Available online at www.sciencedirect.com
SCIENCE DIRECT®
ELSEVIER
Synthetic Metals 153 (2005) 369–372
SYNTHETIC METALS
www.elsevier.com/locate/synmet

Fermi Surface Study of β"-(BEDT-TTF)(TCNQ) by Magnetooptical Measurements

M. Kimata[a],* Y. Oshima[b], K. Koyama[c], H. Ohta[d], M. Motokawa[c], H. M. Yamamoto[e, f] and R. Kato[e, f]

[a]*The Graduate School of Science and Technology, Kobe University, Nada, Kobe 657-8501, Japan*
[b]*National High Magnetic Field Laboratory, Florida State University, Tallahassee, Florida 32310 USA*
[c]*High Field Laboratory for Superconducting Materials, Institute for Materials Research, Tohoku University, Katahira, Sendai 980-8577, Japan*
[d]*Molecular Photoscience Research Center, Kobe University, Nada, Kobe 657-8501, Japan*
[e]*RIKEN (The Institute of Physical and Chemical Research), Hirosawa, Wako 351-0198, Japan*
[f]*Japan Science and Technology Corporation, Kawaguchi 332-0012, Japan*

Abstract

We have performed magnetooptical measurements of β"-(BEDT-TTF)(TCNQ) using a cavity perturbation technique with a millimeter vector network analyzer (MVNA) and a 14 T superconducting magnet at IMR, Tohoku University. The results of ADMRO and SdH measurements at low temperatures are not consistent with the band calculation result. Therefore, we are interested in Fermi surfaces at low temperatures and have performed magnetooptical measurements of β"-(BEDT-TTF)(TCNQ). Although the band calculation and ADMRO results suggest the existence of Q1D FS, our results suggest the existence of Q2D FS only. From the results of magnetooptical measurements, we discuss the FS topology of this system at low temperatures.

Keywords: Transport measurements, magnetotransport, Organic conductors based on radical cation and/or anion salt, Other phase transition

1. Introduction

Organic conductors have been studied intensively because of their various interesting properties, owing to their low dimensionality. To have a complete understanding of organic conductors, the study of Fermi surfaces (FSs) is very important. In general, FS topology has been studied using various techniques such as de Haas-van Alphen (dHvA), Shubnikov-de Haas (SdH) [1], angular dependent magnetoresistance oscillation (ADMRO) techniques. Another useful technique is magnetooptical measurement [2,3]. For example, the conventional cyclotron resonance (CR) is a well-known method for studying semiconductors. Recently, the novel CR-like resonance, which is essentially different from the conventional CR, has been observed in quasi-one-dimensional (Q1D) and quasi-two-dimensional (Q2D) organic conductors [4-7]. This magnetooptical resonance is the so-called Q1D or Q2D periodic orbit resonance (POR), and has attracted great attention because of its potential application for the precise determination of the important parameters of FS (e. g, Fermi velocity and effective mass of carriers).

In this study, we used magnetooptical measurement in probing the FS topology of β"-(BEDT-TTF)(TCNQ) at low temperatures. This salt has recently been synthesized by Yamamoto *et al.* [8]. The unit cell contains two molecules: BEDT-TTF and TCNQ. The donor BEDT-TTF and acceptor TCNQ molecules form 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*-axes, respectively. Therefore, this salt is expected to have a very exotic FS at room temperature in such a way that two Q1D-FSs cross each other at right angle. One FS associated with BEDT-TTF layers is composed of parallel sheets normal to the k_a-axis, while another FS associated with TCNQ layers has parallel sheets normal to the k_c-axis.

*Corresponding author. Tel: +81-78-803-5654; Fax: +81-78-803-5770;
E-mail: kimata@phys.sci.kobe-u.ac.jp

0379-6779/$ – see front matter © 2005 Published by Elsevier B.V.
doi:10.1016/j.synthmet.2005.07.308

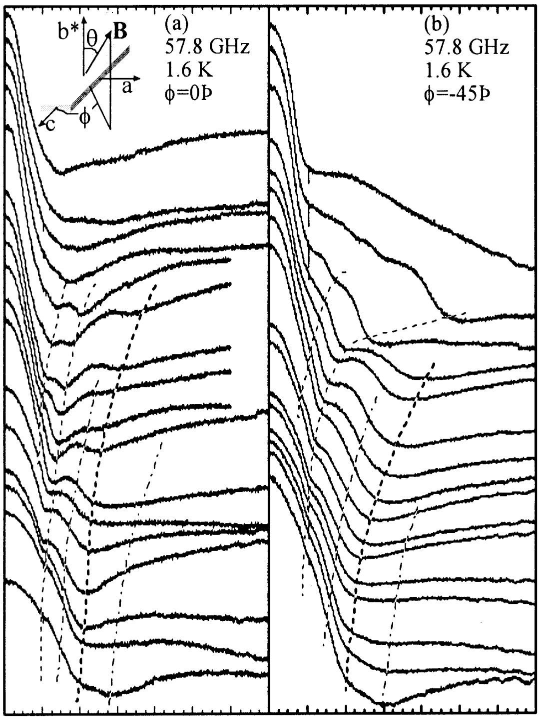

Fig.1. θ dependences of absorption lines at ϕ=0° and -45°. Broken lines are guide to the eyes. Bold lines correspond to n=1 (fundamental) resonances.

The temperature dependence of the resistivity of this salt exhibits metallic behavior down to 0.5 K [9]. In addition, three anomalies at 175 K, 80 K and 20 K were also observed [8]. The origin of these anomalies is not clear at the moment but is considered to be associated with the nesting of Q1D FS. The anisotropy of resistivities for the *a*-, *b*- and *c*-axes has a ratio of 15:1:480 at room temperature [9].

ADMRO and SdH measurements at an ambient pressure have been performed by Yasuzuka *et al.* [10]. When the magnetic field was rotated in the *b*c*-plane (i. e, parallel to Q1D BEDT-TTF FS), Lebed resonance was observed clearly, but the dip positions are not consistent with the conventional resonant conditions. On the other hand, when the magnetic field was rotated in the *b*a*-plane (i. e, parallel to Q1D TCNQ FS), Lebed resonance was not observed. These results suggest the existence of BEDT-TTF Q1D-FS and the absence of TCNQ FS. The SdH measurement suggests the existence of several small Fermi surface pockets. The areas of these pockets are a few % of the first Brillouin zone. To clarify the FS of this salt β"-(BEDT-TTF)(TCNQ), we performed a magnetooptical measurement.

2. Experimental

The experiments have been performed using cavity perturbation techniques with a millimeter vector network analyzer (MVNA) and a 14 T solenoid type superconducting magnet in the High Field Laboratory for Superconducting Materials, IMR, Tohoku University [11]. A new rotational resonant cavity system was used in this experiment [12]. This new cavity is a cylindrical type (diameter:7 mm, length:6 mm) and the cylindrical axis is perpendicular to the solenoid axis. The sample was mounted at the point of the polyethylene pillar (~2 mm), and the pillar was mounted at the center of the end plate of the cylindrical cavity. The sample has a platelike shape with dimensions of 0.7×0.4×0.1 mm^3. 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 3000~5000, respectively. The magnetic field was rotated in the b^*c-plane with angle θ and in the b^*a-plane with angle ϕ, where θ and ϕ are measured from the b^*- and c-axes, respectively.

3. Results and Discussion

Fig. 1 shows the θ dependence of absorption lines at ϕ=0° and -45°. Several resonances, which are shown by dashed lines, are observed. The observed resonances shift to the higher field as θ increases. The resonance fields are almost symmetrical for θ=0°, and independent of ϕ. There are some small resonances that look like the harmonic resonance of the major resonance (see Fig. 1).

The ADMRO measurements performed at ϕ=0° (i. e, when the magnetic field was rotated in the b^*c-plane) show the Lebed structure. This result suggests the existence of BEDT-TTF Q1D FS [10]. If we assume the existence of Q1D FS observed in ADMRO, we can calculate the model of Q1D-PORs and their θ dependences at ϕ=0° are shown in Fig. 2. Fig. 3 shows the observed result of ν/B_{res} versus θ at ϕ=0°, where ν is the frequency used in the experiment and B_{res} is the resonance field in Fig. 1. Solid triangles correspond to the data at 57.8 GHz (TE_{011}) and open circles correspond to the data at 71.9 GHz (TE_{012}). However, the θ dependence shown in Fig. 3 is completely different from that sown in Fig. 2. The θ dependence shown in Fig. 3 is typical behavior of POR in the Q2D system. In other words, the Q1D FS associated with the BEDT-TTF layer was not observed in our measurements. The resonance condition of Q2D-POR [13] can be written as

$$\frac{\nu}{B_{res}} = \frac{ne}{2\pi m^*}\cos\theta, \quad n = 1,2,3..., \qquad (1)$$

where m^* is the effective mass of the carrier. We can fit the data points in Fig. 3 using Eq. (1). The obtained effective masses are 1.8 m_e and 2.8 m_e , which are close to those