Supplementary Information for
Thermoelectric signature of quantum critical phase
in a doped spin liquid candidate

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Supplementary Note 1. Crystal structure and band filling

Most κ-type BEDT-TTF compounds have the composition of κ-(BEDT-TTF)₂X with anion, X. κ-(BEDT-TTF)₂X has a quasi-two-dimensional structure with conducting BEDT-TTF layers and insulating X layers alternately stacked. In the conducting layer, BEDT-TTF dimers form an isosceles triangular lattice characterized by two different transfer integrals, t and t'. In the insulating layer, anion, X, electronically has a closed shell structure with a valence of -1. Therefore, the valence of BEDT-TTF is +0.5; namely, one hole resides per one dimer site so that the band is half-filled in most κ-(BEDT-TTF)₂X systems.

κ-(BEDT-TTF)₄Hg₂.89Br₈ is exceptional in that the lattice periodicity of Hg ions in the insulating layer is incommensurate with that of BEDT-TTF molecules in conducting layers¹,², resulting in a deviation from the half filling of the band³. The incommensurability, namely, the nonstoichiometry in κ-(BEDT-TTF)₄Hg₂.89Br₈ is not varied but fixed by chemistry to give 11% hole doping to half-filling. Note that mercury-included compounds often have incommensurate structures⁴.
The ratio, \(t'/t\), in \(\kappa\)-HgBr is 1.02 according to Ref.\(^5\), where the \(t\) and \(t'\) values are evaluated by tight-binding approximations based on the extended Hückel calculations of molecular orbitals.

**Supplementary Note 2. Temperature dependence and contour plot of \(-S/T\) of samples #1 and #2**

We measured \(-S/T\) for two separate samples #1 and #2. Supplementary Figures 1a and 1b show the \(-S/T\) data for the two samples separately and Supplementary Figures 1c and 1d show their contour plots on the temperature-pressure plane. The results of the two samples coincide with each other.

**Supplementary Note 3. Temperature dependences of the resistivity and \(-S/T\) at 0.4 and 1.5 GPa**

Supplementary Figures 2a and 2b show the temperature dependences of \(-S/T\) and the resistivity of sample #2. The resistivity is normalised to the value at 15 K. The Seebeck coefficient and resistivity were measured simultaneously at the same setup. At 1.5 GPa, the resistivity shows the squared temperature dependence and \(-S/T\) is nearly constant at low temperatures, confirming the Fermi liquid behaviour. At 0.4 GPa, the resistivity shows linear-in-temperature dependence down to \(T_c\) and \(-S/T\) exhibit the logarithmic divergence down to \(T_c\).

**Supplementary Note 4. Definition of \(T_c\) and \(T^*\)**

Supplementary Figure 3 shows the temperature dependence of \(-S/T\) at zero field under 0.3 GPa for sample #1. The uppermost and lowermost values and the midpoint defined in Fig. 3b correspond to the upper and lower bounds of the error bar. The onset temperature, \(T^*\), is defined as the temperature at which \(-S/T\) begins to deviate from the high-temperature normal-state behaviour.

**Supplementary Note 5. Pressure dependence of \(|\gamma'|\) and \(T_c\)**

Supplementary Figure 4 shows the pressure dependences of \(|\gamma'|\) and \(T_c\). The \(\gamma'\) value was evaluated by fitting the form, \(S/T = \gamma'\ln(T_0/T)\), to the experimental data. The \(T_c\) is the midpoint as described above.

**Supplementary Note 6. \(\gamma'\) values of \(S/T = \gamma'\ln(T_0/T)\) for other materials**
Supplementary Table 1 lists the $|\gamma'|$ values reported in literatures$^{6-18}$, where $\gamma'$ values were evaluated from the behaviour of $S/T = \gamma'\ln(T_0/T)$.

**Supplementary Note 7. Estimation of the coefficient of $T$-square term in resistivity**

The coefficient, $A$, in the form of $\rho = \rho_0 + AT^2$ was evaluated by fitting the temperature dependence of the resistivity at low temperatures (Supplementary Figure 5a). The fitting temperature regions were represented in Supplementary Figure 5b.
Supplementary Figure 1. Temperature dependence and the contour plot of\(-S/T\) in \(\kappa\text{-HgBr}\) for samples \(\#1\) and \(\#2\). The open circles in Supplementary Figures 1c and 1d indicate the superconducting transition temperature.
Supplementary Figure 2. Temperature dependences of the $-S/T$ and resistivity simultaneously measured for sample #2 at 0.4 and 1.5 GPa. a. Temperature dependence of $-S/T$ in the logarithmic temperature scale. At 0.4 GPa, $-S/T$ exhibits $\ln T$ behaviour. At 1.5 GPa, however, the Fermi liquid behaviour of $-S/T=\text{constant}$ was observed. b. Temperature dependence of the resistivity normalized to its value of 15 K. The linear- and quadratic-in-temperature resistivity above $T_c$ under 0.4 and 1.5 GPa indicate non-Fermi liquid and Fermi liquid behaviours, respectively.
Supplementary Figure 3. Definition of $T_c$ and $T^*$. The definitions of bulk $T_c$, its error, and the onset, $T^*$, are shown for the 0.3 GPa data of $-S/T$ of sample #1, for example. The bulk $T_c$ is defined by the midpoint. The upper and lower bounds of $T_c$ indicated in the figure determine the error bar of $T_c$. $T^*$ is defined as the temperature at which $-S/T$ starts to deviate from the normal-state behaviour. The inset shows the enlarged view of the $-S/T$ behaviour in the vicinity of $T^*$. 
Supplementary Figure 4. Pressure dependence of $|\gamma'|$ and $T_c$ in $\kappa$-HgBr. The $|\gamma'|$ and $T_c$ values for samples #1 and #2 are plotted as a function of pressure. The squares and circles correspond to the data of samples #1 and #2, respectively. The blue and red symbols indicate $|\gamma'|$ and $T_c$, respectively.
Supplementary Figure 5. Resistivity versus squared temperature in ranges of (a) 0-1000 K$^2$ and (b) 0-300 K$^2$. The resistivity $\rho$ follows the Fermi liquid behaviour of $\rho = \rho_0 + AT^2$ at low temperatures and the values of the coefficient $A$ plotted in the inset of Fig. 3a were determined from the data shown in Supplementary Figure 5b.

Supplementary Table 1. $|\gamma'|$ values in the logarithmic part of $-S/T$ in some materials. The $|\gamma'|$ values in the behaviour of $S/T = \gamma' \ln(T_0/T)$ for other materials$^{6-18}$ are listed in $\mu$V/K$^2$.

| Material                        | Slope value | Material                      | Slope value |
|---------------------------------|-------------|--------------------------------|-------------|
| La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ (Nd-LSCO ($x = 0.24$))(ref.6) | 0.11        | UCoGe ($H=11.1$ $T$)(ref.12)  | 2.3         |
| La$_{1.8-x}$Eu$_{0.2}$Sr$_x$CuO$_4$ (Eu-LSCO ($x = 0.24$)) (ref.7) | 0.16        | YbRh$_2$Si$_2$ (ref.13)       | 4.5         |
| Bi$_2$Sr$_2$−$x$La$_x$CuO$_{6+\delta}$ (Bi2201 ($\rho = 0.39$)) (ref.8) | 0.05        | CeCu$_5$Au$_{0.1}$ (ref.14)  | 6.2         |
| Pr$_{2-x}$Ce$_x$CuO$_4$ (PCCO ($x = 0.16-0.19$)) (ref.9) | 0.012-0.038 | Ce$_2$PdIn$_8$ (ref.15)      | 1.6         |
| La$_{2-x}$Ce$_x$CuO$_4$ (LCCO ($x = 0.15-0.17$)) (ref.9) | 0.0095 − 0.049 | YbAgGe (ref.16)           | 4.7         |
| [BiBa$_{0.66}$K$_{0.36}$O$_2$]CoO$_2$ (ref.10) | 0.62        | YbPtBi (ref.17)              | 0.25        |
| Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ ($x=0.022-0.13$) (ref.11) | 0.32 - 0.895 | EuFe$_2$(As$_{1-y}$P$_y$)$_2$ ($y=0.26, 0.36$) (ref.18) | 0.077, 0.16 |
Supplementary References

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