Critical Behavior in Doping-Driven Metal–Insulator Transition on Single-Crystalline Organic Mott-FET

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We present the carrier transport properties in the vicinity of a doping-driven Mott transition observed at a field-effect transistor (FET) channel using a single crystal of the typical two-dimensional organic Mott insulator $\kappa$-(BEDT-TTF)$_2$CuN(CN)$_2$Cl ($\kappa$-Cl). The FET shows a continuous metal–insulator transition (MIT) as electrostatic doping proceeds. The phase transition appears to involve two-step crossovers, one in Hall measurement and the other in conductivity measurement. The crossover in conductivity occurs around the conductance quantum $e^2/h$, and hence is not associated with “bad metal” behavior, which is in stark contrast to the MIT in half-filled organic Mott insulators or that in doped inorganic Mott insulators. Through in-depth scaling analysis of the conductivity, it is found that the above carrier transport properties in the vicinity of the MIT can be described by a high-temperature Mott quantum critical crossover, which is theoretically argued to be a ubiquitous feature of various types of Mott transitions. [This document is the unedited Authors version of a Submitted Work that was subsequently accepted for publication in Nano Letters, copyright ©American Chemical Society after peer review. To access the final edited and published work see http://dx.doi.org/10.1021/acs.nanolett.6b03817]

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Strongly correlated electrons confined in two-dimensional materials exhibit a rich variety of anomalous phases, such as high-$T_c$ superconductivity, Mott insulators, and quantum spin liquids. Enormous effort has been devoted to understanding the mechanisms of the phase transitions between them, which are driven by the complex interplay between many-body interactions and charge–spin fluctuations. Mott insulators are particularly remarkable in viewpoints of nanoscience as well as fundamental physics since they are not only mother materials for a diverse range of strongly correlated systems, but also serve as a molecular or atomic limit for coherently coupled arrays of single electron transistors (SETs). In Mott insulating state, strong Coulomb repulsion between electrons in half-filled band leads to carrier localization, which can be viewed as a situation that the collective Coulomb blockade confines an electron to the individual subnanosized SET. The electronic/magnetic properties of Mott insulators are governed by quantum mechanics, and indeed various types of unconventional phase transitions occur due to imbalance in the particle–wave duality of electrons which is triggered by charge doping, a magnetic/electric field, pressure, or introduction of disorders. An important issue is the possibility of quantum phase transitions in Mott insulators, where quantum fluctuations play an essential role in the transitions with the emergence of quantum critical points at 0 K. This would hold true for a coherently coupled array of SETs, although it has not yet been recognized.

Even at finite temperatures, quantum fluctuations with an energy scale overwhelming the temperature $k_B T$ give rise to a critical crossover feature observed as a power-law singularity in physical quantities in the vicinity of the quantum critical point. It has been argued that the unconventional non-Fermi liquid behavior commonly observed near the Mott transition should pertain to such quantum criticality, although there is no general consensus on its origin.

To properly investigate the doping-driven Mott transition, it is required to dope carriers in a clean manner without introducing impurities since the modification of the local electronic environment by such disorders should destroy the intrinsic many-body states. Electrostatic doping techniques using the electric double-layer transistor (EDLT) or field-effect transistor (FET) configurations are promising for such investigation. Combined with recent advances in fabrication technology for crystalline nanosheets and interfaces, these techniques not only enable fine control of the charge density as an independent tunable parameter but also extend the possibility of novel device applications that utilize abrupt changes in physical properties triggered by a phase transition.

The organic Mott insulator $\kappa$-(BEDT-TTF)$_2$CuN(CN)$_2$Cl, referred to as $\kappa$-Cl, is one of the
two-dimensional materials suitable for the channel of electrostatic doping devices. The crystal structure of κ-Cl consists of alternately stacked π-conducting \([(BEDT-TTF)_2]^+\) cation layers and atomically thin insulating polyanion layers (Figure 1b), showing excellent crystallinity\(^{23,24}\). An effectively half-filled band is formed by a carrier density of one hole per BEDT-TTF dimer. As the areal carrier density (\(\sim 10^{14} \text{ cm}^{-2}\)) is almost an order of magnitude lower than those of inorganic Mott insulators, a large region of the phase diagram can be investigated by electrostatic doping. Indeed, we have demonstrated that by charge doping up to a density of \(\sim 10^{14} \text{ cm}^{-2}\) using an EDLT, both electron- and hole-doping-driven Mott transitions were successfully realized on a single κ-Cl crystal\(^{25}\). Nonetheless, the EDLT technique is inconvenient for fine-tuning of the band filling at low \(T\) owing to freezing of the ionic liquid\(^{26}\). On the other hand, a solid-gate FET is highly advantageous in terms of the applicability of various micrometer-scale interface engineering methods as well as the high controllability of the doping level in real time at low \(T\).

In this Letter, we report a doping-driven Mott MIT on a solid-gate FET using a single crystal with a highly flat surface as well as a gate dielectric covered with self-assembled monolayers (SAMs). We selected hydrophobic 1H,1H,2H,2H-perfluorodecytriethoxysilane (PFES) and octyltriethoxysilane (OTS) as SAM reagents, which are widely used to improve device performance and are also expected to eliminate the adsorbed solvents on the surface\(^{27,28}\). By micropatterning these SAMs\(^{29,30}\), we successfully induced a continuous MIT on a κ-Cl single-crystal channel. Owing to the high resistivity of the bulk channel below \(T \sim 50 \text{ K}\), the observed MIT was governed by carriers confined in two dimensions at the interface, exhibiting sheet conductivity equal to \(e^2/h\) (\(e^2/h = 3.874 \times 10^{-5} \Omega^{-1}\)) on the metal–insulator crossover (MIC) line. Through in-depth analysis of the conductivity on the verge of the MIT, we demonstrated the quantum critical feature of the doping-driven Mott transition in the intermediate-\(T\) crossover region. This is distinct from the quantum criticality in the low-\(T\) regime for high-\(T_c\) cuprates\(^{21,18,19}\) and follows the high-\(T\) Mott quantum criticality expected to be universal to various types of Mott transition\(^{33,34}\). On the basis of the scaling plot for the quantum criticality, we propose a candidate crossover line that delineates the boundary between metallic and insulating regions.

The FET device was fabricated on a SiO\(_2\) (300 nm)/\(p^+\)-Si substrate with a micropatterned SAM (area: \(50 \times 50 \mu\text{m}^2\)), as schematically shown in Figure 1a. We fabricated four devices: one with gate surface modification by PFES (device F#1) and the other three with gate surface modification by OTS (devices C#1–#3), details of which are shown in Table S1. We found that the difference in SAM species did not noticeably influence the overall MIT behaviors, as discussed in Supporting
Information D. The patterning of the SAMs and the attachment of electrodes were performed using a conventional photolithography technique (Supporting Information A). Then thin crystals of $\kappa$-Cl (thickness: 40 nm for device F#1 and 80 nm for device C#2) were laminated on top of the substrate. Through the improvement of the electrochemical crystal growth process (Supporting Information B), we successfully obtained thin crystals of $\kappa$-Cl with a flat surface. Figure 1c shows an atomic force microscopy (AFM) image of the surface of the typical $\kappa$-Cl crystal laminated on the substrate. Over micrometer-scale areas, the roughness of the surface is below 1.5 nm, or the thickness of one BEDT-TTF molecular layer (Figure 1b). For comparison, shown in Figure S3 is an AFM image of a typical thin crystal of the sister material $\kappa$-(BEDT-TTF)$_2$CuN(CN)$_2$Br ($\kappa$-Br), which was synthesized in accordance with a previous method reported elsewhere. One can recognize the steps of the BEDT-TTF molecular layers and many small vacancies for $\kappa$-Br. The availability of the single crystal with perfect surface flatness for $\kappa$-Cl is a major advantage for precisely investigating the MIT without a significant effect of disorder.

Figure 1e shows the transfer curves of device F#1, which has a partly covered PFES layer and provides both $\kappa$-Cl/PFES/SiO$_2$ and $\kappa$-Cl/SiO$_2$ interfaces on an identical single crystal. Both on the SiO$_2$ and PFES regions, the sheet conductivity ($\sigma$) at low $T$ is suppressed to below $10^{-4} e^2/h$ by applying a negative gate voltage ($V_g$), whereas a positive $V_g$ induces a continuous increase in $\sigma$. This “n-type polarity” stems from the natural n-type doping for $\kappa$-Cl surface (typically density of $\sim 10^{12} - 10^{13}$ cm$^{-2}$) and implies that electron doping breaks the strong carrier localization of Mott insulator state. This description is also supported by the carrier density estimated from Hall effect measurements (Figure 1f): $n = \sigma B/e \tan \theta_H$, where $B$ and $\theta_H$ are the magnetic field and Hall angle, respectively. The carrier density is nearly zero at negative $V_g$, corresponding to the gapped incompressible state (small $|dn/d\epsilon|$, with $\epsilon$ being the chemical potential) characterizing Mott insulators. Upon electron doping by as small amount as $\sim 10^{12}$ cm$^{-2}$, however, hole carriers with the density of $1.3 \times 10^{14}$ cm$^{-2}$ (corresponding to the cross-sectional area of 75% of the first Brillouin zone) rapidly recovered, evidencing a compressible state with a large Fermi surface (large $|dn/d\epsilon|$). This change in the Hall carrier density indicates an abrupt increase in the mobile carrier spectral weight accompanied by closing of the Mott gap upon doping. Plus, continuous increase in $\sigma$ after establishment of large Fermi surface state with almost a constant hole carrier density is also a unique feature of nonrigid bands of strongly correlated electrons, on which doping enhances carrier coherence through modification of the charge mass or lifetime.

A stark difference between the PFES and SiO$_2$ substrate areas manifests itself at a highly posi-
Figure 1. Schematic of the Mott-FET device and the doping-driven metal–insulator transition of $\kappa$-Cl. (a) Schematic of FET device with the micropatterned SAM interface. (b) Crystal structure of $\kappa$-Cl with alternately stacked cationic dimer $[(\text{BEDT-TTF})_2]^+$ layers and anionic insulator layers. (c, d) AFM topographical image measured on the surface of the $\kappa$-Cl crystal. (e) Transfer curves for various $T$ (device F#1) on a PFES-covered substrate (colored circles) and on bare SiO$_2$ (gray squares). Inset: device mobility $\mu_{FE} = (1/c_g) d\sigma/dV_g$ ($c_g = 7.0 \times 10^{14}$ cm$^{-2}$V$^{-1}$; areal capacitance of back gate) as a function of $T$ in the highly doped regime ($V_g = 116$ V). (f) Gate voltage dependence of carrier density $n_{\text{hole}}$ (device F#1) for PFES device area determined from Hall coefficients ($B = 8$ T). In the highly doped regime where the MIC occurs, $n_{\text{hole}}$ is almost independent of $V_g$ with $n_{\text{hole}} \sim 1.3 \times 10^{14}$ cm$^{-2}$, corresponding to the cross-sectional area of $\sim 75\%$ of the first Brillouin zone. Error bars were calculated from the standard deviation of the Hall angle. The solid line (light blue) is a guide to the eye.

On the PFES region, $\sigma$ exceeds $3e^2/h$ and its behavior becomes metallic (i.e., increasing $\sigma$ with decreasing $T$). Importantly, an MIC in the conductivity occurs at $V_g = 75$ V (the point of zero activation energy in Figure 2), whereas the crossover between an incompressible Mott insulator state and a compressible state with a large Fermi surface occurs at $V_g < 0$ (the jump of the $n_{\text{hole}}(V_g)$ curve in Figure 1f). This is in contrast to the MIC of other two-dimensional electron systems, where continuous changes in the carrier density and metallic transport (i.e., $d\sigma/dT < 0$) take place at the same time.\textsuperscript{38,39} The conductivity at the MIC is $\sigma = 1.4e^2/h$, which agrees with the Mott–Ioffe–Regel (MIR) limit in two dimensions (minimum conductivity anticipated within semiclassi-
Figure 2. Characteristics of activated transport in insulating regime (device F#1). (Main panel) Gate voltage dependence of the transport gap $\Delta$ in the insulating regime for the PFES-covered (red) and bare SiO$_2$ substrate (gray) areas. (Inset) Arrhenius plots of sheet conductivity for different gate voltages on the PFES-covered and bare substrate. Symbols are experimental results; dotted lines are fits using $\sigma_{\text{ins}}(T) = \sigma_\infty \exp(-\Delta/(2k_BT))$. Each step is $\Delta V_g = 8$ V.

The device mobility $\mu_{\text{FE}} = (1/c_g) \frac{d\sigma}{dV_g}$ ($c_g = 7\times10^{10}$ cm$^{-2}$V$^{-1}$; areal capacitance of back gate) also becomes very high, reaching 220 cm$^2$ V$^{-1}$ s$^{-1}$ at 5 K. On the SiO$_2$ region, however, $\sigma$ remains insulating with a value below $e^2/h$, and the maximum mobility is 70 cm$^2$ V$^{-1}$ s$^{-1}$, much lower than that on the PFES region (Figure 1c inset).

On the other hand, focusing on the insulating regime the conductivity is well expressed by activated-transport behavior modeled with $\sigma_{\text{ins}}(T, V_g) = \sigma_\infty \exp(-\Delta(V_g)/(2k_BT))$ irrespective of SAM functionalization (Figure 2, inset), where $\Delta$ is the transport gap and $\sigma_\infty (\approx 1.4e^2/h)$ denotes the high temperature limit of activated-transport conductivity. In the insulating limit ($V_g \sim -120$ V, Figure 2), $\Delta$ ($\approx 35$ meV) is comparable to that of bulk $\kappa$-Cl (Supporting Information E), which confirms that the original electronic properties of $\kappa$-Cl are retained through the device fabrication processes. On the other hand, electron doping continuously reduces $\Delta$ toward zero, which occurs more rapidly on the PFES-patterned area than on the bare SiO$_2$ area (Figure 2). The difference in $d\Delta/dV_g$ cannot be explained solely by the doping effect from the SAM, which suggests that the Mott transition is sensitively affected by interfacial disorder conditions such as impurities, the
contamination by the solvent, and charge traps.

Figure 3a,b displays the detailed $T$-dependence of the conductivity near the MIC, which is measured on the SAM-patterned area of device C#2. Similar $\sigma(T)$ behaviors were observed for all the devices on the SAM-patterned areas, and hereafter we concentrate on device C#2 (for devices C#1, C#3, and F#1, see Figure S8). The conductivity curve for each $V_g$ is continuous without jumps or kinks, and reproducible without hysteresis even after sweeping many times, indicating that no structural transition occurs. For $22 \text{ K} \lesssim T \lesssim 36 \text{ K}$, a well-defined MIC behavior is observed; a crossover line exists with $T$-independent conductivity $\sigma^* = \sigma_{\text{MIC}} = 1.35e^2/h$ at $V_g^* = V_{\text{MIC}} = 51 \text{ V}$, which separates the metallic and insulating regions in the $\sigma$ versus $V_g$ plane. (Hereafter, variables with asterisks represent those on the crossover line.) In the lightly doped region ($V_g < V_{\text{MIC}}$), all the $\sigma(V_g, T)$ curves gradually decrease with decreasing $T$. On the other hand, the heavily doped region ($V_g > V_{\text{MIC}}$) shows nonmonotonic behavior with conductivity maxima, below which the transport becomes insulating. The low-$T$ insulating transport for $\sigma \gtrsim 0.5e^2/h$ can be scaled linearly with log $T$. Negative magnetoresistance is also observed in the highly doped regime below $\sim 7 \text{ K}$, which indicates that the linear-in-log $T$ insulating behavior is explained by weak localization correction.\[^{[1]}\] Interestingly, $V_{\text{MIC}}$ is a device-dependent value, whereas $\sigma_{\text{MIC}}$ is almost identical for each device (see Figures 1 and S8). This means that the conductivity in the vicinity of the MIC is not affected by the variation in the thickness of $\kappa$-Cl crystals, and thus the MIC is clearly caused by carriers confined in two dimensions at the interface. Notably, $\sigma_{\text{MIC}}$ corresponds to the minimum metallic conductivity, and hence one cannot identify a bad-metal (BM) feature, that is, metallic transport that violates the MIR limit over the entire $T$ range. (Note that above 40 K, it is difficult to accurately extract the carrier transport on the interface from the observed conductivity because of the dominant contribution of the undoped bulk channel. Nevertheless, one cannot clearly observe BM behavior at high $T$. See Supporting Information E.) This is in contrast to Mott transitions in bulk $\kappa$-Cl at half-filling\[^{[23],[29]}\] or those in inorganic Mott insulators.\[^{[43],[45]}\]

Recently, theoretical studies based on dynamical mean-field theory (DMFT)\[^{[33],[34],[46]}\] have advocated that the high-$T$ MICs associated with Mott transitions bear a type of quantum criticality. This high-$T$ quantum criticality is expected to be observed on any Mott systems because it is governed by high-energy-scale quantum fluctuations due to the competing electron–electron interaction and Fermi energy on Mott insulators. Although DMFT is exact in the limit of infinite dimensions, it becomes a reasonable approximation at high $T$ even for low-dimensional system
Figure 3. Criticality in the vicinity of the MIC point (device C#2, on OTS-covered area). (a) $\sigma - T$ plot for various $V_g$ ($= 120, 112, \cdots, -54, -60$ V). For $21 \, \text{K} < T < 36 \, \text{K}$, there is a distinct MIC point at $V_{\text{MIC}} = 51 \, \text{V}$ where the sheet conductivity becomes $T$-independent, namely $\sigma(V_{\text{MIC}}T) = \sigma_{\text{MIC}} = 1.35e^2/h$ (thick line in panel b). On the metallic side ($V_g > V_{\text{MIC}}$), $\sigma(T)$ curves exhibit maxima at certain temperatures, below which $\sigma(T)$ decreases in a linear-in-log $T$ manner. On the other hand, for $V_g < V_{\text{MIC}}$, $\sigma$ monotonically decreases with decreasing $T$. (b) Magnified view of $\sigma - T$ plot for various $V_g$ (in increments of 1 V) in the vicinity of the MIC. Dots represent measurement points. (c) Color-coded plot of conductivity with the trajectory of the optimized crossover line (orange solid line) under the assumption of high-$T$ Mott quantum criticality (candidate QWL, see text for details). For comparison, $V_g = V_{\text{MIC}}$ is marked with a dashed line (green). (d) Conductivities on the optimized crossover line (orange) and the $V_g = V_{\text{MIC}}$ line (green). (e) Quantum critical scaling plot of $\sigma$ normalized by $\sigma^*(T)$. For $T > 7 \, \text{K}$, all of the conductivity data on the insulating and metallic sides collapse onto two different branches of the universal scaling function (eq. (1)). The critical exponent is $\nu_z = 1.55$. 
such as κ-Cl. Indeed, it has been experimentally demonstrated that various half-filled organic Mott insulators including κ-Cl show quantum critical nature in the high-$T$ crossover region for a pressure-driven Mott transition.\(^{23}\) The experimental and theoretical phase diagrams around the quantum critical Mott crossovers are summarized in Figure S6. In brief, despite the complexity of the low-$T$ material-specific phases, the half-filled organic Mott insulators show material-independent rapid crossovers at high $T$ with the carrier transport continuously changing from insulating to BM behavior. At these crossovers, quantum criticality is commonly observed with the crossover conductivity much smaller than $e^2/h$ (i.e., BM behavior). These behaviors are completely consistent with the quantum criticality suggested by DMFT.\(^{34,46}\) Therefore, the high-$T$ Mott quantum critical crossover scenario is appropriate for describing the continuous MIC in the conductivity, at least for a pressure-driven Mott transition. The following question arises: does this hold true for a doping-driven Mott transition? According to the theory,\(^{33}\) the quantum criticality of a doping-driven Mott transition should have notable features that have not been observed in a pressure-driven Mott transition: (1) A continuous crossover is sustained even in the low-$T$ region (well below 1% of the half bandwidth $D$ with $D \sim 0.25$ eV for κ-type BEDT-TTF compounds\(^{27}\)). (2) The crossover conductivity coincides with the MIR limit at high $T$, whereas it may deviate at low $T$. (3) The MIT can show two-step crossovers at finite $T$; one is a rapid change in charge compressibility $|dn/d\epsilon|$ and the other is a sign change in $d\sigma/dT$ (Supplemental Material in ref. \(^{33}\)). These three points appear to be consistent with the present experimental results, and the third point can be observed in the discrepancy between the jump of the Hall carrier density and the MIC in the conductivity (Figure 1e,f). Thus, the high-$T$ quantum critical crossover in the doping-driven Mott transition is a reasonable scenario that explains the observed MIC.

According to DMFT studies,\(^{33,34,46}\) the MIC of a Mott transition can be well described within the framework of finite-size quantum critical scaling theory.\(^{19,48,50}\) This theory predicts that the conductivity is amenable to a scaling law written as\(^{51}\)

$$\sigma(T, \delta) = \sigma^* F_z \left( \frac{|\delta|}{T^{1/v_z}} \right),$$

where $\delta$ is the distance from the crossover line and the power exponents $\nu$ and $z$ are called the correlation exponent and dynamical exponent, respectively. In contrast to magnetic/charge ordering transitions and superconducting phase transitions, the MIT generally has no obvious order parameter. Yet, in the present doping-driven Mott transition, it is natural to regard $V_g$ as the
scaling variable, that is, \( \delta = V_g - V_g^* \). \( F_+(y) \) and \( F_-(y) \) are the universal scaling functions for the metallic and insulating regimes, respectively, satisfying \( F_\pm(y) \to 1 \) and having mirror symmetry \( F_+(y) = 1/F_-(y) \) for \( y \to 0 \). Importantly, the crossover line in eq. (1) should be provided by the “quantum Widom line (QWL)” on which the system becomes the least stable. For scaling analysis, we attempt to propose a possible crossover line to find the QWL in the following.

In analogy with the Widom line in the supercritical region of the classical gas-liquid-type phase transition, the location of the QWL in the \( T \) versus \( V_g \) plane is generally dependent on temperature. Thus, the crossover line \( V_g^*(T) \) should have a complex form and deviate from the constant-\( V_g \) crossover line widely adopted for conventional doping-driven MITs. Because it is fundamentally difficult to identify the location of the QWL from the transport properties, we instead derive a candidate crossover line through a simple optimization calculation (for details, see Supporting Information). In short, the crossover line is taken as \( V_g^* = V_{MIC} \) for \( T > 25 \) K to satisfy the theoretically suggested high-\( T \) feature of the QWL, \( \sigma_{high-T}^* = e^2/h \) (constant). Then it is smoothly extended to the low-\( T \) region so that the renormalized conductivity data \( \sigma(T, V_g)/\sigma^*(T) \) converge onto the universal scaling curves (eq. (1)) with the highest scaling quality. If the resultant crossover line is appropriate for the candidate QWL, the quantum critical scaling along the crossover line should hold for sufficiently low \( T \).

Figure depicts the results of the scaling analysis based on the high-\( T \) Mott quantum critical crossover scenario, which is obtained with \( \nu_z = 1.55 \). It is found that the quantum critical scaling law persists over three decades of \( \sigma/\sigma^* \) and that the critical region extends down to \( T \sim 7 \) K. As mapped in Figure, the optimized crossover line exhibits a bow-shaped trajectory in the \( T \) vs \( V_g \) plane and deviates from the conventional critical crossover line with \( V_g = V_{MIC} \) at low \( T \). The crossover conductivity \( \sigma^* \) falls from \( \sigma_{MIC}^* \) with decreasing \( T \) (Figure), which is consistent with the theory. In the lower-\( T \) regime of \( T < 7 \) K, it turns out that the data for \( \sigma/\sigma^* \) are no longer scaled by the universal curves. This can be understood from two possible points of view. First, the quantum critical description of the continuous MIT is obscured because of the weak localization in the low-\( T \) region. Second, since long wavelength (low energy) fluctuations should come to play a significant role in low dimensional systems at a sufficiently low \( T \), the high-\( T \) Mott critical crossover scenario derived from a local mean field approximation such as DMFT may be no longer appropriate. We emphasize that the quantum critical crossover with similar critical exponents
down to $T \sim 10$ K was also confirmed in devices C#1, C#3 and F#1 (Figure S8), even though details of the carrier transport characteristics varied from device to device. These findings manifest the essence of the present phase transition that the critical behavior is independent of specific details of the system. Furthermore, we also attempted the scaling analysis using conventional crossover lines (i.e., constant-$V_g^*$ lines), as displayed in Figures and [S7 and S9]. Our attempt to fit eq. (1) with a constant $V_g^*$ was only successful in a limited $T$ range, which indicates that the crossover line in Figure 3d is appropriate for describing the observed quantum criticality. Taken together, the proposed crossover line meets the requirements of the QWL, and the quantum critical crossover region spreads over a wide range in the $T$ vs $V_g$ plane.

One might suspect the possibility of other types of MICs associated with, for example, a percolation transition$^{54}$ or a Mott MIC with classical criticality.$^{24,55}$ However, the continuous MIC in the present study is inconsistent with criticalities derived from these mechanisms, indicting the uniqueness of the quantum critical scaling of the charge transport in the intermediate-$T$ crossover region of the doping-induced Mott transition. Applicability for classical criticality is discussed in detail in Supporting Information [H].

Finally, we discuss the lack of BM behavior in the observed doping-driven Mott transition. In a wide variety of high-$T_c$ superconductors as well as heavy Fermion systems, metallic transport without a coherent Drude peak and with high resistivity exceeding the MIR limit is observed in the crossover region between the localized and delocalized states without showing the saturation observed for conventional metals.$^{12,56}$ This BM behavior is intuitively understood as collective carrier transport retaining the strongly correlated feature of the localized state (e.g., Mott insulator), whereas its origin remains to be fully understood. Early studies based on a quantum critical model$^{57}$ did not appear to explain the BM behavior, promoting various theoretical considerations such as those based on marginal Fermi liquid theory$^{58}$ and holographic duality.$^{59,60}$ A DMFT study$^{33}$ pointed out that the BM behavior is not contradictory to quantum criticality; the linear-in-$T$ resistivity beyond the MIR limit can result from the inclined quantum critical region in the $T$ versus doping density plane. In other words, the emergence of BM behavior depends on the relationship between the characteristics of the QWL and the material-specific band dispersion. Taking into account the remarkable similarity between the high-$T$ criticality in the present experiment and the DMFT study, we speculate that the lack of BM behavior in the present experiment is due to the disordered environment and the two-dimensional band structure that can modify the QWL. So far, BM behavior has been believed to be an inherent property of Mott insulators. However, our
results provide experimental counterevidence, which indicates that the non-Fermi-liquid feature in the crossover region should be understood in terms of the high-$T$ Mott quantum criticality. Optical conductivity measurements may reveal a clearer picture of the quantum critical region of the doping-driven Mott transition in future.

In summary, we successfully obtained single-crystal Mott-FETs with high performance, the device mobility exceeding $\sim 200 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at low temperatures by a combination of thin $\kappa$-Cl crystals with a flat surface and interfacial modification of the device using SAMs. Fine control of the charge doping via gate voltage revealed two-step metal–insulator crossovers in the intermediate-temperature region: a rapid increase in the hole carrier density, followed by inversion of the sign of the temperature coefficient of the conductivity at $\sim e^2/h$ without the occurrence of BM behavior. The high device quality also provided an opportunity to accurately examine the transport properties in the vicinity of the MIC, which revealed quantum criticality in the conductivity. The quantum criticality was retained down to $T \sim 7 \text{ K}$, whereas at lower temperatures metallic transport became insulating owing to a disorder-induced interference effect (weak localization). A bow-shaped crossover line of the MIT with the high temperature limit of $\sigma_{\text{high-}T}^* \sim e^2/h$ was successfully proposed, which implies that the origin of the observed criticality was the high-$T$ Mott quantum critical crossover, a ubiquitous feature for various types of Mott transition suggested on the basis of DMFT. This work provides a key insight into the criticality in the doping-driven Mott transition, as well as its relationship with the pressure-driven Mott transition at half-filling and the interaction-driven MIT in two-dimensional electron gas systems.

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Supporting Information A: Preparation of SAM-patterned substrate

Highly \( p \)-doped Si wafers with a 300 nm thick thermally grown SiO\(_2\) layer were obtained from a commercial source and used as the gate substrate. First, the Ti (20 nm)/Au (180 nm) electrodes were formed on top of the SiO\(_2\) wafers by photolithography (NanoSystem Solutions, DL-1000RS) and thermal deposition (Katagiri engineering). The wafers were cleaned by the oxygen plasma and UV/ozone treatments, followed by a second photolithographic process to make micropatterns for subsequent SAM formation. After short time immersion in alkaline hydrogen peroxide (2:1:1 solution of 30\% w/w aqueous H\(_2\)O\(_2\), alkaline photoresist developer (AZ developer, Microchemicals), and pure water), the wafers were immersed for 2–3 days in 2\% toluene/hexane solution of silylating agents: OTS (octyltrithoxysilane, TCI) or PFES (1H,1H,2H,2H-perfluorodecyltrithoxysilane, TCI). The wafers were sequentially rinsed by \( o \)-xylene, 2-propanol/acetone, and alkaline hydrogen peroxide, followed by sonication in 2-propanol/acetone for 2 minutes. The dried substrates were stored in N\(_2\). The completion of SAM functionalization was confirmed by wetness against pure water. Contact angles were 99° and 96° for the PFES and OTS patterned areas, respectively (Figure S1a,b). Non-contact mode atomic force microscopy (AFM, Nanonavi II, SII) revealed that residues of the photography process were hardly seen on the substrate surface (Figure S1c,d).

Supporting Information B: Crystal growth and lamination process

Growth processes of the \( \kappa \)-(BEDT-TTF)\(_2\)Cu[N(CN)\(_2\)]Cl (\( \kappa \)-Cl) crystal were performed in a similar manner described in the previous reports\(^{40,61}\) although slight modifications were included to efficiently obtain a quantity of thin crystals with good surface flatness. The mixture of bis-(ethylenedithio)tetrathiafulvalene (BEDT-TTF; 24 mg), CuCl (52 mg), TTP·[N(CN)\(_2\)] (TTP = tetraphenylphosphonium ; 200 mg), and TPP·Cl (90 mg) was dissolved in 1,1,2-trichloroethane (10\% v/v ethanol) solution (50 mL), and transferred to the specially-designed electrochemical cell after the residues were filtered. Electrochemical oxidation of BEDT-TTF was performed in Ar by applying galvalnostatic current (5 \( \mu \)A) for 120 hours at 30 °C. Then the electrochemical cell was opened to air and kept at 20 °C for several hours until a large number of rhombic thin-layer crystals were precipitated. Particularly thin \( \kappa \)-Cl crystals (30–150 nm thick) with a well-defined shape were selected using optical microscopy and pipetted into a petri dish filled with pure 2-propanol
Figure S1. **Estimation for SAM-patterned substrate surfaces.** (a, b) Wettability test by water droplets (side view). The right halves of the SiO$_2$/Si substrate (6 mm × 6 mm × 0.3 mm) are functionalized by PFES (a) or OTS (b) through the same process as the SAM patterning of the channel areas. The $\theta_c$ denotes the contact angle. (c, d) AFM topographic images for the substrate surface measured on the PFES-patterned area (c) and the bare SiO$_2$ area (d). (e–m) Pre-lamination testing for wettability of the SAM-patterned substrate. Shown is the same substrate as is designed for conductivity measurements, on which Au/Ti microelectrodes (orange colored) and a SAM-functionalized area (PFES; 90 µm × 98 µm in dimension, as shown in panel (n)) are patterned. After dipping the substrate into the lamination solvent (2-propanol), the droplet of 2-propanol is left on the substrate ((e), bright part). During several seconds, 2-propanol is vaporized and the droplet gradually shrinks. When the edge of the droplet goes through the channel area, 2-propanol on the SAM-patterned area is immediately repelled, which can be observed as deformation of the droplet edges (i, j). (n) Magnified view of electrode/SAM-pattern design around the channel area. SAM pattern is located between the electrodes labeled as drain 1 and source (highlighted by a red square).

for rinsing. After the single crystal was gently transferred on an electrode-patterned substrate in 2-propanol, the substrate was carefully dried to complete lamination of $\kappa$-Cl. In order to confirm that the SAM-patterned area is steadily formed on the substrate, we performed a simple wettability test using 2-propanol *just before* transferring $\kappa$-Cl onto it (Figure S1[e–m]). Solvents and reagents were used as received from commercial sources.
Figure S2. **Optical photo images of Mott-FETs for various thicknesses of κ-Cl single crystals.** Thickness: 30 nm (a), 40 nm (b), 50 nm (c), 65 nm (d), 79 nm (e), 100 nm (f), 120 nm (g), and 141 nm (h), corresponding to 20, 27, 33, 43, 53, 67, 80, and 94 layers of BEDT-TTF, respectively. Apparent colors of κ-Cl crystals are dependent on their thicknesses, which is likely to be due to the optical interference at κ-Cl/(SAM)/SiO$_2$/p$^+$-Si interfaces. Scale bar: 100 µm.

**Supporting Information C: Assessment of thin-layer crystal channel**

Thickness of the laminated κ-Cl crystal was estimated by a surface profiler (P-15, KLA-Tencor). Figure S2 displays microscope images of the laminated thin-layer κ-Cl crystals with various thicknesses. Isolated crystals became almost colorless and transparent when their thicknesses are below ~100 nm. However, by lamination on the (SAM)/SiO$_2$/p-doped Si substrate, the thin-layer κ-Cl crystals became clearly visible and differently colored depending on their thickness, probably due to an optical interference effect$^{62,63}$ (note that the SAM functionalization of the interface did not alter apparent colors of the κ-Cl crystals). Thus, one can estimate the thickness as well as the cleanness of the crystal by optical microscopy, which enables one to immediately screen out samples unsuitable for subsequent measurements. The selected devices were further analyzed by using AFM, and we eventually obtained κ-Cl single crystals without any grains on the surface and with the flatness of the single BEDT-TTF molecular layer level (1.5 nm; see Figure S3). After forming a Hall-bar geometry by the laser ablation of the κ-Cl crystal, carrier transport measurements were performed.
Figure S3. **AFM images of the surface for κ-Cl and κ-Br.** (a,b) Topography for the surface of typical κ-Cl crystals used in the present study (the same images as those shown in Figure 1c,d in the main part). Scale bar: 500 nm. (c,d) Topography for the surface of κ-Br crystals, which are fabricated in accordance with the method previously reported. As contrasted with κ-Cl, the surface of κ-Br has many pits and molecular steps, the height of which corresponds to the thickness of one layer (1.5 nm) or two layers (3 nm) of BEDT-TTF.

Table S1. Device characteristics.

| Device            | F#1 | C#1 | C#2 | C#3 |
|-------------------|-----|-----|-----|-----|
| SAM species       | PFES| OTS | OTS | OTS |
| Crystal thickness of κ-Cl [nm] | 40  | 140 | 80  | 120 |
| $V_{MIC}$ on SAM-covered area [V] | 76  | 76  | 51  | 48  |
Figure S4. OTS-SAM functionalization effect on carrier transport properties of a $\kappa$-Cl Mott-FET and comparison with the PFES-SAM functionalization effect. Transfer curves of device C#1 for various $T$ on a OTS-covered substrate (a) and on bare SiO$_2$ (b). Temperatures are 7 to 35 K by 3.5 K steps. Dashed lines mark $\sigma = 0.1e^2/h$. The insets of (a) and (b) indicate the $T$-dependence of the onset voltage $V_{\text{onset}}$, where the transfer curves crosses the $\sigma = 0.1e^2/h$ line. (c) Onset voltage shift due to SAM functionalization, $\Delta V_{\text{onset}}$, as a function of $T$ for devices F#1 and C#1. Dotted lines are guides to the eye.

Supporting Information D: SAM species dependence on transport properties

In the main part, we demonstrated that the metal–insulator transition (MIT) in $\kappa$-Cl FET is facilitated by chemical functionalization of the channel/gate dielectric interfaces using hydrophobic SAMs. To elucidate the interfacial SAM functionalization effect in depth, we present dependences of carrier transport properties on the SAM species in this section.

Figure S4 shows the transfer curves for device C#1, which has the same device geometries as device F#1 (Figure 1a in the main part) but OTS is used as SAM instead of PFES. Since device C#1 provides both $\kappa$-Cl/OTS/SiO$_2$ and $\kappa$-Cl/SiO$_2$ interfaces on an identical single crystal $\kappa$-Cl, one can extract effects of the OTS functionalization on transport properties. For the OTS-covered area (Figure S4a), a clear MIT is observed, where $\sigma$ increases (decreases) with decreasing $T$ on the highly (lightly) doped regime. One can recognize that the series of transfer curves are similar to that for the PFES-functionalized area on device F#1 (Figure 1b in the main part); indeed, gate voltages of the crossover point ($V_{\text{MIC}}$) are identical (Table S1), and the device mobility for device C#1 ($\sim 180 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for $V_g = 120 \text{ V}$ at 7 K) is comparable to that for F#1 ($\sim 200 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for $V_g = 120 \text{ V}$ at 7 K). The bare SiO$_2$ area exhibits insulating behaviors with $n$-type FET characteristics in the whole $V_g$ range (Figure S4b), which is qualitatively similar to the bare SiO$_2$ area on device F#1. However, the magnitude of conductivity on bare SiO$_2$ area of device C#1 is suppressed to lower than that of device F#1; for example, $\sigma(V_g = 120 \text{ V})$ at 31.5 K is $1.0e^2/h$ for
device F#1, but only 0.16e²/h for device C#1. This discrepancy implies that there is a difference in the intrinsic doping levels of κ-Cl between devices C#1 and F#1; that is, κ-Cl on device C#1 should be at deeper insulating (less doped) region in the phase diagram in the absence of the extrinsic doping by the gate electric field or SAMs. The origin of the device-dependent intrinsic doping remains unclear, yet it is possibly due to charge imbalance at the crystal surface, which is commonly observed in crystalline materials consisting of cation/anion stacking layers.

In order to investigate the effect induced by SAMs, let us define the onset voltage \(V_{\text{onset}}\) of transfer curves as the gate voltage at which \(\sigma\) exceeds 0.1e²/h. (Note that the magnitude of the threshold conductivity is arbitrary, and choice of conductivity other than 0.1e²/h does not change the results qualitatively.) The \(V_{\text{onset}}\) shown in the insets of Figure S4a,b serves as an indicator of the boundary between the strongly localized regime and the relatively conductive regime. By taking the difference of \(V_{\text{onset}}\) between the SAM-covered region and the bare SiO₂ region in an identical single-crystal device, \(\Delta V_{\text{onset}} = V_{\text{onset}} (\text{SAM}) - V_{\text{onset}} (\text{SiO₂})\), one can cancel out intrinsic doping effect of κ-Cl and extract the SAM functionalization effect. Figure S4c compares \(\Delta V_{\text{onset}}\) for devices C#1 and F#1. Over the entire \(T\) range, device C#1 (OTS) shows more negative \(\Delta V_{\text{onset}}\) than device F#1 (PFES). The tendency of \(\Delta V_{\text{onset}} (\text{PFES}) > \Delta V_{\text{onset}} (\text{OTS})\) is consistent with the polarities of SAMs revealed in the previous semiconductor-based device experiments; that is, the PFES SAM induces positive charge on the FET channel due to the electric dipole moment of PFES molecules, whereas the OTS SAM serves as a neutral one. On the other hand, large negative offsets for \(\Delta V_{\text{onset}}\) on both devices imply other dominant effects by SAM functionalization than the simple doping in the present Mott FETs. At the present stage it is difficult to determine the origin of the dominant factors due to lack of experimental methods that can access the buried interface underneath the several tens of nm thick κ-Cl crystal. Nevertheless, we can speculate that reduction of interfacial contaminants by hydrophobic SAMs will bring about apparent negative \(\Delta V_{\text{onset}}\).

It is widely known that the SAM functionalization at the interface effectively minimizes effects of charge traps, impurity-induced potential fluctuations, and atmospheric doping (e.g., oxygen or moisture) on the SiO₂ substrate surface. Furthermore, poor wetness of the SAM-covered region is favorable to reduce contamination during the κ-Cl lamination process. In the lamination process (see Supporting Information B), trace amounts of ingredients used for electrochemical synthesis of κ-Cl such as neutral BEDT-TTF molecules, Cl⁻ and TTP⁺ are unavoidably contained in the droplet of 2-propanol solvent even after careful rinsing. Such ingredients are to be concentrated during evaporation of the solvent, and eventually left as residues in the interface. This
type of contamination would be avoided on hydrophobic SAMs because their low wettability to 2-propanol results in quick elimination of solvent before the ingredients are fully concentrated (Figure S1). Because the Mott transition is particularly sensitive to disorder, cleaner interfaces expected on the hydrophobic-SAM-covered area facilitates the MIT with low-level electron doping, leading to significant negative shift of $V_{\text{onset}}$. As a consequence, the charge doping by SAMs, the most prominent effect of SAM functionalization on conventional semiconductor devices, is considered to be merely a minor effect on the present Mott FET.

**Supporting Information E: High-\(T\) transport**

In the main part, scaling analysis was performed for \(T < 40\) K. This is due to the fact that the charge transport above \(T > 50\) K no more exhibits the quantum critical behavior described by eq. (1). In this section, we briefly consider the cause for such deviation.

The electronic and magnetic properties of bulk \(\kappa\)-Cl have been intensively studied including the pressure effects.\(^{42,71-73}\) At ambient pressure, bulk \(\kappa\)-Cl is a strong insulator showing basically activation-type dc-transport characteristics. In decreasing \(T\), the system undergoes three transport regimes: (i) the high-\(T\) phase above \(T \sim 50\) K, with relatively small transport gap, (ii) the paramagnetic Mott insulator phase (PMI, \(T > 25\) K), in which the electron correlation becomes significant and \(d\sigma/dT\) exhibits a larger transport gap, (iii) the antiferromagnetic Mott insulator phase (AFI) at low \(T\). Figure [S5] compares the in-plane conductivity of \(\kappa\)-Cl at ambient pressure, which were reported in literature.\(^{67-70}\) Although the absolute values of the conductivity are different, semiconductor–PMI crossovers are commonly observed near \(T \sim 50\) K. The transport gap \(\Delta\), which is related to the conductivity by \(\sigma \propto \exp(\Delta/2k_BT)\), is \(\sim 20\) meV in the semiconductor regime, whereas that in the PMI regime depends on samples taking the values within 50–100 meV.

We can estimate the sheet conductivity \(\sigma_{\text{layer}}\) per layer from the in-plane conductivity \((\sigma_{3D} = L/WDR\), where \(R\) is resistance and \(L, W, D\) are the channel length, width, and thickness, respectively) and the thickness of one layer (1.5 nm), as indicated in Figure [S5]. As is mentioned previously, the typical thicknesses of thin-layer \(\kappa\)-Cl crystals used in our FET devices are in the range of 40–140 nm, corresponding to 26–93 BEDT-TTF layers. Since the observed \(\sigma\) is sum of conductances of the internal and surface channels, the contribution from the non-doped internal channel becomes significant in the carrier transport on FET in the high-\(T\) regime.\(^{35}\) On the other hand, the rapid decrease of \(\sigma_{\text{layer}}\) from \(\sim 10^{-3} e^2/h\) with lowering \(T\) is favorable to precisely evaluate the
Figure S5. **Carrier transport in the bulk channel of κ-Cl single crystal and its contribution to the FET conductivity at high $T$ ($T > 50$ K).** (a) Temperature dependence of the in-plane conductivity in bulk κ-Cl single crystals found in literature. Data are adapted from Refs. [67-70]. Left axis ($\sigma_{\text{layer}}$) indicates the conductivity per one BEDT-TTF conducting layer (thickness: 1.5 nm), which is calculated from three dimensional conductivity (right axis). (b) Sheet conductivity of the FET device at high $T$ plotted for various gate voltages (from $V_g = -120$ V to 120 V with the step of 40 V, colored solid lines). The MIC point in the intermediate-$T$ region ($T = 25 - 40$ K) was $V_{\text{MIC}} = +56$ V ($\sigma_{\text{MIC}} = 1.4e^2/h$), and the crystal thickness was 120 nm (80 layers). Dashed line provides $V_g$-independent part of sheet conductivity. For comparison, shaded area shows the expected sheet conductivity of the bulk κ-Cl crystal, which is obtained by conversion of the three dimensional conductivities (panel a) into the values for the thickness of 120 nm (i.e., $\sigma = 80 \times \sigma_{\text{layer}}$). Inset: $V_g$-dependent part of the high-$T$ sheet conductivity as a function of $T$ for varying $V_g$.

Critical phenomena around the MIC on the surface channel occurring at $\sigma \sim [10^{-2} - 10^0]e^2/h$.

Figure S5b represents the high-$T$ sheet conductivity of the κ-Cl FET (thickness of κ-Cl: 120 nm) for various gate voltages. Note that here the thin-layer κ-Cl crystal is laminated on the OTS/SiO$_2$/doped-Si substrate and is subject to tensile strain upon cooling.$^{74}$ A metal–insulator crossover (MIC) is observed at $V_{\text{MIC}} = 56$ V in the intermediate-$T$ region around 30 K (not shown). In increasing $T$, $V_g$-dependence of $\sigma$ becomes smaller and $\sigma(T, V_g)$ curves converge to the insulating one for $V_g = 0$ V. To roughly estimate the surface-channel contribution on $\sigma$, we plot the $V_g$-dependent part of $\sigma(T, V_g)$ ($\Delta\sigma$, difference between the observed $\sigma$ and the $V_g$-independent part of conductivity indicated by dashed line in Figure S5) for varying $V_g$. The metallic transport ($d\Delta\sigma/dT < 0$) is limited to $\Delta\sigma \gtrsim e^2/h$; therefore no hallmarks of bad-metal (BM) behavior were found up to 70 K. The apparent transport gap of the FET in the high-$T$ limit is $\sim 50$ meV, which is
more than two times higher than that for bulk \( \kappa \)-Cl crystal. This could be due to the fact that, for the FETs, the thin-layer \( \kappa \)-Cl crystal laminated on the silicon substrate is subject to tensile strain (effectively negative in-plane pressure) by decreasing \( T \) because of the large difference in the thermal compressibility between the soft \( \kappa \)-Cl crystal and the incompressible substrate. Such an effective pressure is expected to reduce the transfer integrals between BEDT-TTF dimers, enhancing the insulating character of the internal channel of the laminated \( \kappa \)-Cl of FET than the purely bulk crystal. On the other hand, the sheet conductivity of FET exceeds \( 10e^2/h \) at 250 K, which corresponds to \( 0.2e^2/h \) per one-layer BEDT-TTF channel, and is nearly an order of magnitude higher than the conductivity expected for a bulk crystal. It is unclear what makes this discrepancy; one possibility is the difference in the measuring geometry (the aspect ratio of the channel and the wiring geometries) between the bulk and FET samples.

**Supporting Information F: Phase diagrams of doping/pressure-driven Mott transitions**

Here we map out the 3D phase diagrams around a Mott insulator based on both experiments and theories. Figure S6a shows the experimental mapping of the phases and the crossover lines. The phase diagram for a doping-driven metal–insulator transition (MIT), shown in the \( T – V_g \) plane, is determined from the present experiment on a single-crystal \( \kappa \)-Cl FET (device C#2). The conductivity of bulk \( \kappa \)-Cl at half-filling (the \( T – P \) plane) is converted to that per layer (\( \sigma_{\text{layer}} \), defined in Supporting Information F) for comparison with the sheet conductivity obtained from measurement on the FET. The MIC lines are denoted by dashed lines. Taking into account the \( n \)-type characteristics and the tensile effect for the \( \kappa \)-Cl FET fabricated onto the SiO\(_2\)/Si substrate, the coordinate origin for the doping-driven MIT is shifted toward the negative \( P \) and positive \( V_g \) directions. The quantum critical scaling exponents for the crossover regions are \( v_z = 0.49 \) (ref. 23) and 1.55 for the pressure-driven and doping-driven transitions, respectively.

Figure S6b shows the theoretical phase diagram calculated by a DMFT method. The conductivity is represented in units of the Mott–Ioffe–Regel limit \( \sigma_{\text{MIR}} \approx \sqrt{2\pi e^2/h} \) for quasi-2D systems, which is obtained by the condition of \( l \approx a \) (\( l \): mean free path of carrier, \( a \): lattice spacing). The low-\( T \) coexistence domes reside at the boundary of the Mott insulator and Fermi liquid (FL) phases, which accompany high-\( T \) quantum critical regions (QCR, the areas enclosed by the orange dotted lines) along the quantum Widom lines (QWL). Those phases continuously connect in the \( U – \epsilon – T \) space, yielding similarity between the phase diagrams for the pressure-
Figure S6. Phase diagrams for doping-driven and pressure-driven metal–insulator transitions around a Mott insulator. (a) Experimentally determined pressure–gate-voltage–temperature ($P – V_g – T$) phase diagram for \(\kappa\)-Cl with contour maps of conductivity in units of $e^2/h$. The phase diagram for a pressure-driven MIT at half-filling (left half of the panel) is determined from measurement for a bulk \(\kappa\)-Cl single crystal (adapted from ref. 23). (b) Interaction–chemical-potential–temperature ($U – \epsilon – T$) phase diagram based on DMFT solutions of the maximally frustrated Hubbard model, which is adapted from refs. 33, 46. $U$ and $D$ represent on-site Coulomb repulsion energy in the Hubbard Hamiltonian and the half bandwidth, respectively, and $\epsilon$ is the chemical potential measured from the half-filled level.

As described in the main text, the quantum phase transition (QPT) is a phase transition between competing ground states, which can be tuned by a non-thermal parameter of the quantum systems. In contrast to classical phase transitions driven by the thermal fluctuation, the QPT is driven by...
the quantum fluctuation stemming from Heisenberg’s uncertainty principle (or, more specifically, non-commutativity between the kinetic and potential operators in the Hamiltonian). Therefore, the QPT is intrinsically characterized by coupling of the spatial (static) and temporal (dynamical) fluctuations. Mathematically, these fluctuations are formulated as the correlation length $\xi$ in the spatial direction and the dynamical correlation length $\xi_\tau$ in the (imaginary) time direction, respectively. As the system approaches the critical point by tuning external parameters (e.g., magnetic/electric field, pressure, etc.), the two characteristic lengths diverge in coupled power-law forms of $\xi \propto |\delta|^{-\nu}$ and $\xi_\tau \propto \xi^z$, where $\delta$ is the distance from the critical point, and the power exponents $\nu$ and $z$ are the correlation exponent and the dynamical exponent, respectively. At finite $T$, $\xi_\tau$ is cutoff by $\hbar/k_B T$, so that the characteristics of the system are described by one parameter $|\delta|^{1/\nu}$ in the QCR (i.e., $T \gtrsim c|\delta|^z$). Applying this finite-size QPT theory to the MIT, one can obtain the expression of eq. (1) in the main text.

In the simplest one-parametric scaling on two-dimensional MITs, the conductivity of the crossover line, $\sigma^*$ (corresponding to the ‘separatrix’ in the nomenclature of renormalization group theory), is a constant of $\sigma_{\text{MIC}} \sim e^2/h$, and the critical point ($\delta = 0$) is defined on the constant

Figure S7. **One-parameter scaling at intermediate $T$ for device C#2.** Family of conductivity data plotted as a function of $|V_g - V_{\text{MIC}}| T^{-1/\nu z}$. Conductivity at the point of $|d\sigma/dT|_{V_g} = 0$ (i.e., $\sigma_{\text{MIC}}$) is marked by a dashed line. $\nu z = 1.41$ is estimated from a least-square analysis.
scaling variable line in eq. (1), which results in

\[ \sigma = \sigma_{\text{MIC}} F_\pm \left( \frac{|V_g - V_{\text{MIC}}|}{T^{1/n}} \right), \] (S1)

for the MIC induced by gate-electric-field doping. In fact, eq. (S1) does describe the conductivity for a narrow \( T \)-range of \( 22 \, \text{K} \leq T \leq 35 \, \text{K} \), at which one can see an obvious crossover from insulating to metallic regimes with a conductivity plateau \( |d\sigma/dT|_{V_g=V_{\text{MIC}}} = 0 \) in between (Figure 3b in the main part). In Figure S7 family of \( \sigma(V_g, T) \) for device C#2 are re-plotted with respect to \( |V_g - V_{\text{MIC}}| T^{-1/n} \). All the points including both the metallic (\( \sigma/\sigma_{\text{MIC}} > 1 \)) and insulating (\( \sigma/\sigma_{\text{MIC}} < 1 \)) sides collapse onto two branches by providing an exponent as \( n = 1.4 \pm 0.2 \). In terms of the form of the universal scaling function, the metallic side can be approximated by a linear function: \( F_+(y) = 1 + Ay \) with \( A = 0.14 \), that is, the metallic conductivity is given by the sum of a constant and a term proportional to \( T^{-1/n} \), which represents the non-Fermi liquid behavior in the vicinity of the MIC. For the the insulating side, the universal scaling function has a mirror symmetry of \( F_-(y) = 1/F_+(y) \) for \( \sigma \sim e^2/h \), yet in the deeper insulating regime, it has an exponential asymptotic form of \( F_-(y) \approx \exp(-By^C) \) with \( B = 0.12 \) and \( C = 1.3 \). Using \( n = 1.4 \), one obtains \( \sigma_{\text{ins}} \approx \sigma_{\text{MIC}} \exp[-(\Delta/2T)^{0.92}] \), which can be approximated by the activated-transport behavior \( \ln \sigma \propto 1/T \) above the intermediate-\( T \) regime (Figure 2 in the main part).

In the framework of renormalization group theory, however, \( \sigma^*(T) \) is not necessarily identical to the naïve definition of the MIT, \( \sigma^* = \sigma_{\text{MIC}} \). That is, metal and insulator are distinguished by behaviors of the dimensionless conductance on the scale transformation of the length scale and the imaginary time scale (or temperature); the conductance goes to zero in the limit of \( L \to \infty \) or \( T \to 0 \) through renormalization procedures for insulator, whereas not for metal. The ’separatrix’ is the line which delimits the boundary between metal and insulator regimes in the flow diagram of renormalization, and often has a \( T \)-dependent form of \( \sigma^* \) in practice. Therefore, it is essential for a scaling analysis to adequately identify the location of the ’separatrix’ or the crossover line in the phase diagram depending on the system under study.

**High-T Mott quantum critical scaling (HT-MQCS).** —As discussed in the main text, it is generally expected for Mott transitions that the MIC in the conductivity behavior exhibits quantum criticality along a crossover line referred to as ‘quantum Widom line (QWL)’. The QWL may show a bow-shaped trajectory in the phase diagram, and should smoothly converge to a critical
end point at a low $T$ (if present). For the pressure-driven Mott transition at half-filling, where the almost perfect mirror symmetry $\mathcal{F}_+(y) = 1/\mathcal{F}_-(y)$ holds for the wide conductivity range in the crossover region, one can identify the QWL simply by tracing the inflection points of conductivity, $d^2 \log \sigma/dP^2$ ($P$: pressure). For the doping-driven Mott transition in the present study, however, such a straightforward identification of the QWL was found to be impossible due to various factors (i.e., poor mirror symmetry, weak localization at low $T$, and lack of the critical end point in the range of observations). Therefore, we figured out a possible candidate for the QWL by an optimization calculation with the following procedure:

1. Setting series of initial $\{\sigma(T_i)\}$ for each temperature point $T_i$.

2. Calculating $\{V_g(T_i)\}$ by linear interpolation from $\sigma(T_i)$ and the adjacent experimental data points for $\sigma(V_g, T_i)$.

3. Obtaining the above-mentioned approximate/asymptotic forms for the universal scaling function $\mathcal{F}_\pm(|V_g - V_g^*|/T^{1/\nu_z})$.

4. Executing a least squares calculation under a constraint condition (see below) so that the family of conductivity data are converged to $\sigma^*(T) \times \mathcal{F}_\pm(|V_g - V_g^*|/T^{1/\nu_z})$, with a single critical exponent $\nu_z$. Here, the fitting parameters are $\nu_z$, series of $\{\sigma^*(T_i)\}$, and the variables included in the approximate/asymptotic forms of $\mathcal{F}_\pm$. The series of $\{V_g^*(T_i)\}$ are updated (procedure 2) for every optimization calculation loop.

**CONSTRAINT CONDITION:** $\sigma^*(T) = \sigma_{\text{MIC}}$ for $T > 23$ K (for definition of $\sigma_{\text{MIC}}$, see Figure S7 and the main text). This condition is introduced for consistency with the theoretically suggested QWL for a doping-driven Mott transition, that is, at high $T$, the conductivity on the QWL becomes independent of $T$ and coincides with the Mott–Ioffe–Regel limit.

Consequently, one can obtain $\nu_z$ giving the best scaling, as well as the optimized crossover line $V_g^*(T)$.

Figure S8 shows the result for analysis of HT-MQCS for devices C#1, C#3, and F#1, following the above procedure. As the result for device C#2 shown in the main part, one can recognize good convergence of family of conductivity data on the two branches of the universal scaling function for a wide range of conductivity. The critical exponents $\nu_z$ are 1.66, 1.41, and 1.35 for devices
Figure S8. **Transport characteristics in the vicinity of the doping-driven metal–insulator transition for devices C#1, C#3, and F#1.** (a–c) Temperature dependence of sheet conductivity as a function of $V_g$ for devices C#1 (a), C#3 (b), and F#1 (c). Single crystals of $\kappa$-Cl are laminated on the OTS/SiO$_2$ (300 nm)/doped-Si substrates (C#1 and C#3) or the PFES/SiO$_2$ (300 nm)/doped-Si substrates (F#1). Solid black lines represent $V_g = V_{\text{MIC}}$ lines giving $|d\sigma/dT|_{V_g=V_{\text{MIC}}} = 0$ in the intermediate-$T$ regime ($T \sim 25 – 35$ K). Dashed lines highlight the crossover lines $\sigma^*$ between the metallic and insulating regimes (possible QWL), by which the quantum critical scaling (eq. [1] in the main text) is retained in the widest $T$ range with $d\sigma^*/dT = 0$ at intermediate $T$. (d–f) Scaling plots of conductivity normalized by $\sigma^*(T)$ for devices C#1 (d), C#3 (e), and F#1 (f), yielding the critical exponents $\nu_z = 1.61, 1.44,$ and $1.35,$ respectively. Insets: Trajectories of $\sigma^*$ on the $V_g$ versus $T$ plane.

C#1, C#3, and F#1, respectively, being close to that for device C#2, $\nu_z = 1.55$ (Figure 3 in the main part). For all devices, the crossover conductivities $\sigma^*(T)$ are increasing functions with $T$ at low $T$, which is consistent with DMFT.\[33\]
Quantum critical scaling with a conventional crossover line.—To validate the HT-MQCS using an optimization calculation described above, let us consider a conventional approach to experimentally identify the crossover line of the MIC within the one-parametric scaling. The crossover line is expected to fulfill two conditions as follows:

1. $\sigma^* = \sigma(V_g = V_c, T)$. Namely, the crossover line is defined on a constant doping-density (critical density) line, $V_g^* = V_c$ ($V_c$: critical gate voltage). It is implicitly postulated that the gate voltage (or the doping density) should be a good scaling variable.

2. $\sigma^* \sim T^p$. The power-law in the crossover conductivity reflects the self-similarity nature of underlying physics at the critical point. The specific case of $p = 0$ corresponds to the naïve definition of the MIC with $d\sigma^*/dT = 0$, and is adopted in some systems exhibiting doping-driven metal–insulator (or superconductor–insulator) transitions. Nonzero $p$ is also, indeed, applicable to the case of low-density two-dimensional electron gas systems.

Figure S9 displays the results of the quantum critical scaling under various $V_c$ ranging from $-24$ V (insulating regime) to $101$ V (metallic regime at intermediate $T$). The characteristics of scaling sensitively change depending on the choice of $V_c$ as follows. First, the critical exponent $\nu_z$ for $V_c = V_{MIC}$ is 1.5, being close to the value under the HT-MQCS, but it becomes smaller (larger) for higher (lower) $V_c$, ranging from 0.9 to 2.2 (Figure S9c). Second, the quality of the conductivity scaling also depends on $V_c$; the scaling is successful in the widest $T$ range at $V_c \sim V_{MIC}$ (solid lines in Figure S9a,b), whereas the conductivity data convergence becomes worse as $V_c$ is away from $V_{MIC}$. Even for $V_c \sim V_{MIC}$, poor data convergence below $T \sim 12$ K becomes prominent particularly for the insulating regime (inset in Figure S9c-iv), which is in contrast to the HT-MQCS yielding successful scaling for $T > 7$ K. The $T$-dependence of the crossover conductivity deviates from the power-law form irrespective of $V_c$ (Figure S9b), which means that no crossover lines meet the condition 2 above mentioned. Indeed, the accurate low-$T$ behavior of the crossover line remains unclear in doping-driven Mott transitions, and awaits further theoretical investigation beyond DMFT. Nevertheless, these tendencies imply that HT-MQCS is essential for the doping-driven MIC in Mott insulators.
Figure S9. **Quantum critical scaling along the crossover lines with constant critical gate voltage, $V_g^* = V_c$.** Scaling analyses were performed for varying $V_c$: i) $-24$ V, ii) 1 V, iii) 26 V, iv) 51 V ($= V_{MC}$), v) 76 V, and vi) 101 V. (a) Location of the crossover lines in the $V_g - T$ plane. (b) Crossover conductivities $\sigma^*$ as a function of temperature in the log-log plot. For (a) and (b), solid (dashed) lines represent the $T$ regions where the scaling of the conductivity data is successful (failed) for each $V_c$. (c) Scaling plots in the $T$ region where the HT-MQCS is successful (Figure 3 in the main part). The inset of panel (c-iv) highlights the poor scaling quality in the insulating side of $\sigma/\sigma^* \gtrsim 1$ at low $T$. All data were collected for device C#2.
Supporting Information  H: Classical critical scaling

A number of previous studies for Mott insulators have revealed that the pressure-driven Mott transitions at half-filled show phase diagrams analogous to the liquid–gas transition in a classical fluid. Around a critical end point (CEP) at a finite $T$, physical quantities such as electrical conductivity were recognized to exhibit classical critical behaviors driven by thermal fluctuations, whereas transient from classical to quantum critical behaviors occur at a higher $T$. In this section, we verify the applicability of the classical phase transition for the present system. First, we present the formalism we use for the classical critical scaling analysis. Starting from a fundamental homogenous equation describing the classical criticality, two different interpretations for conductivity scaling are introduced. Then we compare the derived theoretical scaling formula with the experimentally observed conductivity behavior around the doping-driven metal–insulator transition in the present study.

Figure S10 shows the general phase diagram of the gas–liquid-type phase transition. The transition is controlled not only by $T$ but also by $H$, the field conjugate to the order parameter $\phi$ in the Hamiltonian. At a low $T$, the dense “liquid” and dilute “gas” phases are separated by a coexistence line. In going across the coexistence line, the thermal fluctuation drives the phase

![Figure S10. General phase diagram for liquid–gas transition of classical fluids.](image)

There is a critical end point at $(H_c, T_c)$, below which the liquid-like and gas-like phases are separated by a coexistence line. Above $T_c$, the liquid–gas crossover is characterized by the Widom line. Here, these two lines are collectively expressed as $H_c(T)$. 
transition with a discontinuous jump of $\phi$. The coexistence line terminates at a CEP $(H_c, T_c)$. Above $T_c$, it is possible that the system undergoes a continuous crossover between the liquid-like phase to the gas-like phase, which are delineated by a crossover line referred to as Widom line$^{52}$. With regard to the difference from the quantum phase transition, the most prominent feature in the classical phase transition is the singularity of the thermodynamic quantities when the system approaches to the finite-$T$ CEP: $(H, T) \rightarrow (H_c, T_c)$. The critical behaviors can be described by scaling laws, which are derived as a consequence of a renormalization group analysis around the CEP.

In the scaling theory, it is assumed that the singular part of the free energy density $f_s$ is characterized by the correlation length $\xi$ in the vicinity of the CEP. Given the distances from the coexistence line ($T < T_c$) or the crossover line ($T > T_c$) as $h = (H - H_c(T)) / H_c(T)$ and $t = (T - T_c) / T_c$, the renormalization group theory states that $f_s$ and $\xi$ are formulated as homogeneous forms using a rescaling factor $b (> 1)$:

\begin{align}
L^d f_s(h, t) &= (L/b)^d f_s(b^s h, b^t t), \\
\xi(h, t) &= b \xi(b^s h, b^t t),
\end{align}

(S1a) (S1b)

where $L$ and $d$ are the system size and the dimension, respectively. These equations represent the relation when the system is seen from two different length scales, i.e., the original scale $x$ and the $b$-fold coarser scale $x' = x/b$. Integrating out the short-range fluctuation contribution in the latter scale leads to renormalization for the parameters as $h' = b^s h$ and $t' = b^t t$, by which the free energy density can be expressed by the same function as that in the original scale, $f_s(h', t')$. Since the scale transformation also reduces the localization length and the system volume to $\xi' = \xi/b$ and $(L')^d = (L/b)^d$, respectively, the above two equations are yielded. Note that for the classical liquid–gas type transition, both $h$ and $t$ are relevant parameters, and thus the exponents $s_h$ and $s_t$ are positive. Choosing $b^s = 1/|h|$ in eqs. (S1a) and (S1b), one can obtain the following two scaling relations:

\begin{align}
f_s &= |h|^{d/s_h} f_s(\pm 1, t/|h|^{s_t/s_h}) \equiv |h|^{d/s_h} g_{f_{1, h\pm, t\pm}} \left( \frac{h}{|h|^{s_t/s_h}} \right), \\
\xi &= |h|^{-\nu_c} g_{\xi_{1, h\pm, t\pm}} \left( \frac{h}{|h|^{s_t/s_h}} \right),
\end{align}

(S2a) (S2b)
where $\Delta_c = s_h/s_t$ is called as gap exponent and $\nu_c = 1/s_h$ is the critical exponent for correlation length. Similarly, choice of $b^c = 1/|t|$ in eqs. (S1a) and (S1b) leads to

$$f_s = |t|^{d/s_t} f_s (h/|t|^{s_h/s_t}, \pm 1) \equiv |t|^{d/s_t} g_{f_2, h, \pm, t_s} \left( \frac{h}{|t|^{\Delta_c}} \right),$$

(S3a)

$$\xi (h,t) = |t|^{-\nu} g_{\xi 2, h, \pm, t_s} \left( \frac{h}{|t|^{\Delta_c}} \right),$$

(S3b)

with $\nu = 1/s_t$. In each equation, the single-parameter function $g_{x, h, \pm, t_s}$ is a universal scaling function, where the pair of symbols $\pm$ denotes four branches defined on the sectors with different signs of $t$ and $h$.

Scaling relations for physical quantities can also be derived from the homogeneous formula for the free energy density (eq. (S1a)). For example, the thermally averaged order parameter $m = \partial f_s / \partial h$ (corresponding to magnetization in a magnetic phase transition) satisfies

$$m (h, t) = b^{s_h-d} m (b^{s_h} h, b^s t).$$

(S4)

With the proper choice of $b$ as above, one can obtain a pair of scaling relations:

$$m (h, t) = |h|^{d/\Delta_c} g_{m1, h, \pm, t_s} \left( \frac{h}{|t|^{\Delta_c}} \right),$$

(S5a)

$$m (h, t) = |t|^{d/\Delta_c} g_{m2, h, \pm, t_s} \left( \frac{h}{|t|^{\Delta_c}} \right),$$

(S5b)

which are akin to the scaling relations for the free energy density except that the exponents of $|h|$ and $|t|$ are different. In general, an arbitrary physical quantity should be amenable to a similar scaling relation as a function of $h/|t|^{\Delta_c}$.

Critical exponents of the transition can be straightforwardly obtained by taking the limit of $h \to 0$ or $t \to 0$ for the above scaling relations. Relations between the critical exponents and the rescaling factors are summarized in Table S2.

From an experimental standpoint, the Mott critical behaviors have been investigated mainly by precise measurements of the dc conductivity in the vicinity of the pressure-induced Mott transition at half-filling. A conductivity scaling identical to the form of eqs. (S2a), (S2b), and (S5a) has
Table S2. Classical scaling exponents.

| exponent | representation | description |
|----------|----------------|-------------|
| $\alpha$ | $2 - \frac{d}{s_t}$ | $C \propto |t|^{-\alpha}$ for $h = 0$ (C: specific heat) |
| $\beta$ | $\frac{d}{s_t} - \Delta_c$ | $m \propto |t|^{\beta}$ for $h = 0$ and $t < 0$ |
| $\gamma$ | $2\Delta_c - \frac{d}{s_t}$ | $\chi \propto |t|^{-\gamma}$ for $h = 0$ and $t > 0$ ($\chi = \partial m/\partial h$: susceptibility) |
| $\delta$ | $\frac{s_h}{d-s_h}$ | $m \propto |h|^{1/\delta}$ for $t = 0$ |
| $\nu$ | $\frac{1}{s_t}$ | $\xi \propto |t|^{-\nu}$ for $h = 0$ |
| $\nu_c$ | $\frac{1}{s_h}$ | $\xi \propto |h|^{-\nu_c}$ for $t = 0$ |

$^a$ The exponent identities are deduced from above representations: $\alpha + 2\beta + \gamma = 2$ (Rushbrooke’s identity), $\beta(\delta - 1) = \gamma$ (Widom’s identity), and $\alpha = 2 - \nu d$ (Josephson’s identity).

$^b$ $\Delta_c = s_h/s_t = \beta\delta = \gamma\delta/(\delta - 1)$.

commonly observed around the CEP ($H_c, T_c$) in various Mott insulators, which read[24,55,79]

$$\sigma_{metal}(h, t) - \sigma^*(T) = |h|^{s_{crh}} g_{crh,\xi} \left( \frac{h}{|h|^{\Delta_c}} \right), \quad \text{(S6)}$$

where $\sigma^*(T)$ is the $T$-dependent crossover conductivity, and pressure $[P - P_c(T)]/P_c(T)$ is taken as $h$ for pressure-induced Mott transition. The $\sigma_{metal}$ indicates that the scaling holds in the metallic region ($H > H_c$). This type of conductivity scaling is also confirmed by theoretical studies based on DMFT calculation. Although the conductivity scaling seems convincing, the universality class for the Mott transition at half-filled is still controversial. This is attributed to the fact that all the physical quantities exhibit mathematically identical scaling formula as shown above, and one cannot know solely from the conductivity scaling which physical quantity should be assigned to the experimentally observed $\sigma_{metal} - \sigma^*$. Therefore, as one interpretation, it is postulated that $\sigma_{metal} - \sigma^*$ is proportional to $m$ (i.e., the density of doublon–holon pairs). The scaling formula for $m$ then can be applicable to $\sigma_{metal} - \sigma^*$, leading to $s_{crh} = 1/\delta$, $\Delta_c = \beta\delta$, and

$$\sigma_{metal}(h = 0, t < 0) - \sigma^*(T) \sim |t|^\delta, \quad \text{(S7)}$$

$$(\partial/\partial h) [\sigma^*(T) - \sigma_{metal}(h = 0, t > 0)] \sim |t|^{-\gamma}, \quad \text{(S8)}$$

$$\sigma_{metal}(h, t = 0) - \sigma^*(T) \sim |h|^{1/\delta}. \quad \text{(S9)}$$

On the basis of this analysis, $(V_{1-x}Cr_x)_2O_3$, a three-dimensional Mott insulator, gives $\beta \approx 1/2,$
\( \delta \approx 3, \gamma \approx 1, \) and \( \Delta_c \approx 3/2, \) which correspond to the mean-field values for the Ising universality class.\(^{29}\) On the other hand, a two-dimensional organic Mott insulator \( \kappa - \text{Cl} \) provides \( \beta \approx 1, \delta \approx 2, \gamma \approx 1, \) and \( \Delta_c \approx 2, \) which cannot be accounted for by any conventional universality classes.\(^{24}\)

On the other hand, it was recently proposed that the conductivity behavior reflects the singularity of \( \xi \) around the CEP rather than the order parameter itself. According to the experimental argument by Abdel-Jawad et al.,\(^{55}\) \( \sigma_{\text{metal}} - \sigma^*(T) \propto 1/\xi \) holds at \( T = T_c. \) Extension for \( T \neq T_c \) can be readily done by assuming

\[
\sigma_{\text{metal}} - \sigma^*(T) = \frac{1}{\xi(h,t)} \vartheta \left( \frac{h}{|t|^\Delta_c} \right),
\]

(S10)

where \( \vartheta(y) \) is a universal function satisfying \( \vartheta(\infty) = 1 \) and \( \vartheta(0) = 0. \) Using eq. (S1b), we obtain

\[
\sigma_{\text{metal}} - \sigma^*(T) = |h|^\nu^r g_{\sigma r^+} \left( \frac{h}{|t|^\Delta_c} \right),
\]

(S11a)

\[
\sigma_{\text{metal}} - \sigma^*(T) = |t|^\nu^r g_{\sigma r^+} \left( \frac{h}{|t|^\Delta_c} \right).
\]

(S11b)

The first equation has the same form as eq. (S2b), and we obtain \( s_{\sigma r^h} = \nu_c \) and \( \Delta_c = \nu/\nu_c = \beta \delta. \) In this description, scaling analysis for \( (V_{1-x} \text{Cr}_x)_2 \text{O}_3 \) yields \( (\nu, \nu_c, \Delta_c) \approx (1/2, 1/3, 3/2), \) which again agrees with mean-field value for the Ising universality class. On the other hand, the critical exponents for two-dimensional organic Mott insulators (\( \kappa - \text{Cl} \) and \( \text{EtMe}_3\text{P[Pd(dmit)]}_2 \)) are \( (\nu, \nu_c, \Delta_c) \approx (1, 1/2, 2), \) which are close to theoretical values for \( \text{two-dimensional} \) Ising systems, \((\nu, \nu_c, \Delta_c)_{\text{theo}} = (1, 1/2, 15/8).\)\(^{55}\)

We now examine whether the scaling theory for classical liquid–gas-type transition is applicable to our experimental results for doping-induced metal–insulator transition on the two-dimensional Mott insulator \( \kappa - \text{Cl}. \) In analogy with paired scaling relations about the free energy density (eqs. (S2a) and (S3a)), we consider eq. (S6) and its counterpart:

\[
\sigma_{\text{metal}}(h,t) - \sigma^*(T) = |t|^{s_{\sigma r^+}} g_{\sigma r^+} \left( \frac{h}{|t|^\Delta_c} \right),
\]

(S12)

where \( s_{\sigma r^h} = s_{\sigma r^h} \Delta_c \) and \( g_{\sigma r^+}(0) = \left. \frac{dg_{\sigma r^+}(y)}{dy} \right|_{h=0} = 0 \) are satisfied. In our case, we take the gate voltage as a scaling variable, defining \( h = \left( V_g - V_g^* \right)/V_g^* \). The most significant parts lacking for the quantum critical scaling (eq. (1) in the main part) are the prefactors \( |h|^{s_{\sigma r^h}} \) and \( |t|^{s_{\sigma r^+}}, \) which yield...
non-monotonic behaviors in conductivity around the CEP. Indeed, taking the derivative of eqs. (S6) and (S12) with respect to \( h \), one can immediately obtain

\[
\frac{d\sigma_{\text{metal}}}{dh}\bigg|_{h=0} \sim |h|^{s_{\text{eh}}-1},
\]

(S13a)

\[
\frac{d\sigma_{\text{metal}}}{dh}\bigg|_{h=0} \sim |t|^{s_{\text{cr}}-\Delta_c},
\]

(S13b)

which lead to warping of \( \sigma(h,t) \) unless \( s_{\text{eh}} = 1 \). In our results, however, \( V_g \)-dependence of conductivity is quite monotonic in the vicinity of crossover line without any points showing \( d\sigma_{\text{metal}}/dh = 0 \) or \( d\sigma_{\text{metal}}/dh \to \infty \) in the measurement \( T \) range (see Figure 1 in the main part). Therefore, we have only to concentrate on two possibilities for the classical transition: (i) the scaling exponents satisfy \( s_{\text{eh}} = 1 \) and \( s_{\text{cr}} = 1/\Delta_c \), or (ii) \( T_c \) is well below the measurement \( T \) range, i.e., \( T_c < 2 \text{ K} \).

As for case (i), the scaling was found to fail for any choice of \( T_c \) and \( \sigma^*\left(V_g^*\right) \). Figure S11 shows examples of the scaling plots for provisionally assumed \( T_c = 25 \text{ K} \). (Note that other choice for the provisional \( T_c \) in the intermediate \( T \) range does not affect the conclusion.) For \( s_{\text{eh}} = \Delta_c = 1 \) (Figure S11a), the data are completely dispersed, and no universal curves can be identified. By assuming \( s_{\text{eh}} = \Delta_c \gg 1 \) (Figure S11b,c), the scaling for \( T > T_c \) (the lower branches
Figure S12. **Classical critical scaling for** $T_c \to 0$ **in the metallic regime.** A constant conductivity line ($\sigma^* = 1.38 e^2/h$) was taken as the crossover line. Best quality of scaling was obtained for $s_{rh}, s_{rt} \to 0$ and $\Delta_c = 0.65$, while the scaling gradually breaks down in decreasing $T$ ($T < 15$ K). Since the plot of $(\sigma_{\text{metal}} - \sigma^*) / (T - T_c)^{s_{rt}}$ (eq. (S12)) is almost identical to that of $(\sigma_{\text{metal}} - \sigma^*) / (V_g - V^*_g)^{s_{rh}}$ (eq. (S6)), only the latter is shown here.

of Figure S11b) is improved, whereas the data for $T < T_c$ (the upper branches of Figure S11b) is still scattered from a single curve. Hence the possibility for the universality class giving $s_{rh} = 1$ should be ruled out.

On the other hand, under assumption of (ii) $T_c < 2$ K better scaling is possible for $T_c \to 0$ as shown in Figure S12. Because the scaling quality is not sensitive to the way of choice for the crossover line $V^*_g (T)$ in this case, the constant conductivity line $\sigma^* = \sigma_c = 1.38 e^2/h$ was taken for the crossover line for simplicity. The scaling analysis leads to $s_{rh}, s_{rt} \to 0$, and a finite gap exponent $\Delta_c = s_h/s_{rh} = s_h/s_t \approx 0.65$. Namely, the conductivity scaling formula is given by $\sigma_{\text{metal}}(h, T) - \sigma^*(T) = g_{rt, \pm} (h/T^{\Delta_c})$, which is almost equivalent to the quantum critical scaling formula (eq. (1) in the main text). The meaning of these exponents depends on how to associate the conductivity to relevant physical quantities. If we adopted the $\sigma - \sigma^* \sim 1/\xi$ correspondence (i.e., $\nu_c = s_{rh}$ and $\nu = s_{rt}$), the exponents in the homogeneous scaling formula (eqs. (S1a) and (S1b)) would take physically inadequate values: $s_h, s_t \to +\infty$. Therefore, the $\sigma - \sigma^* \sim m$ correspondence
(namely, $1/\delta = s_{\text{eh}}$ and $\beta = s_{\text{or}}$) is the only acceptable interpretation for our results. From eqs. (S5a) and (S5b), $s_h = d = 2$ and $s_t = d/\Delta_c \approx 3.1$ are obtained, yielding the critical exponents of $\beta = 0$, $\gamma = \Delta_c \approx 0.65$, $\delta = \infty$, and $\nu = \Delta_c/d \approx 0.33$, which cannot be explained by any known universality classes.

To summarize, attempts for the classical critical scaling of the conductivity data are generally unsuccessful in our doping-driven Mott transition experiments since there is no singularity in the observed $d\sigma/dV_g$ that implies the existence for a finite-$T$ CEP. In the particular case of $T_c \to 0$, the classical critical scaling is possible although the scaling quality is worse than that for the quantum critical scaling. It is noteworthy that the classical scaling on the Mott transition has been found to hold within a narrow $T$ range around the CEP, empirically $T \lesssim 2T_c$.\textsuperscript{23,46} Hence one should be cautious about accepting the above discussion in which metallic conductivity data are scaled for a wide $T$ range of $10 \text{ K} < T < 40 \text{ K}$ in spite of the very low $T_c$. We emphasize that the observed $T_c \to 0$ behavior with wide-$T$-range scaling rather implies the zero-temperature critical point for quantum phase transitions driven by the high-energy scale fluctuations. Reminding the fact that in the quantum critical description the insulating regimes can also be scaled with an identical critical exponent (Figure 5 in the main part), it is more plausible to consider that the continuous conductivity crossover at an intermediate $T$ is brought about by a quantum phase transition.

**Cover Image**
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