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The Fermi Surfaces of $\beta$-(BEDT-TTF)$_2$X

J. Wosnitza (1,*), G. Goll (1,2), D. Beckmann (1), S. Wanka (1), D. Schweitzer (3) and W. Strunz (4)

(1) Physikalisches Institut, Universität Karlsruhe, 76128 Karlsruhe, Germany
(2) Grenoble High Magnetic Field Laboratory, Max-Planck-Institut für Festkörperforschung and Centre National de la Recherche Scientifique, BP 166, 38042 Grenoble, France
(3) 3. Physikalisches Institut, Universität Stuttgart, 70550 Stuttgart, Germany
(4) Anorganisch-chemisches Institut, Universität Heidelberg, 69120 Heidelberg, Germany

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Abstract. — We present de Haas-van Alphen (dHvA) measurements of the isostructural quasi-two-dimensional (quasi-2D) organic superconductors $\beta$-(BEDT-TTF)$_2$IBr$_2$ and $\beta_H$-(BEDT-TTF)$_2$I$_3$ with superconducting transition temperatures of $T_c \approx 2.5$ K and $T_c \approx 7$ K, respectively. For both salts almost the same dHvA frequency and a beating behavior of the dHvA signal are found which reflect the similarity of the Fermi surface (FS) topology in both materials. The FS can be well described by a slightly corrugated quasi-2D cylinder. The corresponding overlap integral $t_{\perp}$ between the highly conducting organic layers could be determined with high precision. $t_{\perp}$ is larger for $\beta_H$-(BEDT-TTF)$_2$I$_3$ although this salt has the larger interlayer distance. The effective cyclotron masses are very similar in both materials ($m_c \approx 4 m_e$). On the other hand, the observed spin-splitting zeros suggest a rather large difference and large absolute values of the conduction electron $g$-factor which seems to reflect the importance of electron-electron interactions in the $\beta$-phase salts.

1. Introduction

Organic charge transfer salts of the $\beta$-(BEDT-TTF)$_2$X type, where BEDT-TTF stands for bisethylenedithio-tetrathiafulvalene (also abbreviated by ET) and X for a monovalent anion, were among the first discovered quasi-two-dimensional (quasi-2D) superconductors [1]. Special interest was focused on the different superconducting states of $\beta$-(BEDT-TTF)$_2$I$_3$ which has a superconducting transition temperature $T_c = 1.3$-1.5 K at ambient pressure [2] and a so-called high-$T_c$ state with $T_c = 7$-8 K [3]. The latter state can be established by application of small pressure of the order 300 bar when the sample is cooled down below $\sim 125$ K [4]. Thereby a structural transition at $\sim 175$ K [5], where the I$_3$ anions become distorted [6] and a modulated superstructure is formed [7], is suppressed and the high-$T_c$ ordered state is stabilized.

$T_c$ of the superconducting $\beta$-phase salts is proportional to the anion length of the linear anions X = IBr$_2$, AuI$_2$, and I$_3$ (in the $\beta_H$ phase) [8]. The question arises whether this $T_c$ dependence

(* )Author for correspondence (e-mail: Wosnitza@fphvax.physik.uni-karlsruhe.de)
is caused by differences in the electronic band structure (driven by small modifications of the crystal structure) or by changed many-body effects, i.e., electron-phonon or electron-electron interactions. From band-structure calculations practically identical electronic structures of all $\beta$-phase salts and phases are obtained [9]. This has led to the proposal that phonon softening is responsible for a changed electron-phonon interaction and the different $T_c$ in these isostructural salts.

First investigations of the $\beta$-phase Fermi surfaces (FS) by Shubnikov-de Haas (SdH) experiments have been reported already a few years ago (see Refs. in [10]). For $X = I_3$ a very low SdH frequency of $F_{\text{low}} \approx 110$ T [11,12] and two frequencies of $F_{\text{low}} \approx 50$ T and $F_{\text{high}} \approx 3900$ T for $X = IBr_2$ were found [12-14]. Only the latter value is consistent with the predicted FS area [9,15]. In a further experiment fast SdH oscillations with $F = 3730$ T have been observed in the high-$T_c$ state of $\beta$-(BEDT-TTF)$_2$I$_3$ [16]. The two investigated samples showed huge SdH amplitudes and very strong anharmonicities. In another experiment at $\sim 1.8$ kbar ($T_c = 6.8$ K) SdH oscillations with smaller amplitude and $F = 4120$ T are reported [17]. The origin of the low SdH frequency and for the different SdH frequencies in the two phases of $\beta$-(BEDT-TTF)$_2$I$_3$ are unknown. A more detailed comparative study with a different technique was desirable.

Here, we report on the first observation of dHvA oscillations in $\beta_H$-(BEDT-TTF)$_2$I$_3$ and compare the results with dHvA studies of $\beta$-(BEDT-TTF)$_2$I$_2$IBr$_2$ which are partially reported in [18] and which are extended to fields up to 23 T. The FS topology has been mapped out in detail and the interlayer transfer integrals could be determined with high accuracy. The effective cyclotron masses have been extracted at many different angles. The found up to four spin-splitting zeros suggest considerably enlarged $g$-values compared to ESR results.

2. Experimental

Single crystals of $\beta$-(BEDT-TTF)$_2$I$_3$ and $\beta$-(BEDT-TTF)$_2$IBr$_2$ (called $\beta$-I$_3$ and $\beta$-IBr$_2$ hereafter) were grown by the usual electrocrystallization technique which is described in detail in [19]. The investigated platelike samples typically had areas of $\sim 0.7 \times 0.7$ mm$^2$ and a thickness of $\sim 0.2$ mm. The magnetization of the $\beta$-IBr$_2$ sample was measured with a capacitance cantilever torquemeter in fields up to 23 T at the Grenoble High-Field Laboratory. These results are presented together with previously obtained modulation-field data [18]. The dHvA effect of $\beta_H$-I$_3$ was measured by field-modulation technique in fields up to 13 T. The modulation frequency was $\sim 78$ Hz and the maximum modulation amplitude was $\sim 20$ mT peak to peak. In order to avoid a field-dependent sensitivity of the pick-up signal the modulation-field amplitude was changed with the square of the applied external quasi-static field (see [20]). Both experimental setups were mounted in $^3$He cryostats on gear-driven rotation mechanisms where the samples could be rotated around one axis with a resolution of better than 0.1°.

The high-$T_c$ phase of $\beta$-I$_3$ was prepared by cooling the sample down to liquid nitrogen temperature under $\sim 1$ kbar He pressure, then the pressure was released and the sample mounted under liquid nitrogen into the dHvA probe which was then quickly transferred into a toploading $^3$He cryostat. Thereby, the sample was kept below $\sim 125$ K to prevent the transition into the modulated superstructure. The high-$T_c$ state of the sample was proven by ac-susceptibility measurements which resulted in $T_c \approx 7$ K. More details will be presented in a separate publication [21].
3. Results and Discussion

For both samples dHvA oscillations could be detected starting around 9 T in an angular range of up to approximately $|\Theta| = 60^\circ$, where $\Theta$ denotes the angle between the applied field and the normal to the BEDT-TTF planes. The torque signal of $\beta$-IBr$_2$ between 11 and 23 T is shown in Figure 1 for two different field orientations $\Theta$. In the shown field range at $\Theta = 12.2^\circ$ clearly two nodes of the dHvA signal are visible which can also be seen as two close peaks around 3900 T in the Fourier transformation (upper inset in Fig. 1). This beating behavior results from the existence of two extremal FS areas of similar size and has been observed previously in SdH [13,14] and dHvA experiments up to 15 T [18]. The average frequency at $12.2^\circ$ is $F_{av} \approx 3930 T$ and the frequency difference $\Delta F = 47 T$. Systematic measurements at different angles showed the usual $1/\cos \Theta$ dependence of $F_{av}$ (see Fig. 2) and a distinctive angular shift of the beating nodes. The minimum frequency at $\Theta = 0^\circ$ obtained from a $1/\cos \Theta$ fit is $F_0 = 3840 T$ (dotted line in Fig. 2). Some apparent deviations from this 2D behavior for $\beta$-IBr$_2$ around $\Theta = 0^\circ$ will be discussed later. The lower panel of Figure 1 shows the dHvA signal at $\Theta = -8.3^\circ$ where no nodes in the measured field range are observed. The Fourier transformation of the data between 19 and 23 T (lower inset of Fig. 1) reveals a high harmonic content of the dHvA oscillations. In previous low-field dHvA measurements on different crystals additional nodes were observed in the oscillating signal which only could be understood by the existence of four dHvA frequencies [18]. This hints to a possible twinning of these samples, although the angular dependence of the frequency difference and the nodes of exactly zero dHvA amplitude are hard to explain by twinning. For the investigated sample presented here we only observed two frequencies which are not due to twinning as we will show in the following.

A very similar behavior of the dHvA signal has been observed for $\beta$-H-I$_3$. Figure 3 shows dHvA measurements for two angles. At $\Theta = -4^\circ$ a beating pattern with an average frequency $F_{av} \approx 3800 T$ and a frequency difference $\Delta F = 57.3 T$ are found. This is in reasonable agreement with
Fig. 2. — Angular dependence of the average dHvA frequency in \( \beta-(\text{BEDT-TTF})_2\text{IBr}_2 \). The closed circles are from modulation-field data between 13 and 15 T [18], the open circles from torque data between 17 and 20 T. The solid and dashed lines represent the angular dependence of the two extremal areas of the warped FS. The dotted line is the fitted \( 1/\cos \Theta \) dependence with \( F_0 = 3840 \) T. The inset shows the angular dependence of the effective cyclotron mass for all investigated angles.

The mentioned SdH results where \( F_{av} \approx 3730 \) T and \( \Delta F = 74 \) T at \( \Theta \approx 0^\circ \) are reported [16]. In contrast to the SdH signal, however, the dHvA oscillations close to \( 0^\circ \) show neither asymmetry nor unusually large harmonic content (see the Fourier transformation of the data at \( \Theta = -4^\circ \) in the upper inset). This changes drastically at those angles where the beating nodes vanish as shown in the lower panel of Figure 3 for \( \Theta = 30^\circ \). The Fourier transformation at this angle reveals peaks up to the fifth harmonic. In common with the \( \beta\text{-IBr}_2 \) salt, the angular dependence of the average frequency can be described by \( F_{av} = F_0/\cos \Theta \) with minimum frequency \( F_0 = 3810 \) T at \( \Theta = 0^\circ \).

The angular dependence of \( \Delta F \) extracted from the observed nodes is shown in Figure 4a for both \( \beta \)-phase salts. It has to be noted, that for some angles only one node could be resolved in the measured field range. For these angles the most plausible node number \( n \) obeying the relation \( \Delta F = B_n(n - 1/4) \) was chosen [22]. Thereby, \( B_n \) is the field where the \( n \)th node is located. For \( \beta\text{-IBr}_2 \) the angular dependence of \( \Delta F \) has been measured for three different rotation planes. The in-plane angle \( \varphi \) as defined in Figure 4b describes the orientation of the field rotation plane with respect to the \( a' \) direction, that is perpendicular to \( b \). The data for azimuthal angles \( \varphi = 0^\circ \) and \( \varphi = 90^\circ \) were obtained in a previous modulation-field study [18] where the investigated sample was oriented with a four-circle X-ray diffractometer. The data for \( \varphi = 32^\circ \) are from the high-field torque measurement. This latter \( \varphi \) is estimated from the sample morphology and from the theoretical consideration of the angular dependence of \( \Delta F \) as explained below.

The observed angular dependence of \( F_{av} \) and \( \Delta F \) can perfectly be described by assuming an almost 2D cylindrical FS with a small corrugation. This warping leads to a distinctive angular dependence of \( \Delta F(\Theta) \) which was first derived for a simple case by Yamaji [23] and later refined for a lower symmetry FS by Kartsovnik et al. [24]. The energy dispersion for a quasi-2D metal
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is usually written as

$$\epsilon(k) = \epsilon_{\parallel}(k_x, k_y) - 2t_{\perp} \cos(\mathbf{h} \cdot \mathbf{k}),$$

where $\epsilon_{\parallel}(k_x, k_y)$ describes the in-plane dispersion, $\mathbf{h} = (u_x, u_y, c')$ is the direction vector of the interlayer transfer energy $t_{\perp}$, and $c'$ is the spacing between adjacent layers. For the low-symmetry triclinic crystal structures of the $\beta$-phase salts all components of $\mathbf{h}$ are non-zero. In reasonable agreement with the data $\mathbf{h}$ is found to coincide with the crystal lattice parameter $c$. This means that the warped structure of the FS is tilted with respect to the normal of the conducting planes. Except for two special azimuthal angles $\varphi$ the angular change of $\Delta F$ is asymmetric and for $t_{\perp} \ll \epsilon_{\parallel}$ is given by

$$\Delta F \cos \Theta = \Delta F_{\text{max}, \varphi} J_0(c' k_B^{\text{max}} \tan \Theta - u k_{\parallel}),$$

where $J_0(x)$ is the Bessel function of zeroth order, $\mathbf{u} = (u_x, u_y, 0)$ is the in-plane component of $\mathbf{h}$, and $k_{\parallel}$ is the $\varphi$-dependent Fermi wave-vector component in the BEDT-TTF plane whose projection $k_B^{\text{max}}$ to the field rotation plane is maximal. In the following we assume for simplicity $|k_{\parallel}| \approx k_B^{\text{max}}$. The prefactor in (2) for $J_0(x) = 1$ is given by

$$\Delta F_{\text{max}, \varphi} = 4 F_0 \frac{t_{\perp}}{\epsilon_F} \frac{J_0(u k_{\parallel})}{J_0(3 u k_{\parallel})},$$

where $\epsilon_F$ is the Fermi energy. At the special angles $\Theta$ where the Bessel function is zero [25] the average electron velocity perpendicular to the BEDT-TTF planes tends to zero and, hence, the resistivity $R_{zz}$ becomes maximal. This effect has been exploited to reconstruct the in-plane shape of the FS by measurements of the angular dependence of magnetoresistance oscillations (AMROs) [24]. The result of this study is depicted in Figure 4b. From AMRO experiments the principal presence of a warped FS and the in-plane component $k_B^{\text{max}}$ can be detected. However, the value of the transfer integral $t_{\perp}$ is not extractable. This is possible by a proper

Fig. 3. — dHvA signal of $\beta$-(BEDT-TTF)$_2$I$_3$ at $T \approx 0.5$ K for $\Theta = -4^\circ$ (upper panel) and $\Theta = 30^\circ$ (lower panel). The insets show the Fourier transformations of the data.
Fig. 4. — a) Angular dependence of the frequency difference for three different rotation planes in \(\beta\)-(BEDT-TTF)\(_2\)IBr\(_2\) (upper panel) and for one plane \((\varphi \approx 40^\circ)\) in \(\beta_{1r}\)-(BEDT-TTF)\(_2\)I\(_3\) (lower panel).
b) In-plane FS of \(\beta\)-(BEDT-TTF)\(_2\)IBr\(_2\) obtained from AMRO experiments [24]. The solid squares are values extracted in this work.

Analysis of the angular dependence of \(\Delta F\). The solid lines in Figure 4a are fits using (2). For \(\beta\)-IBr\(_2\) at \(\varphi = 0^\circ\), that is for \(B \perp b\), the maximum asymmetry and consequently the maximum \(\Delta F_{\text{max,} \varphi}\) is found. This gives the fit parameter \(t_1/\epsilon_F = 1/(280 \pm 10)\). In order to describe correctly the observed angles where \(\Delta F = 0\), \(c'k_{\|}(0^\circ) = 5.0\) and \(uk_{\|} = 0.72\) is obtained. The direction of the transfer integral \(h\) coincides nicely with the triclinic angle \(\arctan(u/c') \approx 8^\circ\) of the \(c\) axis with respect to the BEDT-TTF planes in agreement with the result obtained
from the AMRO measurements [24]. The value \( c'k_j(0^\circ) = 5.0 \) is within 2% what would be obtained for a circular in-plane FS with \( k_j = k_F = \sqrt{2eF_{\text{av}}/h} \). For the two other field rotation planes the respective in-plane Fermi wave vector had to be adjusted to \( c'k_j(32^\circ) = 5.3 \) and \( c'k_j(90^\circ) = 4.8 \). Our values of \( k_j(\varphi) \) (solid squares in Fig. 4b) are in reasonable agreement with the FS in-plane topology extracted out of the AMRO experiment [24]. The vanishing \( \Delta F \) at some special angles was noted already in previous SdH experiments [14] and are (for the shown angles) in good agreement with our dHvA results.

For the \( \beta_{H-I_3} \) sample only one field rotation plane was investigated. For the measured angle \( \varphi \) an asymmetry was found which is somewhat less than expected for \( \mathbf{h} \) parallel to \( \mathbf{c} \). Therefore, if for \( \beta_{H-I_3} \) \( \mathbf{h} \) coincides with \( \mathbf{c} \), as for \( \beta_{-IBr_2} \), the rotation plane in the experiment was tilted away from \( \mathbf{u} \) by \( \varphi \approx 40^\circ \). For the measured angle \( \Delta F_{\text{max},\varphi} = 80 \) T is obtained which means that the maximum value is \( \Delta F_{\text{max},0} \approx 87.5 \) T. This results in a transfer integral of \( t_{\perp}/\epsilon_F = 1/(175 \pm 10) \), somewhat larger than for \( \beta_{-IBr_2} \) in spite of the fact that for \( \beta_{H-I_3} \) the interlayer distance \( c' \) is larger (see e.g. [19]) and naively rather a smaller overlap integral \( t_{\perp} \) would be expected. To adequately describe the angular dependence of \( \Delta F \) in \( \beta_{H-I_3} \) an elongated \( k_j \), 15% longer than for a circular in-plane FS, had to be assumed. This hints to an elliptical shape of the in-plane FS similar to the \( \beta_{-IBr_2} \) one.

At the angles where \( \Delta F = 0 \) the amplitude of the dHvA oscillations considerably enlarges [26]. At these angles the FS cross section \( q_r \) does not change along \( \mathbf{B} \), i.e., the curvature factor \( \partial^2 q_r / \partial \kappa^2 \)^{-1/2} of the Lifshitz-Kosevich (LK) formula becomes maximal [20]. Therefore, all electrons contribute to the dHvA oscillations. In addition, for \( \beta_{H-I_3} \) around these angles extreme unharmonicitics of the dHvA signal (see lower panel of Fig. 3), hysteretic effects, and sudden collapses of the oscillation amplitudes during the field sweeps (not shown here) were observed [21]. The origin of this unusual behavior is not clear. A possible explanation might be the formation of so-called Condon domains due to magnetic interaction [20].

From Fourier transformations the temperature dependence of the fundamental dHvA amplitude was extracted and the effective cyclotron masses, \( m_e \), were determined for both materials over a broad angular range. Within experimental resolution we found the same effective masses from the temperature dependence of the second-harmonic amplitude. The inset of Figure 2 shows the extracted values of \( m_e \) for all investigated angles for \( \beta_{-IBr_2} \). The solid line is a fit expected for 2D materials of the form \( m_e = m_{e0}/\cos \Theta \) with \( m_{e0} = (4.0 \pm 0.1)m_e \). This is somewhat less than \( \sim 4.5m_e \) reported in [14]. For \( \beta_{H-I_3} \) the 2D 1/\cos \Theta dependence of \( m_e \) was found as well. In spite of the largely different superconducting transition temperatures a very similar effective cyclotron mass of \( m_{e0} = (4.2 \pm 0.2)m_e \) was obtained. This is in reasonable agreement with the SdH value of \( (3.7 \pm 0.3)m_e \) reported in [16]. For experiments under pressure an increase of \( m_e \) up to 5.9 \( m_e \) at \( \sim 1.8 \text{kbar} \) and 7.1 \( m_e \) at \( \sim 3.5 \text{kbar} \) was reported [17].

Due to the large values of \( m_{e0} \) in both materials and the experimentally verified 1/\cos \Theta dependence of \( m_e \) a large number of spin-splitting zeros within a relatively small angular range are anticipated and indeed observed. For \( \beta_{H-I_3} \) three and for \( \beta_{-IBr_2} \) four definite angles with almost vanishing fundamental dHvA amplitudes were found. At these angles the spin-splitting factor \( R_s = \cos(r\pi g\mu_b/2) \) is close to zero for \( r = 1 \), i.e., the fundamental dHvA frequency [20]. \( g \) is the conduction electron \( g \) factor and \( \mu_b = m_{b}/m_e \) is the band-structure mass in relative units averaged over one cyclotron orbit. The factor \( g\mu_b \) is not renormalized by electron-phonon interaction. Therefore, if the \( g \) value including a possible electron-electron renormalization factor is known the electron-phonon coupling constant \( \lambda \) can be estimated by \( m_e = m_{b}(1 + \lambda) \) [27]. Because of the 1/\cos \Theta dependence of the effective mass the \( n \)th
Fig. 5. — Observed spin-splitting zeros of the fundamental and the second harmonic dHvA amplitude for six different organic metals vs. $1/\cos \Theta$. The solid lines are fits described in the text.

The spin-splitting zero, i.e., the angles $\Theta_n$ where $R_S = 0$ (for $r = 1$), is given by

$$n = \frac{g\mu_B}{2} \cdot \frac{1}{\cos \Theta_n} + \frac{1}{2},$$

(4)

Figure 5 shows a plot of $n$ vs. $1/\cos \Theta$ for all observed spin-splitting zeros (including zeros of the second harmonics) of six different organic conductors. The slopes of the linear fits (solid lines) directly give $g\mu_B/2$. The intercepts are forced to go through $1/2$ at $1/\cos \Theta = 0$ and determine the absolute values of $n$. The result can be divided into two classes: (i) For one group of materials, namely $\beta$-I$_3$, $\beta$-IBr$_2$, and $\kappa$-(BEDT-TTF)$_2$I$_3$ with $m_{e0} = 3.9m_e$, larger slopes $g\mu_B/2$ of approximately 6.0, 4.5, and 4.3 are found, respectively. If no mass enhancement due to electron-phonon interaction is taken into account ($\lambda = 0$) lower limits of $g$ up to $\sim 2.85$ (for $\beta$-I$_3$) are obtained. The real $g$ values might be even much larger because $\lambda$ is expected to be larger than zero. Since ESR measurements give $g$ close to the free-electron value of $\sim 2$ [28] the larger values from our dHvA experiments hint to an appreciable electron-electron interaction. This was also inferred from the $T^2$ dependence of the electrical resistivity in the $\beta$-phase materials [29] and from cyclotron resonance experiments of some other BEDT-TTF materials [30]. (ii) For the other group of organic metals, namely $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ ($m_{e0} = 3.3m_e$), $\alpha$-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ ($m_{e0} = 2.6m_e$), and $\alpha$-(BEDT-TTF)$_2$TIHg(SeCN)$_4$ ($m_{e0} = 2.0m_e$), the values for $g\mu_B/2$ are compatible with the ESR value of $g \approx 2$. With the assumption that $g \approx 2$ as found in ESR experiments reasonable $\lambda$ values between $\sim 0.1$ and $\sim 0.3$ are obtained [10]. Of course, the $g$ values could also be enhanced in these latter materials which in consequence would mean that the extracted $\lambda$ are only lower limits. Larger $\lambda$ values would be in line with indications for strong coupling suggested by specific-heat results and a variety of other experiments [31]. It should be noted that all the compounds of the first group reveal a strongly enhanced harmonic content of the dHvA signal compared to the predictions of the 3D LK formula. This can be seen for example in the Fourier transformations shown in Figures 1 and 3 where the higher harmonics are considerably larger than calculated with the LK formula for the known parameters. Therefore, it could be speculated that the extraction of $m_e$ out of the LK formula might result in wrong values. This deviation from the standard LK behavior might be due to the pronounced 2D FS topology of the salts of the first group which
essentially only have one 2D band. In contrast, the second group, that is the \( \alpha \)-phase salts and \( \kappa \)-(BEDT-TTF)\(_2\)Cu(NCS)\(_2\), have additional 1D bands. Hence, in these latter materials the chemical potential can remain constant with changing field since the 1D bands act as electron reservoirs. For these materials, therefore, only at considerably higher fields deviations from 3D LK behavior might be expected [32]. Contrary, for the one-band materials especially at the angles where \( \Delta F \) is close to 0 the chemical potential jumps each time a Landau level crosses the Fermi energy. Consequently, the dHvA signal is more unharmonic than predicted by the 3D LK formula. The influence of this deviation from 3D behavior on the effective-mass determination is not exactly clear. A numerical model predicts lower effective masses than the values extracted with the usual LK formalism [32]. This, however, would lead to even larger \( g \) values.

As the final point we want to discuss the dHvA signal of \( \beta \)-IBr\(_2\) close to \( \Theta = 0^\circ \) where this material exhibits a spin-splitting zero (see Fig. 5). Therefore, the dHvA signal is weak and effective-mass determinations are difficult for this angular range. A typical dHvA measurement at \( T = 0.4 \text{ K} \) between 17 and 20 T is shown as the upper trace in Figure 6. Due to the presence of two extremal FS areas two dHvA frequencies, \( F_1 \) and \( F_2 \), with slightly different effective masses exist. Therefore, the spin-splitting condition \( g\mu_b = 2n - 1 \) (for the fundamental) is fulfilled only for one orbit, e.g. for the orbit with frequency \( F_1 \). The second harmonics, on the other hand, are present for both orbits. Consequently, in the dHvA signal (Fig. 6) an apparent field dependent shift of the average second harmonic, \( (2F_1 + 2F_2)/2 \), with respect to the only partially suppressed fundamental signal \( F_2 \) occurs. The lower trace in Figure 6 is a simulation of the dHvA signal using the LK formula with the known parameters \( F_1 = 3840 \text{ T}, F_2 = 3893 \text{ T}, m_{c1} \approx m_{c2} \approx 4m_e, T = 0.4 \text{ K}, \) and the Dingle temperature \( T_D = 1 \text{ K} \). In order to reproduce the correct amplitude ratios of the fundamental and the second harmonics the values \( g\mu_{b1} = 8.995 \) and \( g\mu_{b2} = 8.99 \) werechosen as fit parameters. This is a very small mass difference (assuming \( g \) is the same for both orbits). In first approximation one would expect a proportionality between
the mass and the extremal area, i.e., \( m_{b1}/m_{b2} = F_1/F_2 \). However, values for \( m_{b1} \) and \( m_{b2} \) with this ratio cannot reproduce the measured high harmonic content of the dHvA signal correctly. Therefore, the above stated fit parameters are a reflection of the strong unharmonicities and again show the deviation from simple 3D LK behavior. Alternatively, smaller damping factors for the second harmonics in the LK formula have to be assumed in order to describe the observed dHvA signal. The simulation almost perfectly describes the observed dHvA signal. Especially the node around 18.6 T and the oscillation shape is well reproduced. Since the frequencies \( F_1 \) and \( F_2 \) are calculated with (2) our result further verifies the warped FS model.

A somewhat surprising result for the dHvA frequency was obtained from the Fourier transformation (FT) of the measured and simulated data. In spite of the expected average frequency \( F_{\text{av}} = 3867 \text{T} \) the FT gives \( \sim 3780 \text{T} \), which obviously is an artifact of the FT for this particular field range where the spin-splitting node occurs. This effect is also reflected in the extracted \( F_{\text{av}} \) data close to \( \Theta = 0^\circ \) which are shown in Figure 2 for both low-field (13-15 T) and high-field (17-20 T) data. The frequency determination for the low-field data (filled circles) was done by fitting the dHvA oscillation maxima vs. \( 1/B \) taking into account the phase jumps at nodes, whereas the high-field data (open circles) represent the FT values. The deviation of \( F_{\text{av}} \) from the \( 1/\cos \Theta \) dependence (dotted line) has been observed previously in SdH experiments [13,14]. As we have shown explicitly for the data at \( \Theta = 6.6^\circ \) this reflects not a real topological anomaly of the FS cylinder but rather an FT artifact due to the unique occurrence of spin-splitting zeros for one out of two very similar dHvA frequencies.

4. Conclusions

We presented a comparative dHvA study of the two \( \beta \)-phase organic superconductors \( \beta \)-(BEDT-TTF)\(_2\)IBr\(_2\) and \( \beta_H\)-(BEDT-TTF)\(_2\)I\(_3\). Both materials have a very similar cylindrical FS with a small corrugation along the \( c \) direction. Surprisingly the interlayer transfer integral is larger for \( \beta_H\)-I\(_3\), the material with larger interlayer distance. Contrary to previous SdH results no indications for small portions of the FS could be observed in our dHvA experiments. The effective cyclotron mass for \( \beta_H\)-I\(_3\) is only slightly larger than for \( \beta\)-IBr\(_2\). The observed spin-splitting zeros suggest a large enhancement of the conduction electron \( g \) factor which is much larger for \( \beta_H\)-I\(_3\) than for \( \beta\)-IBr\(_2\). This might indicate a strong electron-electron interaction. This effect hampers a possible extraction of the electron-phonon coupling constant out of the dHvA data.

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References


[22] The relation can be derived easily from the Lifshitz-Kosevich formula taking into account the proper phase factors for a maximal and minimal extremal area. See Ref. [20].


[25] The zeros of the Bessel function $J_0(x)$ are approximately given by $x = \pi(n - 1/4)$.


