# Out-of-plane dielectric constant of $\theta$-(BEDT-TTF)$_2$RbZn(SCN)$_4$ single crystal

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**Abstract**

The out-of-plane dielectric constant $\varepsilon$ of the quasi-two-dimensional organic conductor $\theta$-(BEDT-TTF)$_2$RbZn(SCN)$_4$, which exhibits a metal-insulator transition at 190 K, was measured and analyzed from 100 to 300 K in the frequency range $10^3$-$10^8$Hz. Most unexpectedly $\varepsilon$ is found to show a significant dielectric relaxation above 190 K, which suggests that insulating and metallic phases coexist above 190 K.

*Key words:* dielectric constant, metal-insulator transition, organic conductor

## 1. Introduction

The organic conductor (BEDT-TTF)$_2$X constitutes a large family of layered organic systems, in which the conducting organic donor molecule BEDT-TTF and the insulating monovalent anion X alternately stack to form a layered structure. Among them, $\theta-$(BEDT-TTF)$_2$RbZn(SCN)$_4$ is particularly interesting, in that it exhibits a metal-insulator (MI) transition at $T_{\rm MI}$=190 K with the lattice modulation [1]. This should be different from a conventional Mott transition, because the valence band of this compound is not half-filled, but quarter-filled. Miyagawa *et al.* [2] first pointed out through an NMR experiment that this transition is a charge ordering, and that a charge inhomogeneity grows above $T_{\rm MI}$, possibly as a precursor of the charge ordering.

We have studied the dielectric response of the parent insulator of the high-temperature superconductors (HTSC) $Bi_2Sr_2Dy_{1-x}Er_xCu_2O_8$, and have revealed that $\varepsilon$ shows a relaxation behavior which was attributed to some kind of charge inhomogeneity in the $CuO_2$ plane [3]. Thus it would be tempting to compare $\varepsilon$ of HTSC with $\varepsilon$ of a material showing a "well-defined" charge ordering.

Here we report on measurement and analysis of the out-of-plane dielectric constant of $\theta$-(BEDT-TTF)$_2$RbZn(SCN)$_4$. The observed dielectric spectrum is explained with the Debye description of dielectric relaxation. The most important aspect is that $\varepsilon$ above $T_{\rm MI}$ is essentially similar to that below $T_{\rm MI}$, implying the existence of the charge inhomogeneity above $T_{\rm MI}$.

## 2. Experimental

Single crystals were prepared using a galvanostatic anodic oxidation method, and the detailed growth conditions and their characterization were described in [1]. Complex impedance from $10^3$ to $10^8$ Hz was measured using a two-probe method with a similar technique to Böhmer *et al.* [4], which was described in detail in [3]. Since the physical properties of this material are very sensitive to the cooling rate [1], we slowly cooled the samples at a rate of less than 1 K/min.

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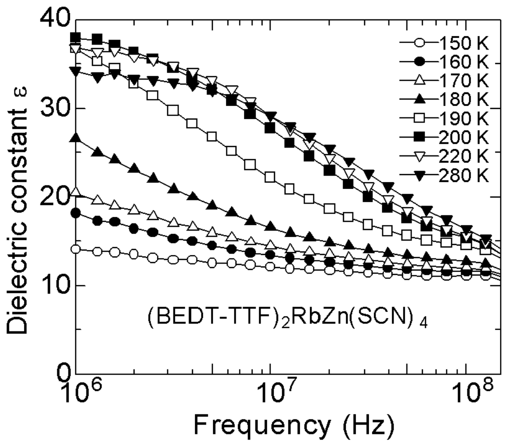


Fig. 1. The out-of-plane dielectric constant of $\theta$−(BEDT-TTF)$_2$RbZn(SCN)$_4$ from $10^6$ to $10^8$Hz

## 3. Result and Discussion

Fig. 1 shows $\varepsilon$ from $10^6$ to $10^8$ Hz. $\varepsilon$ is large and positive, and decreases with frequency to reach a small constant value. This behavior is qualitatively understood in terms of Debye-type dielectric relaxation given as

$$\varepsilon(\omega) = \varepsilon_{\mathrm{HF}} + \frac{\varepsilon_{\mathrm{LF}} - \varepsilon_{\mathrm{HF}}}{1 + i\omega\tau} \qquad (1)$$

where $\varepsilon_{\mathrm{HF}}$, $\varepsilon_{\mathrm{LF}}$ and $\tau$ are the dielectric constant in the high-frequency limit, the dielectric constant in the low-frequency limit, and the relaxation time, respectively. While $\varepsilon$ is weakly dependent on temperature above $T_{\mathrm{MI}}$, it rapidly decreases with decreasing temperature as soon as the MI transition sets in.

The dielectric behavior below $T_{\mathrm{MI}}$ is similar to that for $K_{0.3}MoO_3$ [5] and $Pr_{1-x}Ca_xMnO_3$ [6], which means that charge ordered states cause a dielectric relaxation. We should note that the origin of the charge ordered states are different for different materials. The electron-phonon interaction causes the charge-density wave for $K_{0.3}MoO_3$, whereas the interplay between the orbital/spin ordering and the double exchange causes the charge ordering for $Pr_{1-x}Ca_xMnO_3$. For $\theta$-(BEDT-TTF)$_2$RbZn(SCN)$_4$ intersite Coulomb interaction is considered to be an origin for the charge ordered state, as is already reported in other organic salts[7]. According to eq.(1), $1/\tau$ is roughly estimated as the frequency at an inflection point in Fig.1, and then $\tau$ is found to increase rapidly with decreasing temperature below $T_{\mathrm{MI}}$.

Next we will discuss the dielectric response in the metallic state above $T_{\mathrm{MI}}$. We should emphasize that $\varepsilon$ above $T_{\mathrm{MI}}$ is essentially the same as that below $T_{\mathrm{MI}}$. This is seriously incompatible to the dielectric response for usual metals, where $\varepsilon$ is negative below the plasma frequency. The positive $\varepsilon$ indicates that the screening motion of charged carriers does not work well. As mentioned above, the NMR experiment suggests the existence of the charge inhomogeneity as a preformed charge ordering. If so, the metallic region and the charge-ordered region are randomly distributed, where the dc current is carried by a percolation path of the metallic region. Then $\tau$ is dominated by the randomness (percolation path), and can be weakly dependent on temperature, which is in agreement with the temperature dependence of resistivity above $T_{\mathrm{MI}}$[1].

## 4. Summary

We measured the out-of-plane dielectric constant of $\theta$-(BEDT-TTF)$_2$RbZn(SCN)$_4$ from $10^3$ to $10^8$Hz. Above the metal-insulator transition at 190 K, the dielectric constant is positive with relaxation behavior, which is very similar to the dielectric response of the parent insulator of high-temperature superconductors. We suggest that insulating and metallic phases coexist in the metallic state of this compound.

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