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1994 J. Phys.: Condens. Matter 6 L539

(http://iopscience.iop.org/0953-8984/6/37/001)

J. Phys.: Condens. Matter 6 (1994) L539–L547. Printed in the UK

LETTER TO THE EDITOR

Fermi-surface reconstruction in the organic conductor (BEDT-TTF)$_2$TlHg(SCN)$_4$

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Received 28 June 1994, in final form 21 July 1994

Abstract. Shubnikov–de Haas (SdH) measurements of the two-dimensional organic conductor (BEDT-TTF)$_2$TlHg(SCN)$_4$ have been performed in order to clarify the Fermi surface. At 0.05 K, in addition to the closed orbit and the magnetic breakdown orbit in the original Fermi surface, eight different SdH oscillations are found. All the frequencies follow the same angular dependence. The results are well explained in terms of the reconstruction of the Fermi surface due to spin density wave formation.

Charge transfer salts consisting of (BEDT-TTF) molecules have been extensively studied in an effort to understand the ground states in low-dimensional systems. Among them, the two-dimensional (2D) organic conductor (BEDT-TTF)$_2$MHg(SCN)$_4$ (M = K, Tl, Rb, NH_4) salts have been of particular interest due to their low-temperature properties. The NH_4 salt shows superconductivity at about 1 K [1]. However, the other salts (M = K, Tl, Rb) are metallic down to 0.1 K, and show a magnetic phase transition at low temperatures (T_N = 8–12 K) [2–6]. The ground states of these salts (M = K, Tl, Rb) have been suggested to be spin density wave (SDW) states on the basis of magnetic susceptibility [7, 8] and electron spin resonance measurements [9]. The SDW phase is removed by a high field of about 23 T H_K), where the kink behaviour in the magnetoresistance is seen [2–4, 10–14]. Recently, the possibility of filamentary superconductivity in the SDW phase has been reported for the K and Rb salts [15, 16].

According to band structure calculation, (BEDT-TTF)$_2$MHg(SCN)$_4$ has both 1D and 2D Fermi surfaces (FSs) in the most conducting plane (*ac* plane) as shown in figure 1(a) [17]. For the NH_4 salt, the standard angle-dependent magnetoresistance oscillation (ADMRO) has been found [18]. The observed ADMRO is well explained by the Yamaji effect, which arises from the geometrical property of the single weakly corrugated cylindrical FS. The results are consistent with the observation of a single Shubnikov–de Haas (SdH) oscillation coming from the cylindrical FS for this material. However, for the K and Tl salts, an ADMRO quite different from that observed for the NH_4 salt is found in the SDW phase below T_N [18–21]. It is proposed that the two sheets of the original 1D FS are nested and that the different 1D FS is formed in the new Brillouin zone modified by the superlattice structure due to the nesting [18–21]. The ADMRO results in the SDW phase for the K and Tl salts are interpreted

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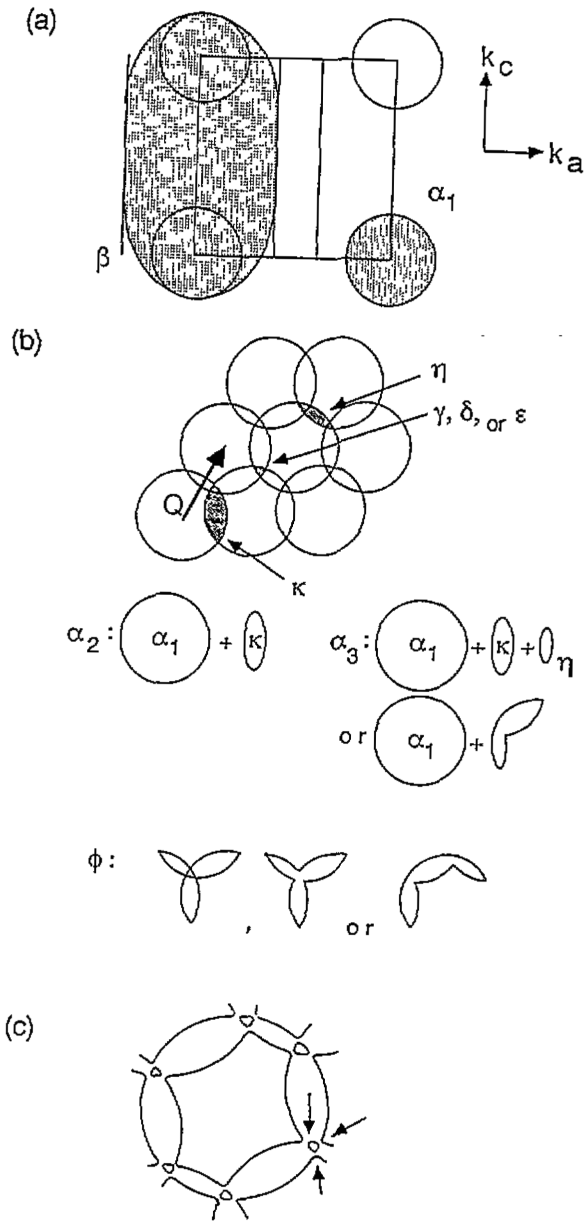

Figure 1. (a) Fermi surface in the normal metallic phase. The shaded parts show the cross-sectional areas corresponding to the α_1 and β oscillations. (b) Schematic picture of the proposed reconstructed Fermi surface in the SDW phase. The nesting vector Q and the orbits assigned to the observed SdH oscillations are shown. (c) Expanded picture of the reconstructed Fermi surface. The arrows show the MB points.

in terms of the drift motion of the conduction electrons on the 1D FS. The striking feature is that the 1D FS in the SDW phase is predicted to be tilted by 20–30° from the b^*c plane [18–21] whereas the original 1D FS in the normal metallic phase above T_N is parallel to the b^*c plane. Such reconstruction of the FS in the SDW phase forms additional closed orbits, whose existence can be verified by quantum oscillation measurements.

To investigate the reconstruction of the FS in the SDW phase, we have made extensive SdH measurements on the $(BEDT\text{-}TTF)_2TlHg(SCN)_4$ salt over a broad range of magnetic field. By applying a field modulation technique, we have found ten different SdH oscillations in the SDW phase. A model of the reconstructed FS is proposed on the basis of the results. Our results suggest that open sheets of FS are absent in the SDW phase, in contrast to the predictions by some groups [18–21].

Figure 2 shows the SdH oscillations for $H \parallel b^*$ at 0.05 K. In the highest-magnetic-field region (figure 2(a)), two fundamental oscillations α_1 and β are evident. The α_1 oscillation ($F(\alpha_1) = 644$ T), which is in good agreement with the reported value [20], is assigned to the closed orbit on the 2D FS predicted by the band calculation (figure 1(a)). The β oscillation arises from the magnetic breakdown (MB) orbit (figure 1(a)) [22, 23], whose cross-sectional area corresponds to ~100% of the Brillouin zone in the *ac* plane. The SdH oscillations observed in the highest-field region are consistent with our previous results [22]. However, below 10 T (figure 2 (b) and (c)), we find new oscillations α_2, α_3, η, κ and ϕ. The amplitudes of these oscillations observed by the field modulation technique are much smaller than those of α_1 and β at higher fields. This is the main reason why they have not been detected by the high-field measurements. In figure 2(d), the low-frequency oscillations δ and ϵ are presented. The power spectrum is calculated by the maximum-entropy method (MEM) [24] to improve the resolution (inset of figure 2(d)). The lowest-frequency oscillation γ, which was reported previously [22], is not evident for this sample. The relative amplitudes of all the oscillations are found to be sample dependent, but the frequencies are sample independent. The sample dependences of the amplitudes may be due to the sample quality and/or the local stress due to the sample cooling process. Similar new oscillations have been found for the K and Rb salts, but not for the NH_4 salt. The new oscillations are observable only for the salts which have the SDW ground states. Therefore, this shows that the observed new oscillations are closely related to the reconstruction of the FS due to the SDW formation.

Table 1. Parameters for the SdH oscillations in $(BEDT\text{-}TTF)_2TlHg(SCN)_4$. $F \parallel b^*$: SdH frequency for $H \parallel b^*$ axis; S/S_{BZ}: ratio of the cross-sectional area to the original Brillouin zone; m_c/m_0: effective mass ratio.

Oscillation	$F \parallel b^*$ (T)	S/S_{BZ}(%)	m_c/m_0
γ	1.2[a]	0.028[a]	0.23 ± 0.3[a]
δ	11.2	0.26	—
ε	12.6	0.30	—
η	38	0.89	—
κ	69	1.6	—
ϕ	183	4.3	2.0 ± 0.1
α_1	664	15.6	1.4 ± 0.1
α_2	720	16.9	1.6 ± 0.1
α_3	768	18.0	2.0 ± 0.1
β	4261	100	3.8 ± 0.1

[a] From [22].

The cyclotron masses of the oscillations were determined by measuring the temperature dependences of the amplitudes in the range between 0.05 K and 0.7 K. The results are listed in table 1. The masses of δ and ϵ cannot be determined because the frequencies are temperature dependent [22]. The amplitudes of η and κ are very small, so reliable values of their masses have not been obtained.

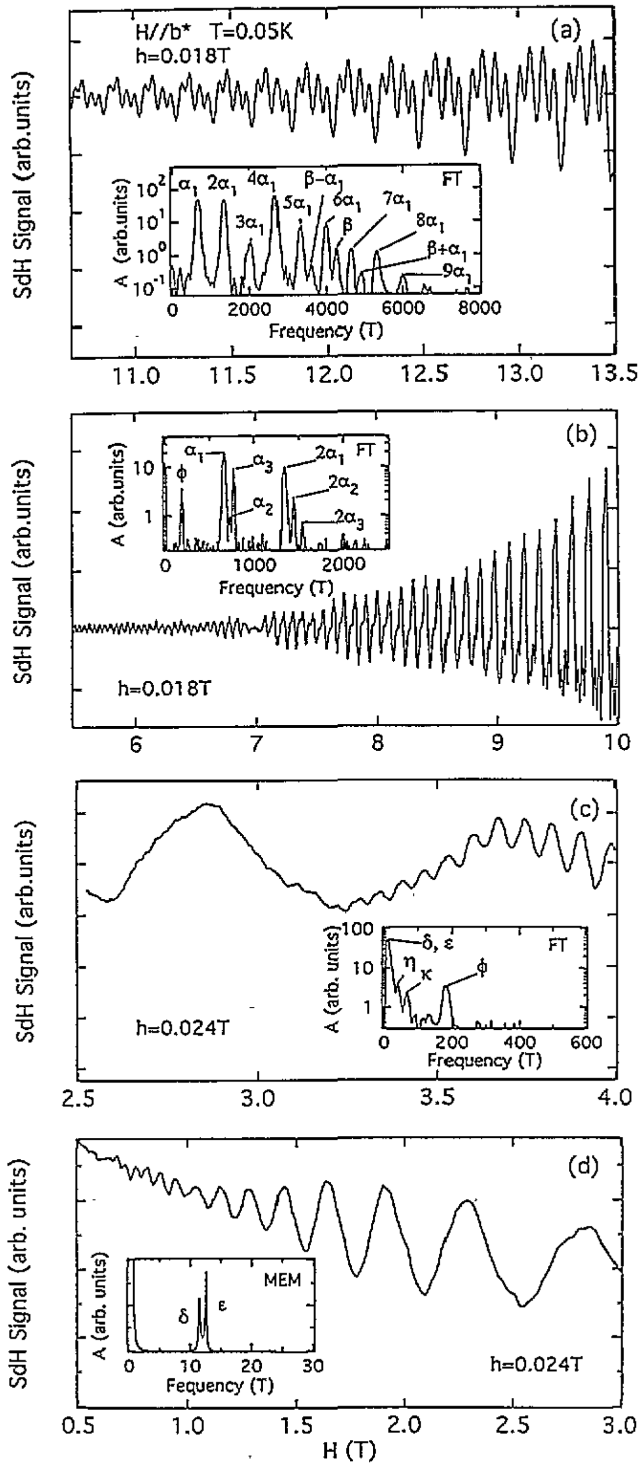

Figure 2. SdH signals at 0.05 K in various magnetic field ranges. The magnetic field is applied parallel to the b^* axis. The modulation field h is 0.018 T or 0.024 T. The insets show the FT or MEM spectra of the signals.

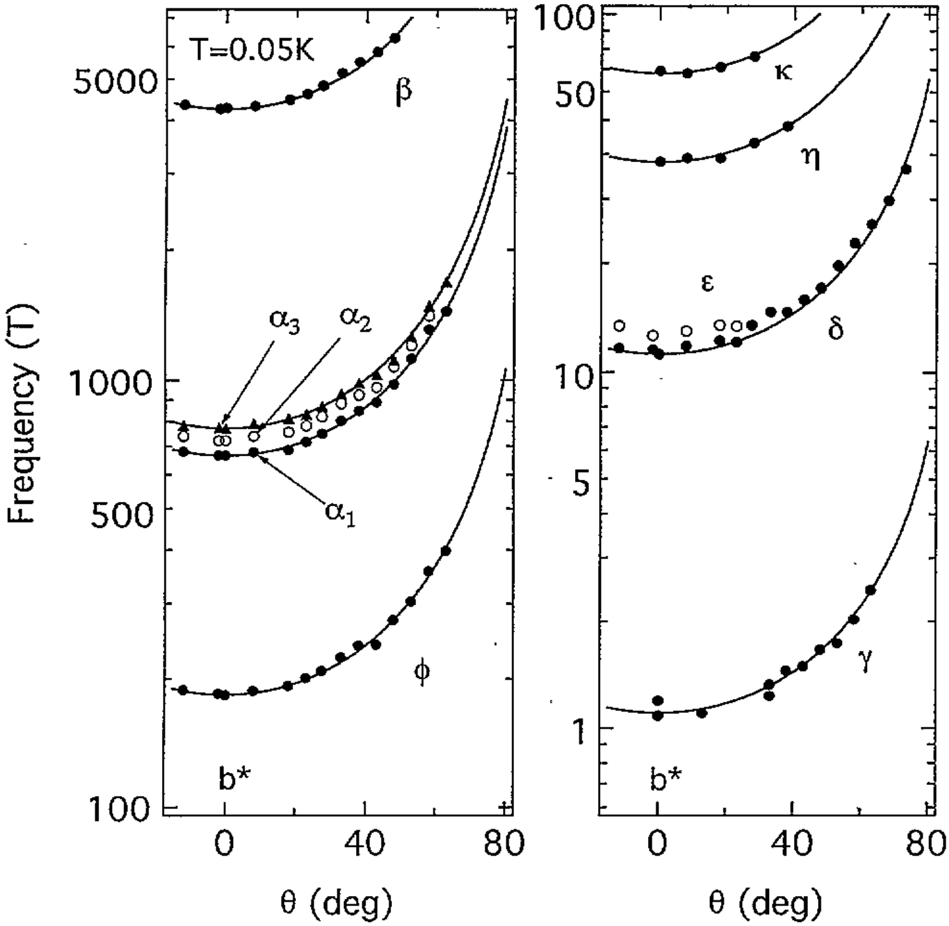

Figure 3. Angular dependences of the SdH frequencies at 0.05 K. The result of γ is from [22].

The angular dependences of all the frequencies are presented in figure 3. We find that all of them follow the $1/\cos\theta$ dependence, where θ is the angle between the magnetic field and the b^* axis. The b^* axis is perpendicular to the conduction plane. The fairly good agreement with the experimental results shows that all the FSs corresponding to the oscillations have an almost perfect cylindrical shape.

Here we note that there are significant relations between the observed frequencies: $F(\alpha_2) \simeq F(\alpha_1) + F(\kappa)$, $F(\alpha_3) \simeq F(\alpha_1) + F(\eta) + F(\kappa)$ and $F(\phi) \simeq F(\eta) + 2F(\kappa)$. The first relation, for instance, suggests that the cyclotron orbit corresponding to the α_2 oscillation is formed by the combination of those corresponding to α_1 and κ. The observation of α_2, α_3 and ϕ is not ascribed to the magnetic interaction effect because other possible oscillations with the combination frequencies $F(\alpha_1) - F(\kappa)$, $F(\eta) + F(\kappa), \ldots$ are not observed. Taking account of the above frequency relations, we propose a possible structure of the reconstructed FS as shown in figure 1(b). The smallest closed orbit is assigned to one of γ, δ and ϵ. The six orbits corresponding to $\alpha_1, \alpha_2, \alpha_3, \eta, \kappa$ and ϕ are the MB orbits. There are two possible orbits for α_3 and three possible ones for ϕ. This reconstruction of the FS is consistent with the fact that all the oscillations have the same angular dependence. For simplicity, the original 1D FS is not shown in figure 1(b). As long as the nesting of the 1D FS is imperfect, small additional closed orbits on the imperfectly nested 1D FS are formed and they should be observed. Therefore, we expect that two of γ, δ and ϵ, which cannot be assigned to the orbits in figure 1(b), arise from the small closed orbits formed on the imperfectly nested 1D FS [22].

For the K salt, Pratt *et al* reported two oscillations with $F_1 = 200$ T and $F_2 = F(\alpha_1) + 50$ T $\simeq F(\alpha_1) + 200$ T, which are sample or cooling method dependent, in addition to α_1 [14]. The two additional oscillations may correspond to ϕ and α_3 in our case. They interpret two of the three oscillations, α_1 and another one with F_2, as arising from two magnetic subbands separated by a magnetic band gap with a size of the order of the exchange energy between the conduction electrons and the SDW. However, we observe the three different oscillations (α_1, α_2 and α_3) with similar frequencies in the same magnetic field range and many other oscillations as shown in figure 2. It is difficult to explain these oscillations solely in terms of their magnetic subband model.

On the other hand, Kartsovnik *et al* reported $\simeq$200 T and $\simeq$860 T oscillations in addition to α_1 in the magnetic field range between 9 T and 14 T for the Tl salt [20]. The 200 T oscillation in Kartsovnik's data seems to correspond to ϕ in our data, but the 860 T oscillation is not seen in our data. They proposed a model of the reconstructed FS via the nesting of the original 1D FS to explain their SdH and ADMRO results. According to their model, the reconstructed FS has open sheets which are tilted by $\simeq 25°$ from the ab^* plane. Similar conclusions have been obtained by a few groups for the K salt on the basis of the ADMRO results [18, 19, 21]. However, it is impossible to explain the oscillations α_2, α_3, γ, δ, ϵ, η, κ and ϕ solely using their nesting model.

Here, we consider the origin of the β oscillation. The β oscillation is observed with α_1, α_2, α_3 and ϕ above $\simeq$7.5 T. The observation of β is evidence that the original 1D FS shown in figure 1(a) still exists in this magnetic field range. Therefore, it is expected that at least two phases, the normal metallic phase having the original FS (figure 1(a)) and the SDW phase having reconstructed FS (figure 1(b)), coexist above $\simeq$7.5 T. For the K salt, an additional phase line in the main SDW state is proposed to exist at ~7 T from the observation of the large hysteresis in the magnetization and magnetoresistance above $\simeq$7 T [3, 14]. Pratt *et al* point out that the degree of magnetoresistance hysteresis correlates with the additional oscillations for the K salt [14]. A similar hysteresis in the magnetoresistance at high fields is reported for the Tl salt [25]. The additional phase line may be understood in terms of a wide distribution of the local stress in the sample. Since the SDW phase is suppressed by pressure [4, 11, 26], a stress-free part of the sample is considered to have the highest critical field ($H_K \simeq 23$ T). A large hysteresis may accompany the transition. On the other hand, another part of the sample may have a lower critical field because of the local stress. If the local stress is distributed over a wide range, we can expect that the hysteretic behaviours in the magnetoresistance and the magnetization will appear in a wide field range from a certain low field up to H_K. In this case, the additional phase line corresponds to the field where the normal phase partially appears. Above the additional phase line, the two phases coexist, which explains the observation of β with $\alpha_1, \alpha_2, \alpha_3$ and ϕ above $\simeq$7.5 T.

The negative slope of the magnetoresistance in the conduction plane above ~10 T for the K salt has been explained in terms of the MB effect on the original FS [23]. A similar negative-slope magnetoresistance is also observed above $\simeq$11.5 T for the Tl salt. The resistance in the normal phase is expected to be smaller than that in the SDW phase. Therefore, if the two phases coexist above the additional phase line, the negative slope may be understood in terms of both effects, the field dependence of the volume ratio of the normal phase to the SDW phase and the MB effect. It is likely that the sample dependences of the field where the magnetoresistance has the maximum and the relative amplitudes of the SdH oscillations are due to the sample-dependent local stress effect.

There is some ambiguity in the proposed FS and the nesting vector shown in figure 1(b), because the precise shapes of the 1D and 2D FSs are still unknown. Recently, the band structure of this system was re-examined in the tight-binding approximation by Ducasse and Fritsch [27]. They point out that the FS is very sensitive to slight changes of the crystal structure. This fact suggests that a band calculation based on the low-temperature crystal structure is necessary to elucidate the FS in the SDW phase in detail.

The important feature of our model is that the reconstructed FS in the SDW phase has no open sheet. The anomalous ADMRO has been observed at fields from 1 T to $\simeq H_K$ for the K and Tl salts [18–21]. The results suggest that the ADMRO comes from the SDW phase and that the main part of the sample is in the SDW phase up to H_K. The models proposed by some groups [18–21], in which the 1D FS in the SDW phase is tilted by 20–30° from the b^*c plane, is not consistent with our reconstruction model. When the reconstructed FS is not simple, a complicated ADMRO is expected to be observed. Further study will be necessary for the understanding of the observed ADMRO results in these salts.

Next, we turn to the field dependences of the oscillation amplitudes. The amplitudes of $\alpha_1, 2\alpha_2$ (second harmonic of α_2), α_3, β and ϕ above 4 T are obtained by performing FT within a 0.17 T^{-1} range of the inverse magnetic field and then are corrected by the Bessel function factor J_2 because of the second-harmonic detection of the modulation frequency [28]. The results are presented in figure 4. The α_2 oscillation cannot be well resolved from α_1 and α_3 because of the limited FT field range. Therefore, the result of the second-harmonic oscillation is plotted for α_2. For γ, δ, ϵ and ϕ below 4 T, the oscillation amplitudes of the recorder trace corrected by J_2 are plotted. The higher-field data for γ, δ and ϵ cannot be obtained because of the steep decrease of the amplitudes due to the factor J_2. It is not clear whether or not the amplitudes of γ, δ and ϵ continue to increase up to higher fields with increasing field.

A few characteristic features are evident in the results. With decreasing field, the amplitude of α_1 shows a steep decrease below $\simeq$7 T and seems to saturate below $\simeq$6 T. For α_3, such anomalies are not so evident. On the other hand, the amplitude of ϕ steeply decreases at $\simeq$7 T and $\simeq$4 T again. The amplitudes of $2\alpha_2$ and β become under the noise level below about 7.5 T. The frequencies of all the oscillations show no appreciable change within the experimental error of 2%. The results suggest that the cross-sectional areas of the FSs corresponding to the oscillations are almost independent of field.

The probability P that the electrons travel along the cyclotron orbit corresponding to each oscillation is written as a function of the MB probability $\exp(-H_0/H)$, where $H_0 = m_c c E_g^2/eh E_F$[28]. E_g and E_F are the energy gap separating the energy bands and the Fermi energy, respectively. The observation of α_1 and ϕ assigned to the MB orbits down to 4 T suggests that H_0 is comparable to 4 T. It is very difficult to calculate the field dependence of the amplitude of each oscillation because the prefactor of the oscillatory term of the conductivity, the Dingle temperature reduction factor and the spin splitting reduction factor [28] are unknown parameters. Since the prefactors change smoothly with magnetic field, the anomalous behaviours of the amplitudes below 7 T cannot be reproduced by the simple Lifshits–Kosevich formula [28] nor by the MB effect. An anomalous field dependence of the amplitude for the additional oscillation is also reported for the K salt [14].

It is well known that the optimum nesting vector in the SDW phase is field dependent, reflecting the instability under the magnetic field [29]. If the nesting of the original 1D FS changes with field, the energy gap E_g also changes. Since the frequency change is not appreciable, the change of the nesting must be small. However, the field dependence of

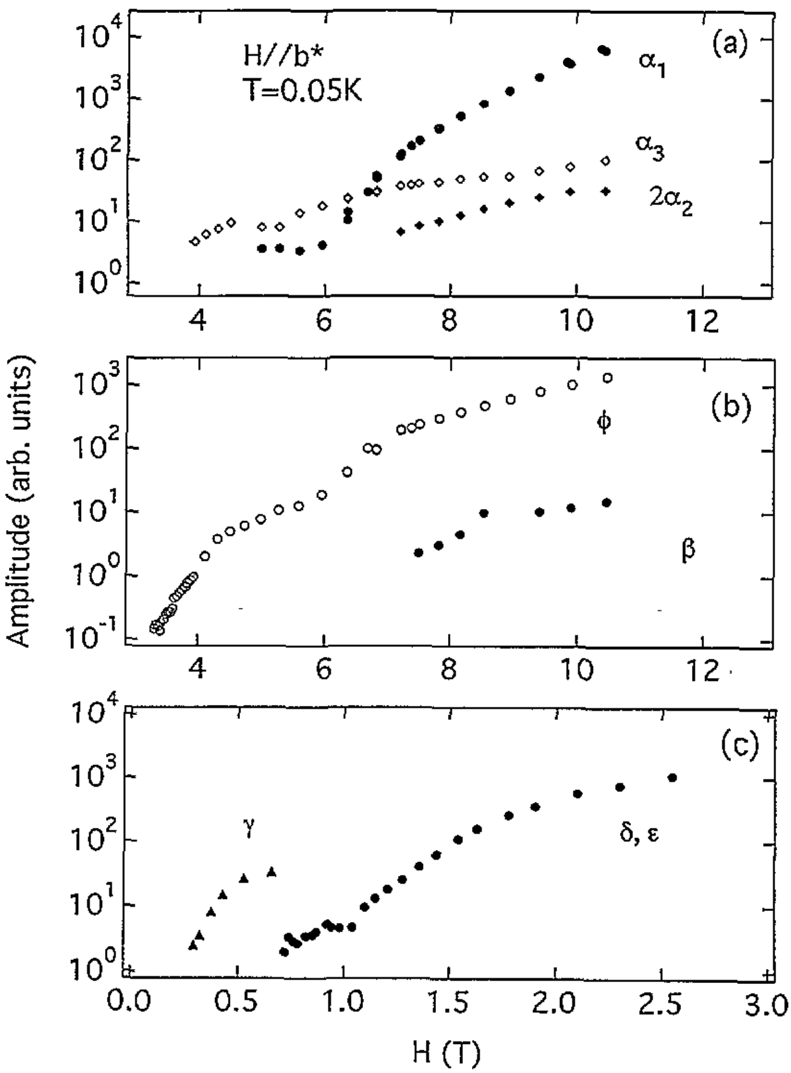

Figure 4. Magnetic field dependences of the oscillation amplitudes for $\alpha_1, 2\alpha_2, \alpha_3, \beta, \phi, \gamma, \delta$ and ϵ. The result for γ is from [22].

the nesting causes a significant variation of the oscillation amplitudes because P depends exponentially on the energy gap E_g. Therefore, the anomalous field dependences may result from the change of the nesting. Anomalous behaviours of the amplitudes of α_1 and ϕ at $\simeq$7 T are probably not related to the additional phase line because both amplitudes increase with increasing field.

We have not found any anomalies in the magnetoresistance at 4 T or 7 T. This result probably suggests that the oscillation amplitudes are much more sensitive to the change of the nesting than the non-oscillatory background of the magnetoresistance.

In conclusion, we proposed the model of a reconstructed FS in the SDW phase for $(BEDT\text{-}TTF)_2TlHg(SCN)_4$. There are no open sheets of the 1D FS in the SDW phase. The SdH oscillations at high fields where β is evident are explained by assuming the coexistence of two phases, the normal metallic phase and the SDW phase. Anomalous field dependences of the oscillation amplitudes are found at ≃4 T and ≃7 T. The behaviours may be related to the instability of the SDW phase under a magnetic field.

The authors appreciate valuable discussion with Dr T Ohno and Dr T Sasaki. This work was done at the High-Magnetic-Field Research Station in NRIM and at Boston University (NSF DMR 92-14889).

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