Large dielectric constant and giant nonlinear conduction in the organic conductor \(\theta-(\text{BEDT-TTF})_2\text{CsZn}(\text{SCN})_4\)

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The dielectric constant and ac conductivity have been measured for the layered organic conductor \(\theta-(\text{BEDT-TTF})_2\text{CsZn}(\text{SCN})_4\) along the out-of-plane direction, which show a relaxation behavior similar to those in the charge-density-wave conductor. Most unexpectedly, they exhibit a large bias dependence with a hysteresis, and changes in magnitude by 100-1000 times at a threshold. These findings are very similar to the collective excitation of the charge density wave. \(\theta-(\text{BEDT-TTF})_2\text{CsZn}(\text{SCN})_4\) has collective excitations associated with charge ordering, though it shows no clear indication of long range order.

I. INTRODUCTION

There appear growing interests in the self-organization phenomena of conduction electrons, known as charge inhomogeneity. A prime example is the high-temperature superconducting copper oxides, in which the nanoscale phase-separation of doped carriers is observed in scanning-tunnel-microscope experiments [1]. Another example is the charge order (CO) in the transition-metal oxides [2, 3], where spins and holes are alternately aligned to form a charge-density-wave (CDW) order and a spin-density-wave (SDW) order simultaneously. These have been regarded as a new aspect of strongly correlated electron systems, because the electrons tend to be self-organized in order to minimize the magnetic energy of the background spins. Thus we expect that such a self-organization will happen regardless of the degrees of disorder, and call it “intrinsic inhomogeneity”. In real materials, however, disorder-driven effects cannot be ruled out, owing to unavoidable solid solutions and defects. But rather, there increase a number of papers reporting that the charge order is quite susceptible to disorder [4]. Thus, to search for intrinsic charge inhomogeneity, we need a charge-ordered system in a very clean solid.

Organic salts are a good candidate for the clean system. They are grown in organic solvent at room temperature, where the thermal energy is small enough to suppress point defects. Actually most of the metallic salts exhibit the Shubnikov-de Haas oscillation at low temperatures [5]. This means that the mean free path of the conduction electron is very long, and that the crystal is free from impurities. In particular, the BEDT-TTF [bis(ethylenedithio)-tetrathiafulvalene] salts are most suitable for the study of the strong correlation in two dimension, and the \(\kappa-(\text{BEDT-TTF})\) salts have been most thoroughly investigated among them, where a superconductor-antiferromagnetic insulator transition takes place by changing hydrostatic pressure, anion species, and deuterium substitution [6, 7].

\(\theta-(\text{BEDT-TTF})_2\text{M}(\text{SCN})_4\) \((\text{M}=\text{Rb and Cs})\) is another interesting family of the BEDT-TTF salts [8, 9], where the BEDT-TTF molecules form a distorted triangular lattice. The BEDT-TTF layer acts as a two-dimensional conducting layer, and owing to the triangular stacking, a single elliptic Fermi surface is calculated in the tight-binding approximation. This makes a remarkable contrast with the \(\kappa\)-type salts, in which the BEDT-TTF molecules form a dimer to make two Fermi surfaces (the electron pocket and the one-dimensional sheet). In this sense, the \(\theta\)-type salts are truly two-dimensional system with a moderate in-plane anisotropy. Another feature is that they are quarter-filled (one hole for the two BEDT-TTF molecules), which means that the system is far from the Mott transition that occurs near half-filling [10]. The most interesting feature is that they are unstable against CO along the \(c^*\) axis [11]. A metal-insulator transition due to CO occurs at \(T_{\text{MI}}=190\ \text{K}\) for \(\theta-(\text{BEDT-TTF})_2\text{RbZn}(\text{SCN})_4\) \((\text{M}=\text{Rb salt})\) [8].

The dielectric constant is a good probe for the charge inhomogeneity. We have studied the dielectric response of strongly correlated systems, and have found a large dielectric constant above \(T_{\text{MI}}\) for the \(\text{M}=\text{Rb salt}\) [12]. The frequency dependence is quantitatively explained in terms of a generalized formula of Debye's dielectric relaxation, which strongly suggests that fractions of CO already exist even at room temperature. Such a picture is consistent with other measurements such as NMR [13].

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and optical reflectivity \cite{14}. This is a piece of evidence that the charge inhomogeneity can occur in a homogeneous system. In this paper, we report on the dielectric response and the nonlinear conductivity of $\theta-(\text{BEDT-TTF})_2\text{CsZn(SCN)}_4$ (the $M=$Cs salt) single crystals. We have found anomalously large nonlinear conductivity just like the sliding of CDW, although there is no sign for CDW/SDW transitions at all temperatures. This implies that the $M=$Cs salt exhibits a collective excitation without long range order.

II. EXPERIMENTAL

Single crystals with a typical dimension of $2\times0.2\times0.2$ mm$^3$ were prepared using a galvanostatic anodic oxidation method, and the detailed growth conditions and their characterization were described in \cite{8}.

The resistivity was measured by a four-probe and two-probe method from 4.2 to 300 K in a liquid He cryostat. The contact resistance was 10-50 $\Omega$ at room temperature. Thus the resistivity obtained by the two-probe method was accurate within an error bar of 1% at all the temperatures for the out-of-plane direction (the $b$-axis direction), and below 10 K for the in-plane direction (the $a$-axis direction).

The dielectric constant ($\varepsilon_b$) and ac conductivity ($\sigma_b$) along the out-of-plane direction (the $b$ direction) were measured with a parallel plate capacitor arrangement using an ac two-contact four-probe method with an LCR meter (Agilent 4284A, and 4286A) from $10^3$ to $10^6$ Hz. The dc-bias dependence of $\varepsilon_b$ and $\sigma_b$ was also measured in the static dc voltage. The in-plane dielectric constant was not measured, because it had too low resistance for us to measure $\varepsilon$ precisely. Owing to the two-contact configuration, the contact resistance and capacitance might have affected the measurement. As mentioned, the contact resistance was negligible along the out-of-plane direction, which was verified by the fact that the observed $\sigma_{ac}$ was quantitatively consistent with the observed dc resistivity for $\omega \to 0$. Though we did not evaluate the contact capacitance, we can employ an evaluated value of 500 pF for La$_2$CuO$_4$ from Ref. \cite{15}, because the contact capacitance is primarily determined by the area of the contact. It gives a reactance of $3\times10^5 \Omega$ at 1 kHz, which is $10^4$ times larger than the contact resistance. As a result, we can safely assume that our contact is primarily a resistive coupling, and can neglect the contact capacitance. We should further note that we tested different contact configurations of other samples, and found that the results were reproducible within experimental errors.

III. RESULTS

Figure 1(a) shows $\varepsilon_b(\omega)$ of the $M=$Cs salt. Reflecting the insulating behavior, the dielectric response is far from Drude-like: $\varepsilon_b$ shows a positive sign, and decreases with increasing frequency. The magnitude is as large as 90 at low frequencies, and such a large $\varepsilon$ is rarely observed in conventional insulators. The large value of 90 is gradually relaxed to a small value of 40 at high frequencies, which can be explained in terms of a generalization of Debye’s dielectric relaxation (the Havriliak-Negami formula \cite{14}) given by

$$\varepsilon(\omega) = \varepsilon_{HF} + \frac{\varepsilon_{LF} - \varepsilon_{HF}}{[1 + (i\omega\tau)^\beta]^\alpha}$$

\hspace{1cm}(1)

where $\alpha$ and $\beta$ are dimensionless parameters, and satisfy $0 < \alpha, \beta < 1$. $\varepsilon_{LF}$ and $\varepsilon_{HF}$ are the low- and high-frequency dielectric constants, and $\tau$ is the relaxation time. The fitting results are shown by the solid curves in Fig. 1(a), which satisfactorily fit the measured data. All the parameters except for $\tau$ are independent of temperature, which will be discussed in the next section.

Figure 1(b) shows $\sigma_b(\omega)$ of the $M=$Cs salt. As is similar to $\varepsilon_b$, the frequency dependence suggests relaxation behavior: A small value of $\sigma$ at low frequencies gradually increases to a higher value at high frequencies. Note that $\sigma_b$ has a finite value for $\omega \to 0$, which cannot be ascribed to the dielectric relaxation. In general, $\sigma$ is expressed as the sum of the dc and ac parts given by

$$\sigma(\omega) = \sigma_{dc}(T) + \sigma_{ac}(\omega, T).$$

(2)

$\sigma_{dc}$ is derived from the overdamped Drude contribution. As is well known, the Drude conductivity $\sigma_D$ is written
by
\[
\tilde{\sigma}_D = \frac{\varepsilon_0 \omega_p^2 (\gamma_D + i\omega)}{\omega^2 + \gamma_D^2},
\]
(3)

where \(\omega_p\) and \(\gamma_D\) is the plasma frequency and the damping rate, respectively. In the low-frequency limit \(\omega \ll \gamma_D\), the above equation reduces to
\[
\sigma_D = \frac{\varepsilon_0 \omega_p^2 \gamma_D}{\omega^2 + \gamma_D^2} = \sigma_{\text{dc}}.
\]
\[
\varepsilon_D = \varepsilon_0 (1 - \frac{\omega_p^2}{\gamma_D^2}).
\]
(4)
(5)

Since \(\omega_p^2/\gamma_D^2\) is independent of frequency and small for the overdamped condition \(\gamma_D \gg \omega_p\), contribution of \(\sigma_D\) to \(\varepsilon\) is just a small constant shift of the order of unity to \(\varepsilon_{\text{LF}}\) and \(\varepsilon_{\text{HF}}\). Thus we can safely assume that the contribution of the Drude conductivity appears only in the dc limit of the conductivity. In the inset of Fig. 1(a), \(\sigma_\alpha(\omega) - \sigma_\beta(0)\) is fitted with the Havrilliak-Negami formula with the same parameters as used in \(\varepsilon_\beta(\omega)\) through the relation of \(\sigma_\alpha(\omega) - \sigma_\beta(0) = -\omega \text{Im} \varepsilon_\beta\). The fitting curves are in excellent agreement with the measured \(\sigma_\alpha(\omega) - \sigma_\beta(0)\), which consolidates the validity of the Havrilliak-Negami fitting.

Most unexpectedly, we have found anomalously large bias dependence of \(\varepsilon_\beta\) and \(\sigma_\beta\), as shown in Fig. 2. They exhibit a jump to higher values at a critical threshold \(E_T\) of 450 V/cm with increasing electric field. The jump is so remarkable that \(\sigma_\beta\) increases by the two orders of magnitude, and \(\varepsilon_\beta\) also increases up to a large value of 200 simultaneously. Furthermore, there appears a clear hysteresis with decreasing electric field.

We should emphasize that an artifact such as heating cannot cause the abrupt jump with the significant hysteresis. We took the data with the sample immersed in liquid \(^4\text{He}\), and carefully evaluated the Joule heating to be a few \(\mu\text{W}\) at the threshold, which is much smaller than a cooling power of liquid \(^4\text{He}\) (typically more than 1W). We should further note that a preliminary measurement using a pulse voltage technique gives identical I-V curves to those taken in static dc bias. The pulse width was changed from 2 to 50 ms with a duration time of 500 ms, and the voltage was swept from 0 to 10 V. The jump in the dc conductivity was clearly observed above 6-7 V, and the conductivity above \(E_T\) was independent of the pulse width \(17\). This further supports that the heating effect is negligible above \(E_T\).

The temperature dependence of nonlinear \(\varepsilon_\beta\) and \(\sigma_\beta\) is shown in Fig. 3. The data for increasing electric field are shown, (the hysteresis in Fig. 2 rapidly disappeared above 4.3 K). The large bias dependence is clearly observed up to 20 K, which suggests that the collective excitation survives at least up to 20 K. The threshold field gradually decreases with increasing temperature, but still remains finite at 20 K. Although \(E_T\) is not the
phase boundary, it can be a measure for a boundary between the metallic and insulating states, which extends to much higher temperatures. Another notable feature is that the metallic state (high-bias state) has essentially temperature-independent $\varepsilon_b$ and $\sigma_b$. In particular, $\sigma_b$ above $E_T$ is as high as $\sigma_a(0)$ at 50 K.

In order to examine whether the large bias dependence exists only along the $b$ direction or not, we measured the non-linear dc conductivity $\sigma(0)$ for the in-plane ($a$ axis) direction as well as along the $b$ direction. As clearly shown in Fig. 4, $\sigma_a(0)$ and $\sigma_b(0)$ exhibit a large non-linearity, although $E_T$ is quite different between the two. Note that $E_T$ along the $b$ direction is lower than that in Fig. 2, because the sample in Fig. 4 is different from the sample in Figs. 2 and 3. $E_T$ varies from sample to sample, but nevertheless the order of the threshold is independent of samples, which is in the range of 1-10 and 100-1000 V/cm for the $a$ and $b$ directions, respectively. Another notable difference is that $\sigma_a(0)$ does not show an abrupt jump, but a steep increase near $E_T$. It does not show a hysteresis either.

It should be emphasized that the observed giant non-linear conduction cannot be assigned to a single particle excitation. In general, nonlinear response is observed when the electric energy gained by the applied electric field $E_{ext}$ exceeds thermal energy $k_BT$. Then a characteristic length scale $L$ can be estimated from the inequality $eE_{ext}L > k_BT$. In the in-plane conduction, for example, $L$ is estimated to be $k_BT/eE_T \sim 1 \mu$m ($T=4$ K and $E_T=5$ V/cm), which is much longer than the mean free path of the conduction electron. It is thus reasonable to assign the giant nonlinear conduction to a collective excitation with a coherent length scale of 1 $\mu$m.

![FIG. 4: Nonlinear dc conductivity of $\theta$-(BEDT-
TTF)$_2$CsZn(SCN)$_4$ for (a) the in-plane direction and (b) the out-of-plane direction at 4.2 K plotted as a function of electric field. The data are measured with increasing electric field.](image)

IV. DISCUSSION

A. Dielectric response

As mentioned in the previous section, the large $\varepsilon_b$ of the $M=$Cs salt is expressed by the dielectric relaxation. A similar $\varepsilon$ is seen in CDW conductors such as $K_{0.3}$MoO$_3$, in which CDW oscillates around pinning centers by an ac electric field to induce a large dielectric response. The dielectric relaxation of CDW is phenomenologically derived from the overdamped Lorentz oscillator given by

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{f}{\omega_0^2 - \omega^2 + i\gamma\omega} \quad (\gamma \gg \omega_0), \quad (6)$$

where $f$, $\omega_0$, $\gamma$ are the oscillator strength, the resonance frequency, and the damping rate, respectively. Equation 6 reduces to the Debye model of dielectric relaxation in the low frequency limit $\omega_0 \gg \omega$ as

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{f/\gamma}{\omega_0^2} \frac{1}{1 + i\omega\tau} \quad (7)$$

where $\tau = \gamma/\omega_0^2$. In real materials, $\tau$ distributes significantly owing to many relaxation channels, and $\varepsilon$ obeys the Havriliak-Negami formula given by Eq. (1).

Accordingly $(\varepsilon_L - \varepsilon_H)/\varepsilon_0$ is equal to $f/\omega_0^2$, and thus the large $\varepsilon_L - \varepsilon_H$ implies a large $f$ and/or a small $\omega_0$. In the case of the pinned CDW, $f$ and $\omega_0$ correspond to the Drude weight ($\varepsilon_0\omega_D^2$) of the order of 1 eV and the pinning potential of the order of 1 meV, respectively, which makes $(\omega_p/\omega_0)^2$ as huge as $10^6$. A more precise treatment is to solve the equation of motion for the phase of the CDW order parameter [10], which also gives the overdamped Lorentz oscillator [20].

From the viewpoint of charge density modulation, difference between CDW and CO is very subtle: the former shows a sinusoidal-wave modulation, and the latter shows a square-wave modulation. In either case, the dielectric response can be described in terms of the overdamped Lorentz oscillator. In fact, charge-ordered materials such as LuFe$_2$O$_4$ [21, 22] and Pr$_{1-x}$Ca$_x$MnO$_3$ [23] show a large $\varepsilon$ described by Eq. (1). Previously we attributed the large $\varepsilon$ of the $M=$Rb salt to the collective motion of CO [12]. An important difference from CDW conductors is that the dielectric relaxation survives in the "normal...
state far above $T_{MI}$. This means that the fluctuation of the pre-formed CO coexists with the unbound carriers that do not participate in CO. In this sense, we can say that the electrons in the $M$ =Rb salt are self-organized above $T_{MI}$ to make its density inhomogeneous.

Since the $M$ =Cs salt is regarded as a system for $T_{MI}$ → 0, $\varepsilon_0$ in Fig. 1 should be compared with $\varepsilon_0$ of the $M$ =Rb salt above $T_{MI}$. This picture is consistent with the x-ray diffraction study by Nogami et al. [24], where they succeeded in observing that a diffuse spot near (0, k, 1/2) grows below 50 K. This indicates that the short range order of CO along the c* direction grows within the BEDT-TTF layer, but no three dimensional order is realized down to 4 K. We should also note that the specific heat [25] and the susceptibility measurements [3] did not detect any jump or kink down to 4 K, implying no second-order phase (CDW/SDW/CO) transitions.

The fitting parameters of Eq. (1) are listed in Table 1. Except for $\tau$ and $\rho$, the obtained parameters are of the same order, meaning that the dynamics of CO is roughly identical. In the overdamped Lorentz model, $\tau$ is associated with the damping rate as $\tau = \gamma / \omega_0^2$. If the damping rate is determined by the scattering between CO and the unbound conducting electron, we can expect $\gamma \propto \rho$, which roughly explains the different magnitudes of $\tau$ and $\rho$ between $M$ =Cs and Rb. We further note that $\varepsilon_{LE}/\varepsilon_0$ for $M$ =Cs is larger than that for $M$ =Rb, implying the smaller pinning frequency $\omega_0$ as was reported in the impurity-doped CDW material [24]. This is consistent with the large bias dependence seen in Figs. 2-4. We did not see any significant nonlinear conduction for the $M$ =Rb salt.

### B. Nonlinear conduction

Although nonlinear conduction is observed in various electron systems, it is rarely observed in homogeneous bulk materials, except for the flux flow resistivity of superconductors, the sliding of CDW/SDW, and the negative resistance in high-mobility semiconductors. Since the nonlinear conduction in Figs. 2-4 occurs in a low electric field of 1-100 V/cm, it should not come from a single particle excitation, but from a collective excitation. Actually the data in Fig. 4 is similar to the nonlinear conductivity of CDW.

Let us list significant differences from the nonlinear conduction of CDW. (i) The conductivity in Figs. 4(a) and 4(b) is highly nonlinear below $E_T$, while most of the CDW conductors show an ohmic conductivity below $E_T$. This suggests that the domain sizes and/or the pinning forces of the pre-formed CO are varied widely in the $M$ =Cs salt. (ii) The CDW conductors show negligibly small nonlinear conduction above $T_c$. Thus it is quite surprising that the $M$ =Cs salt exhibits the huge nonlinearity of 100-1000 times above $T_{MI}$ (< 4 K). (iii) The observed nonlinearity is much larger than that in other two-dimensional conductors [27, 28]. Large nonlinear conduction has been observed mostly in one-dimensional CDW conductors such as $K_3MoO_5$, NbSe$_3$, TaS$_3$ [29, 30]. (iv) The nonlinear conductivity perpendicular to the charge-density modulation is observed for the first time.

Figure 5 shows the dc resistivity along the b axis ($\rho_b$) below $E_T$ plotted as a function of temperature. $\rho_b$ increases with decreasing temperature, the temperature dependence of which is nearly the same as that of the inplane (the a axis) resistivity ($\rho_a$) [8]. This indicates that the insulating state is more or less three-dimensional. The low-temperature upturn in $\rho_b$ has been understood in terms of variable range hopping (VRH) — strongly localized state, but it should be emphasized here that the frequency and temperature dependences of $\varepsilon(\omega)$ and $\sigma(\omega)$ seriously contradict the VRH picture.

**TABLE I: Parameters obtained through fitting to the complex dielectric constant by a generalization of Debye’s dielectric relaxation (the Havriliak-Negami formula)**

|          | Rb ($T > T_{MI}$) | Rb ($T < T_{MI}$) | Cs |
|----------|-------------------|-------------------|-----|
| $T$ (K)  | 200               | 140               | 4.3 |
| $\varepsilon_{LE}/\varepsilon_0$ | 38                | 35                | 90  |
| $\varepsilon_{HF}/\varepsilon_0$ | 12                | 12                | 29  |
| $\alpha$ | 0.23              | 0.23              | 0.35 |
| $\beta$ | 0                 | 0                 | 0.15 |
| $\tau$ (\mu s) | 0.17              | 5.00              | 7   |
| $\rho$ (10$^6\Omega$cm) | 0.016              | 30                | 1   |

**FIG. 5:** Out-of-plane resistivities of $\theta$-(BEDT-TTF)$_2$CsZn(SCN)$_4$ for $E < E_T$ and $E > E_T$ plotted as a function of temperature.
might be associated with the fake insulating state.

The nonlinear conduction below $E_T$ is usually weak in the CDW materials. A homogeneous CDW order has a single coherent length, and the corresponding value of $E_T$ is uniquely determined. On the contrary, the $M =$Cs salt exhibits the nonlinear conduction for $E \ll E_T$ as shown in Fig. 4, which naturally implies a larger scale of the CO domain. Imagine that the CO domains form a percolative network in a fractal dimension. Then a larger cluster can move for a smaller electric field, and the number of the mobile clusters gradually increase with increasing field. This picture is consistent with the charge inhomogeneity in the high-temperature superconductor [1].

The most puzzling finding is the large hysteresis of $\varepsilon_b$ and $\sigma_b$ shown in Fig. 2. At first glance, this resembles the switching behavior of K$_{0.3}$MoO$_3$ [20], which was phenomenologically understood in terms of the damping rate depending on the CDW sliding velocity [23]. However, the electric field was perpendicular to the charge modulation in the present case, and we cannot expect the sliding of CO across the layers. We cannot expect either that a CO domain hops from one layer to other, because it requires enormous numbers of electrons to tunnel simultaneously. We should further note that $\varepsilon$ above $E_T$ decreases with increasing bias voltage for conventional CDW conductors [24], which is seriously incompatible with the data in Fig. 2(a).

Finally we will append a note that we have found a similar bias dependence in the dc conductivity of $\theta-$BEDT-TTF)$_2$CsCo(SCN)$_4$, which has nearly the same structure as $\theta-$BEDT-TTF)$_2$CsZn(SCN)$_4$. This strongly suggests that the giant nonlinear conduction is a generic feature for some $\theta$-type salts [17].

C. Inhomogeneity in a homogeneous system

As mentioned in the Introduction, the organic salts are one of the cleanest solids currently available, in which point defects, dislocations and solid solutions are expected to be negligibly small. Nevertheless, $\rho_a$ for $M =$Cs is 30 m$\Omega$cm at 300 K [8], which is indeed anomalously high. In a two-dimensional conductor, the mean free path $\ell$ is directly evaluated from the in-plane resistivity through the relation

$$k_F\ell = \hbar\epsilon_0/e^2\rho_a,$$

where $\epsilon_0$ is the interlayer spacing [31]. Then 30 m$\Omega$cm gives $k_F\ell$ =0.1, and such a short $\ell$ is difficult to understand in the clean system. The metallic behavior ($d\rho_a/dT > 0$) at 300 K is also difficult to understand, because de Broglie wavelength of the conducting electron is much shorter than $\ell$ for $k_F\ell \ll 1$.

We can clear up these difficulties, if the electronic states of the $M =$Cs salt are spatially inhomogeneous, because Eq. (8) is derived for a homogenous system. Suppose most of the conduction electrons are nearly frozen to form CO domains, and the unbound electrons flow in a percolation network consisting of the domain boundaries. Then the effective cross section for the current path is much smaller than the bulk cross section of the sample, and the “apparent” resistivity would be much higher. If $k_F\ell$ is still larger than unity in the current path, the conduction can be metallic to cause $d\rho/dT > 0$. Judging from the anomalously high in-plane resistivity, we think that the self-organization of the electrons in $M =$Cs readily starts even at room temperature. Then the self-organized CO is gradually frozen with decreasing temperature, and eventually shows the CDW-like collective excitation.

Let us discuss the reason why the self-organized inhomogeneity is realized in the $\theta$-type salts. Mori [35] calculated the ground-state energy for the $\theta$-type salts using the extended Hubbard model, and discussed possible CO patterns at finite temperatures. He found that the on- and off-site Coulomb repulsions are of the same order, and their delicate balance causes various CO patterns. In particular, “horizontal”, “diagonal” and “5-fold” patterns are nearly degenerate in the $M =$Cs salt, and thus it is highly probable that different charge orders coexist. Furthermore, these charge orders would be strongly coupled with phonons, as was evidenced by the strong Fano effects in the infrared spectroscopy [14, 32]. Thus CO domains of different patterns store finite elastic energies, which prevents the domains to grow in size [36]. As a result, homogeneous phase transition is seriously suppressed, and the domains are frozen at low temperatures like polar nano-regions in relaxor ferroelectrics [37]. We therefore expect that the $M =$Cs salt can be a canonical material showing the intrinsic inhomogeneity in the electron density.

A remaining issue is up to what temperature the collective excitation can be observed. Figure 6 shows $E_T$ in Fig. 3 (b) as a function of temperature. Clearly, the phase boundary would exist above 20 K. If we extrapolate the data to higher temperatures as indicated by the dotted line, we can estimate the temperature for $E_T = 0$.
to be 65 K, which is roughly the same temperature at which \(1/\sigma_0(1\text{MHz})\) in the high-bias state is equal to \(\rho_b\) in Fig. 5. To measure \(E_T\) at higher temperatures, we should employ a pulse generator to avoid the Joule heating, which will be tried in future work.

The metallic state above \(E > E_T\) is not yet understood. In CDW conductors, the high-biased state is the state that CDW is sliding with a finite damping from the normal electrons, which has been established from the diffraction experiment in high electric fields. In the present case, the \(M = \text{Cs}\) salt only has the short range order of CO, where the phase of CO acquires no rigidity. Then the sliding of CO is difficult to expect. Another choice is the melting of CO by electric field, like the vortex lattice melting by magnetic field in a type-II superconductor. To clarify this problem, diffraction measurements in electric fields are indispensable, and microscopic theory to explain our findings is also necessary.

V. SUMMARY

We have observed the large dielectric response and giant nonlinear conduction in \(\theta-(\text{BEDT-TTF})_2\text{CsZn(SCN)}_4\). The data resemble those for charge-density-wave (CDW) conductors such as \(\text{K}_0.3\text{MoO}_3\). However, \(\theta-(\text{BEDT-TTF})_2\text{CsZn(SCN)}_4\) shows no second-order transition down to 4 K, which implies that it exhibits CDW-like collective excitations in the “normal” state. Therefore we expect that this material can belong to a new hierarchy of condensed matter, in which conduction electrons are self-organized to have collective modes without long range order.

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