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High-field magnetotransport of organic conductors (BEDT-TTF)$_2$TlHg(XCN)$_4$ with $X = S$ and Se

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Abstract. — High field magnetoresistance of layered organic metals (BEDT-TTF)$_2$TlHg(XCN)$_4$ with $X = S$ and Se has been studied using pulsed magnetic fields. Sharp negative slope and strong hysteresis of the magnetoresistance of (BEDT-TTF)$_2$TlHg(SCN)$_4$ found between about 11 and 27 T suggest a first-order phase transition to occur at these fields. Two different types of angular magnetoresistance oscillations corresponding most likely to different electronic states are observed in the low- and high-field phases, respectively. In contrast, the classical magnetoresistance of the salt with $X = Se$ is found to be smoothly growing with the field increase. The obtained results are compared with those reported earlier on the (BEDT-TTF)$_2$M(Hg(SCN)$_4$ salts with $M = K$ and NH$_4$ and discussed in the framework of the model proposed recently for the low-temperature electronic state of (BEDT-TTF)$_2$TlHg(SCN)$_4$.

Introduction.

The low-dimensional organic conductors (BEDT-TTF)$_2$M(Hg(SCN)$_4$ with polymeric anions M(Hg(SCN)$_4$, where M stands for K, Tl, Rb or NH$_4$, [1-3] have been initially synthesized as a modification of a superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ which has the superconducting critical temperature, $T_c = 10.5$ K, one of the highest among organic metals. It turns out, however, that only the compound with $M = NH_4$ becomes a superconductor above 0.4 K [4], the others undergoing a phase transition at about $T_p = 10$ K which is reflected as a smooth hump in the resistivity versus temperature dependence [2, 5, 6]. (As was recently shown, the

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salts with M = K and Rb exhibit a weak superconductivity below 200 mK [7, 8]. The magnetic susceptibility of (BEDT-TTF)$_2$KHg(SCN)$_4$ was found [9] to be essentially anisotropic below the phase transition temperature, $T_p$. Taking into account results of the band structure calculation [1] predicting the existence of slightly warped open Fermi surface (FS) sheets in addition to the quasi-2D cylindrical ones, the phase transition might be attributed to a spin-density-wave (SDW) instability typical of some quasi-1D organic conductors [10]. NMR studies of (BEDT-TTF)$_2$KHg(SCN)$_4$ [11] revealed some changes in the electronic system below $T_p$, however no direct signs of a magnetic ordering were found. Further investigation should be made to clarify the nature of the low-temperature state of these compounds and the possible competition between the magnetic ordering and superconductivity.

A number of puzzling magnetoresistance anomalies have been reported for the compounds with K [5, 9, 12-14] and Tl [2, 15-17] indicating surprisingly strong effect of magnetic field on their transport properties. Detailed studies of angular-dependent oscillatory magnetoresistance of (BEDT-TTF)$_2$TlHg(SCN)$_4$ [15, 17] give evidence of a commensurate superstructure arising in the electronic system below $T_p$. The superstructure wave vector component, $Q_s$, perpendicular to the plane of the open FS sheets was found to be close to the distance between the sheets, $2k_F$, thus supporting the idea of the Peierls-type density wave formation [17].

As was shown in [2, 17], (BEDT-TTF)$_2$TlHg(SCN)$_4$ shares essentially all the magnetoresistance features with the most popular (BEDT-TTF)$_2$KHg(SCN)$_4$ under magnetic fields up to 14 T. Since a number of important anomalies have been observed in the latter compound in the fields above $15 \pm 17$ T, it would be also interesting to go to the higher fields for (BEDT-TTF)$_2$TlHg(SCN)$_4$.

Here, we present the results of the high-field magnetoresistance studies of (BEDT-TTF)$_2$TlHg(SCN)$_4$. In order to distinguish the specific features related to the low-temperature state of the compound, we have also studied a new conductor, (BEDT-TTF)$_2$TlHg(SeCN)$_4$, which only differs from the former in substituting the S atom by Se in the anion and does not exhibit the low-temperature phase transition [18].

**Experiment.**

Single crystals of (BEDT-TTF)$_2$TlHg(SCN)$_4$ and (BEDT-TTF)$_2$TlHg(SeCN)$_4$ were obtained by electro-crystallization as described elsewhere [2, 18].

The sample resistance was measured perpendicular to the crystal highly-conducting ac-plane in pulsed magnetic fields up to 35 T at temperatures $1.5 \pm 4.2$ K. The samples were glued to 20 µm-diameter platinum wires by a graphite paste. To avoid vibrations during the field pulses, the samples were fixed to a sample holder by a silicon polymer liquid.

The measurements were performed in pulse magnets in the Laboratory of Low Temperature Transport Phenomena (LLTTP) of the A.I. Ioffe Physico-Technical Institute, St.-Petersburg, Russia, and in the International Laboratory of Strong Magnetic Fields and Low Temperatures (ILSMFLT), Wroclaw, Poland.

In LLTTP, the magnetic fields perpendicular to the ac-plane were obtained with either 5 ms total pulse duration (referred below as a short-pulse regime) or 2.5 ms-rise and 25 ms-decrease (a long-pulse regime). The signal from the crystal was amplified by a broad-band amplifier and transferred to an analog-to-digit converter (ADC) with 1 µs gate and 20 µs conversion time.

In ILSMFLT, magnetic fields were generated by a 40 T pulse magnet with the pulse duration of 10 ms. The crystals could be rotated with respect to the magnetic field in a plane perpendicular to the ac-plane. The Shubnikov-de Haas (SdH) oscillations were studied with a dc-method using an analogue second-derivative technique with a broad-band amplifier. The classical magnetoresistance was measured by a standard ac-technique with a narrow-band
amplifier \((f = 50 \text{ kHz})\). The amplified signals were processed by ADC with the digitizing frequency of 300 kHz.

**Results.**

*MAGNETORESISTANCE (MR) FIELD DEPENDENCES. \(H \perp \text{ac-plane.} - \) Figure 1 represents typical MR traces obtained for the \((\text{BEDT-TTF})_2\text{TIHg(SCN)}_4\) crystals at different «short-pulse» sweeps at 1.6 K and 4.2 K. The following important features of the classical part of MR are to be pointed out.

Fig. 1. — Magnetoresistance traces for the Tl-(SCN) salt obtained from a series of successive field pulses at 1.6 K and 4.2 K. \(H \perp \text{ac.} \) The upper curve of each set corresponds to the upward field sweeps while the others represent the downward sweeps from different \(H_{\text{pulse}}\). Inset: Magnetoresistance hysteresis at «long» and «short» pulses. Curve 1 — field up, \(H_{\text{pulse}} = 35 \text{ T} \); curve 2 — field down, long-pulse regime, \(H_{\text{pulse}} = 30 \text{ T} \); curve 3 — field down, short-pulse regime, \(H_{\text{pulse}} = 30 \text{ T} \).

1. Very high MR in the fields up to 13 T is consistent with that observed earlier [2]. At higher fields, MR starts to decrease gradually and then drops rapidly in the interval between 20 and 23 T. Above 27 T it becomes practically field-independent being at least 6 times smaller than the maximum MR. A very similar behavior has been reported by several groups for \((\text{BEDT-TTF})_2\text{KHg(SCN)}_4\) (see e.g. [12, 19, 20]).
2. At $H > H_A = 11$ T a considerable difference arises between the MR traces registered at up and down sweeps of the field (a small difference between the curves at lower fields comes most likely from a slight over-heating of the sample during the pulses, it should be also taken into account that the accuracy of the measurements under high-field pulses is being reduced in their low-field parts). The downward MR traces become lower at increasing the maximum value of the pulse, $H_{\text{pulse}}$, up to $H_{\text{pulse}} = H_B = (27 \pm 1)$ T. Further increase of $H_{\text{pulse}}$, up to $35$ T, does not affect the position of the downward MR curve and no hysteresis is observed above $H_B$. As for the upward MR, it remains reproducible for all the pulses.

3. The magnitude of the hysteresis, $\Delta R(H) \equiv R_{\text{up}}(H) - R_{\text{down}}(H)$, increases rapidly on cooling the sample from $4.2$ K to $1.6$ K. At the same time the changes in the critical fields, $H_A$ and $H_B$, do not exceed $10\%$ within this temperature range.

4. Finally, the hysteresis magnitude, $\Delta R(H)$, appears to depend on the pulse duration. As is seen in the inset in figure 1, the shortening of the field-decay time from $25$ ms to $2.5$ ms (« long » and « short » pulses, respectively, with the same $H_{\text{pulse}}$ value) leads to a significant increase of $\Delta R(H)$.

As concerns the SdH oscillations, we have observed both the fundamental and second harmonics (Fig. 2) with the frequencies $670$ T and $1340$ T, respectively, in agreement with the previous results [16, 17]. Like in (BEDT-TTF)$_2$KHg(SCN)$_4$, the second harmonic is enormously strong above $18$ T, although somewhat weaker than in the latter compound, and drops rapidly at about $H_B$.

![Graph](image)

Fig. 2. — The second derivative of the resistance of the Tl-(SCN) salt for $H \perp ac$, $T = 1.6$ K. The splitting of the oscillations, i.e. anomalously strong second harmonic is most pronounced between about $18$ and $27$ T.

In contrast to (BEDT-TTF)$_2$TlHg(SCN)$_4$, the Se-containing analog (BEDT-TTF)$_2$TlHg(SeCN)$_4$ exhibits rather smooth growth of the classical MR (Fig. 3). No traces of either negative MR contribution or hysteresis have been found. The SdH oscillations are in perfect agreement with those observed in [21] in steady magnetic field $H < 14$ T, with
$F = 650 \text{T}, m = (1.95 \pm 0.05) m_0$ and $T_D \approx 0.6 \text{K}$. Both the fundamental and second harmonic components grow up monotonically with increasing the field their ratio, $A(2F)/A(F)$, not exceeding a few percent at 1.6 K (see inset in Fig. 3).

Angular magnetoresistance oscillations. — Initially, the angular MR oscillations (AMRO) found in (BEDT-TTF)$_2$KHg(SCN)$_4$ at low temperatures [12] were considered to have the same origin as in quasi-2D metals $\beta$-(BEDT-TTF)$_2$IBr$_2$ [22] and $\theta$-(BEDT-TTF)$_2$I$_3$ [23], i.e. associated with closed orbits along the cylindrical FS. However, sharp minima rather than maxima have been noted to be characteristic points of the MR angular dependence. As was suggested in [24], the AMRO reverse their phase around $T_p$ or $H_B$, i.e. the MR minima are transformed into maxima and vice-versa. The reasons for such an abrupt change of the oscillation phase are not clear.

On the other hand, an analysis of AMRO in the low-temperature state of (BEDT-TTF)$_2$TIHg(SCN)$_4$ [15] showed that they must have the nature different from that in $\beta$-(BEDT-TTF)$_2$IBr$_2$ being associated with the existence of a specific plane in the electronic system, most likely, a plane of an open FS. Basing on the oscillation parameters, it was proposed [17] that a Peierls-type commensurate superstructure arises below $T_p$ thus changing essentially the electronic band structure. In this case, the low-temperature-low-field AMRO relevant inherently to the new superstructure should not be related to those existing above $T_p$ and/or $H_B$.

In order to compare the AMRO in the low- and high-field states of (BEDT-TTF)$_2$TIHg(SCN)$_4$, we have measured its magnetoresistance at different field orientations. Figure 4 demonstrates the angular dependences of the magnetoresistance at rotating the sample perpendicular to the highly-conducting ac-plane for two fields, 13 T and 30 T. As is seen in the inset in figure 4, there exist certain orientations for which a considerable growth of MR occurs.
at high fields, instead of the negative slope characteristic of most common orientations. These certain field directions correspond to sharp MR maxima at the high-field angular dependence (curve 2, Fig. 4). The high-field AMRO appear to be periodic in tan $\varphi$ as well as the low-field ones. It is important to note that, despite the apparent closeness of the positions of the high-field MR maxima (downward arrows in Fig. 4 at $\varphi \approx 34^\circ$, $63^\circ$ and $72^\circ$) and the low-field MR minima (upward arrows at $\varphi = 26^\circ$ and $65^\circ$), the difference between them is out of the experimental error range. The same can be said about the oscillation periods: although it is difficult to estimate them correctly due to lack of experimental data, one can see that, anyway, the low-field AMRO have a larger period then the high-field ones.

Discussion.

Basing on the model proposed recently for the groundstate of (BEDT-TTF)$_2$TIHg(SCN)$_4$ [17], one can understand the obtained results as follows.

The low-field (below $H_A$) magnetotransport is determined by the FS magnetic breakdown network formed from the initial cylinders under a periodic density-wave potential. This FS includes new open sheets which may be responsible for high MR at $H < 11$ T. The relatively complicated SdH oscillations, including the enormously strong second harmonic, are due to the breakdown between the open sheets and small cylindrical pockets [18]. The new open sheets are also consistent with the low-field AMRO.

In the high-field range, i.e. above $H_B$, the classical part of MR tends to a constant value at $H \perp ac$ and the SdH oscillations demonstrate extremely strong fundamental frequency, 670 T, and only a small second harmonic contribution consistent with that expected from the Lifshits-Kosevich formula. This is very similar to the case of the TI(SeCN)-salt (Fig. 3) and
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