Magnetoresistance of the organic metal β_L-$(ET)_2I_3$: angular dependence and Shubnikov–de Haas oscillations

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The angular and field dependences of the magnetoresistance of a single crystal of the organic metal β_L-$(ET)_2I_3$ have been studied at $T = 1.45$ K in fields up to 150 kOe. Shubnikov–de Haas oscillations with a frequency of about 1 MG have been observed at field directions close to $H \| c^*$.

At temperatures below 120 K and at standard pressure the organic metal β-triiodide bis(ethylenedithio)-tetrathiofulvalene [β-$(ET)_2I_3$] can be in two different states[1]: in a metastable phase β_L with a superconducting transition temperature $T_c = 1.5$ K and in a phase β_H with $T_c = 8$ K, which can be reached if the crystal is cooled under a pressure $p \gtrsim 400$ to $T \lesssim 125$ K. Judging from measurements of the magnetic susceptibility—which, incidentally, are not totally invulnerable to criticism—the electronic properties of β-$(ET)_2I_3$ change only slightly in the transition from the state β_L to the state β_H (Refs. 2 and 3). The only difference between the crystal lattices is that the β_L phase has an incommensurate superstructure, which is not present in β_H (Refs. 4 and 5). In light of these results, the reason for the fivefold increase in T_c in the transition from β_L to β_H is not completely clear.

An analog of the β_H-$(ET)_2I_3$ phase is the isostructural compound β-$(ET)_2IBr_2$, whose lattice has no superstructural distortions and whose superconducting transition temperature is 2.8 K. Shubnikov oscillations[6,7] and the angular dependence of the magnetoresistance[7] have recently been studied in fairly high-quality crystals of β-$(ET)_2IBr_2$ over a wide range of angles and in various crystallographic planes. These studies have furnished some ideas about the Fermi surface of this compound. In this connection, it is interesting to carry out some similar measurements of single crystals of β_L-$(ET)_2I_3$ in order to identify any possible differences.[2)]

The measurements were carried out in the International Laboratory of Strong Magnetic Fields and Low Temperatures (Wrocław, Poland) in fields up to 150 kOe at a temperature of 1.45 K. A β-$(ET)_2I_3$ single crystal with dimensions of $1.5 \times 0.05 \times 0.02$ mm was studied. The measurement current, 1 mA, was directed along the a axis in all cases.

In fields $H \gtrsim 100$ kOe, for all directions of H, we see a tendency toward a saturation of the magnetoresistance. The angular dependence measured for the resistance at $T = 1.45$ K in a field $H = 150$ kOe lying in the (ac^*), $(b'c^*)$, and (ab) planes (the b' direction is perpendicular to a and c^*) is shown in Figs. 1 and 2. The anisotropy of the relative magnetoresistance does not exceed 2 in any of the cases shown here. In the case of β-$(ET)_2IBr_2$, on the other hand, this anisotropy is an order of magnitude greater[7] in the (ac^*) plane. In β_L-$(ET)_2I_3$ we do not see the angular oscillations of the

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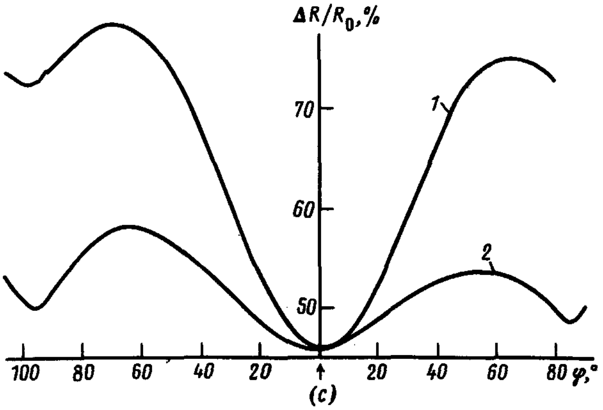

FIG. 1. 1—Angular dependence of the relative magnetoresistance in the (ac^*) plane; 2—in the ($b'c^*$) plane. Here φ is the angle between the field H and the direction corresponding to the minimum of the magnetoresistance. $H = 150$ kOe, $T = 1.45$ K.

resistance which have been observed in β-$(ET)_2IBr_3$ both in the (ac^*) plane[7] and in the ($b'c^*$) plane. The local minima of the magnetoresistance in the (ac^*) and ($b'c^*$) planes in β_L-$(ET)_2I_3$ correspond to the directions $H \| a$ and $H \| b'$, respectively. The absolute minimum arises near the direction $H \| c^*$. The asymmetry of the angular dependence of the resistance with respect to this absolute minimum is a consequence of the triclinic structure of the unit cell of the β-$(ET)_2I_3$ system.[4] In the (ab) plane the minimum of the magnetoresistance corresponds to the direction $H \| b'$, while the maximum occurs $\approx 26°$ from the $H \| a$ direction and apparently corresponds to the direction perpendicular to the ET stacks. In this plane the angular dependence of the resistance is qualitatively the same as in β-$(ET)_2IBr_2$.

At magnetic field directions near $H \| c^*$, oscillations are observed in the resistance which are periodic in the reciprocal field and which are close in frequency to the slow oscillations in β-$(ET)_2IBr_2$ (Ref. 6). Figure 3 shows an example of these oscillations,

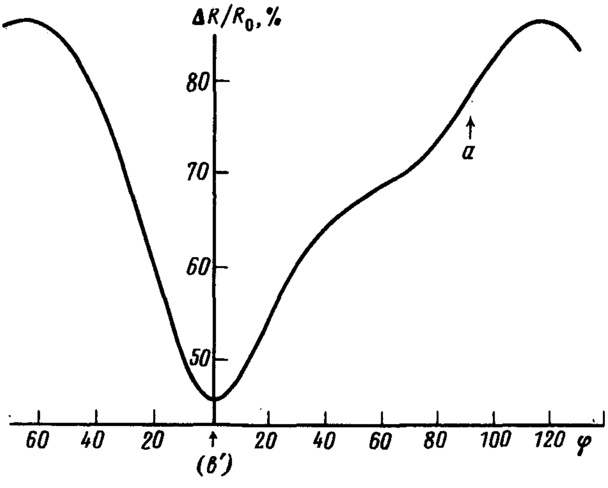

FIG. 2. Relative magnetoresistance versus the angle (φ) between the field H, which lies in the (ab) plane, and the b' direction. $H = 150$ kOe, $T = 1.45$ K.

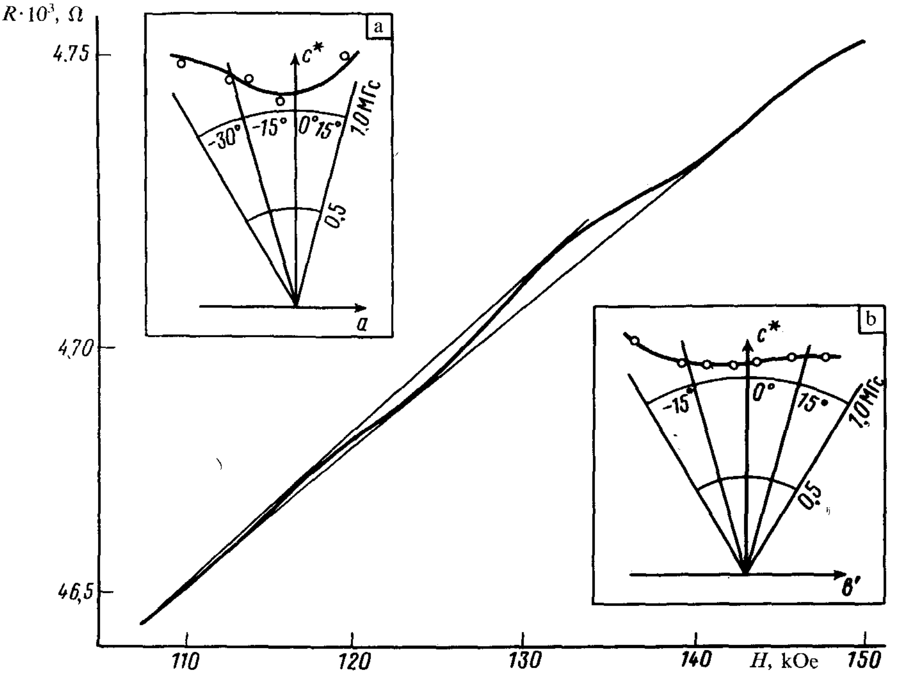

FIG. 3. Oscillations in the resistance for a magnetic field direction close to $H||c^*$. $T = 1.45$ K. The insets show the behavior of the oscillations frequency as the field H is turned away from the direction $H||c^*$, in polar coordinates.

for a direction close to $H||c^*$. The maximum amplitude of these oscillations corresponds to the direction $H||c^*$; at $H = 150$ kOe this maximum amplitude is about 0.2% of the resistance of the sample at $H = 0$. The amplitude falls off rapidly with a deviation from this direction.

The frequency of the oscillations in the case $H||c^*$ is 1.1 MG. The frequency increases with a deviation from this direction (Fig. 3). An estimate of the area of the extremal cross section of the Fermi surface yields ~1% of the cross-sectional area of the Brillouin zone. Consequently, as in[6] β-$(ET)_2IBr_2$, the slow oscillations apparently correspond to extremal cross sections of thin "necks" of the Fermi surface which are oriented along the c^* axis. The cross-sectional area of these necks in β_L-$(ET)_2I_3$, however, is twice that in β-$(ET)_2IBr_2$.

The fast oscillations which are observed in β-$(ET)_2IBr_2$ and which correspond to large extremal cross sections (up to 50% of the cross section of the Brillouin zone) are not seen in β_L-$(ET)_2I_3$. We do not rule out the possibility that the absence of these oscillations, like the absence from the angular dependence of the resistance in β_L-$(ET)_2I_3$ of the oscillations which have been observed[7] in β-$(ET)_2IBr_2$, is a consequence of these compounds. However, a more likely explanation is that the β-$(ET)_2I_3$ crystals are of lower quality than the β-$(ET)_2IBr_2$ crystals. In β-$(ET)_2IBr_2$ we have a ratio $\rho(300\text{ K})/\rho(4.2\text{ K}) = (1.5\text{–}3.0)\times 10^3$, while in β_L-$(ET)_2I_3$ this ratio does not exceed 500–600. The implication is that in order to reach given values of $\omega\tau$ in β_L-$(\mathrm{ET})_2\mathrm{I}_3$, we need fields stronger by a factor of three to six than those required in β-$(\mathrm{ET})_2\mathrm{IBr}_2$. According to the estimates in Refs. 6 and 7, the fast oscillations correspond to electrons with an effective mass $\sim 5m_e$, and the slow oscillations correspond to electrons with $\sim 0.4m_e$. As the field is lowered, the oscillations associated with electrons of the greater mass would usually be damped more rapidly. On the other hand, the oscillations due to the "light" electrons in the narrow necks of the Fermi surface persist down to lower fields. Consequently, if there is no possibility in principle for improving the quality of the β-$(\mathrm{ET})_2\mathrm{I}_3$ crystals (the chances are slim since these crystals contain, in addition to removable impurities, a disorder which is unremovable at standard pressure and which stems from the incommensurate superstructure), the only way to compare the Fermi surfaces in β-$(\mathrm{ET})_2\mathrm{IBr}_2$ and β_L-$(\mathrm{ET})_2\mathrm{I}_3$ would be to substantially strengthen the magnetic fields in a study of the triiodide. A precise answer to the question of whch changes in the Fermi surface accompany the $\beta_L \rightarrow \beta_H$ transition can be found only studying the Fermi surface in the β_L and β_H phases in a common β-$(\mathrm{ET})_2\mathrm{I}_3$ crystal. Such a study is planned for the near future.

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[2)] Shubnikov oscillations in β-$(\mathrm{ET})_2\mathrm{IBr}_2$ and β_L-$(\mathrm{ET})_2\mathrm{I}_3$ were observed in the orientation $H \| c^*$ in Refs. 8 and 9 also; the results of those studies differ substantially from the results of Refs. 6 and 7.

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