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J. Phys. I France **3** (1993) 1187-1199 MAY **1993,** PAGE 1187

Classification
Physics Abstracts
72.15G — 73.20D — 74.70K

Magnetotransport investigation of the low-temperature state of $(BEDT\text{-}TTF)_2TlHg(SCN)_4$: evidence for a Peierls-type transition

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(Received 22 December 1992, accepted 26 January 1993)

Abstract. — Detailed magnetoresistance measurements have been performed on $(BEDT\text{-}TTF)_2TlHg(SCN)_4$. An analysis of angle dependent semi-classical magnetoresistance oscillations is presented in connection with possible changes in the electron spectrum at low temperatures. A spin density wave is proposed to arise leading to a modification of the Fermi surface so that new open sheets appear, consistent with the observed angular magnetoresistance oscillations. Some details of the Shubnikov-de Haas effect are also presented which can be explained by the existence of a magnetic breakdown network, in agreement with the proposed low-temperature electronic state of the compound.

Introduction.

An investigation of strong angular oscillations of magnetoresistance found in a layered organic superconductor β-$(BEDT\text{-}TTF)_2IBr_2$ a few years ago [1] has been shown to be very useful in providing quantitative information about the Fermi surface (FS) of organic metals [2]. As it has been shown in [3, 4] such kind of oscillations in Q2D metals with cylindrical FS is associated with the semi-classical part of the magnetoresistance and originates from the fact that the areas of the cyclotron orbits become independent of the momentum component along the FS cylinder axis at certain directions of the magnetic field [5, 2]. Similar oscillations have been observed, so far, in a number of other organic conductors [6-9]. However, our recent studies of the angular magnetoresistance oscillations (AMRO) in $(BEDT\text{-}TTF)_2TlHg(SCN)_4$ [8] revealed substantial differences in their behavior in comparison with those in β-$(BEDT\text{-}TTF)_2IBr_2$. We argued that in $(BEDT\text{-}TTF)_2TlHg(SCN)_4$ the AMRO are associated with some marked plane in the reciprocal lattice, e.g. a plane of open orbits, and not with closed orbits as in β-$(BEDT\text{-}TTF)_2IBr_2$.

In this paper we present a detailed analysis of the AMRO parameters in the titled compound and discuss how do they reflect its new electron structure occurring below a phase transition near 10 K [10]. We show that a Peierls-type transition can modify the FS so that new open sheets appear which are consistent with the observed AMRO. We also present some details of the Shubnikov-de Haas effect which can be explained by the existence of a magnetic breakdown network, in agreement with the proposed low-temperature electronic state of the compound.

The compound $(BEDT\text{-}TTF)_2TlHg(SCN)_4$ belongs to a family of isostructural organic conductors with mercury-containing polymeric anions, $(BEDT\text{-}TTF)_2MHg(SCN)_4$ where M = K [11], Tl [10], Rb [12] and NH_4 [12]. As was predicted by the extended Hückel model band calculations [11], the FS of these salts combine elements typical of both quasi-2D systems (i.e. a cylindrical sheet) and quasi-1D ones (i.e. slightly warped planes). While $(BEDT\text{-}TTF)_2NH_4Hg(SCN)_4$ is superconducting below 2 K [13], the compounds with M = K, Tl and Rb undergo a phase transition at $T \simeq 10$ K and are not superconducting although remain metallic down to the lowest temperatures. Investigation of their low-temperature state should be important for understanding why these salts are not superconducting, in contrast to their ammonium analog.

The 10 K transition manifests itself in the resistivity *versus* temperature dependence as a small « hump » [10, 14]. The magnetic field above 3 T applied perpendicular to the highly conducting ac-plane leads to a rather sharp increase of the resistivity at cooling below the transition temperature [10, 14]. Besides the giant magnetoresistance, a number of intriguing features of the low-temperature state in $(BEDT\text{-}TTF)_2MHg(SCN)_4$ in the magnetic field have been reported so far, such as strong angle dependent magnetoresistance oscillations [7-9], negative slope and « kink » structure of the $R(H)$ dependence at $H > 22$ T (see, for example [7]), a surprisingly large magnitude of the second harmonic in the SdH oscillation spectra [15-18], etc.

Taking into account the existence of a quasi-1D band and antiferromagnetic ordering more likely occurring below the transition temperature [16], one can suppose that a spin density wave (SDW) transition typical of quasi-1D conductors takes place. Still, the nature of the low-temperature state of these compounds is to be clarified yet, as well as the reasons for the above mentioned anomalies.

Experimental.

For the experiment, we used crystallographically oriented crystals of $(BEDT\text{-}TTF)_2TlHg(SCN)_4$ having a plate-like shape with the dimensions of approximately $1 \times 0.05 \times 0.7$ mm^3. A standard ac-technique ($f \simeq 330$ Hz) was used for the measurements of resistances R_a, R_b and R_c along the **a**-, **b***- and **c**-directions, respectively (**c**′ is directed in the crystal highly conducting **ac**-plane perpendicular to the **a**-axis ; **b*** is normal to the **ac**-plane). The magnetic field up to 14 T was generated by a superconducting magnet. Using a modulation coil ($\Delta H \simeq 0.008$ T ; $F = 20$ Hz) we succeeded in observing the SdH oscillations at temperatures up to 6 K in the $\mathbf{H} \perp \mathbf{ac}$ orientation and at $T = 1.5$ K in fields tilted up to 55° from the **b***-axis.

The samples were mounted into a cell which could be rotated in two planes with respect to the magnetic field direction. This arrangement allowed us to measure angular dependences of the magnetoresistance for the field rotating in any plane perpendicular to the crystal **ac**-plane. The error in determination of the crystal orientation was less than 1° for the angle φ between the field and the **b***-direction and did not exceed ± 2° for the angle θ between the field rotation plane and the **c**′-direction.

Results and discussion.

CLASSICAL PART OF THE MAGNETORESISTANCE. — Figure 1 demonstrates angular dependences of the magnetoresistance of $(BEDT\text{-}TTF)_2TlHg(SCN)_4$ measured at different current directions in the magnetic field $H = 14$ T rotating in a plane which forms an angle $\theta_0 \approx 24°$ with the c'-direction and corresponds to the greatest amplitude of the angular magnetoresistance oscillations (AMRO). Note that all the three dependences were obtained on the same crystal. One can see that while the position of the oscillations is independent of the current direction their amplitude is extremely sensitive to it. The greatest AMRO are observed for the resistance R_b, measured perpendicular to the highly conducting plane, whereas they are much smaller for the R_c and almost vanish for the R_a.

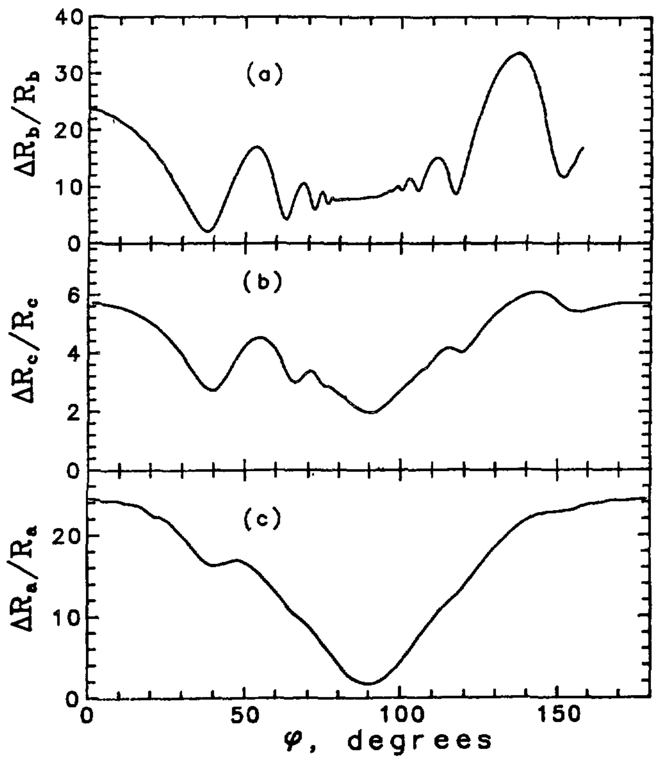


Fig. 1. — Dependence of magnetoresistance measured along (a) **b***-, (b) **c**- and (c) **a**-axes on the angle φ between the magnetic field and **b***. The field, $H = 14$ T, is rotated in the plane corresponding to the maximal AMRO ($\theta = \theta_0 \approx 24°$).

The most distinctive feature of the AMRO in the $(BEDT\text{-}TTF)_2TlHg(SCN)_4$ is that their period varies as $1/\cos(\theta - \theta_0)$ with changing the orientation of the field rotation plane. Such behavior, together with some other specific features, enables us to propose that some marked plane exists in the system and the oscillations are associated with rotating the projection of the magnetic field to this plane [8]. One can naturally suppose that open flat FS sheets may play the role of the marked plane.

Recently, Osada *et al.* [19] have analyzed theoretically the behavior of a quasi-1D metal in a tilted magnetic field and predicted sharp dips of its semi-classical magnetoresistance at certain field directions. This result can be understood qualitatively as follows [8]. Let us consider the following dispersion law near the Fermi level for a quasi-1D system with its 1D-axis along the x-direction :

$$\varepsilon(k) = \hbar v_{\mathrm{F}}(|k_x| - k_{\mathrm{F}}) - \sum_{m,\,n} t_{mn} \cos(m a_y k_y + n a_z k_z), \tag{1}$$

with transfer integrals $t_{mn} \ll \varepsilon_{\mathrm{F}}$. For a general direction of the magnetic field in the plane perpendicular to the 1D-axis, $H = (0, H \sin \varphi, H \cos \varphi)$, the electron trajectory being reduced to the first Brillouin zone (BZ) tends to fill the whole of its area (Fig. 2a). Therefore, the transverse velocity components, v_y and v_z, take all possible values, both positive and negative, and after averaging over the time interval of the order τ (τ is the momentum relaxation time) tend to zero in a high enough magnetic field. However, if the field is oriented so that an electron moves along one of the reciprocal lattice periods, $\mathbf{K} = p\mathbf{K}_z + q\mathbf{K}_y$ (p and q are integers), its trajectory reduced to the first BZ consists of only a finite set of lines as shown in figure 2b. In this case, the transverse velocity takes only a limited set of values prescribed by the electron's initial coordinates in the k-space, and being time averaged it should be non-zero. Therefore, at the special angles, φ_c, between the magnetic field and the k_z-axis, satisfying the condition,

$$\tan \varphi_c = \frac{pK_z}{qK_y}, \tag{2}$$

dips of the transverse resistance components, ρ_y and ρ_z, are expected [19]. In a more general case, the field is rotated in a plane forming an angle θ with the open FS plane, $\mathbf{H} = H(\sin \varphi \sin \theta,\ \sin \varphi \cos \theta,\ \cos \varphi)$, and the directions of $\mathbf{K}_c$ and $\mathbf{K}_b$ vectors do not coincide with the y- and z-axes, $\mathbf{K}_c = (0, K_{cy}, K_{cz})$ and $\mathbf{K}_b = (0, K_{by}, K_{bz})$. Then, since the oscillations are connected with rotating the magnetic field component parallel to the yz-plane, $\mathbf{H}_{/\!/} = H(0,\ \sin \varphi \cos \theta,\ \cos \varphi)$, the condition for the critical angles takes the form :

$$\tan \varphi_c \cos \theta = \frac{pK_{bz} + qK_{cz}}{pK_{by} + qK_{cy}} \tag{3}$$

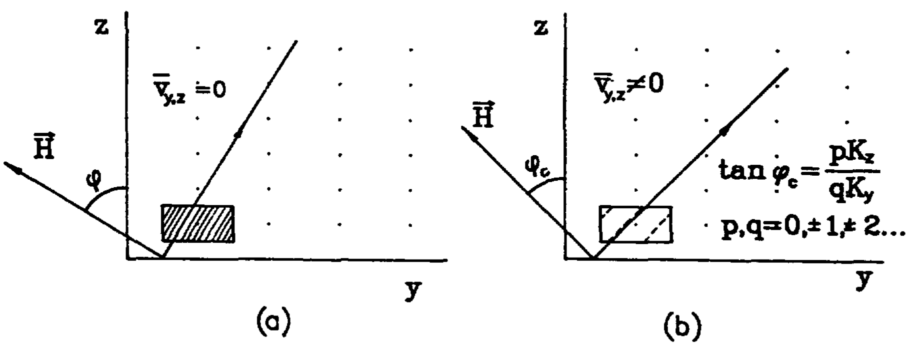


Fig. 2. — Schematic view of the electron motion along a slightly warped FS parallel to yz-plane under tilted magnetic field : (a) general case (incommensurate) ; the electron trajectory reduced to the first BZ tends to fill its whole area (shaded region) ; and (b) commensurate case ; the reduced electron trajectory consists of a finite set of lines only (dashed lines).

or, if $K_{by} = 0$,

$$\tan \varphi_c \cos \theta = \text{cotan}\, \alpha + \frac{pK_b}{qK_c \sin \alpha} \tag{3'}$$

where α is an angle between the $\mathbf{K}_b$ and $\mathbf{K}_c$ vectors.

This model describes both the periodic sharp dips of the magnetoresistance at tilting the magnetic field and the dependence of the oscillation period on the angle θ. Moreover, the relationship between the oscillations amplitude for the resistance R_b, R_c and R_a obtained in our experiment (Fig. 1) is also in agreement with this consideration [19] if we take into account the anisotropy of the compound [20].

There is a number of other theories [21-24] which predict magnetoresistance oscillations for quasi-1D systems rotating in the magnetic field. Some of them are discussed in [25] in connection with angular oscillations in $(TMTSF)_2ClO_4$ which are similar to those described here but much less in the magnitude. All these models being connected with the electron motion along the open FS, lead to the same condition for the critical angles (3), despite proposing different mechanisms for the AMRO. However, the above presented model seems to be the most appropriate in our case since it explains easily not only the periodic sharp dips of the magnetoresistance at the critical angles but also the observed dependence of the oscillations amplitude on the transport current direction. Further experiments are necessary foe establishing the actual mechanisms responsible for the observed phenomenon, e.g. investigation of the magnetic susceptibility or heat capacity as function of the field direction. Anyway, a conclusion can be made that the oscillations are associated with rotating the magnetic field component parallel to the plane of the open FS sheets.

At first sight, one could attribute the AMRO in $(BEDT\text{-}TTF)_2TlHg(SCN)_4$ to the open FS sheets predicted for the $(BEDT\text{-}TTF)_2MHg(SCN)_4$ compounds by the band structure calculations [11] using the room temperature crystal parameters. It is important, however, that these sheets lying in the **b*** **c**-plane do not coincide with the plane of the maximum AMRO making an angle $\theta_0 \approx 24°$ with the latter. It more likely means that some new periodicities in the $(BEDT\text{-}TTF)_2TlHg(SCN)_4$ electron structure appear at low temperatures. One can expect these changes to occur at the phase transition [10]. This suggestion is supported by the temperature dependence of the AMRO amplitude shown in figure 3. The AMRO arise just below the transition temperature, $T_p \approx 9$ K and grow sharply with cooling the sample : the oscillations amplitude at 1.5 K is at least 2 orders of magnitude greater than that at 8.5 K while the mean free path grows not more than four times, if estimated from the sample resistance.

The new periodicity can be determined from the AMRO parameters using the above given conditions (3-3′) for the critical angles φ_c. First of all, we note that, as was established in [8], the critical angles obey the relation (3′). It means that the vector $\mathbf{K}_b$ of the new BZ remains perpendicular to the crystal **ac**-plane as defined at the room temperature. The other vector, $\mathbf{K}_c$, lies in a plane forming an angle $\theta_0 \approx 24°$ with the crystal **b*** **c**-plane and is inclined by $\approx 65°$ with respect to $\mathbf{K}_b$. As will be shown below, a good agreement with the experimental data can be achieved by proposing a commensurate superstructure with a wave vector,

$$\mathbf{Q} = \frac{\zeta}{6}\mathbf{K}_{a_0} + \frac{1}{3}\mathbf{K}_{c_0} + \left(\frac{\eta}{3} - \frac{1}{6}\right)\mathbf{K}_{b_0} \approx (\zeta \cdot 0.11,\ 0.21,\ \eta \cdot 0.11)\ \text{Å}^{-1} \tag{4}$$

where $\mathbf{K}_{a_0}$, $\mathbf{K}_{b_0}$ and $\mathbf{K}_{c_0}$ are the basic vectors of the initial reciprocal lattice deduced from the crystallographic parameters at room temperature [20], $a_0 = 10.05$ Å, $b_0 = 20.55$ Å, $c_0 = 9.93$ Å, $\alpha_0 = 103.6°$, $\beta_0 = 90.5°$ and $\gamma_0 = 93.3°$; the integers ζ and η are equal to -1 or $+1$. The latter uncertainty originates from the fact that the deviation of the real triclinic

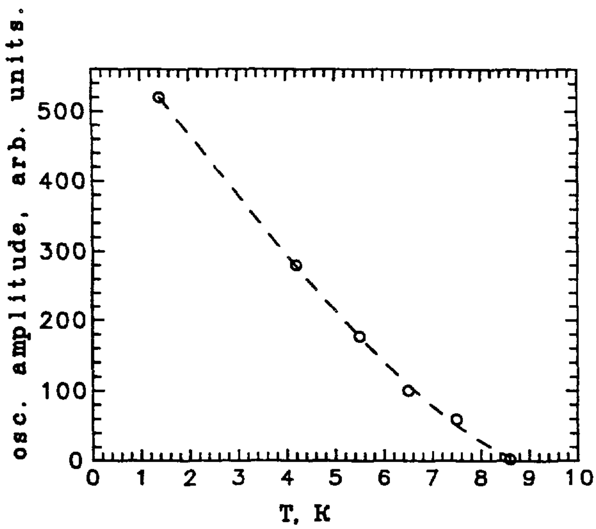


Fig. 3. — Temperatur dependence of the AMRO amplitude. The field is rotated in the plane corresponding to the maximal AMRO, $H = 14$ T.

structure from more symmetrical orthorombic side-centered-A structure with parameters $A = a_0$, $B = 2\, b \sin \alpha_0$ and $C = c_0$ is too small and cannot be resolved within our experimental accuracy. For choosing the correct signs of ζ and η, a precise orientation of the crystal axes in the real coordinate system should be determined.

Up to now, no band structure calculations have been done for the (BEDT-TTF)$_2$TlHg(SCN)$_4$. One can however suggest its FS to be very close to that predicted for the isostructural analog, (BEDT-TTF)$_2$KHg(SCN)$_4$ [11]. In this structure, the value of $2\, k_F^{1D}$ corresponding to the quasi-1D band is about $K_a/6 \simeq 0.1$ Å^{-1} Comparing this value with the Q_x-component (4) and suggesting that a Peierls-type transition (e.g. an SDW transition) occurs at T_p, we naturally propose that

$$Q_x = 2\, k_F^{1D}, \tag{5}$$

so that $\mathbf{Q}$ is the wave vector of the SDW.

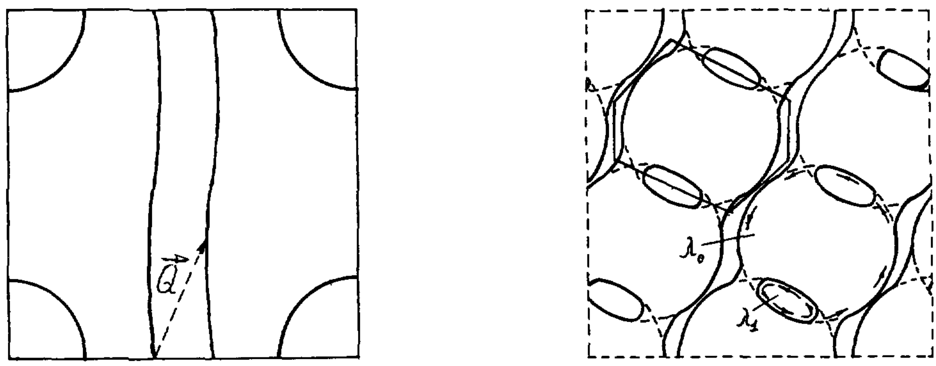
Introducing the vector $\mathbf{Q}$ into the initial reciprocal lattice we obtain a new BZ with the basic vectors,

$$\begin{aligned} \mathbf{K}_a &= \frac{1}{2}\mathbf{K}_{a_0} + \frac{1}{2}\mathbf{K}_{b_0}, \\ \mathbf{K}_b &= \mathbf{K}_{b_0}, \end{aligned} \tag{6}$$

and

$$\mathbf{K} = \mathbf{Q}_c,$$

in which the quasi-1D band disappears under the SDW potential. Taking into account the fact that the periodic SDW potential should also influence the quasi-2D band we find the corresponding transformation of the cylindrical FS sheet. These changes are illustrated in figure 4 in which the FS is shown schematically (a) — above and (b) — below the transition temperature. One can see small « lenslike » cylinders and corrugated planes arising below



$T > T_p$ $T < T_p$

a) b)

Fig. 4. — Transformation of the FS of $(BEDT\text{-}TTF)_2TlHg(SCN)_4$ at the phase transition (schematically). The FS (a) — above and (b) — below the transition ; both figures are in the same scale. The quasi-1D band becomes dielectric below T_p while the cylindrical FS sheet is modified by the superstructure periodic potential to form new open sheets and small cylinders. Thus a magnetic breakdown network with different closed orbits (λ_0, λ_1 and their combinations) arises.

T_p. The new planes lie parallel to the $\mathbf{K}_b$ $\mathbf{K}_c$-plane of the new BZ and form an angle 26° with the crystal **b*** **c**-plane as defined at room temperature. This angle is consistent with the observed $\theta_0 = 24°$ taking into account the experimental error, $\Delta\theta = \pm 2°$, and some variation of the crystal parameters with changing the temperature. The ratio $K_b/K_c = 1.21$ or 1.23 and the angle α between $\mathbf{K}_b$ and $\mathbf{K}_c$ is 64.7° or 67.3° for $\eta = +1$ or -1, respectively. These values are also in agreement with those obtained from the AMRO parameters $(K_b/K_c)_{\text{exp}} = 1.22 \pm 0.02$ and $\alpha_{\text{exp}} = 65° \pm 1°$. So, we conclude that a Peierls-type SDW transition with the commensurate nesting vector **Q** may take place at T_p modifying the electronic structure, so that new open FS sheets arise responsible for the observed AMRO.

It should be noted that an additional series of AMRO has been found, most pronounced at the angles $\theta \simeq \theta_0 \pm 90°$ and $\theta \simeq -25°$ and 155°. In figure 5 the $R(\varphi)$ curve for $\theta \simeq \theta_0 + 95°$ is represented with the upward and downward arrows corresponding to the main and additional AMRO series, respectively. The second series is described by the proposed model if one puts the sign of ζ in (4) to be opposite to that for the main series so that the corresponding superstructure vector, **Q**′, is the mirror reflection of **Q** with respect to the **ab***-plane. The existence of the second AMRO system is most likely associated with the crystal twinning. Such twinning seems to be rather probable since the crystallographic angle β_0 is very close to 90° and the reflection of the **b**-axis with respect to the **ab***-plane is directed approximately along the (**b**-**c**)-direction.

So, the AMRO observed in the $(BEDT\text{-}TTF)_2TlHg(SCN)_4$ indicate a new superstructure in the electronic system arising at low temperatures and most likely associated with the Peierls-type transition (SDW).

Recently an anomalous magnetoresistance kink has been found in $(BEDT\text{-}TTF)_2TlHg(SCN)_4$ at $H > 20$ T [27] very similar to that observed in the K-containing analog [7, 17]. Preliminary measurements of the magnetoresistance anisotropy [27] revealed substantial changes in the AMRO behavior above 20 T. One can suppose that high magnetic field suppresses the antiferromagnetic ordering and restores the high-temperature state. It is worth noting that the extensively investigated $(BEDT\text{-}TTF)_2KHg(SCN)_4$ shares all the characteristic properties with its Tl-containing analog. We believe that more close investigation

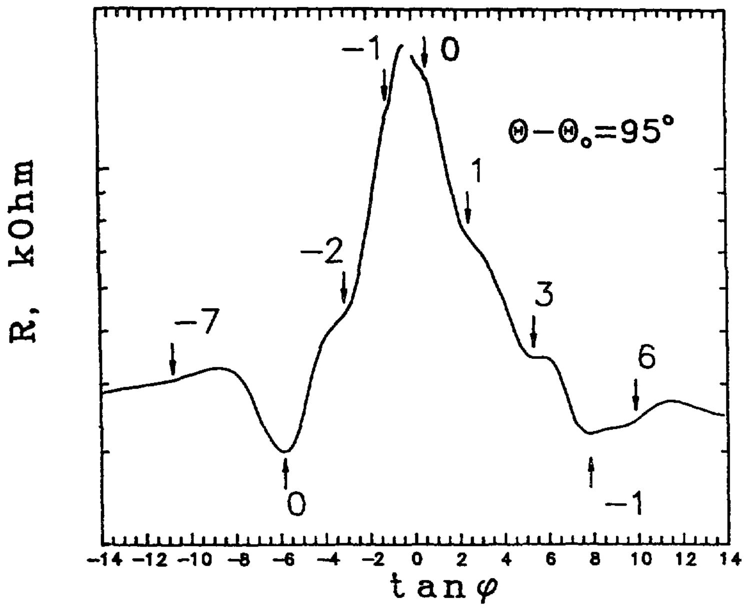


Fig. 5. — Two series of the angular magnetoresistance oscillations at rotating the field in a plane corresponding to $\theta = \theta_0 + 95°$. The up- and downward arrows indicate the main and the additional AMRO systems.

of the AMRO in both compounds in the high-field range is indispensable for clarifying the nature of their electronic state and the unique interplay between the one- and two-dimensional features.

THE QUANTUM OSCILLATIONS OF MAGNETORESISTANCE. — As was reported in [18] the Shubnikov-de Haas oscillations (SdHO) have been observed above 8 T in (BEDT-TTF)$_2$TlHg(SCN)$_4$ corresponding to a weakly corrugated cylinder with its axis along the **b***-direction and the cross-sectional area 6.4×10^{14} cm^{-2} Within our model, the existence of the SdHO below the phase transition may be associated with the magnetic breakdown through small gaps at the edges of the new BZ. In this case, the fundamental harmonic, $F_0 = 670$ T, corresponds to the orbit λ_0 (Fig. 1b).

The breakdown results in a decreasing of the number of electrons moving along the open orbits which are responsible for the AMRO. We observe the saturation of the AMRO amplitude in the field $\geqslant 10$ T. Assuming $H = 10$ T to be breakdown field, we estimate the gap, $E_g \sim 80$ K.

In addition to λ_0, other orbits can exist as well. As was mentioned in [18], surprisingly strong second harmonic, $2F_0 = 1\,340$ T, is observed in the studied compound. The SdHO with highly pronounced second harmonic are represented in figure 6. The frequency of the second harmonic grows perfectly as $1/\cos\varphi$ at tilting the magnetic field (inset b) in Fig. 6). A very similar effect observed in the K-salt was attributed in [15, 16] to the direct observation of the spin-splitting effect. However, the following several facts can hardly be explained within this suggestion :

(i) the direct splitting of the SdH oscillations due to the spin-splitting is usually observed in fields near the quantum limit when the contribution of the highest harmonics becomes significant [26]. In our case the oscillations splitting is observed for both K- and Tl-salts at

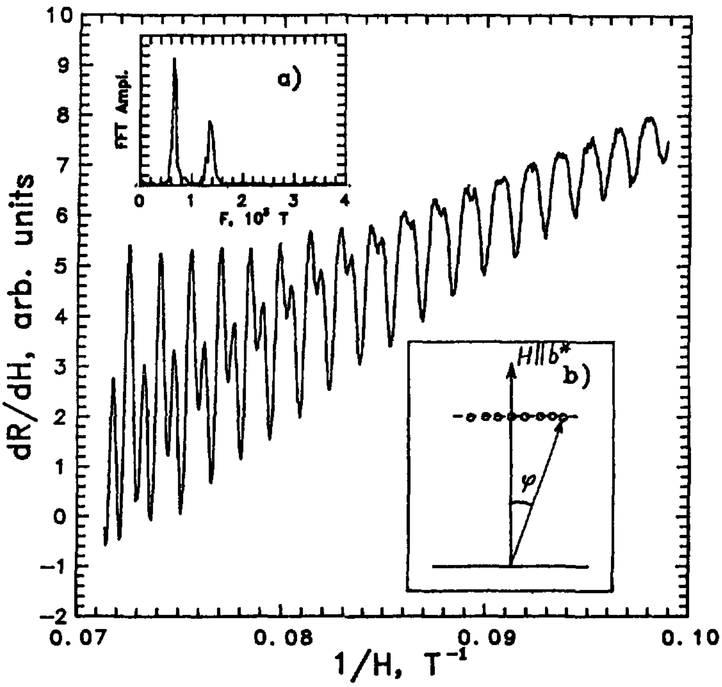


Fig. 6. — Shubnikov-de Haas oscillations at $T = 1.5$ K, the magnetic field is parallel to the **b***-axis. Insets : (a) fast Fourier transform this data and (b) angular dependence of the second harmonic frequency in polar coordinates.

magnetic fields ~ 10 ÷ 15 T i.e. too far from the quantum limit and, despite enormously strong second harmonic, no traces of higher harmonics have been found (see e.g. Fig. 6a) ;

(ii) the distance between the « splitting » oscillation peaks varies as $1/\cos \varphi$ at tilting the field, whereas for the spin-splitting effect one should expect the distance between the peaks corresponding to the opposite spin directions to be independent of the field direction ;

(iii) as is seen e.g. from figure 1 in reference [16] for the K-salt and confirmed by preliminary measurements for the Tl-salt [27], the oscillation splitting is being enhanced up to the field ~ 20 T and diminishes rapidly at further increasing the field, so that no splitting is observed at $H > 23$ T.

The facts (i) and (ii) prove that we deal with the second harmonic of the fundamental frequency, not with direct spin-splitting. One could attribute the low ratio between the fundamental and the second harmonic amplitude to the « spin-splitting zero » of the harmonic ratio [26] as was done in [27] for the $(BEDT\text{-}TTF)_2NH_4Hg(SCN)_2$. Still, in our case the fact (iii) remains unclear within the spin-splitting model.

All the mentioned features can be understood, at least qualitatively, if one considers the magnetic breakdown network presented in figure 4b. The amplitude of the second harmonic of SdHO may be higher for such a network than for the single cylinder because of the additional large orbits, while the next harmonics may be indiscernible due to bigger masses. The increase of the magnetic field results in the increasing probability of the magnetic breakdown and decreasing the contribution of these large orbits. Moreover, as was discussed above fields above 23 T are expected to suppress the low-temperature antiferromagnetic ordering and restore the high-temperature state with the only closed orbit along the cylindrical FS sheet (Fig. 4a).

In addition to F_0 and $2F_0$, another frequency, $F_1 = 860$ T, is observed in the temperature region $2 \div 5$ K. The example of the SdHO with F_1 is presented in figure 7 for $T = 4.2$ K. This frequency may correspond to the sum of the orbits $(\lambda_0 + \lambda_1)$ (Fig. 4b). It should be noted that the amplitude of the harmonic F_1 increase very slowly in comparison with the fundamental with lowering of the temperature, so that its contribution becomes almost negligible at $T < 2$ K (see Fig. 6). This fact can indicate that the corresponding effective mass is very small. The reason for this is not clear yet. Incidentally, the frequency close to F_a was found in the K-containing salt at rather low temperature, $T = 0.5$ K [17], however there was no information about the temperature dependence of the amplitude.

According to the scheme in figure 4b one should expect the SdHO corresponding to the small closed orbits, λ_1. Therefore the small peak at about 200 T in the Fourier transform for 4.2 K (inset in Fig. 7) can be attributed to these orbits. However, the frequency is rather low and the amplitude is too small for an accurate analysis.

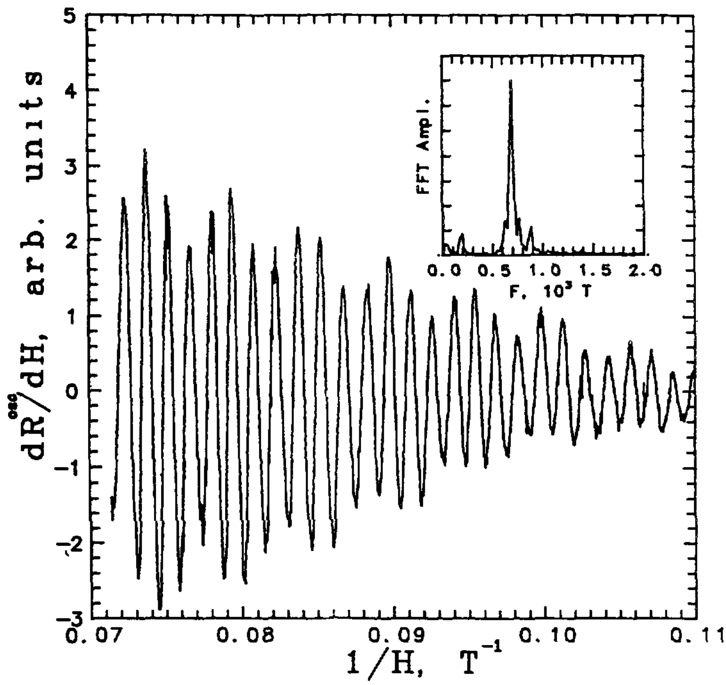


Fig. 7. — Shubnikov-de Haas oscillations at $T = 4.2$ K, magnetic field is parallel to the **b***-axis. The beats are clearly visible, inset represents fast Fourier transform of the data.

Finally, we will consider the dependence of the SdHO amplitude on the angle φ between the magnetic field direction and the **b***-axis. Figure 8 shows the trace of the resistance derivative, $\mathrm{d}R/\mathrm{d}H$, at tilting the field, $H = H_0 + H_1 \cos \omega t$ where $H_0 = 14$ T, $H_1 \approx 8$ mT and $\omega = 120$ Hz, from **b***. The field rotation plane forms an angle $\theta \approx \theta_0 + 90°$ with the **b*** c-plane. (As was mentioned above, this orientation corresponds to the non-oscillating behavior of the semi-classical magnetoresistance.)

The SdHo are clearly observable up to the angles $\varphi = \pm 50°$. The second harmonic contribution is resolved at small φ and decreases rapidly at tilting the field vanishing at $|\varphi| > 18°$. Meanwhile. the fundamental harmonic grows up at these angles and exhibits

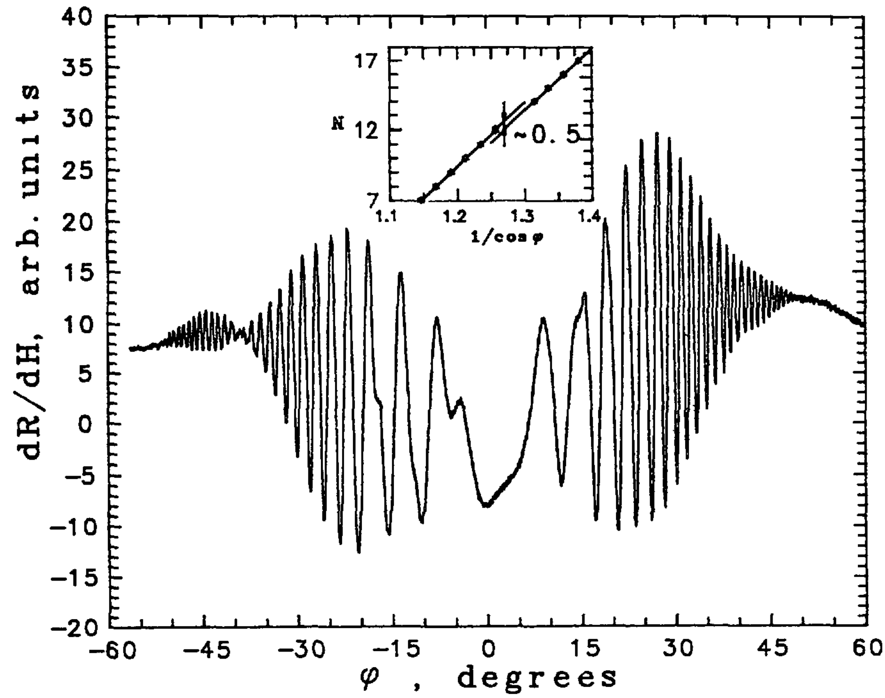


Fig. 8. — Shubnikov-de Haas oscillations at the field, $H = 14$ T, rotating perpendicular to the **ac**-plane, $\theta = \theta_0 + 90°$; $T = 1.5$ K. The inset demonstrates the inversion of the oscillation phase at the node.

maxima at $\varphi \approx \pm 20°$, in agreement with that that was reported in [18]. Besides, a zero of amplitude at $\varphi = -39°$ and a slight bend at $\varphi \simeq 40°$ are observed. The inset in figure 8 demonstrates inversion of the oscillation phase at passing through the amplitude zero. A beating-like behavior is also observed at sweeping the field, for the field directions near $\varphi = -40°$ (Fig. 9). Such a behavior may be considered as a result of a superposition of

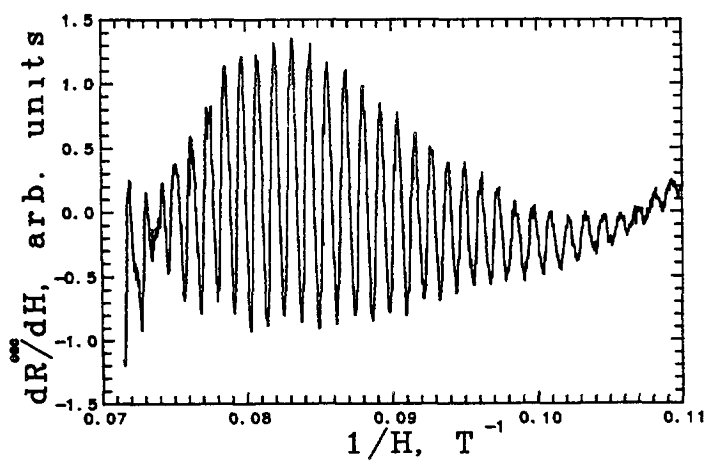


Fig. 9. — The field dependence of the oscillatory part of the resistance derivative, dR/dH, for the field direction corresponding to $\varphi = -38°$ in figure 8, i.e. near the oscillation node.

oscillations with two slightly different frequencies and amplitudes close to each other. If this is the case, the beat frequency, δF, should be rather low. Indeed, at any field direction, we find not more than one node in the whole field interval $(8 \div 14)$ T in which the SdHo are observable. It means that δF cannot exceed $\left(\frac{1}{8}-\frac{1}{14}\right)^{-1} \approx 19$ T that is less than 3 % of the fundamental frequency, F_0. One can naturally attribute these beats to two extreme orbits originated from weak warping of the FS along the $\mathbf{k}_b$-direction. Meanwhile, other possible mechanisms should not be overlooked. In particular, magnetic breakdown circumstances and spin-splitting effect may be essential for the studied compound distinguished for the unusual features of the spin system at low-temperatures.

Summarizing, we have performed detailed magnetoresistance measurements on $(BEDT\text{-}TTF)_2TlHg(SCN)_4$ in the low-temperature state. The analysis of the angle dependent semi-classical magnetoresistance oscillations reveals substantial differences in the electronic structure, in comparison with that expected at room temperature. We argue that a Peierls-type transition with a commensurate wave vector (4) may transform the Fermi surface of the compound, so that new open sheets arise which are consistent with the observed oscillation parameters. Specific features of the Shubnikov-de Haas oscillations, such as extremely strong second harmonic contribution and the existence of an additional harmonic, F_1, appear to be in agreement with the proposed model of the low-temperature state.

Acknowledgements.

We are grateful to Prof. I. F. Schegolev for his support of the work and numerous helpful discussions, Dr. R. P. Shibaeva and Mrs. L. P. Rozenberg for determination of the crystal orientation. We would also like to thank Dr. S. Brazovskii, Dr. M. A. Tanatar, Dr. D. V. Mashovets and Dr. V. N. Laukhin for fruitful discussions.

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