Shubnikov–de Haas oscillations in the new organic metal $(ET)_2TlHg(SCN)_4$

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The Shubnikov–de Haas oscillations have been studied in the new, quasi-2D organic metal $(ET)_2TlHg(SCN)_4$ at temperatures to 1.3 K in fields up to 14 T. The field dependence and the temperature dependence of the amplitude of the Shubnikov oscillations are used to estimate the cyclotron mass and the Dingle temperature in the conducting plane: $m^* \approx 1.4m_0$ and $T_D \approx 2.7$ K. The angular distributions of the oscillation frequency and amplitude support the picture of a closed sheet of the Fermi surface in $(ET)_2TlHg(SCN)_4$ in the form of a slightly corrugated cylinder with an axis perpendicular to the conducting layers.

The isostructural quasi-2D organic metals $(ET)_2MHg(SCN)_4$ (M = k, NH_4, Rb, Tl) with polymer metal-complex anions have recently attracted considerable interest.[1,2] This interest originally arose because the compound $(ET)_2Cu(NCS)_2$, with a superconducting transition temperature $T_c = 10.5$ K—until recently the record high for organic superconductors—was discovered among the salts of ET with polymer metal-complex anions.[3] In the $(ET)_2MHg(SCN)_4$ series, however, only the salt with M = NH_4 turned out to be a superconductor, with a transition temperature[4] $T_c = 0.8$ K. Further research on these compounds focused on several unusual properties, not directly related to the superconductivity. These unusual properties were seen primarily in the behavior of the magnetoresistance.[2,5,6] They are determined by a complex Fermi surface which, according to theoretical predictions,[1] consists of a closed cylinder with axis along $\vec{K}_b$ and two slightly corrugated planes along the $\vec{K}_c\vec{K}_b$ ($\vec{K}_a, \vec{K}_b, \vec{K}_b$ plane are reciprocal-lattice vectors). This circumstance apparently gives rise to both Shubnikov–de Haas oscillations[2,5] and a partial insulating transition at $T_0 \approx 10$ K (Refs. 2 and 6). The metals of this series thus turned out to have a unique combination of quasi-2D and quasi-1D properties.

In this letter we report a study of the Shubnikov oscillations in the organic metal $(ET)_2TlHg(SCN)_4$, which was recently synthesized.

The resistance was measured by the standard ac four-contact method in magnetic fields up to 14 T at temperatures to 1.3 K. The single-crystal $(ET)_2TlHg(SCN)_4$ samples had typical dimensions of $1.0 \times 0.5 \times 0.05$ mm. The Shubnikov–de Haas oscillations were observed when the resistance was measured in the $\vec{a}\vec{c}$ plane, of the conducting layers, and also when it was measured in the $\vec{b}^*$ direction, which is perpendicular to these layers. In most of the measurements the current flowed along the long axis of the crystal, which lay in the $\vec{a}\vec{c}$ high-conductivity plane.

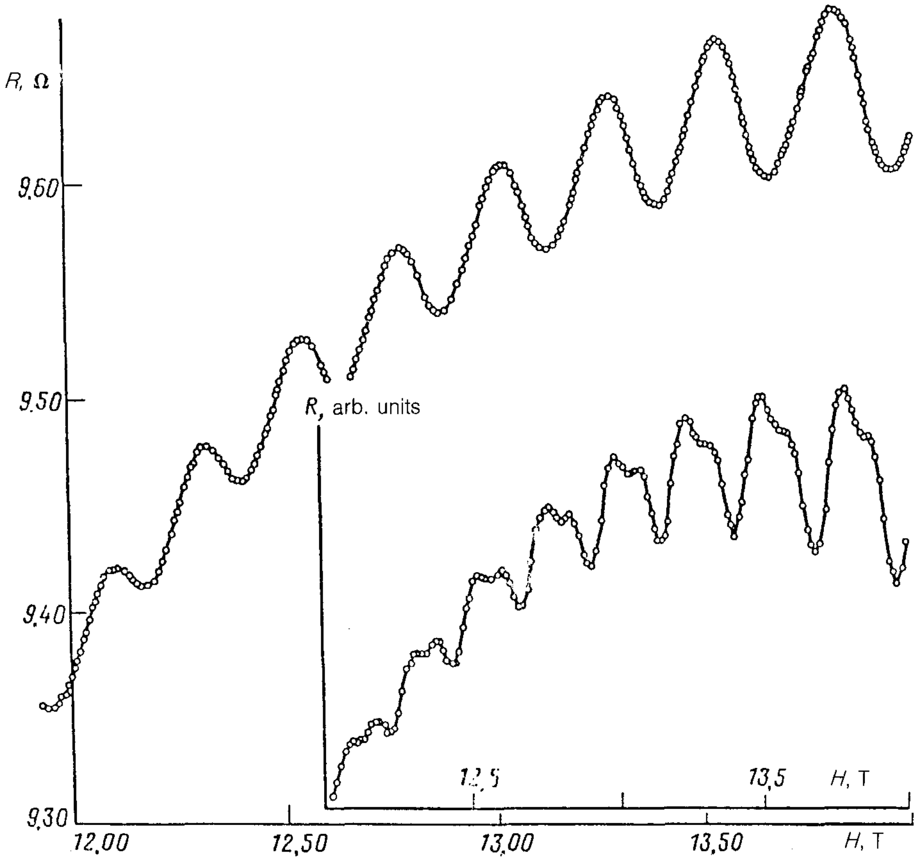

FIG. 1. Shubnikov–de Haas oscillations for the field direction $H\|\vec{b}^*$ at $T=1.3$ K. The inset shows the Shubnikov oscillations for the same field direction and for the same temperature, with a large second-harmonic component.

Figure 1 shows the Shubnikov–de Haas oscillations in the configuration $H\|\vec{b}^*$ at $T=1.3$ K. The frequency of these oscillations for this field direction is 6.7 MHz, which corresponds to an area of 6.4×10^{14} cm^{-2} for the extremal cross section. This area amounts to 16% of the cross-sectional area of the Brillouin zone. The value found is close to the area of the extremal cross section of the Fermi surface in the salt $(ET)_2KHg(SCN)_4$, as determined from the frequency of Shubnikov–de Haas oscillations.[5] It agrees fairly well with the prediction of band-theory calculations.[1] Measurements of the field dependence and the temperature dependence of the amplitude of the Shubnikov oscillations yield estimates of the cyclotron mass and the Dingle temperature: $m^*\approx1.4m_0$ and $T_D\approx2.7$ K. The density of states at the Fermi level can be estimated by approximating this sheet of the Fermi surface by a smooth cylinder: $N(\epsilon_F)\approx2b^*m/(2\pi\hbar)^2\approx2.2\times10^{33}$ erg$^{-1}\cdot$cm^{-3}. Assuming that the dispersion is parabolic, we can estimate the distance from the top of the band to ϵ_F: $\Delta\epsilon\approx50$ meV.

Certain $(ET)_2TlHg(SCN)_4$ crystals in fields with the approximate orientation

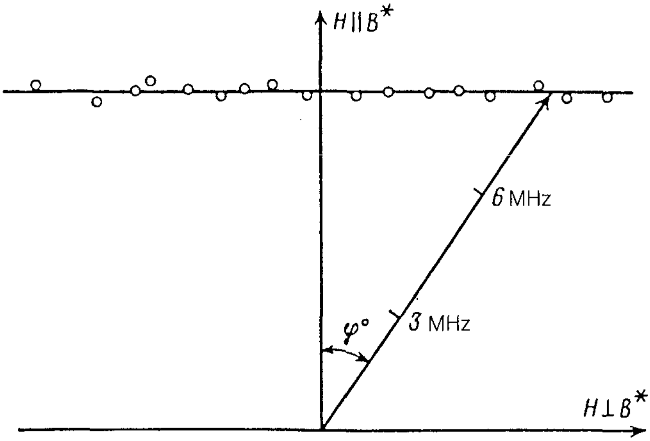

FIG. 2. Angular distribution of the frequency of the Shubnikov oscillations in the $\vec{I}\vec{b}^*$ plane, in polar coordinates.

$H\|\vec{b}^*$ exhibit a large second-harmonic component $F^{(2)} = 13.4$ MHz (see the inset in Fig. 1). A similar effect has been observed in $(ET)_2KHg(SCN)_4$ in strong fields. It has been interpreted as a spin splitting.[7] Since fields $H \sim 14$ T are far from the quantum limit, and since there are no higher-order harmonics in our measurements, that interpretation does not look very convincing to us. The reason for the appearance of the second harmonic thus requires a more careful study.

Figure 2 shows the frequency of the Shubnikov–de Haas oscillations as a function of the angle φ as the field is rotated in the plane $\vec{I}\vec{b}^*$ ($\vec{I}$ is the direction of the current,

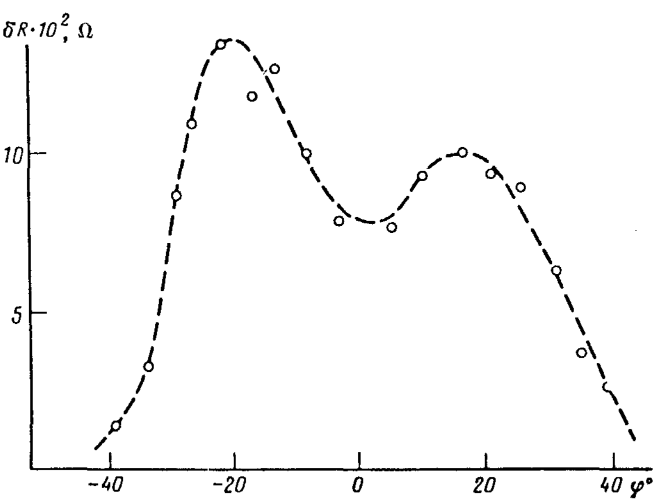

FIG. 3. Angular distribution of the amplitude of the Shubnikov oscillations in the $\vec{I}\vec{b}^*$ plane in a field $H = 14$ T at a temperature $T = 1.3$ K.

which lies in the $\vec{a}\vec{c}$ plane). This distribution can be described well by $F = F_0/\cos \varphi$. This distribution determines a smooth or slightly corrugated cylinder with an axis along $\vec{b}^*$. One might estimate the extent of the corrugation from the beats in the amplitude of the Shubnikov–de Haas oscillations, as has been done[8] for the organic metal $(ET)_2IBr_2$. In the thallium complex, however, no beats were observed in the amplitude of the Shubnikov oscillations in the angular interval studied. In $(ET)_2IBr_2$, a strong dependence of the amplitude of the Shubnikov–de Haas oscillations is also linked with a corrugation of a cylindrical sheet of the Fermi surface.[8] A qualitatively similar behavior is observed in $(ET)_2TlHg(SCN)_4$ (Fig. 3). Quantitatively, on the other hand, the changes in the amplitude in this metal are negligible in comparison with those in $(ET)_2IBr_2$. Consequently, it is quite probable that the deviation of the closed sheet of the Fermi surface from a smooth cylinder in $(ET)_2TlHg(SCN)_4$ is extremely slight.

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