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# Spin splitting at the high-magnetic-field phase transition of the organic conductor $\alpha$ -(BEDT-TTF)<sub>2</sub>KHg(SCN)<sub>4</sub>

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Spin splitting of the de Haas–van Alphen (dHvA) effect is studied in the quasi-two-dimensional organic conductor  $\alpha$ -(BEDT-TTF)<sub>2</sub>KHg(SCN)<sub>4</sub> by means of the magnetic torque method. The magnetic field direction dependence of the dHvA oscillation amplitude shows that the spin-splitting zero angles change at 23 T, where the phase transition magnetic field between the low-temperature, low-magnetic-field phase [antiferromagnetic (AF) phase] and the high-magnetic-field phase (M\* phase) is. This change implies the increase of the  $g(m_c/m_0)$  value in the AF phase, where  $g$  is the  $g$  value,  $m_c$  and  $m_0$  the cyclotron effective and free electron masses, respectively. The enhancement of the electron-electron interaction may be the origin of the large  $g(m_c/m_0)$  value in the AF phase. This result supports a model which requires a modification of the Lifshitz-Kosevich formulation in the AF phase in order to evaluate the effective mass and Dingle temperature correctly. In addition, anomalous structures on the magnetic torque curves are observed in both the AF and the M\* phases. Each structure may correspond to the subsequent phase transitions by changing the magnetic field direction. [S0163-1829(99)13921-3]

## I. INTRODUCTION

An isostructural family of the organic conductor  $\alpha$ -(BEDT-TTF)<sub>2</sub>MHg(XCN)<sub>4</sub>, where BEDT-TTF is bis(ethylenedithio)tetrathiafulvalene, has been the subject of intensive study owing to a variety of the ground states;<sup>1</sup> metals [ $X$ =Se and  $M$ =K (Ref. 2) or Tl (Ref. 3)], superconductor<sup>4</sup> ( $X$ =S and  $M$ =NH<sub>4</sub>) and antiferromagnetic metals<sup>5</sup> ( $X$ =S and  $M$ =K, Tl or Rb). Among them, much attention has been focused on the antiferromagnetic metals which have unique antiferromagnetic [(AF) phase] state below  $T_A = 8 - 10$  K in zero magnetic field and the magnetic-field-induced successive phase transitions.<sup>6</sup> The transition from the Pauli paramagnetic phase (P phase) to the AF phase at  $T_A$  is thought to be a spin-density-wave (SDW) transition.<sup>7,8</sup> The topology of Fermi surface (FS) obtained by the band structure calculations<sup>9</sup> in the P phase, which consists of a pair of open-planer and closed-cylindrical ( $\alpha$ -orbit) parts, is likely to possess a nesting instability for a periodic modulation such as a SDW. The Brillouin zone is reconstructed by the new nesting periodicity,<sup>10</sup> and then the cylindrical  $\alpha$  orbits are also reconstructed to the multiconnected orbit in the extended Brillouin zone. This reconstructed FS is explained well by detailed studies of angle-dependent magnetoresistance oscillations (ADMRO).<sup>10-13</sup> Many experiments on Shubnikov–de Haas (SdH) and de Haas–van Alphen (dHvA) effects also support the reconstruction model.<sup>10,14</sup> The main oscillation frequency of 670 T in the AF phase, which is the same frequency as the  $\alpha$  orbit expected in the P phase, originates in a magnetic breakdown (MB) orbit on the multiconnected  $\alpha$  orbit by the nesting periodicity.<sup>10</sup>

At a high magnetic field of about 23 T in the case of

$\alpha$ -(BEDT-TTF)<sub>2</sub>KHg(SCN)<sub>4</sub>, a sharp kink structure of the magnetoresistance appears.<sup>15</sup> The kink structure has been considered as a phase transition between the AF and P phases, that is, the P phase is restored from the AF phase by applying high magnetic fields.<sup>16</sup> Recent experiments in  $\alpha$ -(BEDT-TTF)<sub>2</sub>KHg(SCN)<sub>4</sub>, however, point out that the high-magnetic field phase above 23 T is an unknown magnetic metal phase (M\* phase) which is different than both the AF and P phases.<sup>17,18</sup> In the M\* phase, the magnetic torque shows hysteresis with sweeping magnetic field and temperature.<sup>19,20</sup> From the view point of the FS topology, ADMRO measurements show that FS in the M\* phase is different than that in the AF phase, and similar to that in the P phase,<sup>13</sup> while SdH and dHvA oscillations have the same main frequency of 670 T in both the AF and M\* phases.<sup>15,17</sup> But it is noted that the magnitude of higher harmonics of the SdH and dHvA oscillations in the M\* phase is rather reduced in comparison with that in the AF phase.<sup>21,22</sup> The change of the effective mass  $m_c$  and/or the  $g$  value in the AF and the M\* phases has been proposed to explain the change of the higher harmonics content. In addition, there is a controversy about  $m_c$  and Dingle temperature  $T_D$  obtained by SdH and dHvA experiments.<sup>23</sup> The effective mass in the AF phase has been reported to be smaller than that in the M\* phase. Recently, it is pointed out that the effective mass in the AF phase might be underestimated<sup>23</sup> because of omitting the additional temperature dependent factor to the standard Lifshitz-Kosevich (LK) formulation.<sup>24</sup> The MB gap in the AF phase, which should have a temperature dependence, is suggested to be the possible origin. In the same way, the reconsideration of the Dingle temperature is required.<sup>17</sup> As is described above, three different phases, the AF, M\*, and P

phases, are recognized as the antiferromagnetic (SDW) metal in low-temperature, low-magnetic field ( $T < T_A, H < 23$  T), unknown magnetic metal ( $T < T_A, H > 23$  T), and paramagnetic metal ( $T > T_A$ ) phases, respectively, in  $\alpha$ -(BEDT-TTF)<sub>2</sub>KHg(SCN)<sub>4</sub>. The microscopic electronic state and parameters such as the effective mass, the  $g$  value and so on, in each phase are not yet well known because of the complex phase transitions.

In this paper, we present the magnetic field direction dependence of the dHvA oscillation amplitude in both the AF and M\* phases of  $\alpha$ -(BEDT-TTF)<sub>2</sub>KHg(SCN)<sub>4</sub>, the P phase of  $\alpha$ -(BEDT-TTF)<sub>2</sub>KHg(SeCN)<sub>4</sub>, and also the P phase of  $\alpha$ -(BEDT-TTF)<sub>2</sub>NH<sub>4</sub>Hg(SCN)<sub>4</sub>, which is measured by the magnetic torque method. On the basis of the spin-splitting-zero (SSZ) analysis, the effective mass  $m_c$  and the  $g$  value are evaluated in each phase. The main finding of this paper is that the  $gm_c$  value in the AF phase is larger than that in the M\* phase of  $\alpha$ -(BEDT-TTF)<sub>2</sub>KHg(SCN)<sub>4</sub>, which is an opposite result compared with the former expectation. This finding suggests that the electron-electron interaction may be enhanced in the AF phase because of smaller carrier density owing to an opening of the SDW gap. Finally, we will mention about anomalous magnetic torque structures observed in addition to the dHvA oscillation. These structures may be related to the subsequent transitions of the magnetic phases or the change of spin structure depending on the magnetic-field orientation.

## II. EXPERIMENT

Single crystals of  $\alpha$ -(BEDT-TTF)<sub>2</sub>MHg(XCN)<sub>4</sub> ( $M=K$  or NH<sub>4</sub>, and  $X=S$  or Se) were grown by the electrochemical oxidation method.<sup>2,17</sup> The crystals, with a typical size of a few mm<sup>2</sup>  $\times$   $\sim$  0.3 mm, were grown on a platinum anode with a constant current of 0.5  $\mu$ A. The well developed facet is the crystallographic  $a$ - $c$  plane.

The magnetic torque was measured with a capacitive cantilever beam torque meter. The capacitor consists of a circular plate (3 mm  $\phi$ ) as the moving electrode, which is suspended by a narrow beam (0.2 mm  $\times$  4 mm) made of thin beryllium copper (50  $\mu$ m), and the sapphire plate with gold film sputtered as the ground plate. The moving electrode and the ground plate are surrounded by a small metallic case as is used for the capacitive guard. This unit can be rotated smoothly in the magnetic field and at low temperature down to about 0.5 K. The typical rotation speed in the present experiment was about 180° per hour. A single crystal was fixed on the moving plate with a small amount of grease. The capacitance was measured by using a decade capacitance bridge (General Radio 1615A) and a lock-in-amplifier (PAR 124A). The magnetic torque data presented in this paper were corrected by subtracting the torque curve in zero magnetic field and at the same temperature. The curve in zero magnetic field shows a  $\cos \theta$  dependence owing to moving of the electrode by the weight of both the sample and the electrode, where  $\theta$  is the angle between the magnetic field direction and normal to the capacitive plate (or the  $a$ - $c$  plane of the single crystal).

Magnetic torque measurements were carried out using 30 T hybrid magnet at the High Field Laboratory, IMR, Tohoku University. The temperature in magnetic field was measured

by using a Cernox thermometer (Lake Shore Cryotronics, Inc.) which was calibrated by a capacitance thermometer.

## III. de HAAS-van ALPHEN OSCILLATIONS ON THE MAGNETIC TORQUE

The magnetic torque per unit volume is given by the expression  $\tau = \mathbf{M} \times \mathbf{H}$ , where the magnetization is related to the susceptibility tensor  $\hat{\chi}$  as  $\mathbf{M} = \hat{\chi} \mathbf{H}$ . The torque in the  $a$ - $b$  plane is described as

$$\tau = \frac{1}{2} H^2 (\chi_{aa} - \chi_{bb}) \sin 2\theta, \quad (1)$$

where  $\chi_{aa}$  and  $\chi_{bb}$  are diagonal elements of  $\hat{\chi}$ . Here, the off-diagonal elements are neglected and the rectangular coordinate axes are applied for simplicity. The magnetic oscillations of the torque appear through the oscillation of  $\chi_{bb}$  in the present case. The first harmonic of the oscillation part of the torque on the basis of the semiclassical Lifshitz-Kosevich (LK) theory<sup>24</sup> is given by

$$\begin{aligned} \tau_{\text{osc}} &= -\frac{1}{F} \frac{dF}{d\theta} M_{\text{osc}} H, \quad (2) \\ M_{\text{osc}} &\propto TFH^{-1/2} \frac{\exp[-\lambda(m_c/m_0)T_D/H]}{\sinh[\lambda(m_c/m_0)T/H]} \\ &\times \cos[\pi g(m_c/m_0)/2] \sin[2\pi(F/H - 1/2) \pm \pi/4], \quad (3) \end{aligned}$$

where  $\lambda \equiv 2\pi^2 m_0 c k_B / e \hbar = 14.69$  T/K,  $c$  is the light velocity,  $k_B$  the Boltzmann's constant,  $\hbar$  the Planck's constant,  $g$  is the electron  $g$  value, and  $T_D$  is the Dingle temperature. The extremal cross-sectional area  $A$  of the FS in the plane normal to the applied magnetic field  $H$  is obtained in the relation  $F = (c\hbar/2\pi e)A$ . In the same way as the magnetic oscillations with the period of  $1/H$ , it is noted that the oscillation with a period of  $1/\cos \theta$  in constant magnetic field are superimposed on the  $\sin 2\theta$  background torque curve in the case of the two-dimensional (2D) FS with  $F(\theta) = F(0^\circ)/\cos \theta$ . This kind of oscillation has the amplitude nodes at the magnetic field directions where the spin-splitting-zero conditions  $g(m_c/m_0) = 2n + 1$  ( $n = 0, 1, 2, \dots$ ) are satisfied. In the case of 2D FS, the SSZ's appear with a period of  $1/\cos \theta$  because the angle dependence of the effective mass is expected as  $m_c(\theta) = m_c(0^\circ)/\cos \theta$ .

## IV. EXPERIMENTAL RESULTS

### A. $\alpha$ -(BEDT-TTF)<sub>2</sub>KHg(SeCN)<sub>4</sub>: Normal metal

Figure 1 shows the magnetic torque curves in a small single crystal (0.52 mg) of  $\alpha$ -(BEDT-TTF)<sub>2</sub>KHg(SeCN)<sub>4</sub> at  $T = 0.52$  K. Background torque curves show the  $\sin 2\theta$  behavior, which is indicated by broken curves. The inset figure demonstrates the  $H^2$  dependence of the torque amplitude of the  $\sin 2\theta$  curves, which are described in Eq. (1). Clear dHvA oscillations are superimposed on the background torque curves, and are periodic in  $(\cos \theta)^{-1}$ . The periodicity in  $(\cos \theta)^{-1}$  results from the cylindrical FS with the cross sectional area corresponding to  $F(0^\circ) = 670$  T. This observation

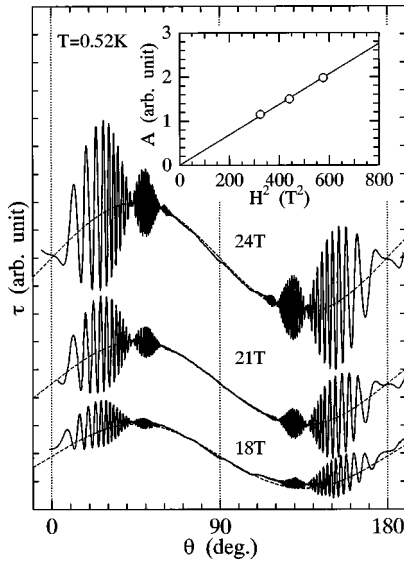


FIG. 1. Magnetic torque curves of  $\alpha$ -(BEDT-TTF) $_2$ KHg(SeCN) $_4$ . The broken curves show the  $\sin 2\theta$  dependence resulting from the anisotropy of the static susceptibility as described in Eq.(1). The inset demonstrates the  $H^2$  dependence of the torque amplitude.

is consistent with band structure calculations<sup>2</sup> and in good agreement with previous SdH (Ref. 2) and dHvA (Ref. 25) measurements. Nodes of the oscillation amplitude are seen at  $\theta = 43^\circ, 58^\circ, \dots, 122^\circ, 137^\circ$ . At these angles, the SSZ conditions mentioned above are satisfied. The SSZ angles do not change with magnetic field. This is quite reasonable because the  $gm_c$  value does not depend on the magnitude of the magnetic field in general.

Figure 2 shows the periodicity of the SSZ angles measured in  $\alpha$ -(BEDT-TTF) $_2$ KHg(SeCN) $_4$  and also in the superconductor  $\alpha$ -(BEDT-TTF) $_2$ NH $_4$ Hg(SCN) $_4$ . The linear dependence of  $(\cos \theta)^{-1}$  for  $2n+1$ , where  $n$  is an integer, represents the two-dimensional angle dependence of the effective mass with  $m_c(\theta) = m_c(0^\circ)/\cos \theta$ . Here we assume that the  $g$  value does not change much with the angle. The values of  $g(m_c/m_0)$  are obtained as 3.63 in  $\alpha$ -(BEDT-TTF) $_2$ KHg(SeCN) $_4$  and 4.48 in

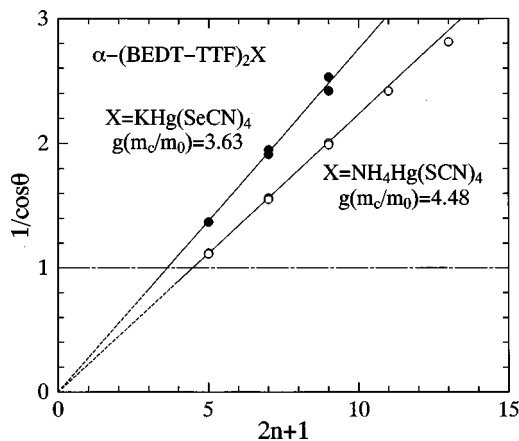


FIG. 2. Integer plots for the spin-splitting-zero angles in  $\alpha$ -(BEDT-TTF) $_2$ KHg(SeCN) $_4$  and  $\alpha$ -(BEDT-TTF) $_2$ NH $_4$ Hg(SCN) $_4$ .

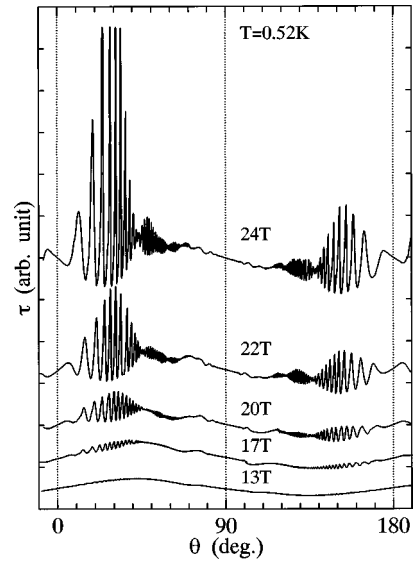


FIG. 3. Magnetic torque curves of  $\alpha$ -(BEDT-TTF) $_2$ KHg(SCN) $_4$ . The curve at 24 T is measured in the M\* phase and other curves are measured in the AF phase.

$\alpha$ -(BEDT-TTF) $_2$ NH $_4$ Hg(SCN) $_4$  from the slope of the straight lines. These values are in good agreement with the previous reports.<sup>25,26</sup>

### B. $\alpha$ -(BEDT-TTF) $_2$ KHg(SCN) $_4$ : AF metal

Figure 3 shows the magnetic torque curves in a single crystal of  $\alpha$ -(BEDT-TTF) $_2$ KHg(SCN) $_4$ . The torque curve at 24 T and in the angle range between  $0^\circ$  ( $120^\circ$ ) and about  $60^\circ$  ( $180^\circ$ ) is considered to be measured in the M\* phase from previous studies on the angle dependence of the magnetic phase diagram.<sup>27-30</sup> Based on the same consideration, other curves below 20 T are measured in the AF phase. The curve at 22 T is expected to be in the boundary region between the AF and the M\* phases. Characteristic features which are not observed in the P phase of the normal metal  $\alpha$ -(BEDT-TTF) $_2$ KHg(SeCN) $_4$  are seen on the torque curves in both the AF and the M\* phases. First, slow oscillatory structures are observed in the  $\pm \sim 30^\circ$  region centered at  $\theta = 90^\circ$ , which were not observed in  $\alpha$ -(BEDT-TTF) $_2$ KHg(SeCN) $_4$  and  $\alpha$ -(BEDT-TTF) $_2$ NH $_4$ Hg(SCN) $_4$ . The angles at which the structures appear shift with magnetic field and do not follow possible trial trigonometrical functions such as  $(\cos \theta)^{-1}$ ,  $\tan \theta$ , and so on. Both features imply that neither ADMRO nor small 2D FS are the possible origin for the structure. We shall return to this point later. Secondly, the SSZ angles are different in the AF and the M\* phases, which is the main experimental finding of this study.

Figure 4 shows the magnetic field dependence of the SSZ angles plotted in  $(\cos \theta)^{-1}$ . In the AF phase, the SSZ angle corresponding to the SSZ condition of  $2n+1=7$ , a number which will be given later in Fig. 5, does not move with magnetic field within our experimental resolution. On the other hand, it is clearly seen that the SSZ angles change at the phase boundary between the AF and the M\* phases. In addition, the SSZ period in  $(\cos \theta)^{-1}$  also changes at the phase boundary. Figure 5 shows the integer plot of the SSZ

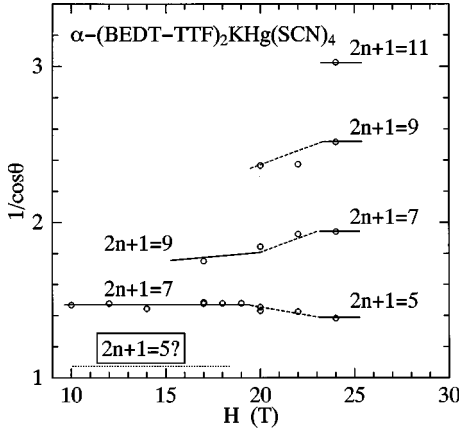


FIG. 4. Magnetic field dependence of the spin-splitting-zero angles of  $\alpha$ -(BEDT-TTF) $_2$ KHg(SCN) $_4$ . The number  $(2n+1)$  at each SSZ position is referred to the integer plots of Fig. 5.

angles in several magnetic fields. The different slopes of two straight lines fitted to the data at 24 T and lower magnetic fields demonstrate the change of the SSZ periodicity at the phase boundary between the AF and the M\* phases. The  $g(m_c/m_0)$  values obtained from the slopes are 3.63 in the M\* phase and 4.7 in the AF phase. The number  $(2n+1)$  indicated at each SSZ angle in Fig. 4 corresponds to the SSZ condition number of this plot. In the AF phase, SSZ corresponding to  $2n+1=5$  could be expected to appear around  $(\cos \theta)^{-1} \sim 1$ , that is,  $\theta \sim 0^\circ$  or  $180^\circ$ . But the present torque experiments do not distinguish the node of the oscillation amplitude around the expected angle because only a few oscillations are observed along these directions. The splitting-wave form, however, has been observed in SdH and dHvA oscillations<sup>22</sup> in the magnetic field perpendicular to the plane ( $\theta \sim 0^\circ$ ). This observation also confirms the reliability of SSZ which occurs at  $\theta \sim 0^\circ$  because the enhancement of the higher harmonics in the oscillations takes place at the SSZ angle in general.

## V. DISCUSSION AND CONCLUSION

We shall discuss the  $gm_c$  value in the AF and the P phases of  $\alpha$ -(BEDT-TTF) $_2$ KHg(SCN) $_4$  in comparison with

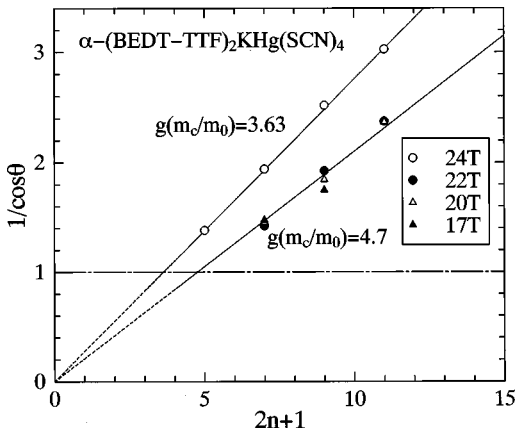


FIG. 5. Integer plots for spin-splitting-zero angles in  $\alpha$ -(BEDT-TTF) $_2$ KHg(SCN) $_4$ . The slope of the straight lines, corresponding to the  $g(m_c/m_0)$  value, are different in the AF (17, 20, 22 T) and M\* phases (24 T).

the value in the P phase of  $\alpha$ -(BEDT-TTF) $_2$ KHg(SCN) $_4$ . The first point to notice is that the same  $g(m_c/m_0)$  value of 3.63 is obtained in the M\* phase of  $\alpha$ -(BEDT-TTF) $_2$ KHg(SCN) $_4$  and the P phase of  $\alpha$ -(BEDT-TTF) $_2$ KHg(SCN) $_4$ . Almost the same value of the effective mass of about  $1.9m_0$  in both phases has been obtained on the basis of the LK analysis in the temperature dependence of the SdH and dHvA oscillation amplitude.<sup>2,17,25</sup> The  $g$  values in both phases are estimated to be about 1.9 from the  $g(m_c/m_0)$  value in the present results and the reported  $m_c$  values. It is worth noting that the  $gm_c$  values obtained with independent methods; the SSZ method and the fitting to the LK formulation, are consistent with each other. This suggests that no additional temperature-dependent factor on the dHvA oscillation amplitude is taken into account in these phases, while such an additional factor may become important in the MB orbits in the AF phase of  $\alpha$ -(BEDT-TTF) $_2$ KHg(SCN) $_4$ . Such an explanation has recently been proposed<sup>23</sup> and will be mentioned later.

Next, we discuss the change of the  $gm_c$  value in the AF and the M\* phases of  $\alpha$ -(BEDT-TTF) $_2$ KHg(SCN) $_4$ . The important point is that the SSZ method and the LK analysis show a different tendency of the enhancement of the  $gm_c$  or  $m_c$  value below and above the phase transition. In the present SSZ method, the  $g(m_c/m_0)$  value ( $\sim 4.7$ ) obtained in the AF phase is larger than that ( $\sim 3.63$ ) in the M\* phase. In contrast, a smaller effective mass ( $m_c \sim 1.6m_0$ ) obtained in the AF phase than that ( $\sim 1.9m_0$ ) in the M\* phase has been reported on the basis of the LK analysis of the temperature dependence of the oscillation amplitude.<sup>17,23</sup> This discrepancy may be explained by a model which requires an additional temperature factor on the LK formulation in the AF phase. In the AF phase, the observed  $\alpha$ -orbit results from the MB effect on the multiconnected FS which is reconstructed by the SDW formation. The MB gap may have a temperature dependence because this gap is opened by the periodic SDW potential which is temperature dependent. In addition, the magnitude of the SDW gap, which is expected to be  $\sim 10$  K, is comparable to the temperature at which the experiments are done. The gap would become small effectively due to the thermal excitation near Fermi level. Here, the MB field is approximated by a polynomial function of  $T$ ;  $H_{MB}(T) = H_{MB}^0(1 - aT \pm bT^2 \pm \dots)$ . The reduction factor<sup>24</sup>  $R_{MB} = \exp(-lH_{MB}/H)$  should be added to the standard LK formulation of Eq. (3), where  $l$  is an integer (2, 3, 4, 5, 6, ...) corresponding to the number [4 (in Ref. 10), 6, 8, 10, 12 (in Ref. 14), ...] of MB points on the MB orbit, respectively. This simple consideration implies that the effective mass  $m_c^{obs}$  and Dingle temperature  $T_D^{obs}$  obtained so far by using the standard LK analysis in the AF phase are underestimated and overestimated with respect to the correct values with the relation of  $m_c^{obs} \approx m_c(1 - laH_{MB}^0/\lambda m_c)$  and  $T_D^{obs} \approx T_D + lH_{MB}^0/\lambda m_c$ , respectively. Here, higher orders of the expansion in  $H_{MB}(T)$  are neglected for simplicity. In short, the reasonable effective mass in the AF phase is obtained by the SSZ method, on the other hand, the underestimated value has been calculated by inappropriate use of the LK analysis in the AF phase.

Let us then consider the enhancement of the  $gm_c$  value in the AF phase on the basis of the results obtained by the SSZ

method. Here we present two possible mechanisms. One is the enhancement of the many-body interactions on both  $g$  and  $m_c$ ,<sup>24</sup> and another is the exchange interaction between the conduction spins through SDW moments.<sup>22</sup> The renormalization by the electron-electron (EE) and electron-phonon (EP) interactions for the effective mass and the  $g$  value is simply  $m_c = m_b(1 + A_0^{\text{EP}})(1 + A_1^{\text{EE}})$  and  $g = g_s / [(1 + B_0^{\text{EE}})(1 + B_1^{\text{EP}})]$ , where  $A_0$ ,  $A_1$ , and  $B_0$  are the coefficients of the Legendre expansion of the spin-symmetric and spin-asymmetric parts, respectively, of the Landau scattering function within the framework of the Landau theory of Fermi liquids. Here  $m_b$  is the band mass calculated from a band structure and  $g_s$  is the  $g$  value measured by spin resonance experiments including spin-orbit coupling but not a many-body effect. The combination value of  $g(m_c/m_0)$ , which is obtained experimentally, depends only on the EE interaction and not on the EP interaction because of  $A_0^{\text{EP}} = B_0^{\text{EP}}$  in general. Thus  $g(m_c/m_0) = g_s m_b (1 + A_1^{\text{EE}}) / (1 + B_0^{\text{EE}})$ , where  $A_1^{\text{EE}} > 0$  and  $B_0^{\text{EE}} < 0$ . This means that the enhancement of the EE interaction would give the larger  $gm_c$  value, although it is difficult to distinguish the respective magnitude of the renormalization on the effective mass and the  $g$  value. It seems reasonable to suppose that the EE interaction is enhanced in the AF phase. The observed SdH (and also dHvA) oscillation frequencies in the AF and the  $M^*$  phases are the same.<sup>15,17</sup> This means that the carrier number corresponding to the observed orbit is the same in two cases of the closed orbit in the  $M^*$  phase and the MB one in the AF phase. On the other hand, the total number of the carrier is expected to be small in the AF phase where the SDW gap is opened on the planer part of FS. The small carrier number in the AF phase is also supported by Hall effect experiments.<sup>31</sup> Thus the effective EE interaction on the carrier orbiting in the AF phase is supposed to be enhanced because of less screening effect on the carrier.

Another possible scenario is the magnetic exchange interaction for the  $g$  value. This effect has been applied to explain a magnetic field dependence of the splitting wave form of the SdH and dHvA oscillations.<sup>22</sup> The result shows that Zeeman splitting of Landau levels is modulated by a magnetic exchange interaction. The effective  $g$  value becomes large and changes with magnetic field as  $\tilde{g} = g + H/H_{\text{ex}}$ , where  $H_{\text{ex}} = \Delta E / \mu_B$ ,  $\Delta E$  is the positive exchange interaction energy and  $\mu_B$  the Bohr magneton. The origin of the exchange energy is expected to be the positive exchange interaction between the SDW moments and Zeeman splitting spins of the conduction carriers. The present SSZ results, however, do not show the visible effect of the exchange interaction because the expected change of the  $g$ -value with magnetic fields is not observed as the change of the SSZ angles in the

AF phase within the experimental resolution. Further experiments are necessary to judge the validity of this model.

Let us return to the problem on the anomalous torque structures appeared around the magnetic field parallel to the 2D plane. As already mentioned, the structure does not relate to a FS effect such as ADMRO and dHvA oscillations. It is worth noting that structures are formed by a reduction from the expected torque value extrapolated from the  $\sin 2\theta$  curve. This implies that the structures appear owing to the reduction of the susceptibility  $|\chi_{aa} - \chi_{bb}|$  in Eq. (1). From this result, we propose a possible model in which the spin structures in the AF and the  $M^*$  phases change successively at characteristic angles, because the reduction of  $|\chi_{aa} - \chi_{bb}|$  in tilted magnetic fields reminds us of the spin-canting, spin-flipping, and so on. The angles would relate to the magnetic anisotropy and magnitude of the magnetic field. In fact, Christ *et al.* reported<sup>19,20</sup> that a series of torque anomalies and hysteresis in sweeping magnetic fields were observed at angles corresponding to the present torque structures. It is expected that the magnetic phase diagram proposed so far in the magnetic field perpendicular to the plane seems to be valid up to  $50^\circ$ – $60^\circ$  from the perpendicular direction to the 2D plane. At larger angles, the phase diagram may change not only below 23 T but also in the higher magnetic fields. We need further systematic studies on the torque anomaly in order to obtain information about the phase diagram and the spin structure in tilted magnetic field.

Finally, we summarize the present study as follows. The observation of SSZ in dHvA oscillations not only in the  $M^*$  phase but also in the AF phase of  $\alpha$ -(BEDT-TTF)<sub>2</sub>KHg(SCN)<sub>4</sub> enables us to directly compare of the  $gm_c$  values in two phases. This method has an advantage because it is not affected by uncertainty in the evaluation process of  $m_c$  in the AF phase in comparison with the LK analysis. The  $gm_c$  value in the AF phase is larger than that in the  $M^*$  phase. The large  $gm_c$  value demonstrates the enhancement of the electron-electron interaction in the AF phase. A more complex magnetic phase diagram is expected from anomalous torque structures in tilted magnetic field. The difference of the phases may arise from the spin structures modified by tilted magnetic field.

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<sup>5</sup>On this subject, there have been many articles published so far. Thus the readers are referred to Ref. 17 containing the references for early works.

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