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Splitting wave form of the magnetic quantum oscillations in α-(BEDT-TTF)$_2$KHg(SCN)$_4$, where BEDT-TTF is bis(ethylenedithio)tetrathiafulvalene

T. Sasaki and N. Toyota
Institute for Materials Research, Tohoku University, Katahira 2-1-1, Aoba-ku, Sendai 980, Japan
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The splitting wave form of Shubnikov–de Haas and de Haas–van Alphen oscillations has been well known to appear in the antiferromagneticlike organic conductor α-(BEDT-TTF)$_2$KHg(SCN)$_4$. We have studied in detail how the separation ratio of the splitting wave form depends on the magnetic field and its orientation. In comparison of these measurements with previously proposed models, it is found that the spin-splitting model taking a positive exchange interaction into account can uniquely explain these results in a consistent way. The physical origin of this positive exchange interaction is discussed from the spin-density-wave picture.

An organic conductor α-(BEDT-TTF)$_2$KHg(SCN)$_4$ was synthesized[1] as a modification of a superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$. However, it was found to show the metallic conduction below 1 K without any superconductivity. The characteristic magnetoresistance measured below 10 K has attracted much attention,[2] which has never been observed in other BEDT-TTF salts. The magnetoresistance, for example, at 0.5 K, rapidly increases with increasing fields and then takes a maximum around 10 T. With a further increase of the field, it decreases continuously up to 23 T where a sharp kink appears, and then increases again.[3,4] The hysteretic behavior is clearly observed for increasing and decreasing fields between 7 and 23 T.[5,6] The anisotropic behavior in the static magnetic susceptibility is observed below 10 K, suggesting that an antiferromagnetic order might develop.[4] This phase transition associates a shoulder-type anomaly in the temperature dependence of the zero-field electrical resistance.[7] We have proposed the magnetic phase diagram on the basis of these measurements.[6]

The Fermi surface is revealed to be a weakly corrugated, cylindrical one by the studies of Shubnikov–de Haas[3–5,8–12] (SdH) and de Haas–van Alphen[10–12] (dHvA) effects at low temperatures. This Fermi surface is considered to be responsible for the quasi-two-dimensional metallic conduction in this salt. The band-structure calculation[13] predicts that a cylindrical Fermi surface is located at the corner of the Brillouin zone and also a pair of corrugated sheets runs along the k_c direction in the k_a-k_c conducting plane. This open sheetlike Fermi surface is likely to be nested together. Therefore, the antiferromagnetic ordering at 10 K has been interpreted as the spin-density-wave (SDW) ordering caused by the nesting.[4]

The splitting wave forms have been observed in SdH (Refs. 4, 5, and 8–12) and dHvA (Refs. 10–12) oscillations below 1 K. In the beginning, the splitting was explained as a *normal* spin-splitting effect[8,9] of the Zeeman splitting of the Landau levels in magnetic fields. However, some curious behaviors were observed,[4] which have not been simply understood. The splitting wave form becomes visible even at relatively low fields where more than fifty Landau levels exist below the Fermi level. This is in strong contrast to the normal spin-splitting effect which is directly observable only near the quantum limit. In addition, it was found in our previous work[4] that the separation ratio δ/Δ, where δ and Δ are the splitting width and the fundamental period of oscillations as a function of H^{-1}, changes in proportion to the inverse of the field. Again, the normal spin-splitting effect hardly explains this field dependence. The following models have been presented to interpret the field dependence. We proposed a model[4] for the spin-splitting effect in terms of modulated Zeeman levels due to an antiferromagnetic exchange interaction. This model could allow for the change of δ/Δ with magnetic fields. A different explanation was proposed by Kang,[14] being based on the quasi-two-dimensional energy dispersion of the system. There exist two extremal (maximum and minimum) orbits on a weakly corrugated cylindrical Fermi surface. The splitting wave form results not from the spin-splitting effect, but from the overlap of two oscillations with slightly different frequencies. Recently, Pratt *et al.*[5] suggested that the SDW might induce a new magnetic superzone along the k_b direction and then two independent cylindrical Fermi surfaces with different cross-sectional areas might appear. The origin of the splitting wave form in this model is the same as with Kang's. The other explanation was proposed by Brooks *et al.*[10] They simulated the Lifshitz-Kosevich formula assuming a field dependence of such parameters as the g factor, the Dingle temperature, and the frequencies.

In the present paper, our results on the field orientation dependence of the separation ratio of the splitting wave forms are discussed with these models.

The single crystals were grown by the usual electrochemical oxidation method.[1,15] The magnetoresistance and magnetization measurements were carried out under static magnetic fields up to 28 T using the hybrid magnet at High Field Laboratory for Superconducting Materials (HFLSM), IMR, Tohoku University. The field orientation dependence of the magnetoresistance was measured using a ^{3}He cryostat with the sample rotation system installed in combination with the 13-T superconducting magnet. For the magnetoresistance measurements, the dc current was applied in the conducting a-c plane. The magnetization was analyzed from the magnetic force measurements on a piece of the single crystal.

Figure 1 shows the oscillatory part of (a) the magnetoresistance (SdH effect) and (b) the magnetization (dHvA

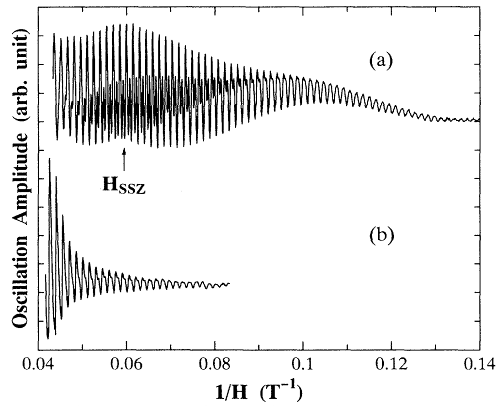

FIG. 1. Splitting wave form of SdH (a) and dHvA (b) oscillations at 0.5 K in the field perpendicular to the plane.

effect) at 0.5 K in the field perpendicular to the a-c plane. Both SdH and dHvA oscillations give the fundamental frequency $F=670$ T exactly equal to each other. The splitting wave forms of both oscillations are clearly visible. A nodelike anomaly is found in the SdH oscillation amplitude at H_{SSZ} of about 16.7 T ($=0.06\ \mathrm{T}^{-1}$).

The characteristic periods Δ and δ are defined as in the inset of Fig. 2; Δ is a fundamental period of oscillations, $0.001\,49\ \mathrm{T}^{-1}$ [$=1/(670\ \mathrm{T})$], and δ is a separation width between the peaks of the splitting wave form. Figure 2 shows the separation ratio δ/Δ as a function of H^{-1}. The open and filled circles denote the data from SdH and dHvA oscillations, respectively. Although the data from dHvA oscillations are fairly scattered, the increase of δ/Δ found in SdH oscillations is reproduced in dHvA oscillations as shown in the figure. The fitting straight line to SdH data below $0.08\ \mathrm{T}^{-1}$ is

$$\delta/\Delta=0.72(\pm 0.01)-3.63(\pm 0.01)/H\ , \tag{1}$$

where H is in units of tesla.

Figure 3 shows SdH oscillations at 0.5 K in magnetic fields tilted by θ from the perpendicular direction to the a-c plane. The splitting becomes unclear with increasing θ and then vanishes around $\sim 25°$. As seen in the upper part of Fig. 4, the fundamental frequency F follows a $1/\cos\theta$ dependence, $F=670/\cos\theta$ (solid line), which is

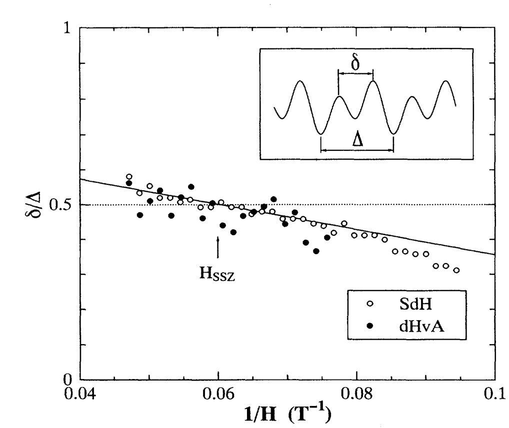

FIG. 2. Separation ratio δ/Δ plotted as a function of $1/H$. The inset shows the definition of δ and Δ.

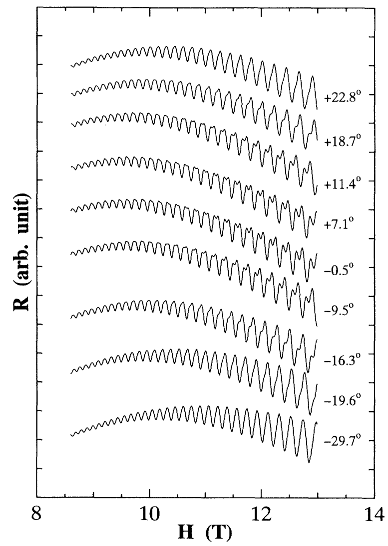

FIG. 3. Field orientation dependence of SdH oscillations at 0.5 K. The angle shown in the figure is measured from the perpendicular direction to the conducting plane.

expected from the cylindrical Fermi surface. In addition, the effective mass m^*, which is obtained from the temperature dependence of the oscillation amplitude, could be described as $m^*=1.4m_0/\cos\theta$ (broken line), where m_0 is the free electron mass.[15] The lower part of Fig. 4 shows the θ dependence of δ/Δ around maximum field 13 T. It is found that δ/Δ becomes small with increasing θ.[16] The solid line shows a least-square-fitting curve with the inverse of $\cos\theta$,

$$\delta/\Delta=1.98(\pm 0.01)-1.55(\pm 0.01)/\cos\theta. \tag{2}$$

With these experimental equations (1) and (2), we shall discuss which model(s) could be consistent with the experiments. First of all, we start to examine the model[5,14] assuming two different closed orbits with very close fre-

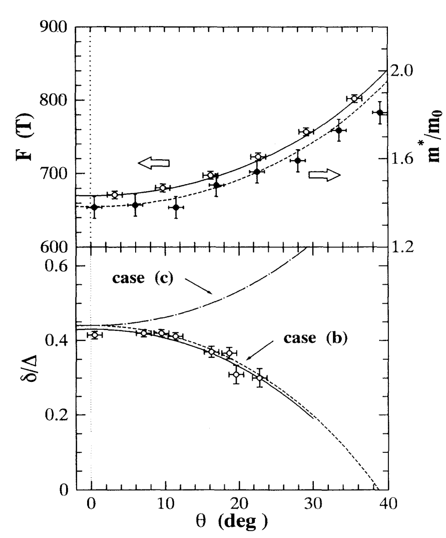

FIG. 4. Field orientation dependence of the SdH frequency, effective mass, and separation ratio. The lines in the figure are described in the text.

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quencies. In this case, the field dependence of δ/Δ can follow a H^{-1} dependence as Eq. (1) and the coefficient will be obtained to be 3.63 when the difference of two frequencies is assumed to be 3.63 T. However, this model does not show any field direction dependence, because both frequencies must follow simultaneously the $1/\cos\theta$ dependence and the g factor is found to be isotropic by electron spin resonance measurements.[17]

Next, we reexamine our previous model[4] of Zeeman splitting of Landau levels and its modulation by a magnetic exchange interaction;[18] normal Zeeman splitting without the modulation [case (a)], Zeeman levels shifted by the positive exchange interaction ($J>0$) [case (b)], and by the negative exchange interaction ($J<0$) [case (c)]. Figure 5 illustrates these three cases. In a magnetic field, each electron energy level (Landau level), separated by $\hbar\omega_c$, splits into a set of spin-up and spin-down levels (Zeeman levels) separated by $g\mu_B H$ $[=(e\hbar H)/(m_0 c)]$. Each of these Zeeman levels gives dHvA or SdH oscillations as they move relative to the Fermi level with field. The oscillations have the same frequency and, generally, the same amplitude, but with phase difference δ/Δ $[=(g\mu_B H)/(\hbar\omega_c)=\frac{1}{2}g(m^*/m_0)]$ which is independent on H. This is just case (a). If we postulate a positive or negative exchange interaction between a conduction electron and magnetic moment, the Zeeman splitting is enhanced or reduced, respectively, by ΔE as shown in Fig. 5(b) or 5(c). As a result, the phase difference between spin-up and spin-down oscillations becomes

$$\delta/\Delta=[2\hbar\omega_c-(g\mu_B H+\Delta E)]/\hbar\omega_c \quad \text{for case (b)} \qquad (3)$$

or

$$\delta/\Delta=[(g\mu_B H-\Delta E)-\hbar\omega_c]/\hbar\omega_c \quad \text{for case (c)}\ , \qquad (4)$$

where δ is chosen to be consistent with the definition as shown in Fig. 2. To discuss the field dependence, we may rewrite them for convenience as

$$\delta/\Delta=2-\tfrac{1}{2}(g+H_{ex}/H)m^*/m_0 \qquad (5)$$

$$=(2-\tfrac{1}{2}gm^*/m_0)-\tfrac{1}{2}(H_{ex}/H)(m^*/m_0) \quad \text{for case (b)} \qquad (6)$$

or

$$\delta/\Delta=\tfrac{1}{2}(g-H_{ex}/H)m^*/m_0-1 \qquad (7)$$

$$=(\tfrac{1}{2}gm^*/m_0-1)-\tfrac{1}{2}(H_{ex}/H)(m^*/m_0) \quad \text{for case (c)}\ , \qquad (8)$$

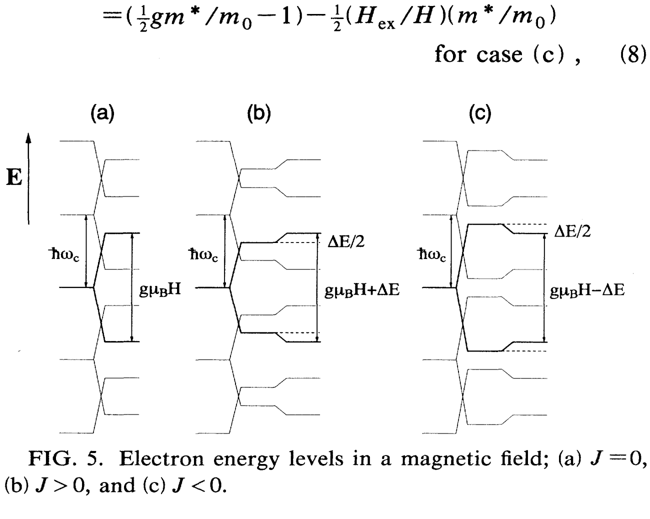

FIG. 5. Electron energy levels in a magnetic field; (a) $J=0$, (b) $J>0$, and (c) $J<0$.

where the exchange field H_{ex} is defined by $H_{ex}\equiv\Delta E/\mu_B$. Both cases (b) and (c) reproduce Eq. (1) when $H_{ex}=5.19$ T and $g=1.83$ [case (b)] or 2.46 [case (c)] using $m^*=1.4m_0$.

The magnetic-field orientation dependence of Eqs. (6) and (8) is attributed to the orientation dependence of the effective mass, $m^*=1.4m_0/\cos\theta$ as shown in Fig. 4. Substituting this relation into Eqs. (5) and (7), we get

$$\delta/\Delta=2-1.56/\cos\theta \quad \text{for case (b)} \qquad (9)$$

or

$$\delta/\Delta=1.44/\cos\theta-1 \quad \text{for case (c)}\ . \qquad (10)$$

These two different curves are shown in Fig. 4 by the broken and dashed-dotted lines, respectively. As clearly seen, case (b) satisfies the experimental formulas (1) and (2) simultaneously. In this case, the effective g factor changes with field as $\tilde{g}=g+H_{ex}/H=1.83+5.19/H$.

In our previous model,[4] only case (c) was discussed because of the lack of data for the field orientation dependence of δ/Δ. As described above, however, the case that the Zeeman levels are modulated by a negative exchange interaction is inconsistent with the orientation dependence.

Using the thus obtained $\tilde{g}$ and m^*, we discuss the spin-splitting-zero condition. In the normal case, the spin-splitting zero can be observed only as a function of the field orientation in a constant field. When gm^*/m_0 is an odd integer, the amplitude A ($\propto\cos[(\pi g m^*)/(2m_0)]$) of SdH or dHvA oscillation becomes zero or small when the spin-dependent scattering leads to the same or different amplitudes, respectively, between spin-up and spin-down components.[19] However, since the g factor depends on H effectively, the spin-splitting zero is expected to be observable also as a function of magnetic field. In our case, it might appear when the equation

$$(g+H_{ex}/H)m^*/m_0=2n+1 \quad (n=0,1,2,\dots) \qquad (11)$$

can be satisfied with a series of $H=H_{SSZ}$ and m^*. In the field perpendicular to the plane, we obtain a series of $H_{SSZ}=16.6$ T ($n=1$), 3.0 T ($n=2$), 1.6 T ($n=3$), ... At the maximum field of 16.6 T where $\delta/\Delta=0.5$, the node with finite amplitude is found on the envelope of SdH oscillations (Figs. 1 and 2). When the oscillatory wave is deformed due to the higher harmonics, the spin-splitting zero does not show the node with zero amplitude. In a constant field, for example, at 13 T, the spin-splitting zero might be observable as a function of field orientation at $\theta=51.4°$ ($n=2$), 63.5° ($n=3$), 69.7° ($n=4$), . . . , where the effective mass varies as $m^*=1.4m_0/\cos\theta$. The SdH oscillations at 13 T (Fig. 2 in Ref. 7) have been observed as a function of θ only up to 40°. Therefore it cannot be checked whether or not the spin-splitting zero could be observed at these characteristic angles. Very recently, Kartsovnik, Kovalev, and Kushch[20] reported the detailed SdH oscillations in isostructural (BEDT-TTF)$_2$TlHg(SCN)$_4$ which showed very similar magnetotransport properties[21] to the present K salt. In the angle-dependent SdH oscillations at 14 T, a clear node where the amplitude is nearly zero is found at $\theta\simeq40°$ and the inversion of the phase is also observed.

Moreover, at this angle, a node of beatlike behavior appears on SdH oscillations as a function of field at about 13.5 T. These observations are consistent with the present spin-splitting-zero model, and the difference in the angle and field might be attributed to the slight difference of the parameters g, m^*, and H_{ex} between two salts.

Finally, the physical meaning of case (b) is considered briefly. The results of the magnetic susceptibility measurements evidence an antiferromagnetic ordering below 10 K with easy axis in the conductive plane.[4] This ordering is most likely due to a SDW caused by the nesting of a pair of planar Fermi surfaces. Recently, we have found from Hall resistance measurements[22] that the carrier density steeply decreases below 10 K. This decrease is consistent with an opening of the SDW gap below the magnetic phase transition temperature. We try to consider the splitting wave form in comparison with other magnetic system. In the dilute Kondo system Cu(Cr),[23] an anomalous field dependence of the amplitude of dHvA oscillations was observed and explained as being due to an antiferromagnetic exchange splitting of Zeeman levels associated with each Landau level. A similar effect was found in Pd(Co) (Ref. 24) with the ferromagnetic exchange interaction. The spin-splitting effect of dHvA oscillations in these systems was analyzed in terms of an *s-d* exchange interaction between the impurity and conduction electrons, which were not directly visible in contrast to the present splitting. In the present system, the SDW moment which lies in the conductive plane in zero field may be considered to induce an internal mean field. The response of the internal mean field to the external field has not been well known. If we postulate that the internal mean field due to SDW could be regarded as an impurity spin in dilute Kondo alloys, the energy ΔE presented here as a parameter might be considered as an exchange interaction energy between SDW and the spin-up and spin-down conduction electrons on the cylindrical Fermi surface. The exchange energy may be written as a form of $J\langle S\rangle\sigma$, where $\langle S\rangle$ and σ are the spatial average of the SDW moment and conduction carriers, respectively. In the magnetic field, σ is parallel or antiparallel to the field. Thus the component of $\langle S\rangle$ parallel to the field may be important. It is derived from our analysis that the parallel component changes little both with field below the magnetoresistance kink field ($H_A \simeq 23$ T) and its direction up to $\theta \simeq 25°$ within an experimental accuracy. This interaction is considered to be isotropic in the conducting plane for the Landau quantized carriers cyclotron orbiting on the cylindrical Fermi surface. It is also noted that the exchange energy ΔE is about 3.5 K, which is of the order of the transition temperature about 10 K.

Still, the detailed magnetic properties in high field need not only further theoretical consideration but also more experiments. Finally, we point out that the magnetic interaction effect[25] must have little influence on the analysis of the oscillation wave form, because the magnetization is so small[4,12] that the magnetic interaction effect could be neglected.

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