Power-law dependence of the optical conductivity observed in the quantum spin-liquid compound $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

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The Mott-insulator $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ is the prime candidate of a quantum spin liquid with puzzling magnetic properties. Our THz and infrared investigations reveal that also the charge dynamics does not follow the expectations for a Mott insulator. The frequency-dependent conductivity exhibits a power-law behavior $\sigma_\omega(\omega) \propto \omega^n$ that grows stronger as the temperature decreases and extends all the way through the far-infrared. With $n \approx 0.8$ to 1.5 we obtain a significantly smaller exponent than predicted by Ng and Lee [Phys. Rev. Lett. 99, 156402 (2007)]. We suggest fluctuations becomes important in the spin-liquid state and couple to the electrodynamic properties differently compared to the antiferromagnetic Mott insulator $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl. We discuss the various possibilities of how charge fluctuations are influenced by the presence or absence of magnetic order.

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I. INTRODUCTION

Among the two-dimensional organic charge-transfer compounds, the $\kappa$-(BEDT-TTF)$_2$X family [where BEDT-TTF stands for bis-(ethylenedithio)tetrathiafulvalene] is of particular interest, because the constituting cationic dimers are arranged in an anisotropic triangular lattice (Fig. 1) with a delicate interplay between electronic correlations (given by the on-site Coulomb repulsion $U$), the effects of low dimensionality and spin frustration. While $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl is a Mott insulator with antiferromagnetic order at low temperatures ($T_N \approx 25$ K) a slight pressure of 300 bar is sufficient to reach the superconducting state with $T_c \approx 12.8$ K. For the Mott insulator $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ hydrostatic pressure of 1.5 kbar is required to enter the superconducting state at 2.8 K.

$\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ triggers particular interest because at ambient pressure no indication of magnetic order could be observed down to lowest temperatures, despite the considerable antiferromagnetic exchange of $J \approx 250$ K within the triangular lattice thus it is considered as the first realization of a quantum spin-liquid state suggested by Anderson 40 years ago. Numerous theoretical and experimental work was performed during the last decade in order to explore mainly the thermodynamic and magnetic properties. In the high-temperature range, the $^{1}H$-NMR relaxation rate shows anomalies around $200$ K; the thermopower at $150$ K microwave experiments exhibit a dielectric anomaly at $113$ K while a peak in $\epsilon'(T)$ occurs below $60$ K in the radio-frequency range. In addition, a low-temperature anomaly near $6$ K has been observed in thermodynamic transport, dielectric and lattice properties that has not been satisfactorily explained.

As far as the charge dynamics is concerned, Ng and Lee suggested that the gapless spinons contribute to the optical conductivity inside the charge gap. In the low-frequency and low-temperature limit, we do in fact observe power-laws, that cross over from a linear to a quadratic dependence. Unexpectedly and even more striking, however, is the experimental evidence of a power-law behavior in the frequency dependent conduc-
tivity of $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ that extends all the way up to the mid-infrared. The exponent $n$ in $\sigma(\omega) \propto \omega^n$, slightly rises from with $n \approx 1$ to 1.5 as the temperature is reduced and then saturates for $T < 50$ K.

II. EXPERIMENTAL DETAILS

Single crystals of $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ (abbreviated as $\kappa$-CN) and $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl ($\kappa$-Cl hereafter) were grown by electrochemical methods and can reach up to $2 \times 2 \times 2$ mm$^3$ in size with nicely shining surfaces. Temperature dependent optical reflection experiments $R(\nu, T)$ were performed in a wide frequency range using several Fourier transform infrared spectrometers ($23 - 12000$ cm$^{-1}$) with an infrared microscope attached to it; in addition the high-frequency optical properties (up to $35000$ cm$^{-1}$) were determined by spectroscopic ellipsometry at room temperature. For $T < 100$ K the optical transmission was measured in the THz range ($18 - 46$ cm$^{-1}$) using a coherent source spectrometer. In all cases, the light was polarized in the two main directions of the crystal planes. In order to perform a Kramers-Kronig analysis the data were extrapolated at low-frequency according to the dc conductivity measured by the standard four-probe method. Alternative approaches by $R(\nu \rightarrow 0) = \text{const.}$ and Hagen-Rubens extrapolation (for elevated temperatures) were tested, but yield only minor changes for $\nu < 30$ cm$^{-1}$ and basically no modifications above.

The temperature dependent resistivity of $\kappa$-CN and $\kappa$-Cl plotted in Fig. 2(a) demonstrates the insulating behavior of both compounds, but also important differences: cooling from room temperature $\kappa$-Cl crosses over from a semiconducting phase to a Mott insulator at $T_M \approx 40$ K and orders antiferromagnetically around $T_N = 25$ K. For $\kappa$-CN the resistivity continuously increases upon cooling without any anomaly. No single activation energy can be extract, but below approximately 100 K the slope of the Arrhenius plot $\rho(T) \propto \exp(\Delta/T)$ corresponds to $\Delta = 200$ K, right in the range of the exchange coupling $J$. Although the in-plane optical conductivity of both compounds [Fig. 2(b)] vanishes for low frequencies and temperatures, the in-gap conductivity is significantly larger for $\kappa$-CN compared to $\kappa$-Cl. When extrapolating $\sigma_1(\omega)$ linearly to zero, we may identify the Mott gap around 500 cm$^{-1}$ for $\kappa$-Cl at $T = 20$ K; however, it is not as well pronounced and with stronger vibronic contributions than reported form early measurements.

III. RESULTS AND ANALYSIS

In Fig. 3 the frequency-dependent reflectivity and conductivity of $\kappa$-Cl is plotted for different temperatures and polarizations. Since the optical properties of $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br$_x$Cl$_{1-x}$ ($0 \leq x < 1$) have been presented and extensively analyzed during the last years, we refer to Refs. 22, 23, 24, 25, 26 for a detailed discussion.

Here we concentrate on the spin-liquid compound. In Fig. 4 the infrared reflectivity and conductivity of $\kappa$-CN are displayed for different temperatures and polarizations. The optical response is governed by a broad peak in the mid-infrared caused by transitions between the lower and upper Hubbard band and intra-dimer excitations; in addition a large number of vibrational modes extend down to 100 cm$^{-1}$. The transition between the Hubbard bands peaks around 2100 cm$^{-1}$ while the intra-dimer excitations center around 3000 cm$^{-1}$ (more pronounced for $E \parallel b$); this is marginally lower in energy than in $\kappa$-
CL. Calculations by density functional theory and tight binding methods yield an intra-dimer transfer integral \( t_1 \approx 170 \text{ meV} \) with a slight increase upon cooling,⁹ in accord with our observations. The Hubbard band, on the other hand, shifts to lower energies as the temperature is reduced.

A. Mott gap

While at first glance the optical spectra of both Mott insulators might look the same at room temperature, there are significant differences as far as the temperature dependence is concerned; Fig. 4 makes it particularly clear. In the case of \( \kappa \)-Cl the Mott insulating state gradually develops upon cooling, seen in Fig. 4(a,b) by the drop of the optical conductivity in the far-infrared range. Accordingly the spectral weight shifts to higher frequencies as the temperature is reduced. The Mott gap increases upon cooling and arrives at \( \Delta \rho \approx 500 \text{ cm}^{-1} \) for \( T = 20 \text{ K} \) and might become slightly larger in the \( T \rightarrow 0 \) limit. Systematic investigations as a function of effective correlations \( \frac{U}{t} \) by applying chemical pressure confirm this assignment.

The optical conductivity of \( \kappa \)-CN, however, exhibits the opposite temperature dependence [Fig. 4(c,d)], and it is not possible to identify a clear-cut energy gap from our optical measurements. While in the low-frequency limit (\( \nu \rightarrow 0 \)) the conductivity decreases upon cooling [according to the insulating behavior observed in the dc resistivity, Fig. 2(a)], above approximately 100 cm\(^{-1}\) the infrared conductivity actually rises. This is in stark contrast to the opening of an energy gap inferred from \( \rho(T) \); instead it indicates that additional excitations develop for low temperatures, which extend down to small frequencies. In cuprates, a pseudogap develops upon cooling even in the superconducting systems. In the present case of the quantum spin-liquid compound \( \kappa \)-CN, however, the conductivity behaves completely different with a strong frequency and temperature dependence of \( \sigma_1(\omega, T) \).

B. Spinon excitations

In order to explain the strong in-gap excitations, Ng and Lee suggested that due to coupling with the internal gauge field, spinons may contribute to the optical
conductivity of a spin liquid. They predict a strongly enhanced conductivity within the Mott gap and a power-law absorption at low frequencies, i.e., for energies smaller than the exchange coupling $J \approx 250$ K. For low frequencies ($h\omega < k_B T$) the optical conductivity $\sigma_1(\omega) \propto \omega^2$, while for $h\omega > k_B T$ the power law should increase to $\sigma_1(\omega) \propto \omega^{3.33}$.

In order to check whether such a behavior can indeed be found in the spin-liquid compound $\kappa$-CN, we first tried to fit our conductivity data directly by the expected power law $\sigma_1(\omega) \propto \omega^n$. The double-logarithmic plot in Fig. 6c allows us to readily determine the exponent $n$: for high temperatures we obtain $n \approx 1$ at small frequencies with a slight increase to $n \approx 1.5$ above 150 cm$^{-1}$. For $T = 12.5$ K, $\sigma_1(\omega)$ rises even slower than linear at low frequencies with a crossover to $n = 2$ for $\nu > 70$ cm$^{-1}$.

The power-law exponents are slightly smaller for $E \parallel c$ compared to $E \parallel b$. While we find a qualitative agreement with theory of optical excitation of gapless spinons, the experimentally obtained power laws are weaker by a factor of two compared to the proposed ones.

C. Disentangling the spectra

As can be seen in Figs. 2 and 3, the spectra of $\kappa$-CN and $\kappa$-Cl are rich in vibrational excitations. Most of them are due to internal vibrations of the BEDT-TTF molecules, with many of them excited via electron-molecular vibrational (emv) coupling which results in asymmetric Fano resonances. They are best taken into account by the cluster model (see for example Rice asymmetric Fano resonances. They are best taken into account by the Lorentz model.

Due to strong electronic correlations, in these Mott insulators itinerant carriers are absent at low temperatures, as discussed in detail in Ref. 25. Thus except for elevated temperatures, no Drude-like contribution is found in the spectra. Transitions between the Hubbard bands can be identified in the optical conductivity as maxima slightly above 2000 cm$^{-1}$. Intradimer transitions cause a band around 3000 cm$^{-1}$ that shows up as a shoulder or separate maximum. Both contributions form the dominant mid-infrared band; we describe these excitations by the Lorentzian model.

The uncoupled vibrational features and electronic excitations were fitted by the Lorentz model

$$\tilde{\sigma}_{\text{Lorentz}} = \sigma_1(\omega) + i\sigma_2(\omega) = \frac{N\varepsilon^2}{m} \frac{\omega}{\omega^2 - \omega_0^2 + \gamma\omega}$$

centered around the eigenfrequency $\omega_0$ with a width $\gamma = 1/\tau$. Depending on the interaction of the vibrational excitations with the electronic background, the modes may become asymmetric and have to be modeled by

$$\tilde{\sigma}_{\text{Fano}} = i\sigma_0(q-i)^2(i+x)^{-1}$$

where $\sigma_0$ is the background, $x = (\omega^2 - \omega_0^2)/\gamma\omega$ ($\gamma$ and $\omega_0$ are the linewidth and the resonant frequency, respectively) and $q$ is the Fano parameter reflecting the degree of asymmetry of the peak. As an example, in Fig. 6 the optical conductivity for $\kappa$-CN is displayed as measured at $T = 12.6$ K for $E \parallel b$; we also plot the background contributions due to intra- and inter-molecular vibrational excitations, due to transitions between the Hubbard bands and intra-dimer excitations.

In order to disentangle the various contributions we simultaneously fitted the measured reflectivity and conductivity spectra in the frequency range below 1100 – 1400 cm$^{-1}$ (depending on temperature and polarization) by

$$\tilde{\sigma}(\omega) = \tilde{\sigma}_{\text{background}}(\omega) + \tilde{\sigma}_{\text{res}}(\omega) = \sum_i \tilde{\sigma}_{\text{Lorentz}}^i + \sum_j \tilde{\sigma}_{\text{Fano}}^j + \tilde{\sigma}_{\text{res}}^j$$

where we used a minimum number of Lorentz terms $\tilde{\sigma}_{\text{Lorentz}}^i$ and of Fano terms $\tilde{\sigma}_{\text{Fano}}^j$. For the residual conductivity contribution we added two power-law terms with different prefactors $A$ and $B$ and exponents $n$ and $m$:

$$\tilde{\sigma}_{\text{res}}^1 = A\omega^n|_{\omega<\omega_c} + B\omega^m|_{\omega>\omega_c}$$

The crossover frequency between the regions of two power-law exponents was found to be at 600 cm$^{-1}$ at $T = 300$ K and increasing to 800 cm$^{-1}$ for low temperatures. The automated procedure was based on the root-mean-square deviation of the fit compared to the experimental data. The uncertainty for the exponent was estimated to be $\pm 0.2$ and indicated by error bars in Fig. 6.

D. Power laws

The residual conductivity $\tilde{\sigma}_{\text{res}}^1 = \sigma_{\text{res}}^1 - \tilde{\sigma}_{\text{background}}$ is plotted in Fig. 7 for several temperatures. The same
FIG. 7: (Color online) Frequency dependent conductivity of $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ after the background due to vibrational and interband excitations has been subtracted shown. The experiments were performed for the polarization $E \parallel b$ at different temperatures as indicated. A power-law behavior $\sigma_1(\omega) = \sigma_1^{\text{background}}(\omega) = \sigma_1^{\text{res}}(\omega) \propto \omega^n$ can be extracted over a large frequency range (indicated by red lines). A change in slope may be identified for higher frequencies (blue lines) that becomes in particular obvious at low temperatures.

analysis can be done for the polarization $E \parallel c$, leading to very similar results. Above 20 cm$^{-1}$ the conductivity follows a power-law $\omega^n$ over a very large range which extends to 600 cm$^{-1}$ at room temperature and up to even 800 cm$^{-1}$ at low temperatures. Above that frequency, a different exponent may be identified in a limited range up to 1200 cm$^{-1}$. Most important, there is definitely no increase in slope observed when going from low to high frequencies; we cannot identify any crossover to occur at a frequency $\hbar \omega \approx k_B T$, i.e. below 200 cm$^{-1}$.

In Fig. 7 we summarize the temperature dependence of the exponent $n$ obtained in the high and low-frequency range for the two directions of polarization. It starts with a linear dependence $\sigma_1^{\text{res}} \propto \omega$ at ambient temperature, but then increases to almost $n \approx 1.5$ when approaching $T \approx 50$ K. A noticeable change is observed when cooled down further: the power-law exponent saturates or even decreases to about 1.25. Taking the uncertainty of determination into account, the low-temperature value is approximately $n = 1.35 \pm 0.1$ for both directions. Interestingly, also the higher-frequency exponent significantly drops for $T < 50$ K, and $\sigma_1^{\text{res}}(\omega) \approx \text{const.}$ below 20 K. At no time the power-law in conductivity approaches a quadratic behavior in frequency or even larger exponents $n$ or $m$.

The situation is completely different for the Mott-insulator $\kappa$-Cl. From Fig. 2(b) it becomes obvious that the optical conductivity is substantially smaller in the spectral range $\nu < 1000$ cm$^{-1}$. The Mott gap is well defined for both polarizations and it continuously increases to 500 cm$^{-1}$ as the temperature decreases [Fig. 7(a,b)]]. The up-shift in spectral weight and the reduced contribution of the conduction electrons below 50 K is rather a consequence of the Mott state than magnetic order.\cite{28,29,36} Tuning the effective correlations $U/t$ by chemical\cite{125,36} and hydrostatic pressure\cite{28} causes spectral weight to shift to lower energies and fill the gap. Although the room-temperature conductivity appears similar for both compounds, the further evolvement is distinct: for $\kappa$-Cl it is not possible to extract a power-law behavior $\sigma_1^{\text{res}}(\omega) \propto \omega^n$ over a sufficiently large frequency range for any temperature $T < 300$ K.

IV. DISCUSSION AND CONCLUSION

The low-frequency behavior highlighted in Fig. 7(e,f) could in fact be caused by optical excitations of gapless spinons, as suggest by Ng and Lee,\cite{23} although the
conductivity should increase much faster and exhibit a stronger frequency dependence than it actually does: the low-temperature exponents extracted for the power law are significantly lower than predicted. At $T = 12.6$ K there is a crossover around 70 cm$^{-1}$ where the linear rise increases to a quadratic behavior in frequency. When taking out the vibrational features and interband transitions, this behavior continues all the way up to the mid-infrared spectral range: this casts some doubt to this explanation. Even more surprising is the fact, that we do observe the power-law behavior up to room temperature where light-induced spinon excitations should not be observable. A final decision of the applicability of the model can only be based on advanced experiments at lower frequencies and lower temperatures.

The dependence $\sigma^{\text{in}}(\omega, T) = A(T) \omega^n$ observed for the conductivity of quantum spin-liquid compound $\kappa$-CN could in fact be caused by fluctuations. The contribution $A(T)$ significantly increases with lowering temperature which infers quantum fluctuations. In the vicinity of a quantum critical point, power-law behavior is often observed and quantum fluctuations become more pronounced when $T$ is reduced. We have to keep in mind, however, that this behavior is observed up to rather high frequencies and at elevated temperatures, far beyond the regime quantum fluctuations should be dominant.

It is also not clear why this behavior saturates around 50 K, since no magnetic, charge, or structural order is revealed at any temperature. It coincides with anomalies in the dielectric properties. Hotta proposed a dipolar-spin liquid assuming quantum electric dipoles on the dimers that interact with each other through the dipolar spin coupling. This would lead to a ferroelectric-like behavior where a soft mode moves to low frequency. However, there are no indications of charge order in these $\kappa$-phase compounds and soft modes and collective charge excitations commonly show up at much lower frequencies. We suggest that charge fluctuations are strongly enhanced in the vicinity of a quantum critical point and correspond to a much larger energy scale than commonly observed in the case of quantum fluctuations in the spin degree of freedom.

Most important, a very similar dielectric anomaly was observed in the in-plane and out-of-plane properties of $\kappa$-Cu$_2$(BEDT-TTF)$_2$Cl$_2$ where the Mott gap is clearly developed at low temperatures and no in-gap contribution are present in the optical conductivity. Tomić and collaborator suggested that grain boundaries, linked to magnetically ordered domains in $\kappa$-Cl might move, thus causing a strong dielectric contribution. However, this argument, however, does not hold for $\kappa$-CN where no magnetic order is present down to lowest temperatures. Pressure dependent investigations on $\kappa$-Cl showed that the bandwidth-controlled Mott criticality involves critical fluctuations in charge, spin, and lattice. Nothing like that is known for the spin-liquid compound $\kappa$-CN for the intermediate temperature range. While this issue can be clarified by further experimental studies, considerable theoretical work is required in order to describe the power-law behavior the dimer Mott insulator with a quantum spin liquid state, in the low-frequency limit as well as in the far-infrared range.

From our optical investigations of two Mott-insulators with very similar triangular structure but different magnetic ground states, we can conclude that only the magnetically ordered organic salt $\kappa$-(BEDT-TTF)$_2$Cu[N-(CN)$_2$]Cl exhibits a well defined Mott gap at low temperatures. The quantum spin-liquid compound $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ exhibits a power-law behavior in the frequency-dependent conductivity that becomes stronger as the temperature decreases. We suggest spin fluctuations get important in the spin-liquid state and couple to the electrodynamic properties differently compared to an antiferromagnetic Mott insulator. The power-law $\sigma_1(\omega)$ observed in an extended range of frequency and temperature remains a puzzle that calls for further investigations of how charge fluctuations are influenced by the presence or absence of magnetic order.

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