Low-Frequency Optical Properties of $\beta''$-(BEDO-TTF)$_5$[CsHg(SCN)$_4$]$_2$: Indications of Electronic Correlations in a 1/5-Filled Two-Dimensional Conductor

N. Drichko$^{1,2}$, K. Petukhov$^3$, M. Dressel$^1$, O. Bogdanova$^3$, E. Zhilyaeva$^3$, R. Lyubovskaya$^3$, A. Greco$^5$, and J. Merino$^5$

$^1$ Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57, 70550 Stuttgart, Germany
$^2$ Ioffe Physico-Technical Institute, 194021 St. Petersburg, Russia
$^3$ Institute of Problems of Chemical Physics, Russian Academy of Sciences, Chernogolovka, Russia
$^4$ Facultad de Ciencias Exactas Ingeniería y Agrimensura e Instituto de Física Rosario (UNR-CONICET), Rosario, Argentina
$^5$ Max-Planck-Institut für Festkörperforschung, D-70569 Stuttgart, Germany

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The polarized reflectivity of $\beta''$-(BEDO-TTF)$_5$[CsHg(SCN)$_4$]$_2$ is studied in the infrared range between 60 cm$^{-1}$ and 6000 cm$^{-1}$ from room temperature down to 10 K. Already at $T = 300$ K a pseudogap in the optical conductivity is present of about 300 cm$^{-1}$; the corresponding maximum in the spectrum shifts to lower frequencies as the temperature decreases. In contrast to quarter-filled BEDT-TTF-based conductors of the $\beta''$-phase a robust Drude component in the conductivity spectra is observed which we ascribe to the larger fraction of charge carriers associated with the 1/5-filling of the conduction band. This observation is corroborated by exact diagonalization calculations on an extended Hubbard model on a square lattice for different fillings. A broad band at 4000 cm$^{-1}$ appears for the electric field polarized parallel to the stacks of the BEDO-TTF molecules is associated to structural modulations in the stacks; these modulations lead to a rise of the dc and microwave resistivity in 100 to 30 K temperature range.

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I. INTRODUCTION

Molecular quasi two-dimensional organic conductors based on BEDT-TTF (where BEDT-TTF stands for bis-ethylenedithio-tetrathiafulvalene) and its derivatives are known to be excellent model objects to study the effects of electron-electron correlations and charge ordering of different nature in two dimensional (2D) systems. According to theoretical studies the electronic parameters which are crucial for the metallic properties and formation of charge ordering (CO) are the filling of the conduction band and the size of the electronic correlation energy compared to the width of conductance band or transfer integrals. In BEDT-TTF-based compounds these physical parameters can be tuned chemically by changing the structure or/and chemical composition of the crystals. The width of the conduction band increases with the orbital overlap of neighboring molecules. The filling of the conduction band depends upon both the anionic charge and upon the ratio of cations and anions. Recently this has been devoted to systematically control the band filling and to study its influence on the physical properties. However, most of 2D organic conductors have quarter-filled bands (charge $+0.5e$ per molecule), or effectively half-filled bands ($+1e$ per dimer) for dimerized structure. The isostructural family $\beta''$-(BEDO-TTF)$_5$[M$\text{Hg(SCN)}_4$]$_2$ ($M = K$, Rb, Cs, NH$_4$, Li) was of interest to us, as these salts crystallize in a 5:2 ratio of cations and anions leading to a 1/5 filling of the conduction band.

It is important for our study, that when going from BEDT-TTF to BEDO-TTF (bis-ethylenedioxy-tetrathiafulvalene) the on-site electron-electron correlation energy does not change considerably. However, the probability to obtain an insulating and superconducting salt among the BEDO-TTF family is lower than among BEDT-TTF based ones. Horiuchi et al. suggested that these differences are due to the fact that the BEDO-TTF molecules tend to form conducting layers which are more two-dimensional. Indeed, within the (BEDO-TTF)$_5$[M$\text{Hg(SCN)}_4$]$_2$ ($M = K$, Rb, Cs, NH$_4$, Li) family most members exhibit a metallic behavior in the highly-conducting plane down to low temperatures. The only exception is $\beta''$-(BEDO-TTF)$_5$[CsHg(SCN)$_4$]$_2$ which shows non-metallic behavior of the dc conductivity in one direction of the plane, while in perpendicular direction conductivity remains metallic for any $T < 4$ K. Previous optical studies of $\beta''$-(BEDO-TTF)$_5$[CsHg(SCN)$_4$]$_2$ in the 700-6000 cm$^{-1}$ region have also revealed an anisotropic temperature behavior of the optical conductivity; the reason for this unusual behavior might be a dimerization in the stacks of BEDO-TTF molecules. These investigations also led to the conclusion that at frequencies below the mid-infrared a non-Drude response has to be expected.

Optical investigations and theoretical studies of BEDT-TTF based materials with a metallic ground state have shown that the effects of electronic correlations lead to deviations from the simple Drude behavior; however, the results strongly depend on the band filling. For the metallic half-filled $\chi$-phase compounds a narrow Drude-peak was observed only at temperatures below 50 K in accordance with dynamical mean-field-theory calculations on a frustrated lattice at half-filling with strong on-site Coulomb repulsion $U \approx W$ ($W$ being the bandwidth). Quarter-filled systems, on the other hand, remain metallic for any $U \gtrsim 3W$ as long as the nearest-neighbor interaction $V$ is negligible; by increasing...
V beyond some critical value $V_c$, charge-ordering phenomena become relevant. In fact this was recently demonstrated in the spectra of BEDT-TTF-based organic conductors with an $\alpha$-structure of the conducting layers.  While $\alpha$-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ shows metallic properties at any temperature, in $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$ a gap opens at about 200 cm$^{-1}$ at temperatures below 200 K with a narrow Drude component remaining. However, in some metallic $\beta''$-packed BEDT-TTF salts which are quarter filled, no Drude peak was observed down to 10 K.

This has motivated us to extend the optical measurements of $\beta''$-(BEDO-TTF)$_3$[CsHg(SCN)$_4$][12] to frequencies well below 700 cm$^{-1}$. We got an opportunity to study the expected deviations from a simple Drude behavior for this 1/5 filled system and to compare them to theoretical expectations and to the low-frequency behavior of a $\beta''$-packed BEDT-TTF based 1/4 filled system. In this first low-frequency optical experiment on a BEDO-TTF salt, we have also investigated the temperature dependence of the anisotropy which characterizes the in-plane conductivity.

II. EXPERIMENTAL

Single crystals of $\beta''$-(BEDO-TTF)$_3$[CsHg(SCN)$_4$][12] were prepared by electrochemical oxidation of BEDT-TTF as described in Ref. [13]. In structure of these crystals the BEDO-TTF molecules form conducting layers in the $ab$-plane and alternate with insulating anion layers along $c$-axis. $\beta''$-phase means that the molecules form stacks parallel to the $2a$-$b$ direction; along the stacks the overlap integrals show some modulation. The overlap integrals to the molecules in the neighboring stacks are nearly two times larger than between the molecules along the stacking direction. According to the stoichiometry, each BEDO-TTF molecule carries an average charge of $+0.4e$, thus the conduction band is 1/5 filled. The crystals grow as black parallelogram-shaped plates, some of them reached $2 \times 1$ mm$^2$ in size. The largest extension of the crystals is parallel to the stacking direction of the BEDO-TTF molecules in the conducting layer, i.e. the $2a$-$b$ direction.

The $\beta''$-(BEDO-TTF)$_3$[CsHg(SCN)$_4$][12] crystals were previously characterized by standard dc transport measurements parallel and perpendicular to the stacks. The values of the room temperature resistivity for the two directions are around 0.2 to 0.5 $\Omega$m. The temperature dependence of the dc resistivity is plotted in Fig. 1. Perpendicular to the stacking direction the resistivity remains metallic down to helium temperatures; parallel to the stacks the resistivity changes only little between 270 K and 50 K indicating a semiconducting behavior, and becomes metallic below 50 K.

In order to investigate the anisotropic transport properties without applying contacts, we performed microwave resistivity measurements by means of cavity perturbation technique. The experimental setup and measurement technique are discussed in detail elsewhere. For the experiments reported here, the sample was fixed to a quartz rod positioned in the electric field maximum of a cylindrical copper resonator which operates in the $TE_{011}$ transmission mode at a resonance frequency of 24 GHz. At any temperature down to 1.5 K the sample could be rotated inside the cavity during the measurement, allowing to determine its anisotropy in situ.

In Fig. 2 the normalized microwave resistivity of $\beta''$-(BEDO-TTF)$_3$[CsHg(SCN)$_4$][12] measured parallel and perpendicular to the stacks is plotted as a function of temperature. Perpendicular to the direction of the stacks the resistivity $\rho(T)$ exhibits a metallic behavior between 300 and 150 K, with only little change on cooling below 150 K. Along the stacks we find a decrease in $\rho(T)$ when going down to 100 K, followed by some insulating be-
behavior; below 50 K a metallic regime is entered again. These results are consistent with dc resistivity measurements displayed in Fig. 1.

The polarized reflectivity $R(\omega)$ spectra of the two main directions in the conducting plane of $\beta''$-(BEDO-TTF)$_3$[CsHg(SCN)$_4$)$_2$, are measured in the frequency range of 60-6000 cm$^{-1}$ using a modified Bucker IFS113v Fourier transform spectrometer equipped with three different light sources and various radiation detectors, including a helium cooled bolometer. As reference we use an aluminum mirror which replaces the sample by translation. With the help of proper polarizers the optical response was measured for the electric field $E$ polarized parallel and perpendicular to the stacks of the single crystal. The sample is cooled down to 10 K in a CryoVac helium bath cryostat with a very slow cooling rate of 1 K/min. The overall behavior of the reflectivity is in good agreement with the previous results obtained in the spectral region of 700-6000 cm$^{-1}$ which were measured by a microreflectance technique. However due to small defects on the surface of the single crystals, our values of reflectivity are slightly lower. The correct absolute values are obtained by normalizing the reflectivity with a constant factor in order to receive perfect agreement with the microscopic measurements at room temperature. Since the low-temperature spectra $E$ parallel to the stacks are very noisy below 300 cm$^{-1}$, for clarity reasons we show a smoothed curve as a dashed line in Fig. 3.

III. RESULTS AND ANALYSIS

The spectra of the reflectivity $R(\omega)$ and the optical conductivity $\sigma(\omega)$ in the two polarizations parallel and perpendicular to the stacks are presented in Fig. 1. For the electric field $E$ of the incident light parallel to the stacks, the room-temperature reflectivity is lower than for the perpendicular polarization, in agreement with the smaller overlap of the BEDO-TTF orbitals in this direction. The reflectivity $R(\omega)$ increases to lower frequencies and the plasma edge is located around 5000 cm$^{-1}$. In the frequency range from 700 to 1600 cm$^{-1}$ weak vibrational features are seen, their detailed assignment to $a_g$ vibrations activated by electronic-molecular vibrational (EMV) coupling is published in Ref. 14. With lowering temperature, $R(\omega)$ grows in both polarizations of the electric field, however in different ways. For $E$ normal to the stacks the reflectivity in the frequency range from 60 to 700 cm$^{-1}$ increases upon cooling down to 150 K, but remains unchanged below this temperature. Parallel to the stacks the reflectivity in the 60-700 cm$^{-1}$ range changes rapidly between 200 and 150 K, and continues to grow slightly below this temperature. There is basically no change of $R(\omega)$ on cooling between 50 and 10 K. At low temperatures the reflectivity along the stacks reaches the corresponding $R(\omega)$ values of $E$ perpendicular to the stacks. With other words, the large room-temperature anisotropy of reflectivity in the conducting plane observed in the spectral range from 60 to 700 cm$^{-1}$ disappears at temperature $T \leq 50$ K.

The optical conductivity spectra are calculated with the help of the Kramers-Kronig transformation. The measured $R(\omega)$ spectra are extrapolated by the Hagen-Rubens formula for $\omega \rightarrow 0$; above 6000 cm$^{-1}$ the results of our room-temperature measurements and extrapolation of $R(\omega) \propto \omega^{-4}$ for $\omega \rightarrow \infty$ are used. The results are displayed in Fig. 3a and d (note the different scales).

In both directions the room-temperature conductivity spectra consist of a narrow Drude-like component

$$\sigma(\omega) = \frac{\omega_p^2 \tau}{4\pi} \frac{1}{1 + \omega^2 \tau^2}$$

(1)

and two broad maxima at higher frequencies. The Drude contribution is described by the plasma frequency $\Omega_p = \omega_p/(2\pi c) = 3550$ cm$^{-1}$ and the scattering rate $\Gamma = 1/(2\pi\tau) = 180$ cm$^{-1}$ for $E \perp$ stacks, and $\Omega_p = 2300$ cm$^{-1}$ and $\Gamma = 140$ cm$^{-1}$ for the polarization $E \parallel$ stacks. An intensive maximum lies at about 700 cm$^{-1}$, correspondingly a pseudogap is observed in the spectra at 300 cm$^{-1}$. At higher frequencies a broad and weak maximum can be seen at about 2600 cm$^{-1}$.

The spectral weight of the optical conductivity

$$\frac{\omega^2}{8} = \int \sigma(\omega) d\omega$$

(2)

shifts to low frequencies when the temperature is reduced. Although the Drude contribution becomes extremely narrow, its plasma frequency increases slightly; at $T = 10$ K the corresponding fit yields $\Omega_p = 4500$ cm$^{-1}$ and $\Gamma = 30$ cm$^{-1}$ for $E \perp$ stacks, and $\Omega_p = 4050$ cm$^{-1}$ and $\Gamma = 45$ cm$^{-1}$ for $E \parallel$ stacks. This temperature behavior is quite common for organic conductors and is observed at the $\kappa$-phase of BEDT-TTF, for instance.

The 700 cm$^{-1}$ peak considerably shifts to low frequencies as the temperature decreases and lies at about 350 cm$^{-1}$ at $T = 100$ K; thus the low-frequency gap becomes less pronounced. The described behavior is basically the same for both polarizations of the electric field, except that for $E$ perpendicular to the stacks the spectral weight of this maximum grows by a factor of 1.5 when going from $T = 300$ K to 100 K while in the direction parallel to the stacks the intensity of this maximum does not change.

An in-plane anisotropy observed in dc and microwave resistivity measurements is also clearly seen in the different temperature behavior of $\sigma(\omega)$ below 100 K. The spectra for $E \perp$ stacks change only slightly between 100 and 10 K, in agreement with the constant microwave resistivity in this temperature range; in particular no changes are noted in the mid-infrared range. Parallel to the stacks a wide band around 4000 cm$^{-1}$ appears at 100 K, as is clearly seen in Fig. 2, where the conductivity spectra of different temperatures are off-set. Simultaneously, the intensity of optical conductivity $\sigma(\omega)$ in the mid-infrared region between 1000-3000 cm$^{-1}$ decreases; also the low-frequency maximum becomes smaller. This corresponds to the decrease of the reflectivity in this range.
to the abrupt increase of the microwave resistivity at 100 K. Although ρ(T) becomes metallic below 30 K, the 4 K resistivity is still higher than that at 100 K, in accordance with our optical results: the 4000 cm⁻¹ is present at the lowest measured temperature of 10 K.

IV. DISCUSSION

The most important observations of our optical experiments on β''-(BEDO-TTF)₅[CsHg(SCN)₄]₂ are an extremely narrow Drude-component and a gap at about 300 cm⁻¹. A similar overall behavior was previously found in BEDT-TTF bases organic conductors with a metallic ground state.¹⁷,²⁶ In particular the fact that the low-frequency spectral weight increases several times when the temperature decreases from T = 300 K to helium temperature, as observed for quite a number of them,¹⁶,¹⁷,²⁶-²⁷,²⁸,²⁹ cannot be simply explained by a reduced scattering rate which leaves the spectral weight conserved.²⁵ Therefore these effects are ascribed to the influence of electron-electron correlations.

It is interesting to compare our findings on β''-(BEDO-TTF)₅[CsHg(SCN)₄]₂ with the optical results obtained on a superconductor β''-(BEDT-TTF)₂SF₅CH₂CF₂SO₃ (T_c=5.2 K) which is a 1/4-filled compound with a similar packing of the cation layers. The reflectivity of β''-(BEDT-TTF)₂SF₅CH₂CF₂SO₃ is lower than that of β''-(BEDO-TTF)₅[CsHg(SCN)₄]₂, although the dc conductivity is of the same order of magnitude. Very similar to our optical data, the spectra of the BEDT-TTF-based compound show two maxima in σ(ω) at about 1000 and 2500 cm⁻¹; the low-frequency maximum grows and shifts to lower frequencies with decreasing temperature, while no Drude peak appears in the investigated temperature and frequency range.²⁵

The structural data on the two discussed β''-salts of the BEDT-TTF and BEDO-TTF families show that although the number of short contacts between the neighboring molecules is larger in the case of β''-(BEDO-TTF)₅[CsHg(SCN)₄]₂, the values calculated for the intra-stack transfer integrals are equal. The inter-stack overlap, on the other hand, is nearly twice larger for the BEDT-TTF-based compound than for the BEDO-TTF one. If we assume that the Coulomb interactions are comparable for both compounds, the ratios of U/t and V/t are estimated to be larger for β''-(BEDO-TTF)₅[CsHg(SCN)₄]₂ compared to β''-(BEDT-TTF)₂SF₅CH₂CF₂SO₃. Since the Coulomb effects do not drive the system insulating, but we observe a Drude peak in our spectra (Fig. 3), additional effects have to be considered.

An essential difference between these two β''-salts is the filling of the conduction band which is 1/4 for the BEDT-TTF-compound and 1/5 for β''-(BEDO-TTF)₅[CsHg(SCN)₄]₂. We suggest that this deviation from the quarter-filled band leads to an appearance of the narrow

![Graph](image-url)
Drude contribution, while the low-frequency gap is still present in the spectra. The following theoretical considerations confirm this idea.

A theoretical model of charge-ordering in layered molecular crystals with quarter-filled conduction bands has been recently put forward\cite{6, 7} to describe this family of $\beta''$-compounds.\cite{6, 7} Assuming a simple square lattice to describe the quarter-filled molecular planes of the crystal, a checkerboard charge-ordered state is induced if values of the on-site $U$ and inter-site Coulomb repulsion $V$ become large enough with respect to the nearest-neighbors hopping integral $t$. Although for certain ratios of $U/t$ and $V/t$ electronic correlations are not strong enough to push the system into a checkerboard insulating state, they nevertheless lead to strong modifications in the distribution of the optical weight: for instance a low frequency peak appears at the edge of a broad mid-infrared band.\cite{6, 7} This is related to the dynamics of quasiparticles interacting with short range checkerboard charge fluctuations. For values of $V \gtrsim V_c \approx 2t$ and sufficiently large $U = 20t$, the Drude weight obtained from Lanczos diagonalization is suppressed signalling the occurrence of a metal-insulator transition (at quarter-filling).

This situation is changed as we dope the system away from one quarter filling. Let us still model the materials by means of an extended Hubbard model on a square lattice at arbitrary filling. A simple understanding of the electronic properties of the system can be obtained by considering the case in which we dope the insulating checkerboard charge-ordered state with only one hole as discussed earlier by Ohta and collaborators.\cite{37} The single hole (which is an empty site in the checkerboard) has non-zero probability ($V > V_c = 2t$ and $U >> t$), to move to a second-nearest neighbor in the diagonal direction by two successive hopping processes through its nearest neighbors. This is of course a virtual second order process because the electron has to hop twice along its nearest neighbors at the expense of some energy. In a subsequent process the electron can hop to its third-nearest neighbors and so on. Hence, quasiparticles can disperse with renormalized hopping along the lattice and the system is expected to be always metallic (at a finite value of $V$). Hence, while at 1/4-filling a metal-insulator transition at a finite $V_c$ is expected, this is not the case at 1/5-filling. This explains why the 1/5-filled salt exhibits a robust Drude component in contrast to the 1/4-filled $\beta''$ salts. The short range checkerboard charge fluctuations which lead to signatures in the dynamical properties of the 1/4-filled system (such as the appearance of the low frequency peak in the optical conductivity) are also expected to be present in the 1/5-filled system. However, these signatures are anticipated to be somewhat suppressed at 1/5-filling with respect to 1/4-filling for a given value of $V$ because in the former case, the checkerboard charge-ordered state is modified by the presence of mobile empty sites. This arguments are consistent with the observation that while the spectral weight of the feature appearing at low frequencies in the optical conductivity increases by factor of 5 as the temperature is reduced from room temperature to the lowest temperature at 1/4-filling, at 1/5-filling this feature is only enhanced by a factor of approximately 1.5.\cite{38}
In Fig. 5 we show the behavior of the optical conductivity and the Drude weight at 1/5-filling for different values of $V$ an $U = 2t$, computed from exact diagonalization of an extended Hubbard model on a square lattice. We observe how, at 1/5-filling, the Drude weight remains finite; the system remaining metallic for any value of $V$. This is in contrast to the metal-insulator transition found at $V \approx 2t$ for the 1/4-filled system. The optical conductivity for typical values of $V/t$ displays a broad band and a feature at low frequencies; much smaller than the one found for the 1/4-filled case. The behavior of the optical spectra at low frequencies when going from the 1/4 to the 1/5-filled system is consistent with exact diagonalization results.

While the features ascribed to the influence of electronic correlations are observed in the spectra of both polarizations, the non-metallic behavior of dc, microwave, and optical conductivity is seen only for $E$ parallel to the stacks and can be ascribed to structural effects. One of the major changes below 100 K affects the broad band at about 2500 cm$^{-1}$.

A wide band in the mid-infrared region is typical for the conductivity spectra of most BEDT-TTF and BEDO-TTF based materials. It can appear due to structural peculiarities: for example, when the filling of the conduction band is not equal to one half, a dimerization in the stacks of the organic molecules can cause the conduction band to split; transitions across this gap result in a maximum in the optical conductivity spectra. Since in the cation layers of $\beta''$-(BEDO-TTF)$_5$CsH$_8$(SCN)$_4$ the molecules are weakly modulated along the stacks already at room temperature, the wide band with a maximum at about 2500 cm$^{-1}$ can be assigned to transitions across the gap caused by this structural modulation. The shift of this band up to 4000 cm$^{-1}$ and the fact that its intensity for the polarization parallel to the stacks grows at temperatures below 150 K points towards a strengthening of the modulation. The spectral weight of this band increases on the expense of the low-frequency feature: below $T \approx 100$ K the intensity of the 350 cm$^{-1}$ maximum is reduced and the 250 cm$^{-1}$ gap becomes more distinct; consequently, the dc and microwave resistivity increase.

Most changes in structure and charge on the BEDT-TTF molecules in the organic conductors can be nicely followed by observing changes of EMV-coupled vibrational features. The $a_g$ vibrations of central C=C bonds of the BEDO-TTF molecule observed at 1619 cm$^{-1}$ is known to be most sensitive to the changes of charge on the molecule. Its position does not show any variations with temperature, which suggests that the charge on the BEDO-TTF molecules does not change considerably. On the other hand, for the polarization parallel to the stacks two $a_g$ vibrational modes at 864 and 1015 cm$^{-1}$ appear simultaneously with the growth of the 4000 cm$^{-1}$ electronic band. The appearance of new vibrational features indicates a lowering of symmetry: a symmetry center disappears. Since $a_g$ features couple to electrons which are localized on clusters of molecules, we can conclude that these changes in the spectra occur due to a strengthening of the modulation within the stacks of BEDO-TTF molecules, which leads to the appearance of clusters (e.g. dimers of BEDO-TTF) and lowers the symmetry. However, for 1/5-filled system the most primitive modifications (dimerization, teramerization, etc.) will not cause a gap at the Fermi-surface and lead to a insulating behavior. Maybe this is the reason why we observe such a complicated behavior in the dc and microwave resistivity instead of a simple metal-to-insulator transition. Interestingly, this non-metallic behavior is absent in the direction of the larger overlap of the neighboring molecule orbitals, i.e. for the polarization $E$ perpendicular to the stacks.

V. CONCLUSIONS

The present optical study of $\beta''$-(BEDO-TTF)$_5$CsH$_8$(SCN)$_4$ in the infrared range between 60 and 6000 cm$^{-1}$ from room temperature down to 10 K allows us to investigate and distinguish between two effects relevant to most of the two-dimensional organic conductors. Electronic correlations lead to the presence of a narrow Drude-like contribution and to a pseudogap at about 300 cm$^{-1}$ in the optical conductivity $\sigma(\omega)$. These features are observed for both principal directions in the conducting plane, parallel and perpendicular to the stacks, and in the whole temperature range, from 10 K up to room temperature. The maximum in the optical conductivity due to excitations across the pseudogap shift to low frequencies as the temperature decreases. Comparing our results on the BEDOT-TTF systems with the optical data of BEDT-TTF at 1/4-filling, we propose that the robustness of the Drude contribution in our spectra is due to the fact that the conduction band is 1/5 filled. We have used an extended Hubbard model with $U \gg t$ on a square lattice at different fillings to model the molecular planes. At quarter-filling there is a critical value, $V_c \approx 2t$, at which a metal-insulator transition to a checkerboard charge ordered state occurs. Optical conductivity close to but in the metallic side of this transition displays a strong suppression of the Drude component and transfer of weight to a broad mid-infrared band. At 1/5-filling, however, empty sites induced by doping the checkerboard propagate along the molecular planes leading to metallic behavior. This explains the strong Drude component observed in the optical conductivity at 1/5-filling. It would very interesting to analyze materials with other fillings so that an understanding of the dynamics of holes in a charge ordered background can be achieved.

At temperatures below 100 K structural modulations along the stacks of BEDO-TTF molecules activate a band at 4000 cm$^{-1}$ and new EMV-coupled features for the polarization $E$ parallel to the stacks. In the dc and microwave resistivity this effect is observed as a non-metallic behavior in this direction.
Acknowledgments

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