

On the Fermi surface of the organic conductor β -(BEDT-TTF) $_2$ I $_3$

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Abstract

Our recent experiments on the Shubnikov-de Haas(SdH) effect in the high T_c phase of β -(BEDT-TTF) $_2$ I $_3$ are reviewed. The founding of extraordinarily large amplitude magnetoresistance oscillations with the frequency of 3730T suggests that the Fermi surface consists of a single cylinder with only a very small warping ($t_a/t_c \simeq 140$).

Recently, very much interest has been concentrated on the study of the Fermi surface of quasi-two dimensional organic conductors and superconductors, (BEDT-TTF) $_2$ X where X is a monovalent anion. The main method has been the measurement of the SdH oscillations and the salient results are concerned with the β -phase salts $X = I_3[1,2]$ and $IBr_2[3,4,5]$, and with the κ -phase salt $Cu(SCN)_2$. [6,7] However, there are still large discrepancies among results from different works, in particular for the last two compounds. For instance, frequencies and amplitudes differ between authors. Although the latter can be attributed to the sample quality, the former, which represents the intrinsic nature of the material, should be the same. For β - IBr_2 , rapid oscillations were reported in addition to the slow oscillations originally reported. [5] Since only very slow oscillations are found in the isostructural compound β - $I_3[1]$ when it is normally cooled, we think that the absence of the rapid oscillations is probably related to the existence of the incommensurably modulated structure. Therefore, it was particularly interesting to try and perform the same experiment in the pressure cycled sample (β_H -phase). We present here some experimental data obtained independently in two samples.

Throughout the experiments, a magnetic field up to 12T was applied normal to the sample plate and current was flowing along the highest conductivity direction. Fig. 1 shows the overall magnetoresistance between 0 and 12T at 380mK. The black envelopes are due to the very rapid oscillations. The origin of the difference between two samples is not clear but can be considered to come from the sample difference and/or from slight misorientation. Even a slight angle difference can affect the data for this nearly

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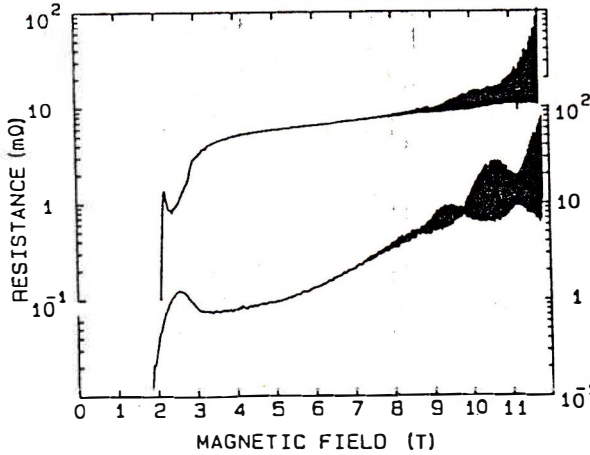


Fig. 1. Magnetoconductance of two samples measured independently at 380mK in logarithmic scale. ($H \perp ab$ -plane, $I \parallel a$)

perfect two-dimensional system. The detailed behavior of magnetoconductance for sample 2 is presented in Fig. 2 where the rapidly oscillating magnetoconductance and the envelopes are clearly visible. In the inset, the positions of the last 133 oscillations are traced as a function of reciprocal field which provides the fundamental field from its slope and also guarantees the quantum origin from its perfect linearity. We deduce 3730T as a fundamental field which corresponds to $3.56 \times 10^{15} \text{cm}^{-2}$ of cross sectional area of Fermi surface or 51.3% of the first Brillouin zone (FBZ; $S_{\text{BZ}} = 6.99 \times 10^{15} \text{cm}^{-2}$). Besides rapid oscillations, Fig. 2 shows also a peculiar beat behavior. From Fig. 1 and Fig. 2 we have three complete oscillation envelopes of frequency 36.8T which will be used to derive the cyclotron mass of the conduction electrons. The evolution of the oscillatory magnetoconductance as a function of temperature is presented in Fig. 3 for sample 1. The oscillations begin

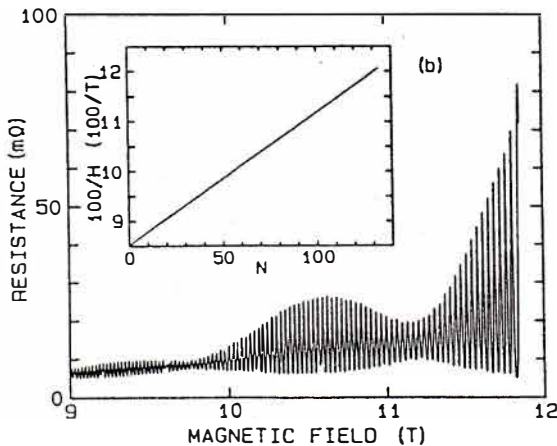


Fig. 2. Magnetoconductance of sample 2 at high field. Inset shows the reciprocal field of each maximum versus integral numbers.

to be visible at high field below 1.2K and evolve slowly down to 0.7K, At further cooling, the amplitude increases extremely fast. Supposing that the three-dimensional formula for oscillatory resistance, which works reasonably well except at very low temperature, is applicable in this case, we derive the effective mass of $m_c \simeq 3.7m_e$. As already said the field dependence of the oscillation especially that of the envelope maxima gives the Dingle temperature of 0.53K.

One of the most remarkable fact in our observation is the development of the anharmonic oscillations at low temperature and high field as shown in Fig. 4. It is extremely sensitive to the temperature. It can be qualitatively understood with the quasi-two dimensional nature of energy bands. However, at least with the experimentally observed parameters, there is no full understanding for the origin of this strong anharmonicity.

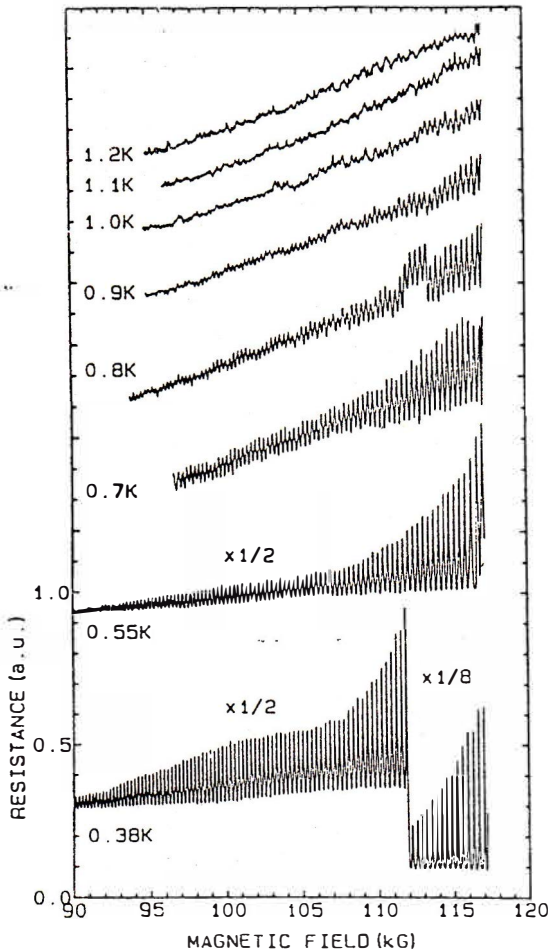


Fig. 3. Temperature dependence of the magnetoresistance oscillations in sample 2. For 0.55K and 0.38K, the amplitudes are scaled by the factors indicated above the curves.

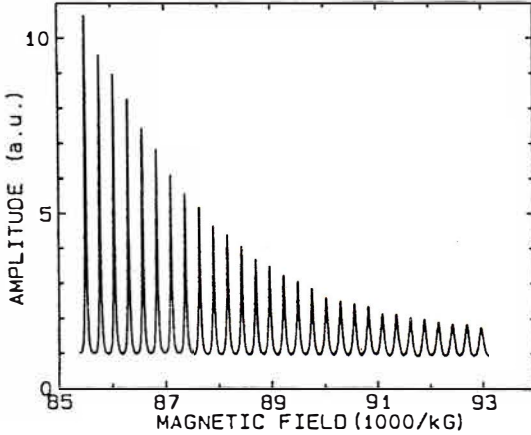


Fig. 4. High field behavior of the magnetoresistance in the function of the reciprocal field. The anharmonicity and the periodicity are clearly visible.

From our observation, we derive two frequencies, 3730T and 36.8T with which we develop a slightly warped cylindrical Fermi surface. We obtain $t_a/t_c \simeq 144$, which means an almost perfect two dimensional system. It is probably this enhanced two-dimensionality which is the cause of the giant amplitude and of the big anharmonicity.

Our experimentally derived Fermi surface structure is in agreement with the band calculations by several authors. Mori et al.[8] and Whangbo et al.[9] reported that it consists of an elongated cylinder. In the recently reported band structure calculation, Kübler and Sommers[10] have derived the closed orbit slightly less than half the Brillouin zone area, but they also found existence of the branches connected in the repeated zone scheme, which are hard to reconcile with the actual observation.

In β_L -phase, we have found neither slow nor fast oscillations. Admittedly, our noise condition was not satisfactory enough to observe the 0.4% oscillations reported in ref. [1]. But, in one pressure cycled sample we have found the oscillations of the same frequency as for the β_H -phase, but with a much smaller amplitude, which probably have the same origin as the resistance drop around 7K as frequently observed in the thermally cycled samples.

In addition to present data, experiments in a tilted magnetic field will be useful to determine the reliability of the band structure calculation. Also the full understanding of the difference between the β_L and β_H phase will be helpful to understand the five-fold difference in T_c .

In conclusion, we have reported an observation of SdH oscillations with a giant amplitude and a large anharmonicity found in β_H - $(\text{ET})_2\text{I}_3$ and we have concluded that it is an almost complete two-dimensional system with the ratio of the transfer integral $t_a/t_c \simeq 144$.

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