Features of interlayer electron transport in quasi-two-dimensional organic metal $(ET)_4HgBr_4(C_6H_4Cl_2)$

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Angular dependences of the interlayer magnetoresistance in quasi-two-dimensional organic metal $(ET)_4HgBr_4(C_6H_4Cl_2)$ (ET is bis(ethylenedithio)tetrathiafulvalene) in magnetic fields up to 15 T in the temperature range 1.5—4.2 K were studied. Interlayer charge transport in this metal can be described within the framework of the model of almost noninteracting metal layers.

Key words: quasi-two-dimensional metals, magnetic field, low temperatures, interlayer transport, magnetoresistance, Shubnikov—de Haas oscillations, Fermi surface.

For nearly all quasi-two-dimensional organic metals based on bis(ethylenedithio)tetrathiafulvalene (ET) and its derivatives known to date a metallic type of the temperature dependence of the in-plane and interlayer resistance was observed at least at relatively low temperatures.[1] Low-temperature metallic state of these compounds was predicted theoretically by calculations and confirmed experimentally by observation of quantum and semiclassical oscillations of the magnetoresistance in a number of compounds.[1] The dependence of the non-oscillating component of the magnetoresistance on the orientation of magnetic field can be significantly different for different metals even with similar electronic structures. Detailed theoretical and experimental studies showed that such differences are to a great extent due to the character of electron transport between conducting layers.[1—4] Three regimes of this process can be distinguished. One of them, the coherent interlayer transport, involves transfer of an electron through some conducting layers of the organic metal without scattering. In this case, the Fermi surface is a corrugated cylinder whose axis is perpendicular to the conducting layers. The temperature dependence of the interlayer resistance has metallic character. The second regime is defined as the weakly incoherent transport. In this case, the momentum of an electron is conserved only on going to the neighboring layer. The Fermi surface becomes a smooth cylinder; however, the temperature dependence of the resistance in zero magnetic field still has a metallic type. The third regime, the incoherent charge transport, assumes a jumpwise character of interlayer transitions without conservation of the momentum and a nonmetallic temperature dependence of the resistance. The Fermi surface is still a smooth cylinder, as in the second case. Recently, a new version of the description of interlayer electron transport was proposed.[5] It assumes (i) no weakly incoherent transport and (ii) coexistence of the coherent and incoherent regimes within the same electronic system. Accordingly, differences in the behavior of different systems are determined by the differences between the contributions of these two regimes only. In this context, studies of metals with the nonmetallic behavior of the interlayer resistance in a wide low-temperature interval (strong indication of incoherent transport) are of particular interest. However, such metals are rare to occur so far (see above). To date, an unrestricted increase in the interlayer resistance down to the lowest temperatures with conservation of the metallic state within the layer was observed only in an organic metal θ-$(BETS)_4HgBr_4(C_6H_5Cl)$ (BETS is bis(ethylynedithio)tetraselenafulvalene).[6,7] A periodic change in the electronic structure of the metallic layers in this compound undoubtedly favors deterioration of conditions for coherent interlayer transport.

In the present study we report the results of the magnetoresistance studies of a new organic quasi-two-dimensional metal $(ET)_4HgBr_4(C_6H_4Cl_2)$ for which the temperature dependence of the resistance is similar in character to that of θ-$(BETS)_4HgBr_4(C_6H_5Cl)$. Analysis of the angular dependences of the magnetoresistance in the polar and azimuthal planes allowed one to unambiguously explain the interlayer electron transport

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in this metal using the model for incoherent electron transport.

Experimental

Single crystals of $(ET)_4HgBr_4(C_6H_4Cl_2)$ having the shape of an irregular parallelepiped (averaged characteristic dimensions 1.5×0.5×0.05 mm^3) were studied. The crystal structure determined by single-crystal X-ray analysis at room temperature corresponds to the tetragonal system.[8] The ET molecules are packed by the θ-type in the conducting layers alternating along the short edges of the samples. The resistance was measured by a conventional a.c. four-point method. All measurements in magnetic field were carried out at current direction perpendicular to the conducting layers. The magnetic field up to 15 T was produced by an Oxford Research superconducting solenoid. The magnetoresistance measurements were carried out in the 1.5—4.2 K temperature range. To change the sample orientation in magnetic field, a double-swivel apparatus was used, which allows one to rotate the sample in the polar plane and in the azimuthal plane parallel to the conducting layers. All experiments were carried out at the International Laboratory of High Magnetic Fields and Low Temperatures (Wroclaw, Poland) headed by Prof. J. Klamut. The equipment used in the experiments can be seen at the Website http://alpha.mlspmint.pan.wroc.pl/ml_ang.html.

Results and Discussion

The temperature dependences of the resistance normalized to its room-temperature value in zero magnetic field are shown in Fig. 1. The in-plane resistance (see Fig. 1, curve *1*) shows a metallic behavior. At the same time, the temperature dependence of the interlayer resistance (see Fig. 1, curve *2*) has a nonmetallic character. The inset in Fig. 1 shows a portion of the temperature dependence of the in-plane resistance which exhibits a feature characteristic of the first-order phase transition. A preliminary X-ray diffraction study revealed a structural phase transition of $(ET)_4HgBr_4(C_6H_4Cl_2)$ at $T < 280$ K.[8] Thus, it is highly probable that a phase transformation accompanied by a change in the crystal lattice type occurs at $T = 285$ K. It should be noted that this transformation was not accompanied by the change in the character of both types of temperature dependences of the resistance.

Figure 2 shows the magnetic field dependence of the interlayer resistance at a magnetic field orientation perpendicular to the conducting plane. The magnetoresistance at $H = 15$ T is $\Delta R(H)/R(0) \approx 10\%$. The Shubnikov—de Haas oscillations at a frequency $F \approx 900$ T are observed in magnetic fields $H > 9$ T. The effective mass m^* of the carriers responsible for the oscillations was estimated from the temperature dependence of the amplitude of oscillations. It was found that $m^* \approx 2.2m_0$, where m_0 is the mass of a free electron. Observation of the Shubnikov—de Haas oscillations is strong evidence showing that the conducting layers contain a metallic system of electrons described in the framework of the Fermi liquid approximation. Therefore, the data shown in Figs 1 and 2 suggest that $(ET)_4HgBr_4(C_6H_4Cl_2)$ has a layered structure with normal metallic layers and nonmetallic type of interlayer transport.

Figure 3 presents the dependences of the interlayer magnetoresistance on the angle θ between the magnetic field direction and the conducting plane in three different polar planes. For convenience, the curves obtained at different values of the azimuthal angle are shifted by 0.5 kOhm relative to one another. As can be seen, the magnetoresistance is almost isotropic in the azimuthal plane and anisotropic in the polar plane; the minimum value is attained when the magnetic field is parallel to the conducting plane while the maximum magnetoresistance is attained at a magnetic field nearly perpendicular to the conducting layers. The anisotropy is $R_{min}/R_{max} \approx 10\%$. The oscillations observed near the maximum are due to the Shub-

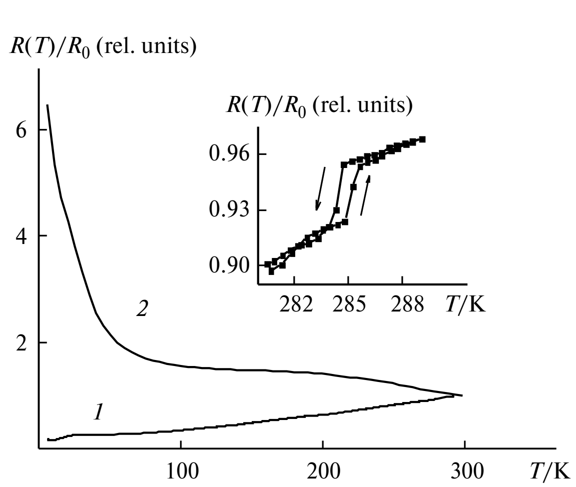

Fig. 1. Temperature dependence of the resistance of $(ET)_4HgBr_4(C_6H_4Cl_2)$: in-plane (*1*) and out-of-plane (*2*) resistance. Inset: a portion of curve *1*.

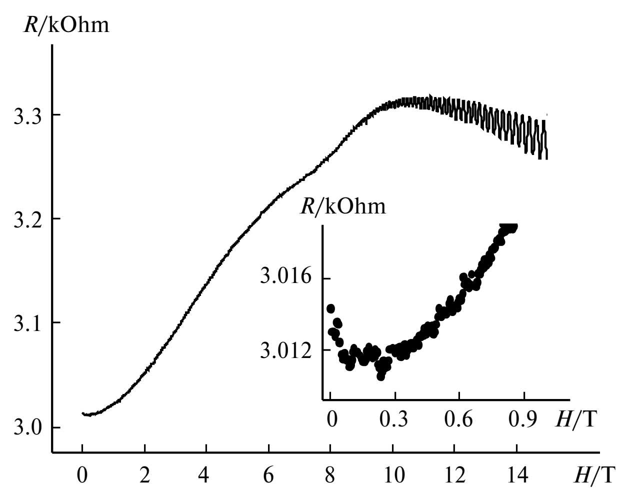

Fig. 2. Interlayer resistance plotted *vs.* magnetic field perpendicular to the conducting layer at $T = 1.5$ K. Inset: initial portion of the $R(H)$ curve.

nikov—de Haas oscillations. In the case of coherent interlayer electron transport, one usually has the reverse situation, *viz.*, the maximum magnetoresistance corresponds to the in-plane orientation of magnetic field while the minimum magnetoresistance is attained if magnetic field is perpendicular to the conducting plane.[1,3,4] This behavior should be a consequence of movement of electrons in this field along open orbits in the momentum space at the in-plane orientation of magnetic field. This movement usually ensures a higher magnetoresistance compared to the magnetoresistance at perpendicular direction of magnetic field when electrons move along closed trajectories. In addition, in the case of coherent electron transport, intracrystalline anisotropy is the reason for a large magnetoresistance anisotropy in the azimuthal plane,[1,4] which was not observed in our experiments. In the case of incoherent or weakly incoherent electron transport, the Fermi surface has the shape of a smooth cylinder, *i.e.*, the electronic system is divided into a series of noninteracting or nearly noninteracting layers and the angular dependence of magnetoresistance in the polar plane will mainly be determined by the projection of the magnetic field on the normal to the conducting plane.[1,4] Consequently, the magnetoresistance in the azimuthal plane should be isotropic. A comparison of the magnetoresistance (see Fig. 2), the magnetoresistance anisotropy (see Fig. 3), and the shape of the curves in Figs 2 and 3 suggests that the results obtained better correspond to this type of behavior. Thus, the angular dependences of the magnetoresistance provide an evidence in favor of incoherent interlayer electron transport in the organic metal $(ET)_4HgBr_4(C_6H_4Cl_2)$. However, yet another issue is to be addressed. A small peak of magnetoresistance is observed at the magnetic field parallel to the conducting layers (see Fig. 3). Usually, this peak (so-called "coherence peak") is due to a feature of the motion of electrons along the corrugated generator of the cylindrical Fermi surface under coherent transport conditions.[1] In this case, the peak amplitude depends on the magnetic field magnitude, the temperature, and the azimuthal angle, whereas the peak width is independent of these parameters, being determined by the shape and size of the corrugation pattern only. Of course, this peak is not observed for incoherent electron transport. The inset in Fig. 3 shows that the peak amplitude remains almost unchanged while the peak width decreases as the magnetic field increases. Therefore, the peak observed is not a coherence peak. Most probably, it corresponds to a weak minimum of unknown nature on the magnetic field dependence of the magnetoresistance at $H \approx 0.2$ T (see Fig. 2, inset).

The interlayer resistance of the new quasi-two-dimensional organic metal $(ET)_4HgBr_4(C_6H_4Cl_2)$ was investigated as a function of temperature and the magnetic field magnitude and direction. It was established that

1) the metallic electronic system in each conducting layer obeys the Fermi liquid model;

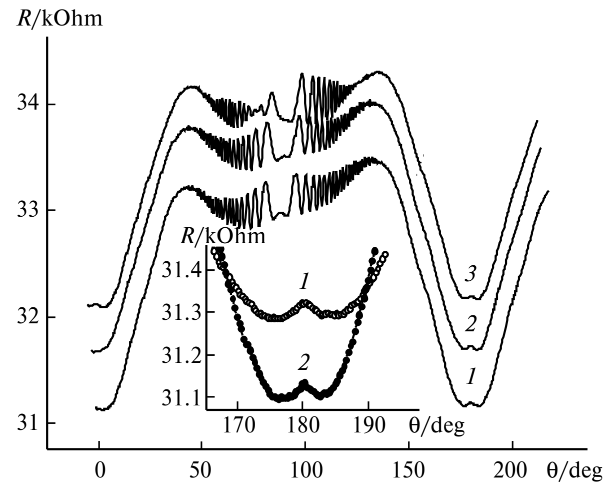

Fig. 3. Angular dependences of the interlayer magnetoresistance in the polar plane at different azimuthal angles Φ equal to 16° (*1*), 52° (*2*), and 82° (*3*) at $T = 1.5$ K and $H = 15$ T. Inset: angular dependences of the interlayer magnetoresistance at $H = 10$ (*1*) and 15 T (*2*) at arbitrary azimuthal angle; $T = 1.5$ K.

2) the Fermi surface of the entire electronic system is a smooth cylinder and it corresponds to a series of almost noninteracting metallic layers; and

3) interlayer electron transport occurs in the incoherent regime.

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