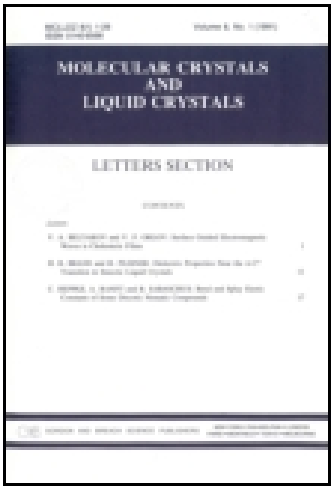

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Transport and Magnetotransport in the New Quasi-Two-Dimensional Organic Metal $(BETS)_2ZnBr_4(C_6H_4Cl_2)$ with Different Electron Structure of Neighboring Cation Layers

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Transport and Magnetotransport in the New Quasi-Two-Dimensional Organic Metal $(BETS)_2ZnBr_4(C_6H_4Cl_2)$ with Different Electron Structure of Neighboring Cation Layers

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Intralayer and interlayer transports and Shubnikov – de Haas oscillations are investigated in the new dual-layered organic metal $(BETS)_4ZnBr_4(C_6H_4Cl_2)$. It is shown that: a) the interlayer resistance behavior corresponds to incoherent transport; b) the behavior of quantum oscillations is well described by the model of a coherent network of magnetic breakdown orbits. Both the interlayer transport and quantum oscillations are in a good agreement with the theoretical calculation of the zone structure.

Keywords Quasi-two-dimensional organic metal; interlayer transport; quantum oscillations

Introduction

Traditional organic quasi-two metals comprising cation-radical salts based on ET (bis (ethylenedithio)tetrathiofulvalene) molecules and its derivatives have been actively investigated for more than a quarter century. Organic synthesis results in layered single crystal structures having cation layers, consisting of ET molecules and exhibiting the metallic type conductivity, sandwiched between insulating anion layers. This creates a well-pronounced layered quasi-two-dimensional organic metal with anisotropic conductivity along and perpendicular to the layers differing by a factor of $10^3 – 10^4$. One of the features of traditional organic metals is that the molecular and electronic structure of the cation layers are the same in each layer. This leads to a single Fermi surface (FS) within each layer and the total FS for the reciprocal lattice as a whole having a shape of a cylinder with the axis perpendicular to the metal layers [1–3]. The second important feature of most traditional organic metals is the reduction in both the intralayer and interlayer resistances under decreasing the temperature, which is characteristic of metallic systems [3]. Relatively recently, a new class of quasi-two-dimensional organic metals, the so-called dual-layered metals has been synthesized [4–9]. In these materials, the electron and molecular structures of adjacent

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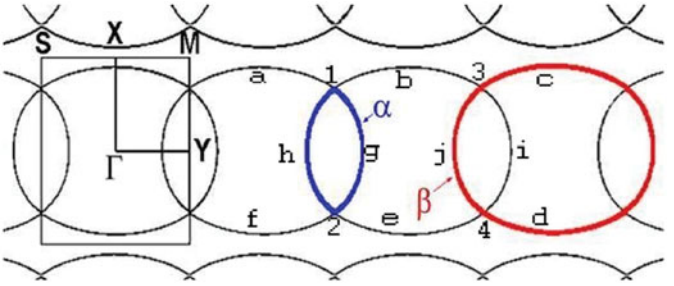

Figure 1. Fermi surface of a metallic cation layer in $(ET)_4CoBr_4(C_6H_4Cl_2)$ [8].

cation layers differ from each other. In other words, the properties of a cation layer, and in particular of FS, are translated over the layer. The structure and properties of the dual-layered quasi-two-dimensional organic metal $(ET)_4CoBr_4(C_6H_4Cl_2)$ were studied in [6, 8]. X-ray analysis showed that this compound contains two different alternating cation layers. Band structure calculations suggested one of the layers to be an insulator with small gap, the width of which does not exceed the accuracy of the calculation. The second layer is a metal with FS shown in Fig. 1. This FS is typical of the θ-type packing of ET molecules in the cation layer [1]. The detailed analysis of quantum oscillations in $(ET)_4CoBr_4(C_6H_4Cl_2)$ showed good agreement of the frequency spectrum of oscillations with theoretical calculations [8]. Similar results were obtained for the isostructural dual-layered organic metal $(ET)_4ZnBr_4(C_6H_4Cl_2)$ [9].

This paper presents the results of studies of intralayer and interlayer transports and Shubnikov - de Haas (SdH) oscillations in another isostructural dual-layered organic quasi-two-dimensional metal $(BETS)_4ZnBr_4(C_6H_4Cl_2)$. The BETS molecule (bis (ethylenedithio)tetraselenofulvalene) differs from the ET molecule in that sulfur atoms are partially replaced by selenium. Taking into account that the conductivity of ET metals is provided by the overlapping of sulfur atomic orbitals, one can expect changes in conducting properties of the BETS metal, even if their crystal structures are identical.

Experiment and discussion

The study of the magnetoresistance of $(BETS)_4ZnBr_4(C_6H_4Cl_2)$ was carried out in single-crystal specimens by the standard four-probe method at liquid helium temperatures in the magnetic fields up to 14 T generated by a superconducting magnet. Figure 2 shows the temperature dependence of the resistance to current in parallel and perpendicular directions to the conducting layers. In the

former case, the conductivity is purely metallic, while, in the latter case, the monotonic non-metallic behavior of the resistance occurs almost down to the temperature of liquid helium. While this behavior is radically different from that observed in the traditional quasi-two-dimensional metals [3], it has a simple explanation. The nonmetallic type of the interlayer resistance corresponds to the incoherent transfer of electrons, which is determined by the time relation

$$\tau_\hbar = h/t >> \tau_c, \tag{1}$$

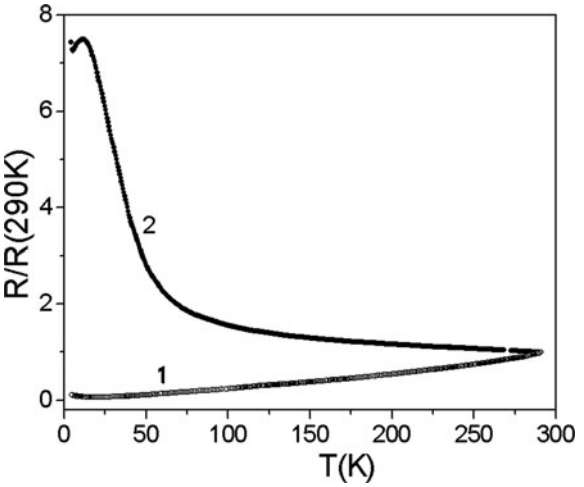

Figure 2. Temperature dependences of intralayer (1) and interlayer (2) resistances for $(BETS)_4ZnBr_4(C_6H_4Cl_2)$.

where τ_h and t are the interlayer transfer time and the interlayer transfer energy (transfer integral), respectively, and τ_c is the electron scattering time in the layer [2, 3, 10–12]. Provided of the identity of the crystal structures in the ET and BETS metals, the transition of an electron from the metal layer to the adjacent layer is considerably eliminated. Therefore, the value of t drops, which leads to relations (1). A similar temperature dependence for the resistance was observed in the ET complex [8]. The maximum interlayer resistance at T $\sim$ 10 K can be associated with a change of the regime of the interlayer transfer from incoherent to weakly incoherent one corresponding to the time relation $\hbar$ / t $\sim \tau_c$, mainly due to increasing the time of the intralayer scattering of electrons at low temperatures. Such a regime is accompanied by the metallic temperature dependence of the resistance [2, 3,].

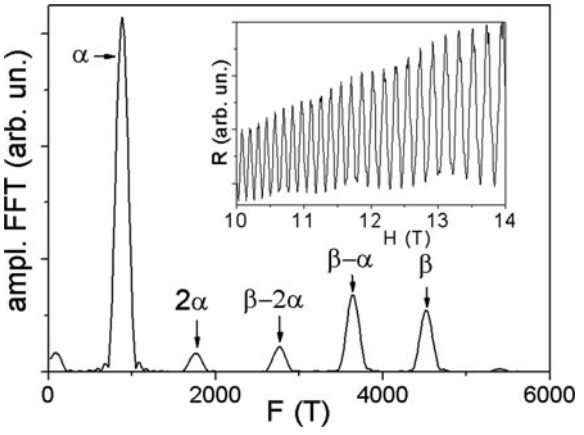

Figure 3. Fourier spectrum of SdH oscillations (presented in the insert) for $(BETS)_4ZnBr_4(C_6H_4Cl_2)$. T = 1.5 K, $\theta = 6^0$.

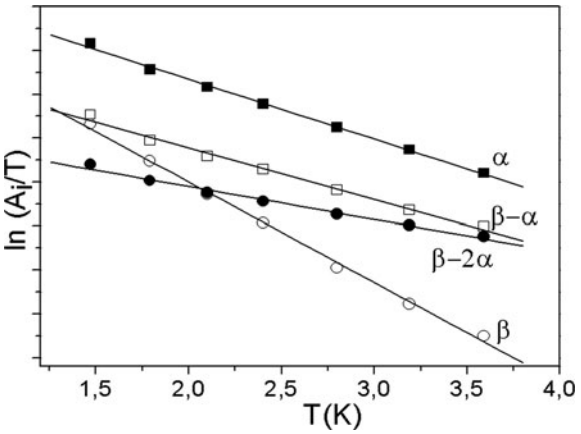

Figure 4. Temperature dependences (mass plot) of the amplitude for oscillations with different frequencies. Solid lines correspond to the LK model.

Figure 3 shows the Fourier spectrum of the SdH oscillations in $(BETS)_4ZnBr_4(C_6H_4Cl_2)$. An example of such oscillations is shown in the inset in Fig. 3. The spectrum is in good agreement

with theoretical calculations of FS for ET salts (Fig. 1). All the frequencies of oscillations depend on the angle θ between the magnetic field and the direction perpendicular to the conducting layers in accordance with the law $F_i(\theta) = F_i(0) / \cos\theta$, where $i = \alpha, \beta, 2\alpha \ldots$. This law defines a cylindrical FS typical of quasi-two-dimensional metallic systems. The spectrum has two fundamental frequencies F_α ($F_\alpha(0) = 875$ T) and F_β ($F_\beta(0) = 4550$ T), which correspond to α and β orbits covering 19% and 100% of the intralayer first Brillouin zone, respectively. The cyclotron masses associated with these frequencies and calculated from the temperature dependence of the oscillation amplitudes are in good agreement with the Lifshits – Kosevich formula (see Fig. 4) and have values: $m_\alpha = (1.3 \pm 0.1)m_0$ and $m_\beta = (2.2 \pm 0.2)m_0$ at the field direction perpendicular to the conducting layers, where m_0 is the free electron mass. The α and β frequencies are due to the existence of a gap at points 1 - 4 (Fig. 1) of the initial FS, covering 100% of the Brillouin zone (two electrons per unit cell). As a result, the closed hole orbit α, (orbit h1g2h in Fig. 1) and two sheets of open electron orbits, (... abc ... and ... def ...) (see Fig. 1) are formed [8]. In a magnetic field, electrons can transfer through the gap due to the magnetic breakdown from the α orbit to open sheets, and the closed β orbit appears.

In addition to the fundamental frequencies, the spectrum contains the combination frequencies $F_{\beta-\alpha}$ and $F_{\beta-2\alpha}$, which have no allowed closed orbits. Their appearance is most likely due to the effect of quantum interference [13]. In this case, the frequency $F_{\beta-\alpha}$ corresponds to the interferometer trajectories *abief* and *agf*, and the frequency $F_{\beta-2\alpha}$ – to the trajectories *abief* and *aghgf* (see Fig. 1). The ratio of cyclotron masses which is a characteristic of the interference process also supports this hypothesis: $m_{\beta-\alpha} = (1.1 \pm 0.2)$ $m_0 \sim m_\beta - m_\alpha$ and $m_{\beta-2\alpha} = (0.6 \pm 0.2)m_0 \sim m_\beta - 2m_\alpha$.

It should be noted that virtually all of the cyclotron masses in $(BETS)_4ZnBr_4(C_6H_4Cl_2)$ are about one and a half times lower than the cyclotron masses in the isostructural complexes $(ET)_4CoBr_4(C_6H_4Cl_2)$ and $(ET)_4ZnBr_4(C_6H_4Cl_2)$ [8, 9]. Most likely, this is due to a significant weakening of many-body interactions in the BETS metal. This weakening was observed in few traditional BETS complexes [14-16]. The reason remains unclear so far.

Summary

The resistance and the magnetoresistance in the new dual-layered qusi-two-dimensional organic metal $(BETS)_4ZnBr_4(C_6H_4Cl_2)$ have been studied. It is established that the temperature dependence of the resistance and the SdH oscillations are close to those in isostructural metals $(ET)_4CoBr_4(C_6H_4Cl_2)$ and $(ET)_4ZnBr_4(C_6H_4Cl_2)$ and are in good agreement with theoretical calculations of the band structure and FS. SdH oscillations can be described by the model of a coherent chain of magnetic breakdown orbits, including the effects of quantum interference. The observed reduction in the cyclotron masses is likely due to a significant weakening of the many-body interactions in the investigated material.

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