Fermiology of the Organic Superconductor β"-(ET)₂SF₅CH₂CF₂SO₃

J. Wosnitza [a], S. Wanka [a], J. Qualls [b], J.S. Brooks [b], C.H. Mielke [c], N. Harrison [c], J.A. Schlueter [d], J.M. Williams [d], P.G. Nixon [e], R.W. Winter [e], G.L. Gard [e]

[a] *Physikalisches Institut, Universität Karlsruhe, Engesserstr. 7, D-76128 Karlsruhe, Germany*
[b] *National High Magnetic Field Laboratory, Florida State University, Tallahassee, FL 32306, USA*
[c] *National High Magnetic Field Laboratory, LANL, Los Alamos, NM 87545, USA*
[d] *Chemistry and Materials Science Divisions, Argonne National Laboratory, 9700 S Cass Ave, Argonne, IL 60439, USA*
[e] *Department of Chemistry, Portland State University, Portland, OR 97207, USA*

Abstract

We present a detailed Fermi-surface (FS) investigation of the quasi two-dimensional (2D) organic superconductor (T_c = 4.5 K) β"-(ET)₂SF₅CH₂CF₂SO₃. In line with previous investigations, de Haas–van Alphen measurements in pulsed fields up to 60 T show a single oscillation frequency, F_0 = 200 T, which corresponds to a FS size of about 5% of the first Brillouin zone. Angular dependent magnetoresistance oscillations (AMROs) are utilized for the exact determination of the in-plane FS, which is found to be a strongly elongated ellipsoid with an axes ratio of about 1:9. Transport measurements in static fields up to 33 T show an unusual temperature dependence of the Shubnikov–de Haas (SdH) signal, i.e., a decrease of the SdH amplitude with decreasing temperature.

Recently β"-(ET)₂SF₅CH₂CF₂SO₃, a new organic superconductor with T_c = 4.5 K (bulk value, diamagnetic onset 5.2 K) based on the donor molecule bis(ethylenedithio)-tetrathiafulvalene (BEDT-TTF or ET for short) and a purely organic discrete anion, was synthesized [1]. Shortly after this discovery we were able to detect first Shubnikov–de Haas (SdH) [2] and later de Haas–van Alphen (dHvA) oscillations [3]. Consistently, we found one single fundamental oscillation frequency F with the expected two-dimensional (2D) angular dependence, i.e., $F = F_0/\cos\Theta$, where Θ denotes the angle between the applied field and the normal to the highly conducting ET planes. These observations are in line with extended Hückel tight-binding band-structure calculations [2] which predict a FS consisting of a small 2D hole pocket and a pair of wavy 1D bands. However, the observed FS area of A_k = 1.9 10⁻² A⁻² (corresponds to F_0 = (1991) T) is approximately a factor of three smaller than the predicted 2D band area of 14.8% of the first Brillouin zone [2]. In addition, the calculations seem to underestimate the ellipticity of the in-plane FS [2].

This work was performed in order to complement the previous results, i.e., to determine the complete in-plane FS topology by angular dependent magnetoresistance oscillations (AMROs) and to extend the field range to higher fields close to the quantum limit of the extraordinary small 2D FS. Indeed, with the available pulsed fields of up to 61 T only three Landau levels (neglecting spin splitting) are still within the FS.

We investigated different pieces of two large single crystals (3.2 mg and 2.4 mg) grown at Argonne by electrocrystallization [1]. The pulsed-field induced magnetization was measured at Los Alamos by use of a pick-up coil system (see [4]). SdH measurements up to |Θ| 74° were done at Tallahassee in static fields up to 33 T. The AMRO experiments were performed in Karlsruhe with a two-axes
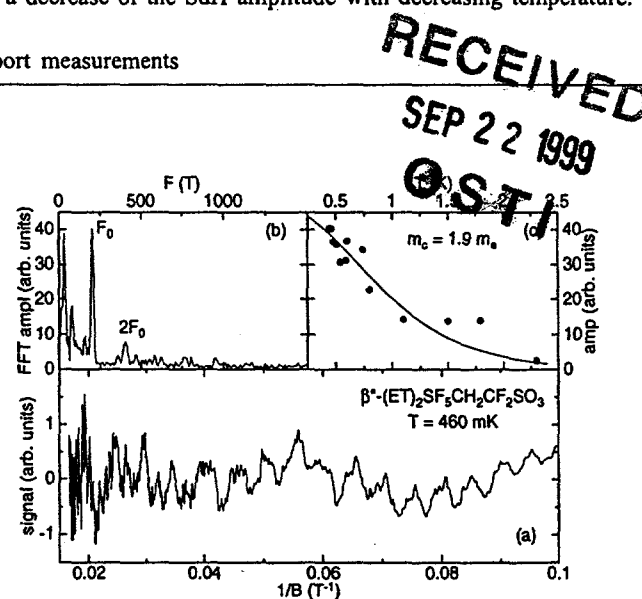

Fig. 1. (a) Raw pick-up signal (proportional to M/t) detected on the rising side of a 61 T pulse. (b) Fourier transformation of the signal with the clearly visible fundamental dHvA frequency, F_0 = 200 T, and the second harmonic $2F_0$. (c) Temperature dependence of the fundamental dHvA amplitude with Lifshitz-Kosevich fit and an effective cyclotron mass m_c = 1.9 m_e.

rotator for two different samples. All measurements were made in different ³He cryostats.

Figure 1a shows the induced voltage on the rising side of a 61 T pulse vs. reciprocal field. A dHvA signal with frequency F = (2001) T is clearly seen in the original data and in the Fourier transformation (Fig. 1b). At higher fields the signal becomes more noisy due to the decreasing sweep rate B/t close to the maximum field. In spite of the low dHvA frequency (the induced dHvA signal is proportional to F^2) the high sensitivity of the setup allowed the detection of the dHvA signal up to about 2.3 K. From the temperature dependence of the signal amplitude an effective mass of m_c = (1.90.2) m_e is

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obtained by using the 3D Lifshitz–Kosevich (LK) theory [5] (Fig. 1c). This value and all other band-structure parameters, too, are in accordance with previous low-field results [2,3].

In [2] AMRO data for only the field-rotation plane perpendicular to k_b^*, i.e., for an azimuthal angle φ 167°, were obtained. By use of a two-axes rotator AMROs for additional 16 angles φ were measured (Fig. 2). The warping of a 2D FS due to a small interlayer transfer integral t along the direction vector h leads to an angular variation of the resistivity with maxima at

$$\tan \Theta_n = [\pi(n1/4) + k^{max} u]/k_B^{max} c', \tag{1}$$

where n counts the maxima, u (0 in our case) is the in-plane component of h, c' is the spacing between adjacent layers, and k^{max} is the Fermi wave vector in the ET plane whose projection k_B^{max} to the field rotation plane is maximal [6,7]. For the upper two traces in Fig. 2 ($\varphi = 170°$ and $\varphi = 95°$) prominent AMRO peaks are visible. For $\varphi = 50°$ only the much smaller SdH oscillations can be seen; the maxima at about $|\Theta| = 75°$ just at the onset of superconductivity give only upper limits for k_B^{max}. Our analysis with (1) results in the points for k_B^{max} shown in the inset of Fig. 2. From these we can reconstruct the in-plane FS (solid line in the inset) which obviously is an extremely elongated ellipse with aspect ratio of 0.2334 Å^{-1} : 0.0264 Å^{-1}. The resulting area $A_k = 1.93610^{-2}$ Å^{-2} is in very good agreement with 1.910^{-2} Å^{-2} extracted directly from F. The warping t itself must be very small since no nodes in the SdH signals can be observed.
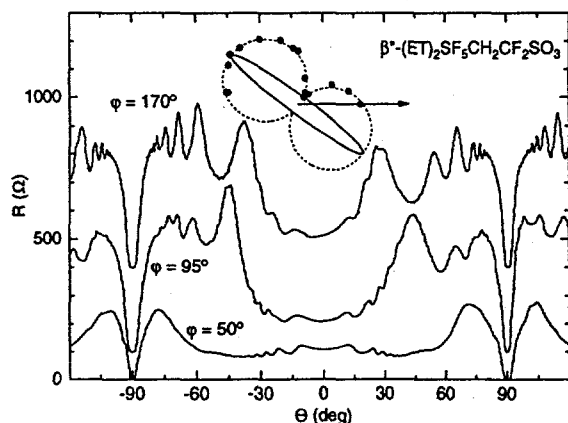

Fig. 2. Angular dependence of the resistance for selected azimuthal angles φ at $B = 10$ T and $T = 0.5$ K. The upper two curves are offset for clarity. The inset shows the extracted in-plane dependence of k_B^{max}. The thick solid line depicts the resulting in-plane FS.

The high degree of two dimensionality and the small FS, i.e., the closeness to the quantum limit already for the used magnetic fields, leads to some remarkable transport properties for currents perpendicular to the ET planes: first, as shown in Fig. 3 the background magnetoresistance grows enormously (by a factor of about 20 from 5 T to 33 T at 0.55 K) with an approximately quadratic field dependence. This effectively leads to an insulating behavior of the longitudinal resistivity at high fields. Second, the SdH signal deviates from the

predicted LK behavior. The analysis of SdH oscillations in terms of usual theories is valid only for the relative conductance oscillations, i.e., $\Delta\sigma/\sigma = 1 - \sigma/\sigma_{steady}$. We fitted the steady part of the conductance, σ_{steady}, by a polynomial. By simple inverting the resistivity we neglected the Hall resistivity which, however, should be zero for our geometry. The resulting SdH signal (inset of Fig. 3) shows a highly unconventional temperature dependence, i.e., the SdH amplitude decreases with decreasing temperature for the highest fields and below about 1 K. A similar effect has been observed for κ-(ET)$_2$I$_3$ (see [4] and references therein) and seems to be related to the 2D nature of the FS. Further transport and magnetization studies at this 30 T field range should help to elucidate these discrepancies to conventional theories.
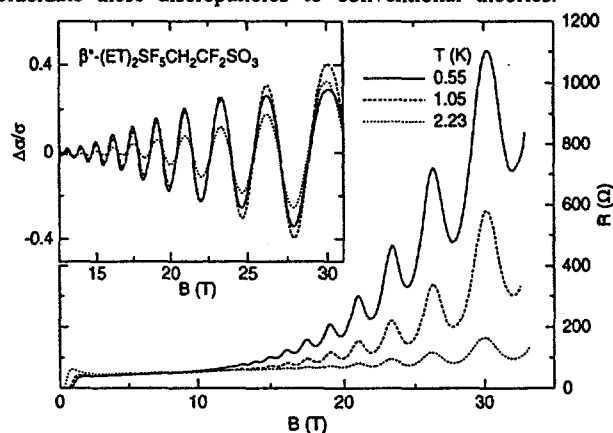

Fig. 3. Field dependence of the resistivity up to 33 T for three different temperatures. The inset shows the SdH signal, i.e., the relative conductance oscillations, which at high fields are decreasing in amplitude with decreasing T.

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