Spin-zero anomaly in the magnetic quantum oscillations of a two-dimensional metal

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New Journal of Physics 10 (2008) 083032 (13pp)
Received 28 April 2008
Published 22 August 2008
Online at http://www.njp.org/
doi:10.1088/1367-2630/10/8/083032

Abstract. We report on an anomalous behavior of the spin-splitting zeros in the de Haas–van Alphen (dHvA) signal of a quasi-two-dimensional organic superconductor. The zeros as well as the angular dependence of the amplitude of the second harmonic deviate remarkably from the standard Lifshitz–Kosevich (LK) prediction. In contrast, the angular dependence of the fundamental dHvA amplitude as well as the spin-splitting zeros of the Shubnikov–de Haas (SdH) signal follow the LK theory. We can explain this behavior of the dHvA signal by small chemical-potential (CP) oscillations and find a very good agreement between theory and experiment. A detailed wave-shape analysis

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of the dHvA oscillations corroborates the existence of an oscillating CP. We discuss the absence of the above spin-zero effect in the SdH signal and argue that in $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ it can be explained by an incoherent variable range hopping interlayer transport which is insensitive to the small CP oscillations.

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1. Introduction

For three-dimensional (3D) metals the well-established theory of Lifshitz and Kosevich (LK) [1] can comfortably be utilized to obtain highly valuable band-structure parameters [2]. The LK theory is well proven and has the advantage of easy applicability to the experimentally measured magnetic quantum oscillations. The situation is considerably less resolved for 2D metals. Both analytical [3] as well as numerical [4] models have been developed which were proven valid somewhat later by experiments (see [5, 6] and references therein). However, in these models not all aspects have been taken into account and they are not as easy to apply as the LK theory. In addition, not all band-structure parameters can be extracted satisfactorily from the existing theories leaving some experimental features unexplained.

Prototypical examples for which the fundamental theoretical predictions can be tested are the quasi-2D organic metals based, e.g. on the organic donor BEDT-TTF (= bisethylenedithiotetraethiafulvalene or ET for short). The de Haas–van Alphen (dHvA) signal in these layered metals is usually easy to detect and it is mostly comprised by only a small number of oscillation frequencies [7]–[9]. Consequently, the Fermi surfaces are relatively simple and in most cases highly 2D, i.e. with almost negligible dispersion perpendicular to the conducting planes. Nevertheless, in dHvA signals only seldom notable deviations from the 3D LK theory appeared

This is remarkably different for the organic superconductor $\beta''$-(BEDT-TTF)$_2$SF$_5$-CH$_2$CF$_2$SO$_3$ which shows a dHvA signal almost perfectly in line with that expected for an ideal 2D metal with fixed chemical potential (CP) [5]. However, some questions remain.

9 Strong deviations from 3D LK behavior were reported for the SdH signals in some organic metals. In magnetotransport data, however, some unique phenomena may obscure the real thermodynamic behavior. See [5, 10].
That is, in order to fix the CP either an usually large additional electronic density of states (DOS), originating from a different band, has to be assumed or some localized states were proposed to be responsible. Another possibility might be the existence of edge states. One should bear in mind, however, that there exists a difference between ideal-2D and quasi-2D conductors which is reflected by different mini-band structures attached to each Landau level. In layered quasi-2D conductors the electron population of these mini-bands easily suppresses the amplitude of the CP oscillations and alters their shape as compared to the ideal 2D case.

Although the dHvA signal in $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ could be described extraordinarily well by theory, small deviations still are visible (see figure 2 below). Here, we prove this latter feature to be valid by careful additional measurements utilizing the modulation-field technique. We further report on an unusual angular dependence of the spin-splitting zeros of the second harmonic. As we will show, both effects reflect the existence of small oscillations of the CP. Especially the spin-zero anomaly of the second harmonic, therefore, offers a definite way to validate these oscillations.

In conventional metals, for which the LK theory initially was developed the spin zeros are the same both for dHvA as well as for Shubnikov–de Haas (SdH) oscillations. Our experimental data reveal, however, that this is not the case, i.e. no unusual spin-zero anomaly of the second harmonic appears in the SdH signal. We show that there is a fundamental physical reason for this discrepancy between SdH and dHvA signals caused by the interlayer incoherence.

For $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$, the value of the hopping integral between the layers is very small, $t \ll \hbar / \tau$. Under this condition, the charge carriers scatter many times within the layer before hopping to a neighboring layer, so that there is no coherent wave function perpendicular to the layers. The concept of interlayer incoherence is well established in anisotropic high-$T_c$ cuprates. For highly anisotropic layered conductors the role of incoherence on the angular-dependent magnetoresistance oscillations (AMRO) was studied and on the SdH signal for transport across the layers in.

Incoherent interlayer transport in $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ most likely is related to the observed anomalous enhancement of the dHvA and SdH oscillations in the superconducting state. The restoration of the interlayer coherence in the superconducting phase explains the observed phenomenon.

In this paper, we show that an incoherent variable range hopping (VRH) interlayer transport might be responsible for the absence of any unusual effect in the spin-splitting zeros of the SdH signal. The paper is organized as follows. In section 2, the observed second-harmonic anomaly in the dHvA data is described and explained by small CP oscillations. In section 3, the SdH measurements are presented and discussed. A summary of the results is given in section 4.

2. Spin zeros in dHvA oscillations

2.1. Experiment

We discuss here results of dHvA experiments that have been described in detail previously. Different high-quality $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ single crystals have been

10 In [11] localized states caused by a density wave were suggested which supposedly comes from a resistive maximum observed during cooling in one sample. In contrast, in all our samples we always observed a metallic resistivity leaving the existence of a density wave highly questionable.
Figure 1. Angular dependence of the fundamental ($A_1$) and second harmonic ($A_2$) of the dHvA signal of $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$. The solid lines are obtained by use of (7) and (8) assuming an oscillating CP. For $A_1$ the same result as in the LK theory is obtained. The dashed line is the behavior of $A_2$ expected from the LK theory.

measured by using a capacitance cantilever torquemeter down to about 0.4 K as well as utilizing the modulation-field technique down to $\sim$30 mK. The SdH data have been extracted from interlayer-transport measurements utilizing the usual four-point low-frequency ac method. Additional experiments have been performed in a $^3$He cryostat equipped with a 15 T magnet measuring torque-dHvA and SdH signals simultaneously. The crystals were grown by electrocrystallization at the Argonne National Laboratory [27].

For $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$, the dHvA (as well as the SdH signal) consists of only one frequency $F = F_0/\cos(\Theta)$, where $F_0 = (198 \pm 1)$ T is the dHvA frequency at $\Theta = 0$, i.e. for magnetic field applied perpendicular to the highly conducting plane [5, 25, 28]. One of the puzzling results we discuss here, is the unusual angular dependence of the second harmonic, $A_2$, of the dHvA signal (figure 1) that does not follow the behavior predicted by the LK theory (dashed line in figure 1). On the other hand, the fundamental amplitude, $A_1$, is completely in line with expectation.

To be more precise, the dHvA amplitudes in the 2D LK theory are given by

$$A_p = M^0 p^{-1} R_T(p) R_D(p) R_S(p),$$

(1)

where the prefactor $M^0 = eAS(\varepsilon_F)/(2\pi^2hm^*)$ is given by the Fermi-surface area $S(\varepsilon_F)$ and the effective cyclotron mass $m^*$; $e$ is the electron charge, $A$ the sample area, $p$ counts the harmonics, and $R_T(p)$, $R_D(p)$, and $R_S(p)$ are the usual damping factors [2]. The term dominating the
angular dependence is the spin-splitting factor given by \( R_S(p) = \cos[\frac{1}{2} p \pi g (m^*/m_e)] \), where \( g \) is the electron \( g \) factor and \( m_e \) is the free-electron mass. Since for the present superconductor \( m^*/m_e = (2.0 \pm 0.1)/\cos(\Theta) \), \( R_S(p) \) repeatedly becomes zero for those angles where the dHvA oscillations of the spin-up and spin-down electrons interfere destructively. This allows to determine \( g m^*/m_e = (3.92 \pm 0.01)/\cos(\Theta) \) from the vanishing of \( A_1 \) quite accurately. The complete angular dependence of \( A_1 \) of the torque signal\(^{11}\) can be well described with a Dingle anomaly are CP oscillations. Although the origin of these small CP oscillations is unknown so far, they cannot be described by the simple model of an ideal 2D electron gas with constant background DOS presented in [29]. Indeed, this model cannot describe the angular dependence of \( A_2 \) and it predicts shifts of the spin zeros opposite to the experimental observation. Besides, the algebraic method used in [29] cannot be generalized to real systems with arbitrary Fermi surface, nonzero temperatures, electron scattering, etc. Thus, it is worthwhile to study the influence of CP oscillations on the dHvA effect in layered conductors within a more general theoretical approach which takes account of the interlayer incoherence and can explain as well the absence of the changes in the spin-zero effect in the SdH oscillations.

We begin with the thermodynamic dHvA effect. As is well established, the oscillations of the magnetization, \( \hat{M} \), in 2D layered conductors and the oscillations of the CP, \( \mu = \mu - \varepsilon_F \), are closely related, i.e. \( \mu \propto \hat{M} \) [4, 12, 30]. The oscillating part of the magnetization can be written as

\[
\hat{M} = M^0 \exp \left[ 2\pi i p \left( \frac{F}{\hbar} + \frac{\tilde{\mu}}{\hbar \omega_c} \right) \right],
\]

(2)

with \( F = S(\varepsilon_F)/(2\pi e\hbar) \), the cyclotron frequency \( \omega_c = eB/m^* \), and \( \tilde{R}(p) = (I(p)R_T(p)R_S(p)R_D(p) \). The factor \( I(p) \) (see below) takes account of interlayer electron-hopping effects which are beyond the LK theory [31].

As we will show later in detail the CP oscillations in \( \beta'' \)-(BEDT-TTF)\(_2\)SF\(_5\)CH\(_2\)CF\(_2\)SO\(_3\) are minute and opposite to the usually assumed ideally 2D sawtooth-like shape. Under these conditions and because we are interested in corrections up to the second harmonic, \( A_2 \), we may take into account only first harmonics in the series for \( \mu \), i.e.

\[
\mu = \varepsilon_F - \eta \sin \left( 2\pi \frac{F}{B} \right) + \cdots.
\]

(3)

\(^{11}\) In torque dHvA measurements the additional factor \( \sin(\Theta)/\cos^2(\Theta) \) occurs for the present 2D Fermi surface leading to zero amplitudes at \( \Theta = 0 \).
Using the notations \( \eta = h \omega_c \hat{R}(1)/\pi, \ z_p = 2\pi p F/B, \ \eta_p = 2\pi p \eta / h \omega_c, \) and the identity

\[
\exp(-i \eta_p \sin z_1) = \sum_{n=-\infty}^{\infty} (-1)^n J_n(\eta_p) \exp(i z_n),
\]

(4)

where \( J_n(\eta_p) \) is the Bessel function of the order \( n, \) one can write the magnetization (2) in the standard form

\[
\tilde{M} = \sum_{n=1}^{\infty} A_n \sin \left( 2\pi n \frac{F}{B} \right).
\]

The amplitudes of the harmonics are given by

\[
A_n = M^0 \sum_{p=1}^{\infty} \frac{(-1)^p}{p} \hat{R}(p) \left[ J_{p-n}(\eta_p) - J_{p+n}(\eta_p) \right].
\]

(6)

The amplitudes \( A_n \) are, therefore, weighted sums of the terms \( \hat{R}(p). \) In addition to the standard LK theory the latter term contains the factor \( I(p) \) that takes into account such effects as interlayer hopping [12, 31] or the dispersion of magnetic-breakdown bands [13, 32]. (For \( \beta^{-}(BEDT-TTF)_2SF_2C_6H_2CF_2SO_3, \) however, magnetic breakdown is irrelevant as has been shown by SdH measurements up to 60 T [33]). Important consequences of (6) are deviations from the usual LK temperature and magnetic-field dependences. This is realized, e.g. in the effective masses which apparently become smaller for each higher harmonic when extracted by use of the LK formula [2]. In the present case, an apparent effective mass of only about 1.5 \( m_c \) is obtained for the second harmonic.

What is of importance here, is that the angular spin-zero positions also differ from those predicted in the LK approach. To show this in more detail we consider the fundamental and second harmonic which can be compared to the experimental data. Since the small correction to the CP, given in (3), oscillates with the frequency of the first harmonic it gives corrections only to the second and higher harmonics. Accordingly, the relevant Bessel functions in (6) can be approximated up to the first-order in \( \eta \) as \( J_0(\eta_1) \approx J_0(\eta_2) \approx 1 \) and \( J_1(\eta_1) \approx \eta_1/2 = \hat{R}(1) \) resulting in

\[
A_1 = -M^0 \hat{R}(1), \quad (7)
\]

\[
A_2 = M^0 \left[ -\frac{1}{2} \hat{R}(2) + \hat{R}(1)^2 \right]. \quad (8)
\]

Therefore, the amplitude of the fundamental, \( A_1, \) basically remains identical to that predicted by the LK formula, whereas \( A_2 \) becomes a linear combination of the damping factors \( \hat{R}(2) \) and \( \hat{R}(1)^2. \) Consequently, care has to be taken when extracting band-structure parameters from the second harmonic. Besides the modified temperature dependence (\( A_2 = a R_T^2(1) + b R_T(2) \) is a linear combination of the two temperature factors), an unusual angular dependence of \( A_2 \) with shifted spin-splitting zeros results. The latter are determined by \( \left[ \frac{1}{2} \hat{R}(2) + \hat{R}(1)^2 \right] = 0. \) With \( R_D(1)^2 = R_D(2) \) and \( R_S(p) \) as stated above, the spin-splitting zeros are given by

\[
\cos \left( \frac{\pi g m^*}{m_c} \right) I(2) + 2 \cos^2 \left( \frac{\pi g m^*}{2 m_c} \right) \frac{R_T^2(1)}{R_T(2)} I^2(1) = 0.
\]

(9)

Thus, the zeros are shifted as compared to the LK theory where the second term is absent. The shift is a weak function of temperature and magnetic field caused by the factors \( R_T(p) \) and \( I(p) = \int g_L(\epsilon) \exp(2\pi i \epsilon / h \omega_c) d\epsilon \) [31].

New Journal of Physics 10 (2008) 083032 (http://www.njp.org/)
Figure 2. Comparison of the measured dHvA data (open symbols) with the calculated signals for a 2D metal with fixed number of charge carriers (dashed line) and for a 2D metal with fixed CP (solid line).

The above equations are valid both for the coherent and incoherent electron hopping across the layers. In the coherent case, the layer-stacking factor $I(p)$ for a simple cosine-like interlayer dispersion $\varepsilon(p_z) = t \cos(p_z a/\hbar)$ can be written as $I(p) = J_0(4\pi tp/\hbar\omega_c)$, where $t$ is the interlayer-hopping integral [31]. For the present 2D superconductor there is no detectable dispersion across the layers [15] and the hopping integral is very small compared to $\hbar\omega_c$. Therefore, one may speculate that the width of the DOS $g_L(\varepsilon)$ is of the order $t$, so that $I(p)$ can be well approximated by 1.

The excellent agreement between our theory and experiment is evident from figure 1 where we used equations (7) and (8) to obtain the solid lines. Since all experimental parameters, $m^*, T, B$ and $T_D$, are well known there is no free parameter except for simple scaling factors.

This result implies that a weak oscillation of the CP exists. Indeed, when analyzing in detail the experimental dHvA wave shape small deviations from the 2D LK behavior for fixed CP can be resolved. In figure 2, it is obvious that the observed steep increase of the dHvA signal cannot be described satisfactorily by the theoretical 2D LK behavior (solid line in figure 2). This corroborates the notion of an oscillating CP. This oscillation, however, must be different from the usually predicted sawtooth-like 2D behavior [3], visualized by the dashed line in figure 2.

Since possible artifacts, such as torque interaction, might obscure the dHvA signal we checked the validity of our torque result by comparing it with modulation-field data (figure 3). For a modulation-field amplitude not too large and signal detection on the second harmonic, the modulation-field data are approximately proportional to the second derivative of the magnetization with respect to $B$ times $B^4$ [2]. The excellent agreement between both signals is evident[12]. This proves the validity of both experimental data and verifies the deviation

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[12] The modulation-field data were taken at 30 mK, the torque data at 0.44 K. Slightly different Dingle temperatures ($T_D = 0.4$ K of the torque sample and $T_D = 0.58$ K for the modulation-field sample) partly compensate this and lead to the particularly good agreement.
from the 2D LK behavior with fixed CP as real. Indeed, for the modulation-field data the deviation appears even more pronounced since the second derivative of $M$ is analyzed (inset of figure 3).

Consequently, the shapes of the modulation-field and torque-dHvA signals substantiate the existence of an oscillating CP. In line with our assumption, this oscillation is small as apparent from the minute wave-shape effects, but must be more elaborate than the simple lowest-order sinusoidal waveform considered in (3). Qualitatively, the waveform of $\tilde{\mu}$ must be fast changing for rising $M$ and slowly varying for decreasing $M$, similar as visualized in figure 1 of [4] for an almost fixed CP.

As shown in [12, 13], CP oscillations in layered metals are much more complex than the usually assumed simple saw-tooth form valid only for an ideal 2D electron gas. This is caused by the electronic mini-band structure attached to each Landau level. This mini-band structure strongly reduces the amplitude of the CP oscillations depending on the DOS within the mini-bands. For an ideal 2D electron gas the Landau levels are flat and at zero temperature $\mu(B)$ jumps between the Landau levels with amplitude $\hbar\omega_c$. For 3D metals the amplitude of the CP oscillations reduces to $\hbar\omega_c/\sqrt{\hbar\omega_c/\varepsilon_F}$. In superlattices and layered conductors the amplitude is between this value and $\hbar\omega_c$. Although the mini-band structure in organic conductors is unknown, the occupied bands below $\varepsilon_F$ stabilize the CP in quasi-2D organic metals [12], such as $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$. Therefore, the observed inverse shape of the CP oscillations is not surprising, since it holds even for the pure 2D lattice in a magnetic field as was shown recently in numerical studies of the Azbel-Hofstadter problem [34].
Figure 4. Angular dependence of the fundamental ($A_1$) and the second harmonic ($A_2$) of the SdH signal. The solid lines show the expected behavior according to the 2D LK theory.

3. Spin zeros in SdH oscillations

3.1. Experiment

The angular dependences of the SdH amplitudes are shown in figure 4. Surprisingly, in view of the preceding discussion, no anomalous shifts of the spin-splitting zeros occur. The LK theory describes the angular dependences very well (except for some $A_2$ points close to $\Theta = 0$). In particular, the zeros of the second harmonic lie exactly at the positions expected for a metal with fixed CP.

As a possible explanation for this difference between dHvA and SdH spin-split zeros, one could think of the electrical leads, necessary for measuring the SdH signal, acting as possible charge-carrier reservoirs. This would lead to a fixed CP and, consequently, to the observed LK behavior. In order to check this assumption we performed additional experiments, measuring dHvA and SdH oscillations on the same sample at the same time. For this, a sample with four gold leads for the transport measurement was placed on a cantilever torquemeter and measured in magnetic fields up to 13 T. Before that, the dHvA signal was measured without leads attached to the sample. As seen in figure 5, also for this sample and for the lower field range the dHvA spin zeros of the second harmonic, $A_2$, cannot be described by the LK theory but by (8), i.e. taking into account an oscillating CP.

If indeed external leads would pin the CP to a fixed value, the spin zeros of $A_2$ would occur at the positions given by the LK theory (dashed line in figure 5). However, our measurements clearly show that even with leads attached to the sample the spin zeros
Figure 5. Angular dependence of the fundamental ($A_1$) and the second harmonic ($A_2$) of the torque dHvA signal of a second $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ sample. The closed circles were obtained from a measurement where no leads were attached to the sample, whereas the open circles were extracted from a measurement where dHvA and SdH oscillations were measured simultaneously. The solid lines are obtained using (7) and (8), the dashed line is the behavior of $A_2$ expected from the LK theory.

of $A_2$ remain unaffected in the dHvA signal (figure 5). The only effect of the leads is an overall smaller signal amplitude due to a reduced sensitivity of the torquemeter. This clearly proves that a fundamental difference between the dHvA and SdH effects exists in $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ and CP oscillations have to be taken into account in the SdH case. This is, however, a challenging task since the incoherent transport across the layers is not fully understood so far. For example, for $\beta''$-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ such unusual effects as the magnetic-field-induced metal–insulator transition [10], the anomalous enhancement of the quantum oscillation amplitudes in the superconducting state [26], etc are yet to be explained in detail [35]. Some progress was achieved recently in the theoretical description of these effects [24]. Based on previously published experimental data [10], here we propose the VRH mechanism as being responsible for these effects as well as for the absence of the spin-zero shift in the SdH oscillations.

3.2. The VRH, scaling, and ineffectiveness of the CP oscillations on the SdH oscillations

The principal difference between dHvA and SdH oscillations in quasi-2D metals is caused by the fact that thermodynamic dHvA oscillations are completely determined by the Landau quantization within the layers. Disorder broadens the Landau states and may result in
localization, which changes dramatically the SdH conductivity across the layers, but hardly influences the dHvA oscillations.

In $\beta''\text{-}(\text{BEDT-TTF})_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$, a metal–insulator transition was found at a critical field of $B_0 = 3.5$ T [10, 35]. Below $B_0$, the background magnetoresistance, $R_b$, shows a metal-like temperature dependence, whereas above $B_0$ the temperature dependence of $R_b$ is insulating. Moreover, the normalized $R_b$ can be scaled onto a universal curve by use of the scaling variable $X = (B - B_0)/T\kappa$, with the critical exponent $\kappa = 0.65$ (see figure 4 in [10]). The resulting curve may be approximated by two (close to straight) lines crossing smoothly at $X = 10$. Below and above this point the data of the background conductivity across the layers can be described by the function

$$\sigma_{zz} = \sigma_{zz}(0) \exp\left(-\sqrt{\frac{T_0}{T}}\right),$$  \hspace{1cm} (10)

which describes a Mott VRH conductivity. Thereby, $T_0 \propto 1/\xi \propto |B - B_0|^\gamma$, where $\xi$ is the correlation length and $\gamma^{-1} = 0.65$ has been taken from experiment [10]. This VRH mechanism would explain nicely the found scaling behavior. A theory of the SdH oscillations in layered organic conductors with incoherent VRH hopping across the layers on a background of the metal-to-insulator transition with the above scaling behavior was developed in [24].

Thus, $R_b = 1/\sigma_{zz}$ displays a crossover between two VRH regimes. Although the nature of this crossover is unclear, it may be caused by intra- and inter-layer hopping. The VRH conductivity (10) is well documented in semiconductor 2D integer quantum Hall effect (IQHE) systems (see [36, 37] and references therein). In $\beta''\text{-}(\text{BEDT-TTF})_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$, the conducting layers are weakly coupled with the CP nearly fixed which favors the IQHE within the layers. Therefore, at low temperatures and high enough fields, one can expect the IQHE within the layers. Because of the interlayer incoherence electrons scatter many times within the layers before hopping to the neighboring layer in this organic conductor. That gives a contribution to the conductivity as described by (10). Electron tunneling across the layers is a 1D VRH process which also yields a conductivity given by (10) but with different coefficients.

At $B_0$, the correlation length $\xi$ has a singularity which can be interpreted as a field-induced transition to a quantum-Hall-insulator state within the layers [38]–[40]. The quantum-Hall-insulator regime assumes the absence of percolation of the Landau orbits drifting along the equipotential contours within the weakly coupled layers. The conductivity across the layers is provided by electron tunneling between equipotential contours belonging to neighboring layers. This explains the nonzero magnetoresistance across the layers, $R_{zz} = 1/\sigma_{zz}$, even if these layers are in the Hall-insulator state (see [24] for more details).

The important point is that the VRH mechanism means that charge carriers hop between localized states within a narrow energy range near the Fermi level (CP). The width of this energy range decreases for $T \to 0$. The exact position of the CP within this range is not important. Therefore, small oscillations of the CP do not change the VRH conductivity across the layers and cannot change the shape of the SdH oscillations in contrast to the dHvA oscillations. This would explain the absence of the spin-zero shift in the SdH oscillations.

4. Conclusions

In conclusion, we observed and explained quantitatively an anomalous angular dependence of the dHvA signal in a 2D organic metal, i.e. in the limit $\hbar\omega_c \gg \hbar/\tau \gg t$. This is shown to be a genuine effect of the two dimensionality that can be utilized as direct proof for an oscillating
CP. In the present case, these oscillations are very small but directly visible in the detailed dHvA wave shape. Our analytical theory explains, in the most general way, the spin-zero anomaly of the second harmonic in dHvA oscillations in layered conductors. In SdH experiments of the same material, no spin-zero anomalies are detected. In a simultaneous torque and transport measurement we have checked that this effect is not caused by the contacts applied to the sample for SdH measurements. The experimental data in [10] suggest that the conductivity in $\beta''-(BEDT-TTF)_2SF_5CH_2CF_2SO_3$ is due to the Mott VRH mechanism. Small CP oscillations do not affect the electron hopping between localized states and do not change the conductivity across the layers. The spin-zero positions in the SdH signal, therefore, would not be affected by CP oscillations as seen experimentally.

Acknowledgments

We thank T Maniv and V Zhuravlev for fruitful discussions. Part of this work was supported by INTAS, project INTAS-01-0791, by EuroMagNET under the EU contract RII3-CT-2004-506239, and by the ESF Scientific Programme on Fermi-liquid instabilities in correlated metals (FERLIN). Argonne, a US Department of Energy Office of Science laboratory, is operated under Contract No DE-AC02-06CH11357. Work at Portland State University was supported by NSF (Che-9904316). VMG is grateful to P Fulde and S Flach for the hospitality at the MPIPKS in Dresden.

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New Journal of Physics 10 (2008) 083032 (http://www.njp.org/)