The Fermi surface in the organic superconductor k-$(ET)_2Cu(NCS)_2$

M. V. Kartsovnik,[1)] P. A. Konovich,[1)] V. N. Laukhin, R. B. Lyubovskii, and S. I. Pesotskii

N. N. Semenov Chemical Physics Institute, USSR Academy of Sciences, Chernogolovka

(Submitted 20 March 1990)
Zh. Eksp. Teor. Fiz. **98**, 708–711 (August 1990)

The behavior of magnetoresistance in single crystals of the organic superconductor k-$(ET)_2Cu(NCS)_2$ has been studied in liquid helium temperatures in fields up to 150 kOe. Shubnikov–de Haas oscillations are observed when the field **H** makes an angle $\varphi = \pm 50°$ with the direction **a*** perpendicular to the plane **bc**. The angular dependence of the magnetoresistance and oscillation frequency indicates that the Fermi surface has an almost smooth cylindrical shape.

The synthesis of high-quality single crystals of organic quasi-2-D superconductors of the system ET [bis(ethylenedithio)tetrathiofulvalene] has lately simulated publication of a number of studies dealing with the electronic structure of these compounds, in particular, of their Fermi surface (FS). The principal method used involved a study of the galvanomagnetic properties and quantum oscillations. Some idea was thus obtained of the form of the FS in such quasi-2-D superconductors as β-$(ET)_2IBr_2$ (Ref. 1), θ-$(ET)_2I_3$ (Ref. 2), and k-$(ET)_2Cu(NCS)_2$ (Ref. 3). As expected from general structural considerations, a consistent feature of the FS in these superconductors is a cylinder with its axis parallel to the direction perpendicular to the conducting layers. However, the detailed form of the FS in different materials may vary greatly, while this common feature is preserved. Thus, in k-$(ET)_2Cu(NCS)_2$, Shubnikov–de Haas oscillations were detected, which, at least as a rough approximation, correspond to a FS in the shape of a smooth cylinder.[3] In θ-$(ET)_2I_3$, the cylinder is represented as being fluted,[2] and in β-$(ET)_2IBr_2$, in addition to the basic corrugated cylinder with a cross-sectional area of the order of one-half the area of the corresponding cross section of the Brillouin zone, small cross sections of FS were found.[1] At the same time, these superconductors are characterized by an appreciable difference in critical temperatures, from 2.8 to 10.5 K. It can be expected that a more detailed study of the FS of these compounds will shed light on the nature of this difference.

This paper deals with the angular dependence of magnetoresistance in the k-$(ET)_2Cu(NCS)_2$ superconductor. The behavior Shubnikov–de Haas oscillations with changing temperature and field orientation is studied. The most probable form of the FS is discussed.

Measurements of the magnetoresistance of k-$(ET)_2Cu(NCS)_2$ single crystals involve the use of the standard four-contact method with a 13.7-Hz alternating current and a magnetic field $H = 150$ kOe, produced by a superconducting solenoid.[1)] The measuring current was always parallel to crystallographic axis **b** of the crystal and amounted to 1 mA. The magnetic field rotated in the **a*b** plane relative to the crystal (the layers of ET molecules were in the plane **bc**; **a*** was the direction perpendicular to this plane). Use was made of measuring modules with low-noise clamping contacts that made it possible to raise the sensitivity of the measurements and consequently, to observe Shubnikov–de Haas oscillations at higher temperatures (1.45–2.0 K) than in studies by other authors.[3,4]

The single crystals studied had average dimensions of $1.2\times0.2\times0.02$ mm^3; room-temperature conductivity $\sigma_{295\,K} = 30\,\Omega^{-1}\,cm^{-1}$; a resistance with a typical temperature dependence having a maximum at 90 K, at which $R_{max}/R_{295\,K} \simeq 2$; and a superconducting transition at $T = 10$ K. Near the transition the resistance of the samples was approximately two orders of magnitude lower than at $T = 295$ K.

Figure 1 shows the angular dependence of the resistance of the k-$(ET)_2Cu(NCS)_2$ crystal at $T = 1.45$ K in a magnetic field of 150 kOe. The vanishing of the resistance for directions of the field near **H**||**b** is associated with the superconducting state, which is preserved because of the high value of the upper critical field H_{c2}^{b} (Ref. 5). In the range of angles where the resistance corresponds to the normal state, the change in resistance is less than 15%. A certain asymmetry of the dependence $R(\varphi)$ relative to the direction **H**||**b** may be due, for example, to the contribution of the Hall emf.

A characteristic feature of the angular dependence of the resistance is that the strong oscillations of the classical part of the magnetoresistance, periodic functions of the tangent of the angle which were recorded in β-$(ET)_2IBr_2$ (Ref. 1), are completely absent. The observed small oscillations at $\varphi = \pm 50°$ are due to the Shubnikov–de Haas effect.

Shubnikov–de Haas oscillations are clearly manifested in the indicated range of angles between **H** and **a*** on the curves showing the field dependence of the resistance. In Fig. 2, these oscillations are shown for **H**||**a*** and $T = 1.45$

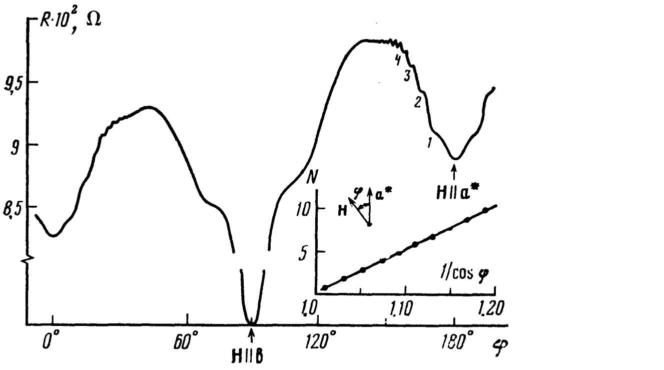

FIG. 1. Angular dependence of the resistance of the k-$(ET)_2Cu(NCS)_2$ crystal in a field $H = 150$ kOe at $T = 1.45$ K. Shown in the inset is the number of Shubnikov–de Haas oscillations in the angular dependence of the resistance as a function of the reciprocal cosine of the angle between **H** and **a***.

 0038-5646/90/080396-02$03.00

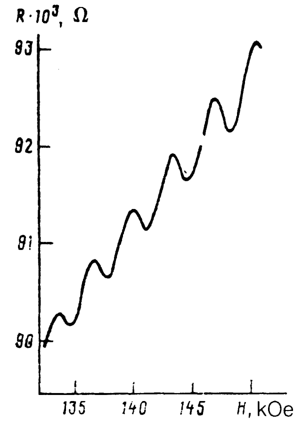

FIG. 2. Example of Shubnikov–de Haas oscillations at $T = 1.45$ K with $\mathbf{H}\|\mathbf{a}^*$.

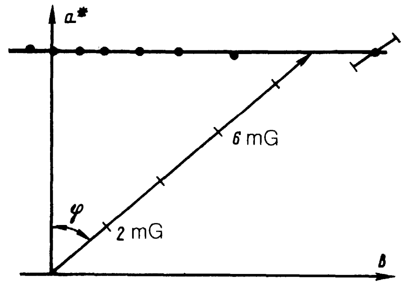

FIG. 3. Angular dependence of the frequency of Shubnikov–de Haas oscillations in polar coordinates (φ being the angle between **H** and **a***).

K. Their frequency is 6.1 mG, which corresponds to the extreme cross section of the FS with an area comprising $\simeq 18\%$ of the area of the corresponding cross section of the Brillouin zone. The oscillation frequency changes as a function of the angle φ between **H** and **a*** in accordance with the law $F \propto 1/\cos\varphi$ (Fig. 3). This frequency dependence indicates that the observed FS sheet is a slightly corrugated cylinder with the axis along **a***. When the field deviates from the direction $\mathbf{H}\|\mathbf{a}^*$, the oscillation amplitude increases slightly and reaches a maximum at $\varphi = -18°$, then rapidly decreases. As was pointed out in Ref. 6, this behavior of the amplitude may be due to a slight corrugation of the cylindrical FS. Oscillations due to cross sections of the FS of a different shape or different size were not observed. As noted above, Shubnikov–de Haas oscillations are also manifested in the angular dependence of the resistance in the form of slight variations of $R(\varphi)$. These angular oscillations are periodic in the secant of the angle (see inset in Fig. 1), which is consistent with the observed dependence $F(\varphi)$.

The deviation of the shape of the FS from an ideal cylinder in $\beta(ET)_2IBr_2$ was indicated by the presence of Shubnikov–de Haas oscillations with different frequencies, and the presence of large angular oscillations of the resistance of a nonquantum nature.[1] Similar angular oscillations were observed in θ-$(ET)_2I_3$ and were also attributed to the distortion of the cylindrical shape of the FS.[2] The absence of nonquantum angular oscillations in k-$(ET)_2Cu(NCS)_2$ crystals as well as the observation in the later of Shubnikov oscillations of only one frequency for any direction of the field constitute important arguments indicating that the FS is in the shape of an almost smooth cylinder.

The extremely slight corrugation of the observed FS sheet is also indicated by the small relative increase of the oscillation amplitude at the maximum as compared to the $\mathbf{H}\|\mathbf{a}^*$ direction: in the compound under consideration, this increase amounts to $\sim 10\%$, whereas for β-$(ET)_2IBr_2$, it amounts to ~ 1.5 orders of magnitude.[1] Thus, the interlayer interaction leading to distortion of the ideal cylindrical shape of the FS is seen as being very weak in k-$(ET)_2Cu(NCS)_2$ in comparison with β-$(ET)_2IBr_2$ and θ-$(ET)_2I_3$. This is also confirmed by the high value of the conduction anisotropy: $\sigma_\perp/\sigma_\| \sim 10^4$ in k-$(ET)_2Cu(NCS)_2$ where ($\sigma_\|$ and $\sigma_\perp$ are the conductivities in the layer and at right angles to it).[7] The analogous quantity in β-$(ET)_2IBr_2$, measured by Montgomery's method, is approximately an order of magnitude smaller.[2]

Using the temperature and field dependences of the amplitude of Shubnikov–de Haas oscillations, one can determine the cyclotron mass and Dingle temperature for k-$(ET)_2Cu(NCS)_2$. They are found in the **bc** plane to be $m^* = 3.5m_e$ and $T_d = 0.5$ K, respectively, is in good agreement with Ref. 3. In the **bc** plane, the cross-sectional area of the FS is $S_F \simeq 4\pi^2\hbar^2 \cdot 0.2/bc$. One can thus estimate the Fermi velocity in this plane: $v_F \simeq 2\hbar(0.2\pi/bc)^{0.5}/m^* = 8\times 10^6$ cm/sec for $m^* = 3.5m_e$. This value is close to that of the Fermi velocity $v_F = 6\times 10^6$ cm/sec obtained from measurements of the upper critical field $H_{c2}^{a^*}(T)$ (Ref. 5) in the "pure" superconductor approximation. The soundness of this approximation is confirmed by the fact that Shubnikov oscillations exist in k-$(ET)_2Cu(NCS)_2$.

The authors are deeply grateful to N. E. Alekseev, T. Palevskii, and E. B. Yagubskiĭ for their steady interest in this work, to N. D. Kushch for synthesizing the samples, and to I. F. Shchegolev for useful discussions.

1) Solid State Physics Institute, Academy of Sciences of the USSR.
1) All the measurements were carried out at the International Laboratory of Strong Magnetic Fields and Low Temperatures (Wrocław, Poland).
2) Data supplied by L. I. Buravov.

[1] I. F. Schegolev, P. A. Kononovich, V. N. Laukhin, and M. V. Kartsovnik, Phys. Scr. **29**, 46 (1989).
[2] K. Kajiti, Y. Nishio, T. Takahasi *et al.*, Solid State Commun. **70**, 1189 (1989).
[3] K. Oshima, T. Mori, H. Inokuchi *et al.*, Synthetic Metals **27**, A165 (1988).
[4] G. Saito, H. Urayama, H. Yamochi, and K. Oshima, Synthetic Metals **27**, A334 (1988).
[5] K. Murata, Y. Honda, H. Anzai *et al.*, Synthetic Metals **27**, A341 (1988).
[6] K. Yamaji, J. Phys. Soc. Jpn. **58**, 1520 (1989).
[7] L. I. Buravov, A. V. Zvarykina, N. D. Kushch *et al.*, Zh. Eksp. Teor. Fiz. **95**, 322 (1989) [Sov. Phys. JETP **68**, 182 (1989)].

Translated by Adam Peiperl