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Far-infrared reflectance of β"-(BEDT-TTF)$_2$AuBr$_2$: co-existence of free carriers and a single-particle gap at 2Δ=130cm^{-1}

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Abstract

We have measured the temperature dependence of far-infrared reflectance of β"-(BEDT-TTF)$_2$AuBr$_2$ to investigate the possibility of SDW transition. The polarized reflectance in two-dimensional BEDT-TTF layers shows a highly anisotropic feature of conduction band. Both the $E\|c$ and the $E\perp c$ reflectance line-shapes, which are Drude-like at room temperature, demonstrate a clear dip below 100 cm^{-1} even at 40 K. The frequency-dependent conductivity at 7 K indicates the presence of a single-particle gap at 2Δ=130 cm^{-1} assignable to the SDW nesting along *c*. The nesting fluctuation exists already at room temperature because of a strongly one-dimensional nature of the band dispersion.

1. INTRODUCTION

Recent studies concerning Fermi-surface topology (so-called *fermiology*) of organic conductors have been extensively developed by electromagnetic measurements such as Shubnikov-de Haas (SdH) and de Haas-van Alphen (dHvA) effect. These phenomena are based on quantized cyclotron motion of electrons[1], so that a closed Fermi surface is at least necessary for the object. Among reported organic conductors (BEDT-TTF)$_2X$ system is very appropriate for this condition because a number of salts are predicted to have a two-dimensional, closed Fermi surface through tight-binding band calculations[2]. In most cases the SdH and/or the dHvA oscillations have been observed in good agreement with the above band picture, suggesting the efficiency of this calculation. Since this approximation, however, completely neglects Coulomb interaction between conduction electrons as well as electron-phonon interaction, the model cannot describe SDW and CDW state, which situations become more conspicuous if the band is highly anisotropic.

The β"-type BEDT-TTF salt, which retains metallic properties down to mK regions, is just a instance for the above case. The band calculation predicts two kinds of Fermi surface[3]: one is based on a one-dimensional band along the Γ→Z direction, and another is a small pocket around the X point in the first Brillouin zone. This picture is inconsistent with the SdH and dHvA results which suggest the presence of three closed parts in the Fermi surface[4-6]. The reconstruction of the Fermi surface is then necessary to explain the quantum oscillations, which driving force should be related to the neglected interactions. We measured the temperature dependence of reflectance of β"-(BEDT-TTF)$_2$ICl$_2$ to investigate the band structure of this type salt: the spectra down to 25 K and 500 cm^{-1} did not show any sign for the occurrence of SDW or CDW transition[7]. We here present the far-infrared results on β"-(BEDT-TTF)$_2$AuBr$_2$. We have found a single-particle gap at 2Δ=130cm^{-1} besides free carrier absorption below 20 cm^{-1}. The details of the electronic structure are discussed in comparison with (TMTSF)$_2X$ salts.

2. EXPERIMENTAL

Single crystals of β"-(BEDT-TTF)$_2$AuBr$_2$ were prepared by a conventional electrochemical method using tetra-*n*-butylammonium dibromoaurate(I) as a supporting electrolyte. Temperature dependence of far-infrared reflectance was measured on Bruker IFS-113v spectrometers (both at UF and IMS *UVSOR* BL6B[8]) with a reflectance unit and a Hi-tran cryostat over the wave-number region of 20-700 cm^{-1} at temperatures among 5-300 K. The absolute reflectivity at a given temperature was determined from four measurements: $R(\omega,T)=[P_s(\omega,T)/P_r(\omega,T)]/[P_{Au}(\omega,300)/P_r(\omega,300)]$, where $P_x(\omega,T)$ is the power spectrum at temperature T and the suffixes x=s, r, Au represent the spectrum from a sample, reference (Au mirror), the sample coated with Au, respectively.

3. RESULTS AND DISCUSSION

In the β"-type crystal BEDT-TTF molecules are arranged uniformly along the *c* axis while dimerized along the *a* axis (nearly ⊥c; β=103°), forming two-dimensional layers parallel to the (010) face. The room temperature reflectance (not presented here) looks like a Drude type for both polarizations $\|c$ and $\perp c$, the latter spectrum having some vibrational structures at 250, 310 and around 450 cm^{-1}. These bands are due to A_g modes of BEDT-TTF molecule resulting from the strong dimerization along the *a* axis.

Figure 1 shows the polarized reflectance at 7.0 K. The data below 20 cm-1 are linear-extrapolated as $R\rightarrow 1$ on $\omega\rightarrow 0$, shown with dotted lines. The $E\|c$ reflectance is high and gradually increases as frequency down to 150 cm^{-1}. The structures appeared at nearly the same positions as in the $E\perp c$ spectrum, are also assignable to A_g modes of BEDT-TTF. This seems unreasonable because the molecules are not dimerized in the $\|c$ direction, that is, A_g modes are infrared-inactive. The x-ray results have also indicated that no structural change corresponding to a formation of CDW occurs down to 4 K[7]. The vibrational structures in the $E\|c$ spectrum are then regarded as the component of $\|a$ polarization, whose direction is not

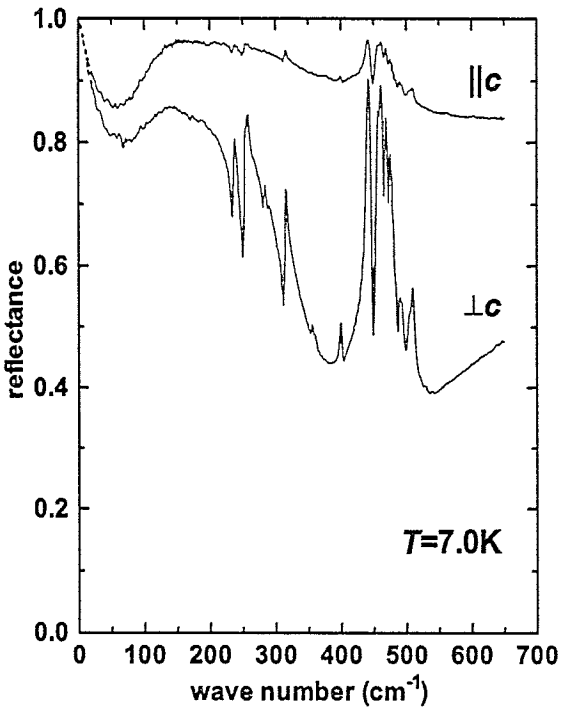

Figure 1. Polarized reflectance at 7.0K.

completely orthogonalized to $\|c$. These vibrational features are extremely pronounced in the $E\perp c$ spectrum at low temperatures.

We now turn our interest to a reflectance dip below 150 cm^{-1} in both spectra. This infers the presence of energy gap, which become more obvious through Kramers-Krönig analysis. Figure 2 shows the $E\|c$ conductivity spectra at temperatures 7.0 K and 300 K (Note that the spectrum of 300 K is drawn in tenfold scale). The conductivity at 7.0 K demonstrates a very strong and sharp peak at 130 cm^{-1}. These spectral features including its line-shape quite resemble the case in $(TMTSF)_2AsF_6$ at 6 K, where the material is in the SDW state[9], suggesting that the peak comes from an optical transition across a single-particle gap on the one-dimensional band. The possibility of a CDW collective mode is denied by the reason mentioned above. Considering the EPR result which indicates the occurrence of magnetic transition below 20 K[10], the origin of the gap is probably due to the SDW nesting along the $\|c$. This assignment also suits the SdH/dHvA results[4-6] because the Fermi surface is predicted to consist of three pockets after the nesting transition[11]. The temperature dependence of the $E\|c$ reflectance among 5-40 K, however, scarcely changed over 20-700 cm^{-1}, meaning that the pseudo-gap exists above the EPR transition temperature. We here remark that the conductivity spectrum at 300 K is far from a typical line-shape of free-carrier absorption: the spectrum demonstrates a broad peak around 170 cm^{-1}, accompanying a dip below 100 cm^{-1}. This suggests that the nesting fluctuation of SDW is present even at room temperature. The similar results have been reported in the chain-axis conductivity of $(TMTSF)_2PF_6$[12].

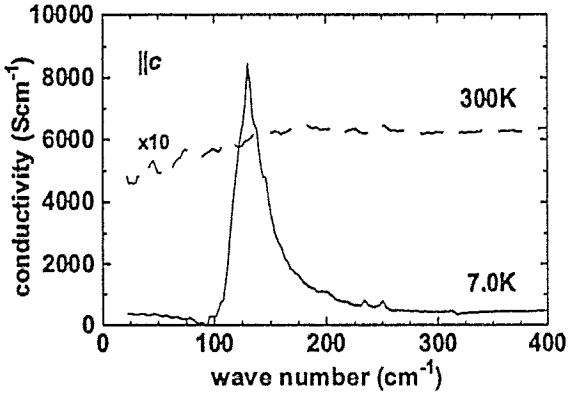

Figure 2. Conductivity sprctra for $E\|c$ at 7.0K and 300K.

As discussed in this section the electronic structure of β"-$(BEDT\text{-}TTF)_2X$ is proved to quite resemble that of $(TMTSF)_2X$ in spite of the large difference of donor molecule arrangement. The calculated band structure of $(TMTSF)_2X$ yields a one-dimensional Fermi surface with small warp[13], which is also very similar to the one-dimensional part in β"-$(BEDT\text{-}TTF)_2X$. In both cases the band dispersion is dominated by the largest overlap(s) along the particular direction ($\|a$ for the TMTSF salt and $\|c$ for the BEDT-TTF salt), the overlap(s) being much stronger than in other directions. The major part of the Fermi surface, as a result, consists of open planes in both salts, and the optical transitions near the Fermi level become similar.

The superconductivity in β"-$(BEDT\text{-}TTF)_2X$ has not been found yet down to 44 mK[6] in spite of the quite resemblance to $(TMTSF)_2X$. This is an interesting problem in the future.

4. CONCLUSIONS

The temperature dependence of polarized far-infrared reflectance has been measured in β"-$(BEDT\text{-}TTF)_2AuBr_2$. A single-particle gap has been found at 2Δ=130 cm^{-1}, which is assignable to the SDW nesting along *c*. The electronic structure has been proved to be similar to $(TMTSF)_2X$.

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