Transverse magnetoresistance and Shubnikov–de Haas oscillations in the organic superconductor β-$(ET)_2IBr_2$

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The transverse magnetoresistance of β-$(ET)_2IBr_2$ single crystals has been studied in fields up to 150 kOe in the orientations $H \| c^*$ and $H \| b'$. Shubnikov–de Haas oscillations have been observed at $T \lesssim 4.2$ K and $H_{\| c^*} \gtrsim 80$ kOe.

Of particular interest among the conducting cation-radical salts of bis(ethylenedithio) tetrathiofulvalene (ET) is the family of compounds $(ET)_2X$, where $X = I_3^-$, IBi_2^-, AuI_2^-, or $Cu(SCN)_2^-$. These compounds are superconductors at atmospheric pressure with superconducting transition temperatures $T_c \approx 1.5$ K and 7.5 K (Refs. 1 and 2), 2–2.7 K (Refs. 3 and 4), 4–5 K (Ref. 5), and 10 K (Ref. 6), respectively. In order to reach an understanding of the superconductivity mechanism in organic metals, it is extremely important to study not only the superconducting characteristics but also the electronic band structure, in particular, the Fermi surface. At the moment, however, there are essentially no experimental data on the Fermi surface of superconductors of this family.

Most of the band-structure calculations have been carried out in the strong-coupling approximation and without consideration of the conductivity across the ET molecular layers, i.e., along the c^* axis.[7] In this case the Fermi surface is represented as a cylinder. Recently published results of calculations of the band structure of β-$(ET)_2I_3$ by the method of a local-density functional[8] indicate that the strong-coupling approximation is not applicable; they point to a more complex structure of the Fermi surface. In particular, these calculations predict the presence of a multiply connected sheet of the Fermi surface with cylindrical necks along the c^* axis and some small electron pockets at the center of the Brillouin zone.

In the present letter we report a study of the transverse magnetoresistance of β-$(ET)_2IBr_2$ single crystals, which have the same structure as the compound β-$(ET)_2I_3$. We also report the observation of Shubnikov-de Haas oscillations in them.[2)]

The measurements were carried out over the temperature interval 1.4–12 K in magnetic fields up to 150 kOe. The sample was oriented in two ways with respect to the field direction: $H \| c^*$ and $H \| b'$ (b' is the direction in the ab plane which is perpendicular to the a axis of the crystal). The error in this orientation was $\pm 5°$. The transverse magnetoresistance was measured along the a axis by a four-contact method with either a direct current or an alternating current $I = 1$ mA. The sample was cemented to platinum electrodes 20 μm in diameter by a conducting graphite paste. For the ac measurements, gold contact strips were first deposited on the sample. The resistance of the contacts was ~ 10–20 Ω. We took measurements from several crystals

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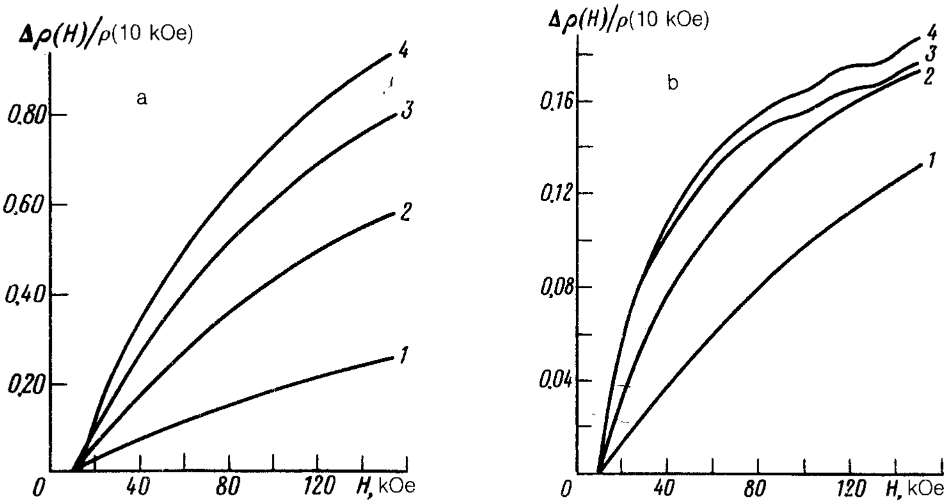

FIG. 1. The relative magnetoresistance $\Delta\rho(H)/\rho(10 \text{ kOe}) = [\rho(H) - \rho(10\text{kOe})]/\rho(10\text{kOe})$ as a function of the field for sample 1 at several temperatures: 1) 12.2; 2) 6.2; 3) 3.2; 4) 1.45 K. a—$H \| b'$; b—$H \| c^*$. Since the sample goes superconducting at $T < 2.5$ K, the value of $\rho(10 \text{ kOe})$ at $T = 1.45$ K was found by extrapolating $\rho(T)$ at $H = 10$ kOe.

of fairly high quality: sample 1, with a ratio $\rho(300\text{K})/\rho(4.2\text{K}) \approx 2000$, and samples 2 and 3, with $\rho(300\text{K})/\rho(4.2\text{K}) \approx 1000$.

Figure 1 shows the results of measurements of the relative magnetoresistance $\Delta\rho(H)/\rho(10 \text{ kOe}) = \rho(\text{H}) - \rho(10 \text{ kOe})/\rho(10 \text{ kOe})$ of sample 1 (the minimum field $H \approx 10$ kOe was a consequence of the remanent field of the superconducting solenoid). The field dependence of the magnetoresistance at essentially all temperatures has a negative curvature, which is reminiscent of a tendency toward saturation. The quadratic $\rho(H)$ dependence characteristic of weak magnetic fields is not observed in fields $H > 10$ kOe even at $T \approx 12$ K. A comparison of the values of $\Delta\rho_{\|}(H)$ and $\Delta\rho_{\perp}(H)$ in fields directed along b' and c^*, respectively, at $T = 1.4$ K yields $[\Delta\rho_{\|}(150 \text{ kOe})/\Delta\rho_{\perp}(150 \text{ kOe})] \approx 5$. The reason for this anisotropy is not clear at this point: It would seem that the quasi-2D nature of the structure of this compound would lead to the opposite relation, $[\Delta\rho_{\|}(H)/\Delta\rho_{\perp}(H)] < 1$.

A tendency toward saturation on the $\Delta\rho(H)$ curve for samples 2 and 3 is observed only in a field $H \| c^*$ at $T \lesssim 4.2$ K. At $T > 4.2$ K for $H \| c^*$ and $H \| b'$, we observe a quadratic increase in the resistance, $\rho(H) \sim H^2$, at $H \lesssim 40$ kOe. In stronger fields we find $\rho(H) \sim H$. The anisotropy in the magnetoresistance remains the same in nature, but it is smaller in magnitude than in sample 1.

These results suggest that at $T < 12$ K and $H > 40$ kOe the condition $\omega_H \tau \ll 1$ does not hold for any of the samples studied; i.e., the magnetic field is not weak. Furthermore, at $T < 4.2$ K and $H \gtrsim 100$ kOe the relation $\omega_H \tau > 1$ probably holds. From the behavior $\Delta\rho(\text{H})$ we conclude that the highest values of $\omega_H \tau$ are reached for sample 1 in the orientation $H \| c^*$ (Fig. 1b). In this case the tendency toward the saturation of

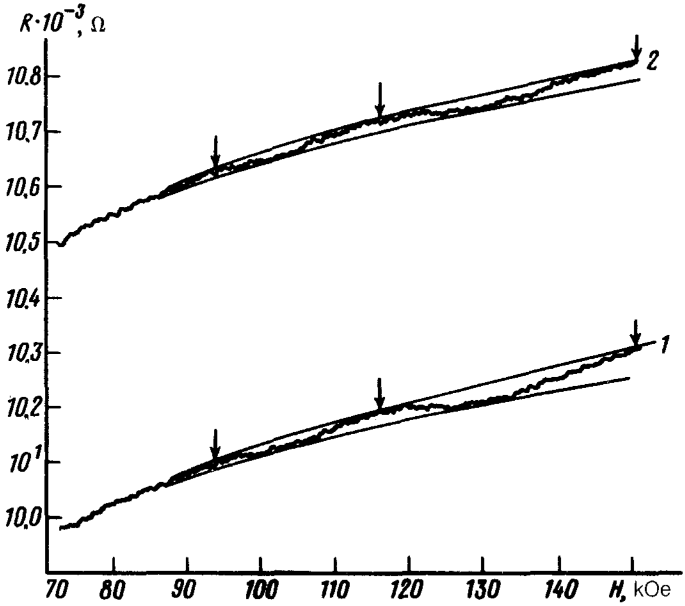

FIG. 2. Oscillations in the magnetoresistance in a field H||c^* for sample 1 at two temperatures: 1—1.45 K; 2—2.90 K.

$\Delta\rho(H)$ is seen most clearly, and oscillations appear at $T \lesssim 4.2$ K and $H \gtrsim 80$ kOe (Figs. 1b and 2).

These oscillations are periodic in the reciprocal field; their phase does not depend on the temperature; and their amplitude increases with decreasing temperature. These aspects of the oscillations support the assertion that the observed deviations from a monotonic behavior are Shubnikov-de Haas oscillations. From the period. $\Delta(1/H) \approx 2\times10^{-6}$ Oe^{-1}, we can estimate the area of the extremal cross section of the Fermi surface, which contributes to the oscillations: $S \approx 2\pi e\hbar/c\Delta(1/H) \approx 5\times10^{13}$ cm^{-2}, or about 1% of the cross-sectional area of the Brillouin zone. An estimate of the effective mass at the extremal cross section from the temperature dependence of the amplitude yields $m^* \approx (0.5 \pm 0.1)m_0$ where m_0 is the mass of the free electron. Working from the field dependence of the oscillation amplitude, we can estimate the Dingle temperature: $T_D \approx 2.2$ K. This temperature corresponds to a relaxation time $\tau_D \approx 3.2\times10^{-12}$ s and to $\omega_H\tau_D \approx 10.2$ at $H = 100$ kOe and $T = 4.2$ K.

The observation of Shubnikov-de Haas oscillations with $H \| c^*$ indicates the existence of closed orbits in the intersections of the Fermi surface with the $K_c = \mathrm{const}$ plane. At first glance, this conclusion would seem to agree with the representation of the Fermi surface as a cylinder.[7] In that case, however, the corresponding cross-sectional area would be $\sim 50\%$ of the area of the Brillouin zone and substantially greater than the area of the cross section responsible for the observed oscillations. In order to reconcile these results, we might represent the Fermi surface as a highly corrugated cylinder, but then the Fermi surface would obviously lose its 2D nature.

The existence of necks and small pockets in the Fermi surface of the complex β-$(ET)_2IBr_2$, like those predicted by Kübler *et al.*[8] for the isostructural compound β-$(ET)_2I_3$, could explain the origin of the observed oscillations. The calculations by Kubler *et al.*[8] predict the formation of open trajectories in the *ab* plane; correspondingly, there should be a quadratic contribution to the magnetoresistance in the configuration $H \| c^*$. Experimentally, on the other hand, we observe a $\Delta\rho(H)$ dependence (Fig. 1) with a clearly expressed tendency toward saturation, which apparently means that open trajectories make only a small contribution to the magnetoresistance of the β-$(ET)_2IBr_2$ crystals.

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