Magnetooptical measurements of \( \beta''\)-\((\text{BEDT-TTF})(\text{TCNQ})\)

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Abstract

Magnetooptical measurements have been performed on \( \beta''\)-\((\text{BEDT-TTF})(\text{TCNQ})\) using the cavity perturbation techniques. \( \beta''\)-\((\text{BEDT-TTF})(\text{TCNQ})\) has very exotic Fermi surface (FS). Several harmonic resonances were observed at various angles. We consider these resonances as q1D-periodic orbit resonance. The obtained Fermi velocity and the scattering time are \(4.4 \times 10^4\) m/s and \(2.1 \times 10^{-11}\) s, respectively. When the magnetic field was rotated in the \(b^*a\)-plane, the similar resonances are observed. According to these results, the FS topology of the system at low temperature is discussed.

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1. Introduction

In general, Fermi surface (FS) topology has been studied using various techniques such as de Haas–van Alphen, Shubnikov–de Haas (SdH) \([1]\), angle-dependent magnetoresistance (ADMRO) and cyclotron resonance (CR) \([2]\). However, quantum oscillations and CR measurements are not applicable for a quasi-one-dimensional (q1D) system, because these measurements are based on the cyclotron motion of carriers. Recently, the novel CR-like resonance, which is essentially different from the conventional CR, has been observed in \(z\)-(BEDT-TTF)\(_2\)KHg(SCN)\(_4\) with q1D-FS \([3,4]\). This magnetooptical resonance is so called q1D-periodic orbit resonance (POR) or fermi traverse resonance, and has attracted a great deal of attention because of its potential application for the precise determination of the important parameters describing the FS topology of the q1D conductors \([5]\).

In this study, we applied the magnetooptical measurement to probe the FS topology of \( \beta''\)-\((\text{BEDT-TTF})(\text{TCNQ})\) at low temperature. This
The salt was synthesized by Yamamoto et al. recently [6]. The unit cell contains two molecules, BEDT-TTF and TCNQ. The donor BEDT-TTF and the acceptor TCNQ molecules form the separate layers parallel to the ac-plane. According to the band calculation, the dominant overlap integral between BEDT-TTF molecules and between TCNQ molecules spread along the a- and c-axis, respectively. Therefore, this salt is expected to have a very exotic FS at room temperature in such way that two q1D-FS are crossing each other at right angle. One FS, associated with BEDT-TTF layers, is composed of parallel sheets normal to the ka-axis, while the other FS associated with TCNQ layers has parallel sheets normal to the kc-axis.

The temperature dependence of the resistivity of this salt exhibits metallic behavior down to 0.5 K [7]. In addition, three anomalies at 175, 80 and 20 K are also detected [6]. The origin of these anomalies is not clear at the moment but is considered to be related to the nesting of q1D FS. The anisotropy of resistivities for the a-, b- and c-axis has the ratio of 15:1:480 at room temperature [7].

ADMRO and SdH measurements at ambient pressure have been performed by Yasuzuka et al. [8]. When the magnetic field was rotated in the b*c-plane (i.e. parallel to the q1D BEDT-TTF FS), Lebed resonances were observed clearly, but the dip position are not consistent with the conventional resonant condition. On the other hand, when the magnetic field was rotated within the b*a-plane (i.e. parallel to the q1D TCNQ FS), Lebed resonances were not observed. These results suggest the existence of BEDT-TTF q1D-FS and the absence of TCNQ one. SdH measurement suggests the existence of several small Fermi surface pockets. The areas of these pockets are a few percent of the first Brillouin zone. In order to clarify the FS of this salt \( \beta \)-BEDT-TTF(TCNQ), we have performed magneto-optical measurements.

2. Experimental

Magneto-optical measurements have been performed using a cavity perturbation technique equipped with a millimeter vector network analyzer (MVNA) at IMR, Tohoku University [9]. The sample used in the present study had typical dimensions of \( 1.0 \times 0.5 \times 0.2 \) mm\(^3\) and was mounted in the end plate of the cylindrical resonant cavity. Due to the TE\(_{011}\) resonant cavity mode, the oscillatory magnetic field was always applied to the sample. The fundamental frequency and the \( Q \) factor of the cavity were 58 GHz and over 3000, respectively. The magnetic field was rotated in the \( b^*c \)-plane with the angle \( \theta \) and in the \( b*a \)-plane with \( \phi \), where \( \theta \) and are \( \phi \) measured from the \( b^* \)-axis.

3. Result and discussion

Fig. 1 shows the angle \( \theta \) dependence of the typical cavity transmission spectra observed at 1.6 K. Two or three resonances are observed at depending on \( \theta \). We plot these resonances in \( v/B_{\text{res}} \) as a function of \( \theta \) as shown in Fig. 2, where \( v \) and \( B_{\text{res}} \) are the observed frequency and the resonance

![Fig. 1. Angular dependence of the cavity transmission spectra of \( \beta \)-BEDT-TTF(TCNQ) observed at 1.6 K, when the magnetic field is rotated in the \( b^*c \)-plane.](image-url)
the Fermi velocity \(E_{\text{F}}\) is defined as an oblique crystal lattice system. Thus q1D-FS for BEDT-TTF is defined as

\[
E(k) = \hbar v_F(|k_\alpha| - k_F) - \sum_{m,n} t_{m,n} \times \cos\{(mc + nd)k_c + nb^*k_b^*\},
\]

where the integers \(m, n\) and \(t_{m,n}\) correspond to the Fourier components of the FS corrugations and its effective transfer integral, respectively. \(b^*, c\) and \(d\) are the inter layer spacing between \(ac\)-plane, the in-plane lattice spacing (parallel to q1D FS) and the oblique parameter, by which the difference from the orthorhombic lattice is characterized, respectively [5].

From the Boltzmann transport equation and the equations of motion for carriers in a magnetic field, we can obtain the frequency-dependent conductivity matrix as follows:

\[
\begin{pmatrix}
\sigma_{yy}(\omega) & \sigma_{yz}(\omega) \\
\sigma_{zy}(\omega) & \sigma_{zz}(\omega)
\end{pmatrix}
= N(E_F) \frac{e^2}{\hbar} \sum_{m,n} \sum_{m,n} \frac{t_{m,n}^2}{1 + \{(\omega - \nu_F G_{mn})\tau\}^2}
\]

\[
G_{mn} = \frac{eB}{\hbar}[(mc + nd)\cos \theta - nb^* \sin \theta],
\]

where \(N(E_F)\) is the density of states per unit volume at the Fermi level, \(v_F\) is the Fermi velocity and \(\tau\) is the scattering time of carriers. When the frequency-dependent conductivity given by Eq. (2) is minimum, the resonance condition is satisfied and therefore, results in \(\omega = v_F G_{mn}\) for a finite \(\omega\). Using Eq. (3), we can obtain the angular dependence for q1D-POR as follows:

\[
\begin{align*}
\frac{v}{B_{\text{res}}} &= \frac{e\nu_F}{\hbar} [(nb^*)^2 \\
+ (mc + nd)^2]^{1/2} |\sin(\theta - \theta_{mn})|,
\end{align*}
\]

\[
\tan \theta_{mn} = \frac{m c}{n b^*} + \frac{d}{b^*},
\]

\[
\theta_{mn} = 0, 1, 2, \ldots, m = 0, \pm 1, \pm 2, \ldots.
\]

Letting the Fermi velocity as a free parameter in Eq. (4), the observed resonances can be fitted as shown in Fig. 2. The best fit was obtained at \(v_F = 4.4 \times 10^4\) m/s. Using \(c/b^* = 0.357\) and \(d/b^* = 0.092\) obtained from the ADMRO results, these q1D-POR curves can be identified into different combination of \((m, n)\) as indicated in the Fig. 2. Since these resonances are caused by the in-plane conductivity (i.e. \(\sigma_{yy}\) in Eq. (2)), the resonance mode corresponding to \(m = 0\) was not observed in the present experiment. When \(m = 0\), the magnitude of the \(\sigma_{yy}\) is very small, because the oblique parameter \(d\) is much smaller than the lattice parameter \(c\) or \(b^*\). Our POR results are almost consistent with the ADMRO results. However, we are missing the resonance mode with odd number of \(m\). The reason is not clear at the moment. Moreover, the lattice parameters used in the POR fitting are different from those obtained at room temperature. This may suggest the occurrence of a structural transformation in this salt at low temperature, caused by the nesting of q1D-FS.

The scattering time \(\tau\) can be also determined from the resonance field and the resonance line width of POR by using the following equation [10]:

\[
\Delta B = \frac{2B_{\text{res}}}{\omega_{\text{res}} \tau}.
\]
The obtained result from the POR is $\tau = 2.1 \times 10^{-11}$s.

We also performed similar measurements within the $b^*a$-plane. Fig. 3 shows the angle $\phi$ dependence of typical cavity transmission spectra observed at 1.7 K. The two resonances were observed and they shifted to higher field as the angle $\phi$ increases. When $\phi = 90^\circ$ ($B||a$-axis), no resonance was observed. This may be attributed to the localization of carriers due to the small closed orbit on the q1D-FS. Fig. 4 shows the angular dependence of the resonance fields. These resonances show $\cos \phi$ dependence. This is reasonably understood by considering the $\cos \phi$ component of magnetic field involved in the resonance condition. The observation of two resonance modes suggests the existence of two Fourier components for the FS corrugations. As the maxima of the curves in Fig. 4 correspond to the same $v/B_{\text{res}}$ curves in Fig. 2, which correspond to $(m,n) = (2,0)$ or $(4,0)$ mode in Fig. 2, we can state that the observed resonance in the $b^*c$-plane and the $b^*a$-plane originate from the same FS (i.e. BEDT-TTF FS). Therefore, the present experimental results suggest the existence of the BEDT-TTF FS. However, the existence of the TCNQ one is not confirmed by our measurements. This is consistent with the ADMRO results and suggests the disappearance of the TCNQ q1D-FS due to the nesting at low temperature.

Moreover, we observed SdH oscillations in the same experiment. Fig. 5 shows the high field region of absorption lines at 57.9 and 55.1 GHz for $\theta = -15^\circ$. The period of the oscillation is about $7.1 \times 10^{-3} \text{T}^{-1}$, i.e. the frequency is about 140 T.

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![Fig. 3](image1.png)

**Fig. 3.** Angular dependence of the cavity transmission spectra of $\beta$-(BEDT-TTF)TCNQ observed at 1.7 K, when the magnetic field was rotated in the $b^*a$-plane.

![Fig. 4](image2.png)

**Fig. 4.** Angular dependence of the resonance fields, when the magnetic field is rotated in the $b^*a$-plane. The solid lines show the $\cos \phi$ dependence.

![Fig. 5](image3.png)

**Fig. 5.** The high field region of the cavity transmission spectra at 55.1 and 57.9 GHz for $\theta = -15^\circ$ at 1.5 K. SdH oscillations are observed and the obtained period from the spacing of the dashed line is about $7.1 \times 10^{-3} \text{T}^{-1}$. 
This value is consistent with the results of SdH measurement by Yasuzuka et al. [8]. As the obtained frequency show 1/cos \( \theta \) dependence as shown in Fig. 6, this FS can be considered to have q2D nature, which may be coming from the small pocket generated by the imperfect nesting of FS responsible for TCNQ.

4. Conclusion

We have demonstrated the results of the magnetooptical measurements on a new organic conductor \( \beta''-(\text{BEDT-TTF})(\text{TCNQ}) \), the FS of which at room temperature are considered to be composed of two q1D-FS’s crossing each other at right angle. The observed resonance at low temperature appears to be q1D-POR from its characteristic angular dependence. Considering together with the results of ADMRO, the observed POR signals can be reasonably attributed to be the BEDT-TTF layer. However the estimated lattice spacing of q1D-FS for the BEDT-TTF molecule does not correspond to the lattice parameters determined at room temperature. This fact may suggest the occurrence of a structural transformation induced by the nesting of the TCNQ FS.

We also observed SdH oscillations with a frequency of about 140 T. From the 1/cos \( \theta \) dependence of the oscillation frequency, the existence of q2D-FS, which may be coming from the TCNQ molecules, is suggested. This result agrees with the presence of small FS pocket reported from various SdH study [8].

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References