Sudden slowing down of charge carrier dynamics at the Mott metal–insulator transition in \( \kappa-(D_8\text{-BEDT-TTF})_2Cu[N(CN)_2]Br \)

Jens Brandenburg\(^1,2\), Jens Müller\(^1,4\) and John A Schlüeter\(^3\)

\(^1\) Institute of Physics, Goethe-University Frankfurt, 60438 Frankfurt (M), Germany
\(^2\) Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany
\(^3\) Argonne National Laboratory, Materials Science Division, Argonne, IL 60439, USA
E-mail: j.mueller@physik.uni-frankfurt.de

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Abstract. We investigate the dynamics of correlated charge carriers in the vicinity of the Mott metal–insulator (MI) transition in the quasi-two-dimensional organic charge-transfer salt \( \kappa-(D_8\text{-BEDT-TTF})_2Cu[N(CN)_2]Br \) by means of fluctuation (noise) spectroscopy. The observed \( 1/f \)-type fluctuations are quantitatively very well described by a phenomenological model based on the concept of non-exponential kinetics. The main result is a correlation-induced enhancement of the fluctuations accompanied by a substantial shift of spectral weight to low frequencies in the vicinity of the Mott critical endpoint. This sudden slowing down of the electron dynamics, observed here in a pure Mott system, may be a universal feature of MI transitions. Our findings are compatible with an electronic phase separation in the critical region of the phase diagram and offer an explanation for the not yet understood absence of effective mass enhancement when crossing the Mott transition.

\(^4\) Author to whom any correspondence should be addressed.
1. Introduction

A key phenomenon in condensed-matter systems with strong electronic correlations is the Mott metal–insulator (MI) transition, i.e. the opening of a gap in charge-carrying excitations due to electron–electron interactions [1]. An important remaining challenge is the understanding of the static and dynamic criticality at the Mott transition and the question of universality [2–4]. Also, compared to the electronic band states in conventional metals, the metallic state induced in a correlated insulator can be radically different, showing for example pseudogap behavior [5, 6] or an intrinsic electronic phase separation on the nanoscale [7]. In particular, superconductivity of an unconventional nature and unusual metallic states are frequently observed in the vicinity of the Mott transition, e.g. in high-$T_c$ cuprates and organic charge-transfer salts. Among the latter, the quasi-two-dimensional $\kappa$-phase salts (BEDT-TTF)$_2X$, where BEDT-TTF (commonly abbreviated as ET) represents bis-ethylenedithio-tetrathiafulvalene and $X$ a polymeric anion, have recently attracted considerable attention as model systems for studying the physics of correlated electrons and the Mott phenomenon in reduced dimensions. In these materials, the Mott MI transition and critical endpoint can be easily accessed either by varying the ratio of bandwidth $W$ to effective onsite Coulomb repulsion $U$ by means of physical or chemical pressure, or by changing the temperature. Accordingly, the ground state of the antiferromagnetic insulator $X = \text{Cu}[\text{N(CN)}_2]\text{Cl}^-$ (denoted as $\kappa$-Cl) with $T_N = 27$ K can be turned into a superconducting state either by applying a moderate pressure of $\sim 300\text{ bar}$ or by modifying the anion: $\kappa-(\text{ET})_2\text{Cu}[\text{N(CN)}_2]\text{Br}(\kappa-\text{Br})$ is a superconductor with $T_c = 11.6$ K. A generalized phase diagram of these materials [8], shown in figure 1(a), is characterized by a critical region around $(W/U)_c$, where the S-shaped first-order MI transition line terminates in a second-order critical endpoint, which is reported to be in the temperature–pressure regime of $(P_0 \sim 200–300\text{ bar}, T_0 \sim 30–40\text{ K})$ [3, 6, 9, 10]. In the vicinity of the critical endpoint $(P_0, T_0)$, unusual metallic behavior characterized by a temperature scale $T^*$, where $T_0 \lesssim T^*(P > P_0) \simeq 35–55\text{ K}$, is associated with pronounced anomalies in transport, thermodynamic, magnetic and elastic properties; see [11] and references therein. A dramatic anomaly of the sound velocity, for example, is interpreted as an incipient divergence of the electronic compressibility caused by the proximity to the Mott transition [12]. A widely discussed explanation for the line of anomalies, $T^*(P)$, is a crossover from a coherent Fermi liquid at low temperatures into a regime with incoherent excitations (bad metal) at high temperatures [6, 13]. Recent thermodynamic investigations suggest that the broadened mean-field-like features at $T^*$ observed on the metallic side far from the critical point [14] develop into a critical behavior when approaching
It is found that the change in volume across the Mott transition shows critical scaling behavior caused by the coupling to the order parameter \[ T^* \] \[ (P_0, T_0) \] \[ 15, 16 \]. The change in volume across the Mott transition shows critical scaling behavior caused by the coupling to the order parameter \[ T^* \] \[ (P_0, T_0) \]. The change in volume across the Mott transition shows critical scaling behavior caused by the coupling to the order parameter \[ T^* \] \[ (P_0, T_0) \].

Although the unusual electronic transport properties have been studied in great detail \[ 3 \], there is only limited information about the dynamical properties of the charge carriers \[ 17 \], in particular at low frequencies. However, drastic changes of these properties are an important hallmark of critical behavior in the vicinity of a Mott MI transition \[ 7, 18 \]. In this paper, we report for the first time a correlation-induced, sudden slowing down of the electron dynamics as a fingerprint of the Mott critical point in \[ \kappa-(ET)_2X \], which may be considered as a universal feature of MI transitions. Furthermore, our results offer an explanation for the not yet understood non-divergence of the effective thermodynamic electron mass at the Mott critical point.

2. Experiment

2.1. Single-crystal growth

Single crystals of \[ \kappa-(ET)_2X \] with \[ X = \text{Cu}[\text{N(CN)}_2]\text{Br}^- \] of plate- and rod-shaped morphology were grown by electrochemical crystallization according to the literature methods \[ 19 \]. In this study, we focus on the fully deuterated variant \[ \kappa-(\text{D}_8-\text{ET})_2\text{Cu}[\text{N(CN)}_2]\text{Br} \] (hereafter denoted as \[ \kappa-\text{D}_8\text{-Br} \]), where all hydrogen atoms in the donor molecule are replaced by deuterium atoms. At ambient pressure, this material is located very close to the Mott critical point in the \[ T(P, X) \] phase diagram \[ 5, 20, 21 \]; see figure 1(a).

2.2. Fluctuation spectroscopy

To experimentalists, noise is often a nuisance rather than a signal. Noise, however, is caused by fluctuations of microscopic entities and its measurement probes the microscopic motion and transitions of particles. By analyzing time-resolved conductivity data, information about the kinetics, i.e. the spectroscopic information, of the relevant microscopic processes in solids can be gained.

In this work, low-frequency fluctuation spectroscopy measurements have been carried out in a five-terminal setup using a standard bridge-circuit ac technique \[ 22 \]. The experiment is described in detail in \[ 23 \]. Care has been taken that spurious noise sources, in particular contact noise, do not contribute to the results. Resistance \( R \) and noise power spectral density (PSD) \( S_R \) have been measured perpendicular to the conducting layers. \( S_R \) is calculated from the voltage noise PSD defined by \[ S_V(f) = \langle |\delta \tilde{V}(f)|^2 \rangle \], where \( \delta \tilde{V}(f) \) is the Fourier-transformed fluctuating voltage drop across the sample and the brackets \( \langle \rangle \) denote the time average. Typical spectra (raw data) are shown in figure 1(b); a discussion of reproducibility and error bars is presented in a previous publication \[ 24 \]. The noise PSD is related via the Wiener–Khintchine theorem to the current–current correlation function (or likewise—in voltage measurements—the voltage autocorrelation function \( \Psi(\tau) = \langle V(t) \cdot V(t + \tau) \rangle \)) of the electronic system \[ 25 \]:

\[
S_V(f) = 4 \int_0^\infty \Psi(\tau) \cos(2\pi f \tau) \, d\tau,
\] (1)
Figure 1. (a) A schematic generalized phase diagram of κ-(ET)$_2$X. $(P_0, T_0)$ denotes the second-order critical endpoint of the first-order Mott MI transition (red line). PI, PM, AFI and SC denote the paramagnetic insulating and metallic, antiferromagnetic insulating and superconducting phases, respectively. $T^*$ characterizes the anomalous normal conducting state and $T_g$ denotes the glass-like ordering temperature of the ET molecules’ terminal ethylene groups. (b) Representative spectra of the resistance noise PSD $S_R$ of κ-D$_8$-Br, exemplarily shown for two temperatures, where both the magnitude and the slope of the $1/f^\alpha$ noise are different. (c) Resistance and integrated resistance noise power versus temperature.

and thus characterizes the microscopic kinetics of the electronic fluctuations. Also, in certain cases, a fluctuation–dissipation relation provides a connection to the imaginary part of the dynamic susceptibility, i.e. $S(\omega) \propto \tilde{\chi}''(\omega)/\omega$.

For the present material, at all temperatures shown here, we have observed excess noise of general $1/f^\alpha$ type in the frequency range of $10 \text{ mHz} < f < 50 \text{ Hz}$, characterizing the intrinsic resistance (conductance) fluctuations. Figure 1(b) exemplarily shows the resistance noise PSD at two different temperatures. At selected temperatures, we have extended the frequency range down to $1.6 \text{ mHz}$ and up to $100 \text{ Hz}$, in which case a full spectrum (50 averages) requires a measuring time of about 8 h. Special measures have been taken to ensure sufficient temperature stability (our setup allows for $\Delta T < 1 \text{ mK}$, e.g. at 200 K) for the time of taking the spectra. The spectral weight of the resistance fluctuations presented in figure 1(c) is calculated...
by integrating the spectra $S_R(f)$ over the measured frequency range, i.e. it represents the variance of the resistance fluctuations in the given bandwidth. The frequency exponent $\alpha(T)$ presented in figure 2(a) has been determined by a linear fit to the spectra taken at different temperatures in a double-logarithmic plot. Down to the lowest frequencies, no cut-off frequency, i.e. crossover from $1/f$-type noise to frequency-independent (‘white’) noise, has been observed. Such long-term-memory correlations are commonly observed in macroscopic condensed-matter systems, even down to the $\mu$Hz regime [26]. Likewise, no high-frequency cut-off, i.e. crossover from $1/f$- to $1/f^2$-noise, has been seen; rather the $1/f$-type spectra drop below the ‘white’ background noise level of our experimental setup at about 100 Hz. The contour plot of the relative noise level $a_R(f, T) = f \times S_R/R^2$ with $S_R \propto 1/f^\alpha$ presented in figure 3 has been calculated from the fits to the raw data (spectra) described above, yielding the magnitude and frequency dependence of the noise shown in figures 1(c) and 2(a), respectively. In the contour plot we show frequencies up to 25 Hz for clarity and have extrapolated the spectra down to frequencies of 1.6 mHz for all temperatures.

3. Results

Representative of our experiments, figure 1(c) compares the resistance $R$ and the normalized spectral weight of the resistance fluctuations, $\langle (\delta R)^2 \rangle / R^2 = \int S_R(f) / R^2 \, df$, integrated over a bandwidth of 0.01–100 Hz for a deuterated sample $\kappa$-D$_8$-Br. The investigated samples are continuously cooled down to 4.2 K with a slow cooling rate of 3 K h$^{-1}$. Noise spectra have been taken while warming up the sample in discrete steps. For control, the resistance values are determined simultaneously and are found to be in good agreement with the continuous resistance measurement (figure 1(c), solid line). The $\kappa$-D$_8$-Br sample investigated here is situated very close to the Mott transition on the metallic side of the phase diagram. The resistance measurements are consistent with the resistivity reported in the literature for pressure-tuned $\kappa$-Cl [10]: at high temperatures, $R(T)$ exhibits a semiconducting behavior down to about 50 K, below which a step-like decrease of the resistance by almost two orders of magnitude indicates the occurrence of a metallic phase. Below 20 K, the resistance shows an increase again (with small hysteresis), which may be caused by electron localization near the Mott transition [27], before the transition into the superconducting state occurs at a temperature $T_c = 12.6$ K (onset). As becomes clear from figure 1(c), the temperature dependence of the spectral weight of the resistance noise $S_R(T)$ looks remarkably different from the resistance $R(T)$. Below a broad, Gaussian-shaped maximum centered around $T \simeq 100$ K, the integrated noise power decreases with decreasing temperature. Superimposed on this general trend is a pronounced anomaly at about 35–36 K, a temperature that is very close to the critical endpoint $(P_0, T_0)$ of the Mott transition, followed by two smaller peaks at lower temperatures.

Besides the noise PSD $S_R$ (magnitude of the fluctuations), the frequency exponent of the $1/f^\alpha$-type noise, $\alpha(T) = -\partial \ln S_R(f, T) / \partial \ln f$, can be extracted from the measured spectra as described in section 2.2. We find a strongly non-monotonic behavior of $\alpha(T)$, shown in figure 2(a), indicating substantial variations of the electrons’ dynamical properties in different temperature regimes.

Our previous studies have revealed that for the (ET)$_2$X salts the temperature dependence of the nearly $1/f$-type noise spectra can be well described by a generalized random fluctuation model, see [23, 24, 28], that was first used by Dutta, Dimon and Horn (DDH) to describe noise processes in metals caused by the diffusion of defects [29]. The phenomenological

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model assumes thermally activated fluctuators—not specified \textit{a priori}—which linearly couple to the resistance. The $1/f^\alpha$-type noise is produced by a large number of fluctuators acting in concert (each contributing a Lorentzian spectrum), whereas the distribution of activation energies $D(E)$ governs both the strong temperature dependence of the magnitude of the noise and any deviations from perfect (i.e. $\alpha = 1$) $1/f$ behavior. In this case, unlike for an individual fluctuator, the kinetics is non-exponential, i.e. the correlation function falls off with time, not exponentially [25], and

$$ S_R(f) = \frac{1}{\pi f} \int_0^\infty dE \frac{D(E, T)}{\cosh[(E - E_\omega)/k_B T]} . $$

Due to the large logarithmic factor in $E_\omega = -k_B T \ln \omega \tau_0$ ($\tau_0$ is an attempt time of the order of inverse phonon frequencies, typically $10^{-12} - 10^{-14}$ s), ordinary activation energies in solids can be accessed with this technique [25]. If $D(E)$ is not an explicit function of temperature, the assumptions of the DDH model can be checked for consistency by comparing the measured frequency exponent $\alpha_{\text{meas}}(T)$ with the predictions of the model [29]:

$$ \alpha_{\text{calc}}(T) = 1 - \frac{1}{\ln \omega \tau_0} \left( \frac{\ln S(f, T)}{\ln T} - 1 \right) . $$

As shown in figure 2(a), the agreement of the measured data with the generalized DDH model is excellent (see footnote 5). In particular, the model calculation provides a good quantitative

![Figure 2](http://www.njp.org/)
Figure 3. Contour plot of the relative noise level $a_R$ versus frequency $f$ and temperature $T$ in an Arrhenius representation. The white line corresponds to the ethylene endgroup activation energy, $E_a = 250$ meV, as extracted from the DDH model.

description of the measured $\alpha(T)$ and accounts for all of the features, i.e. non-monotonic temperature dependences being related to the fingerprints of the fluctuating entities, since for $\alpha$ greater or smaller than 1, $\partial D(E)/\partial E > 0$ and $\partial D(E)/\partial E < 0$, respectively. The striking agreement of the measured data with the model calculation underlines the physical meaning of extracting $D(E)$ from the temperature dependence of the magnitude of the noise by $D(E) \propto S_R/R^2 \times 2\pi f/k_B T$. From this analysis, shown in figure 2(b), we find that $D(E)$ exhibits a pronounced maximum at about $E_\omega \sim 250$ meV, which corresponds to the activation energy $E_a$ of the orientational degrees of freedom of the ET molecules’ terminal ethylene groups, as determined for example by NMR, specific heat and thermal expansion \[14, 30, 31\], i.e. in this case the relevant fluctuators can be identified \textit{a posteriori} \[24\]. Thus, the strong resistance fluctuations at about 100 K originate from the strong coupling of the electronic degrees of freedom to structural excitations related to the ET molecules’ outer ethylene groups.

4. Discussion

The noise results on $\kappa$-D$_8$-Br are summarized in a contour plot of the relative noise level $a_R(f, T) = f \times S_R/R^2$, shown in figure 3 in an Arrhenius representation. The enhanced noise level at elevated temperatures (about 105 K at $f = 1$ Hz) seen in the entire accessible frequency range is due to the coupling of the electronic fluctuations to the above-mentioned conformational motion of the ET molecules’ ethylene endgroups, as seen also in the related $\kappa$-Cl system \[24\]. The main result, however, is the abrupt and strong enhancement of $a_R$ by two orders of magnitude at $T^* \sim 36$ K. The increase of the noise level occurs at the lowest measuring
frequencies and corresponds to a large redistribution of the spectral weight of the electronic fluctuations to low frequencies. This is a signature of a sudden slowing down of the charge carrier dynamics due to the proximity to the critical endpoint at \( T_0 \approx 35–36 \) K.

Viewed from a different perspective, the observed enhancement of the noise level is consistent with the notion of electronic phase separation with metallic and insulating domains, as has been observed for example in NMR experiments [5] and scanning microregion infrared spectroscopy [32] below the temperature of the critical endpoint at 30–40 K. Such an electronically inhomogeneous state results in strongly enhanced local electric fields and current densities (as compared to the average values \( \langle E \rangle \) and \( \langle j \rangle \), respectively) [25]. An inhomogeneous current distribution is equivalent to a reduction of the effective (‘noisy’) volume and results in an enhanced \( 1/f \)-noise level [33]. This behavior of the present material may indicate a percolative nature of the MI transition. It is interesting to note that evidence for such a scenario in the Mott system VO\(_2\) comes from a diverging dielectric function [7], which via fluctuation–dissipation relations is connected to the noise PSD. Importantly, however, a simple percolation scenario does not apply here: firstly, the increase of the noise level occurs in a narrow temperature interval, presumably very close to the critical point. Secondly, we not only observe a strongly enhanced \( 1/f \) noise, but also an abrupt shift of spectral weight, which is not necessarily expected in a pure phase separation scenario, for which—under the simplifying assumption of uncorrelated resistance fluctuations in a random network—the excess noise is given by

\[
\frac{S_R(f)}{R^2} = \tilde{s}(f) \frac{\sum_m i_m^4}{\left(\sum_m i_m^2\right)^2},
\]

where \( i_m \) is the local current passing through the resistance \( r_m \) divided by the total current \( I \), which passes through the sample [34]. The function \( \tilde{s}(f) \) describes the mechanism of fluctuations in the local resistances \( r_m \), which is usually of \( 1/f \) type. Hence, in a simple percolative phase separation scenario, the magnitude of the noise varies depending on the fourth moment of the current distribution, whereas the frequency exponent remains unchanged. In the present case, therefore, a plausible scenario is that in addition to a change in the effective noisy volume on the micro- or nanoscopic level, a change in the intrinsic electron dynamics within these domains occurs. Our results viewed with respect to the reports of electronic phase separation in the literature [5, 32] suggest that the enhanced \( 1/f \)-noise level and critical slowing down are due to Mott physics and that percolation of the metallic domains may occur as a consequence when these grow and connect [7].

A systematic investigation of different compounds located both on the insulating and metallic sides of the generalized phase diagram reveals that the enhanced low-frequency fluctuations for \( \kappa\text{-D}_8\text{-Br} \) are induced by electronic correlation effects, since for the less-correlated metallic systems, a strong (exponential) decrease of the noise level is observed below \( T^* \), consistent with a coherent Fermi-liquid metallic ground state [35]. In contrast, the noise of the more correlated \( \kappa\text{-D}_8\text{-Br} \) shows a pronounced peak anomaly at about 35–36 K and exhibits a roughly linear temperature dependence for \( T < 30 \) K, superimposed to which are smaller peak-like features\(^6\). The relation of these features to the crossing of the low-temperature phase lines (and the upturn of the resistivity below about 20 K), or the pseudogap state recently discussed in NMR experiments [36, 37], is a matter for further investigations. We emphasize that our earlier studies on a pressurized \( \kappa\text{-Cl} \) sample [38] also reveal a correlation-induced enhanced noise

\(^6\) The error bar as determined from the reproducibility of the obtained spectra, see also [38], is here of the order of the symbol size.

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level. However, in view of the results reported here, this sample is located further away from the Mott critical endpoint and, accordingly, the relative enhancement of the electronic fluctuations in the critical temperature region is less pronounced and a sharp and pronounced increase of the frequency exponent $\alpha(T)$ is not observed. For $\kappa$-D$_8$-Br, however, figure 2 reveals that the correlation-induced, strongly enhanced noise level close to $(P_0, T_0)$ goes along with a sudden and substantial shift of spectral weight to lower frequencies. This combination makes evident an abrupt critical slowing down of the time scale of charge fluctuations, which may be explained by correlated transitions of electrons over a large number of metastable states when approaching the critical endpoint both from below and above [18].

Furthermore, the strongly enhanced noise level indicates electronic phase separation into metallic and insulating domains. This may naturally explain the reported inconsistency with the Brinkmann–Rice picture of an effective mass enhancement, which for the present materials is not observed, for example in the Sommerfeld coefficient of the specific heat [21], a fact that has remained a puzzling peculiarity over the years [17, 39]. On the other hand, an enhanced effective mass in the metallic domains would be compatible with the observed increase of low-frequency noise, but might be obscured in averaging macroscopic measurements like the specific heat [7].

Finally, similar observations, i.e. a large rise in the low-frequency noise upon approaching the MI transition—interpreted as a manifestation of the slow dynamics of the fluctuations in the local density of states which are strongly Coulomb correlated—have been made for the critical region of other canonical MI transitions, namely the Anderson–Mott transition in P-doped Si [18], as well as carrier-concentration- and magnetic-field-induced MI transitions in Si inversion layers [40, 41]. Our observations provide further evidence that such correlated dynamics may be considered a universal feature of the MI transition [18], irrespective of the mechanism driving the MI transition or the dimensionality of the system. The results offer the intriguing possibility of exploring the dynamical scaling properties directly at the critical point of the Mott transition, e.g. in pressure-tuned $\kappa$-Cl. Such experiments will also address the debated nature of the transition/crossover line $T^*(p, X)$ characterizing the anomalous metallic state in these materials. DMFT calculations, which can possibly describe the behavior of conductivity fluctuations at the Mott transition, are highly desirable.

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