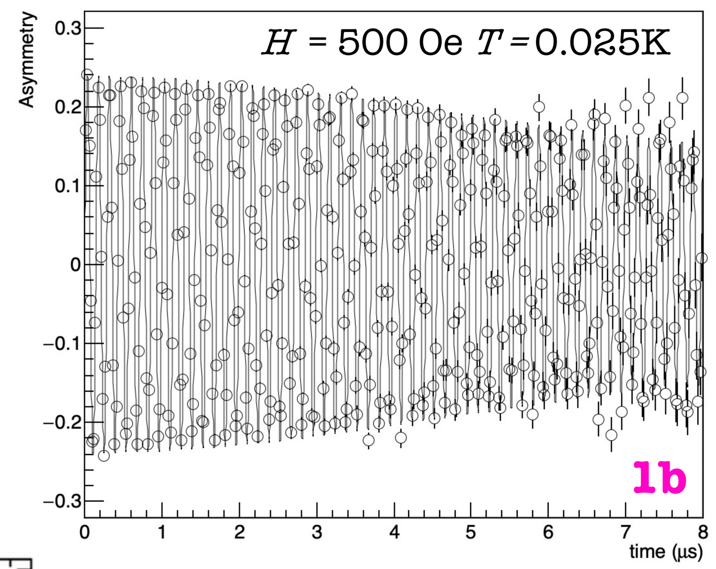


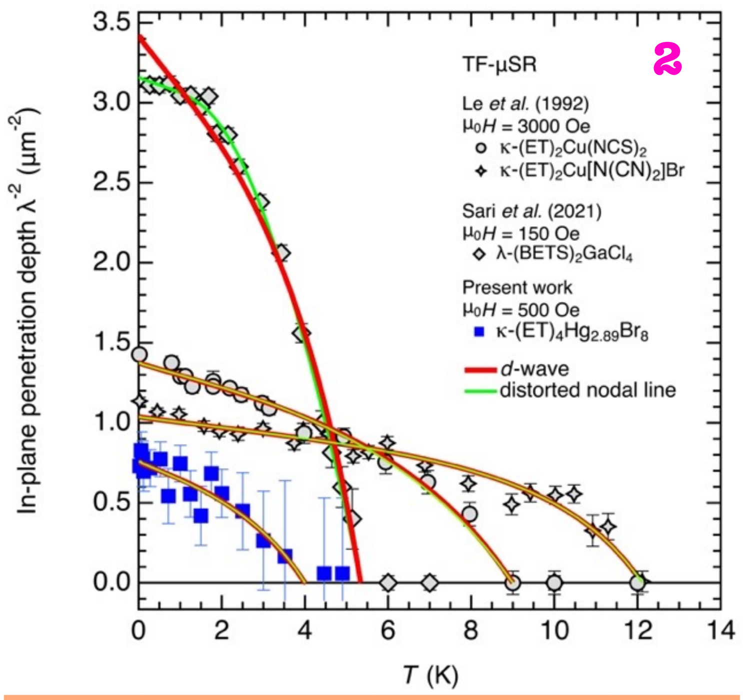
Aligned κ-HgCl crystals, 100 mg

a = 37.524(5) Å
b = 8.7491(11) Å
c = 11.1200(15) Å

Results and Discussion







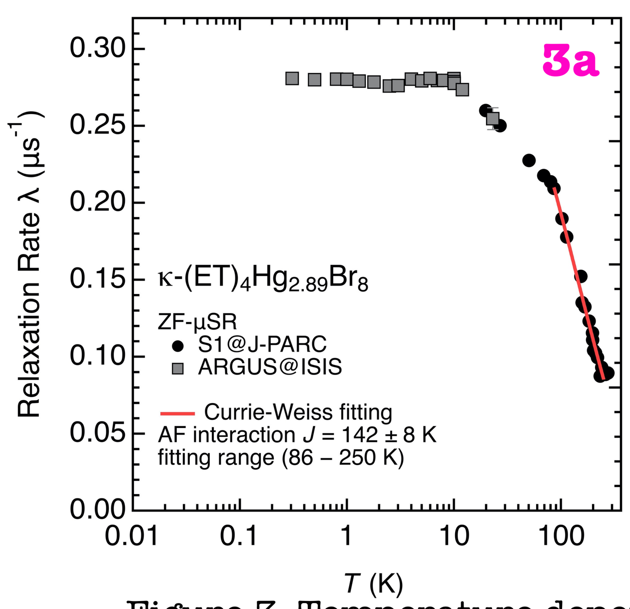
$$A^{TF}(t) = A_1\exp[-\sigma^2t^2]\cos(\gamma_\mu B_1 t + \phi) + A_2\cos(\gamma_\mu B_2 t + \phi),$$

Figure 1-2. TF-μ^+SR time spectra at $T < T_c$ and $T > T_c$ and comparison of several organic SCs [17].

$$\frac{\lambda_{ac}^{-2}(T)}{\lambda_{ac}^{-2}(0)} = 1 + \frac{1}{\pi}\int_0^{2\pi}\int_{\Delta(\phi,T)}^{\infty}\left(\frac{\delta f}{\delta E}\right)\frac{EdEd\phi}{\sqrt{E^2-\Delta_i(\phi,T)^2}}$$

Superconductivity:

1. Temperature independence of ZF μ^+SR from around 10 K crossing superconducting state → preserved time-reversal symmetry of incoherent metal [Ref. 23]
2. Strong coupling superconductor with gap ratio 5.0 ±0.7. Measurement at 1000 Oe gave gap ratio 7.0 ± 1.2 → very similar to SYK-NFL!
3. Well fitted by ***d*-wave** symmetry.
4. The $\boldsymbol{T_c}$ **= 4.3** K but has **smallest superfluid density.**



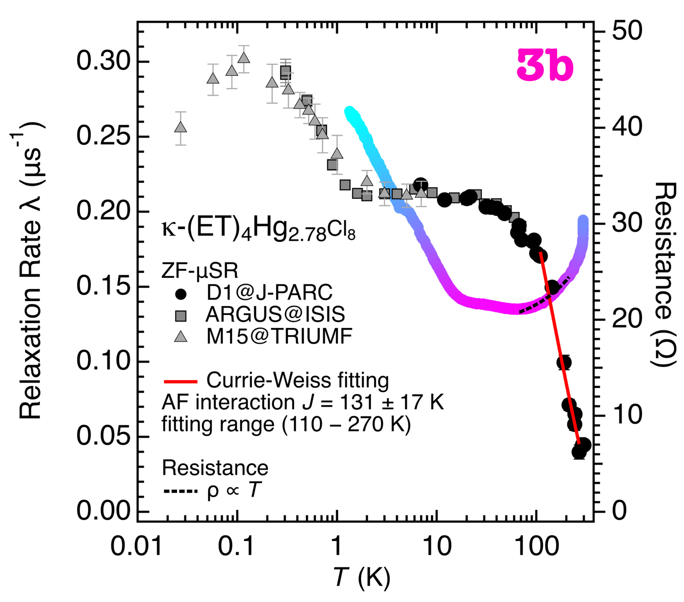
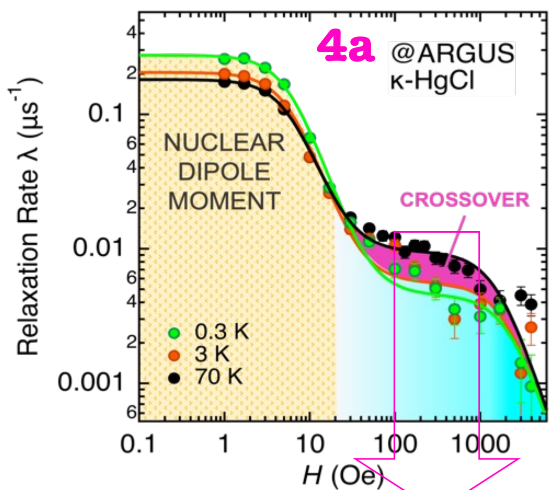
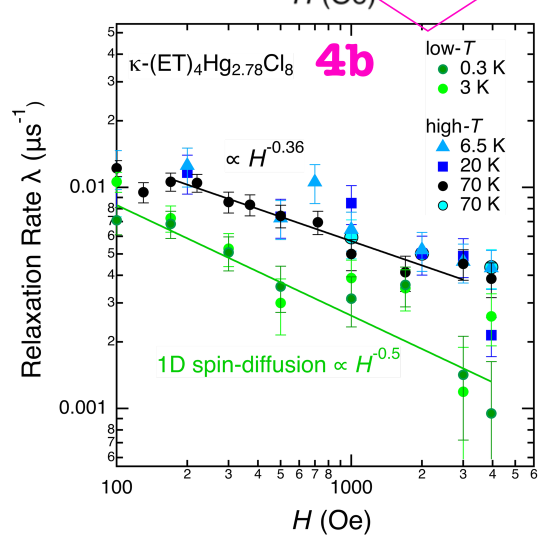
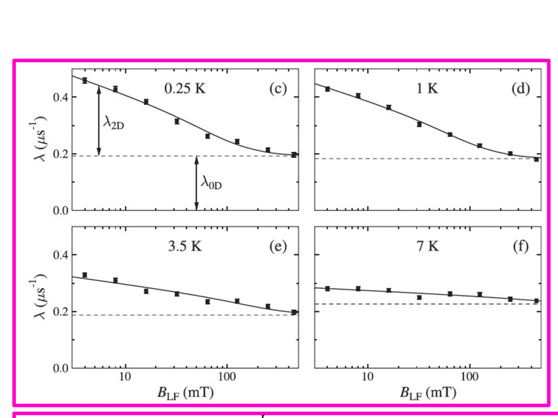
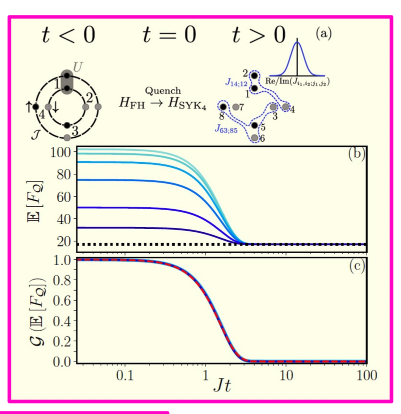


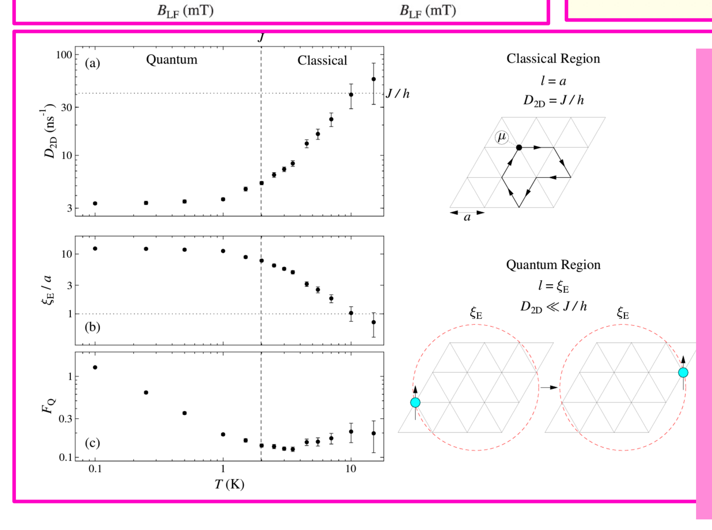
Figure 3. Temperature dependence of ZF-μ^+SR relaxation rate











Comparison with other spin liquid $YbZnGaO_4$ [ ] toward Quantitative analysis of the SYK-model through F_Q (Quantum Fisher Info) [20-22].

Figure 4. LF-μ^+SR relaxation rate at several temperatures in k-HgCl

Spin Liquidity:

1. There is no long-range magnetic order down to 25 mK. Instead, there is a broad peak around 0.1 K → the spin seems to be freezing.
2. There is a plateau region separating M-I transition → QCP?
3. In the metallic state (high-T region) the Antiferromagnetic exchange interaction are confirmed. Cf [Ref. 24]
4. From LF- μ^+SR the spin is more diffused and dynamics at high-T region → Quantum entanglement nearby QCP?

Conclusion: κ-HgBr and κ-HgCl seems to be a good starting candidates for studying holographic superconductor and SYK model in laboratory

References

[1] B. Keimer et al., Nature 518, 179-186 (2015); [2] O. Parcollet and A. Georges, PRB 59, 8 (1998); [G. Grissonnache, et al., Nature 595, 667-672 (2021); [4] S. Sachdev and J. Ye, PRL 70, 3339 (1993); [5] A. Kitaev, J. Suh, JHEP 183 (2018); J. Maldacena and D. Standford, PRD 94, 106002 (2016); [7] S. Hartnoll, P. Herzog, and G. T. Horowitz, PRL 101, 031601 (2008); S. Sachdev, PRX (2015); [8] N. Tsuji and P. Warner, PRB 99, 115132 (2019); [9] I. Esterlis and J. Schmalian, PRB 100, 115132 (2019);[10] G-A Inkof, K. Schalm, J. Schmalian, npj Quantum Materials 7:56 (2022); [11] C. D. Hebert, P. Semon, A.-M. Tremblay, PRB 92, 195112 (2015); [12] H. Taniguchi et al., JPSJ 76, 113709 (2007), H. Taniguchi, et al., JPSJ 76 Suppl. A, pp. 168-171; [13] H. Oike, et al., Nat. Commun. 8, 756 (2017); [14] K. Satoh, et. al., Physica B 404, 597 (2009); [15} Y. Eto, et al., PRB 81, 212503 (2010); [16] A. Naito, et. al., PRB 71, 054514 (2005);[17] D. P. Sari, et al., PRB 104, 224506(2021); [18] A. D. Hillier, Nat. Rev. Method Primer, 2, 4, (2022); [19] Y. Kurosaki, et al., Physica B, 404, 31 (2009); [20] F. L. Pratt, et al., Phys: Cof. Ser. 2462 012038 (2023); [21] F. L. Pratt et al., PRB 10, L060401 (2022); [22] S. Bandyopadhyay, et al., Quantum, 7, 1022 (2023); [23] D. P. Sari et al., J. Phys.: Conf. Ser. 2462 012061 (2023); [24] D. P. Sari, et al., in Solid State Phenomena, 345, 47 (2023) Reference therein.