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Anomalous beating phase of the oscillating interlayer magnetoresistance in layered metals

P. D. Grigoriev,[1,2] M. V. Kartsovnik,[3] W. Biberacher,[3] N. D. Kushch,[4] and P. Wyder[1]
[1]*Grenoble High Magnetic Field Laboratory, MPI-FKF and CNRS, BP 166, F-38042 Grenoble Cedex 09, France*
[2]*L. D. Landau Institute for Theoretical Physics, 142432 Chernogolovka, Russia*
[3]*Walther-Meißner-Institut, Bayerische Akademie der Wissenschaften, Walther-Meißner-Strasse 8, D-85748 Garching, Germany*
[4]*Institute of Problems of Chemical Physics, 142432 Chernogolovka, Russia*

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We analyze the beating behavior of the magnetic quantum oscillations in a layered metal under the conditions when the cyclotron energy $\hbar\omega_c$ is comparable to the interlayer transfer energy t. Using a simple semiclassical model, we show that the positions of the beats in the interlayer resistance are considerably shifted from those in the magnetization oscillations. The shift is determined by the ratio $\hbar\omega_c/t$ that may lead to significant deviations from the conventional periodicity of the beats in the scale of inverse magnetic field. A comparative study of the Shubnikov–de Haas and de Haas–van Alphen oscillations in the layered organic metal β-(BEDT-TTF)$_2$IBr$_2$ is performed and shown to be consistent with the theoretical prediction.

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In the last decade, the de Haas–van Alphen (dHvA) and Shubnikov–de Haas (SdH) effects were extensively used for studying quasi-two-dimensional (Q2D) organic metals.[1–3] Due to extremely high anisotropies of the electronic systems the amplitudes of the oscillations are strongly enhanced in high-quality samples of these materials (see, e.g., Refs. 4 and 5) and often cannot be described by the Lifshitz-Kosevich (LK) formula derived for conventional three-dimensional metals.[6] A number of theoretical works performed in the last years on the dHvA effect in two-dimensional (2D) and Q2D systems[7–13] provide a consistent theory which can be used for a quantitative analysis of experimental data obtained on Q2D systems as long as many-body interactions only lead to constant renormalization effects and magnetic breakdown effects are not concerned.

The theory of the Q2D SdH effect was also of a great interest in recent years[9,14–17] but the situation here is more complicated and even some qualitative questions remain open. One of these open questions is the origin of the phase shift in the beats of the resistivity oscillations with respect to those in the magnetization. The beating behavior of the oscillations in Q2D metals is well known to originate from the slight warping of their Fermi surfaces in the direction normal to the 2D plane. The superposition of the contributions from the maximum and minimum cyclotron orbits is expected to lead to an amplitude modulation of the kth harmonic by the factor $\cos(2\pi k\Delta F/2B-\pi/4)$, where B is the magnetic field and $\Delta F=(c\hbar/2\pi e)(A_{\max}-A_{\min})$ is the difference between the oscillation frequencies caused by the extremal orbits with the k-space areas $A_{\max}$ and $A_{\min}$, respectively.[6] From the beat frequency one can readily evaluate the warping of the Fermi surface (FS) and hence the interlayer transfer integral, $4t \approx \epsilon_F\Delta F/F$ (see, e.g., Refs. 1 and 18). The situation becomes less clear when the warping is so weak that less than one-half of the beat period can be observed experimentally. In principle, an observation of one single node would already be quite informative,[19] provided the phase offset (i.e., the phase of the beat at $1/B\rightarrow 0$) is known. In the standard LK theory this phase offset is strictly determined by geometrical reasons to be equal to $-\pi/4$ for both dHvA and SdH effects.[6]

However, recent experiments on layered organic metals κ-(BEDT-TTF)$_2$ Cu[N(CN)$_2$]Br (Ref. 19) and (BEDT-TTF)$_4$[Ni(dto)$_2$] (Ref. 20) have revealed a significant difference in the node positions of beating dHvA and SdH signals. The respective phase shift in the latter compound was estimated to be as big as $\pi/2$. Noteworthy, in both cases the oscillation spectrum was strongly dominated by the first harmonic when no substantial deviations from the standard LK theory was expected.

Although several potential reasons for this behavior have been outlined in Refs. 19 and 20 the most plausible one seems to be its association with the Q2D nature of the electronic system.[19] However, the limited amount of the reported experimental data is not sufficient for a detailed analysis. Moreover, multiple-connected FS's typical of both compounds might lead to additional complications due to effects of magnetic breakdown, interband scattering, etc.

In order to clarify the problem, we have carried out comparative studies of the oscillating magnetization and interlayer resistivity of the radical cation salt β-(BEDT-TTF)$_2$IBr$_2$. This material exhibits a relatively simple behavior without superstructure transitions or insulating instabilities. It is normal metallic down to low temperatures and undergoes a superconducting transition at $T_c \approx 2$ K.[21] Its electronic properties are basically determined by a single cylindrical FS (Ref. 22) slightly (by $\simeq 1\%$) warped in the direction perpendicular to the highly conducting BEDT-TTF layers.[23,25] Thus, the present compound appears to be an ideal object for our purposes. The experiment was done on a high-quality single crystal at $T \approx 0.6$ K in magnetic field up to 16 T. To assure exactly the same conditions for the dHvA and SdH effects (in particular, identical field orientations are of crucial importance) the measurements were performed in a setup providing a simultaneous registration of the magnetic torque and resistance.[19,26] In the field range between 7 and 16 T we have observed clear beating with several nodes in both dHvA and SdH signals. The positions of the beat nodes in the SdH signal are found to be different from those in the dHvA signal, the difference being dependent on the magnetic field.

Below we propose an explanation of the phenomenon based on the consideration of both density of states (DOS) and Fermi velocity oscillations contributing to the interlayer magnetotransport in a Q2D metal and then compare the theoretical estimations with the experimental results.

We consider a Q2D metal in a magnetic field perpendicular to the conducting layers with the energy spectrum

$$\epsilon_{n,k_z}=\hbar\omega_c(n+1/2)-2t\cos(k_z d), \tag{1}$$

where t is the interlayer transfer integral, k_z is the wave vector perpendicular to the layers, d is the interlayer distance, and $\omega_c=eB/m^*c$ is the cyclotron frequency. Both $\hbar\omega_c$ and t are assumed to be much smaller than the Fermi energy.

The DOS of electron gas with this spectrum can be easily obtained performing the summation over all quantum numbers at a fixed energy:

$$g(\epsilon)=\sum_{n=0}^{\infty}\frac{N_{LL}}{\sqrt{4t^2-[\epsilon-\hbar\omega_c(n+1/2)]^2}}, \tag{2}$$

where N_{LL} is the Landau level degeneracy. The sum over Landau levels can be represented as a harmonic series using the Poisson summation formula.[28] As a result one gets[12]

$$g(\epsilon)\propto 1+2\sum_{k=1}^{\infty}(-1)^k\cos\left(\frac{2\pi k\epsilon}{\hbar\omega_c}\right)J_0\left(\frac{4\pi kt}{\hbar\omega_c}\right). \tag{3}$$

We shall consider the case $4\pi t>\hbar\omega_c$ when the beats can be observed. Then the zeroth-order Bessel function $J_0(\pi k4t/\hbar\omega_c)$ describing the beating of the DOS oscillations can be simplified, as $J_0(x)\approx\sqrt{2/\pi x}\cos(x-\pi/4)$. Further, we consider the limit of strong harmonic damping, retaining only the zeroth and first harmonics. In this limit the oscillations of the chemical potential μ can be neglected. Knowing the DOS we can now obtain the oscillating part of the magnetization as[12]

$$\tilde{M}\propto\sin\left(\frac{2\pi\mu}{\hbar\omega_c}\right)\cos\left(\frac{4\pi t}{\hbar\omega_c}-\frac{\pi}{4}\right)R_T, \tag{4}$$

where R_T is the usual temperature smearing factor. This expression coincides with the result of the three-dimensional LK theory[6] and allows an evaluation of t from the beat frequency.

The interlayer conductivity σ_{zz} can be approximately evaluated from the Boltzmann transport equation,[27] assuming the impurities are pointlike:

$$\sigma_{zz}=e^2\sum_{m\equiv\{n,k_z,k_x\}}v_z^2(k_z)\,\delta(\epsilon(n,k_z)-\mu)\tau(\mu)$$
$$\equiv e^2 I(\mu)\tau(\mu), \tag{5}$$

where v_z is the z component of the electron velocity and $\tau(\mu)$ the momentum relaxation time at the Fermi level. The latter, in Born approximation, is inversely proportional to the DOS: $1/\tau(\mu)\propto g(\mu)$ and oscillates in magnetic field according to Eq. (3). In addition, when the cyclotron energy is comparable to the warping of the FS, the oscillations of the electron velocity summed over the states at the Fermi level, $I(\mu)\equiv\Sigma_{\epsilon=\mu}|v_z|^2$ also become important.[9] To calculate this quantity one has to perform the integrations over k_x and k_z in Eq. (5) and substitute the expression for v_z:

$$v_z(\epsilon,n)=\frac{d}{\hbar}\sqrt{4t^2-[\epsilon-\hbar\omega_c(n+1/2)]^2}. \tag{6}$$

As a result one obtains

$$I(\epsilon)=\sum_{n=0}^{\infty}\frac{N_{LL}d^2}{2\pi\hbar}\sqrt{4t^2-[\epsilon-\hbar\omega_c(n+1/2)]^2}$$
$$=\frac{4N_{LL}d^2t}{\pi\hbar^2}\left[\frac{4\pi t}{8\hbar\omega_c}+\sum_{k=1}^{\infty}\frac{(-1)^k}{2k}\cos\left(\frac{2\pi k\epsilon}{\hbar\omega_c}\right)J_1\left(\frac{4\pi kt}{\hbar\omega_c}\right)\right]. \tag{7}$$

The first-order Bessel function $J_1(4\pi kt/\hbar\omega_c)$ entering Eq. (7) also describes beatings but with a phase different from that of the DOS beatings given by $J_0(4\pi kt/\hbar\omega_c)$ in Eq. (3): at large x, $J_1(x)\approx\sqrt{2/\pi x}\sin(x-\pi/4)$ is just shifted by $\pi/2$ with respect to $J_0(x)$.

The phase shift in the beating of the oscillations in $g(\epsilon)$ and $I(\epsilon)$ is illustrated in Fig. 1 in which these quantities are plotted for two different values of the ratio $4t/\hbar\omega_c$. When $4t/\hbar\omega_c=2.25$ the DOS oscillations have a maximum amplitude of the first harmonic [Fig. 1(a)]. At the same time the oscillations of $I(\epsilon)$ exhibit a nearly zero amplitude of the first harmonic as shown in Fig. 1(c) while their second harmonic (not shown in the figure) is at the maximum. By contrast, when $4t/\hbar\omega_c=1.8$ [Fig. 1(b,d)] the first harmonic of the DOS is at the node whereas that of $I(\epsilon)$ has the maximum amplitude.

The difference between the phases of the beats in oscillating $g(\epsilon)$ and $I(\epsilon)$ leads to a shift of the beat phase of the SdH oscillations with respect to that of the dHvA oscillations. Indeed, taking into account that $\tau\propto 1/g$, substituting Eqs. (3) and (7) into Eq. (5), applying the large argument expansions of $J_0(x)$ and $J_1(x)$, and introducing the temperature smearing, we come to the following expression for the first harmonic of the interlayer conductivity:

$$\tilde{\sigma}_{zz}\propto\cos\left(\frac{2\pi\mu}{\hbar\omega_c}\right)\cos\left(\frac{4\pi t}{\hbar\omega_c}-\frac{\pi}{4}+\phi\right)R_T, \tag{8}$$

where

$$\phi=\arctan(a)\quad\text{and}\quad a=\hbar\omega_c/2\pi t. \tag{9}$$

Comparing these expressions with Eq. (4), we see that the beats in the SdH and dHvA oscillations become considerably shifted with respect to each other as the cyclotron energy approaches the value of the interlayer transfer integral.

The above evaluation is based on the semiclassical Boltzmann equation and certainly is not expected to give a precise result. Nevertheless, as will be seen below, its predictions concerning the phase shift are in good qualitative agreement with the experiment and we therefore believe that it correctly reflects the physics of the phenomenon.

Figure 2 shows the oscillating parts of the magnetization and interlayer magnetoresistance in the normal state of β-(BEDT-TTF)$_2$IBr$_2$ at magnetic field tilted by $\theta\approx 14.8°$ from the normal to the BEDT-TTF layers. The curves have

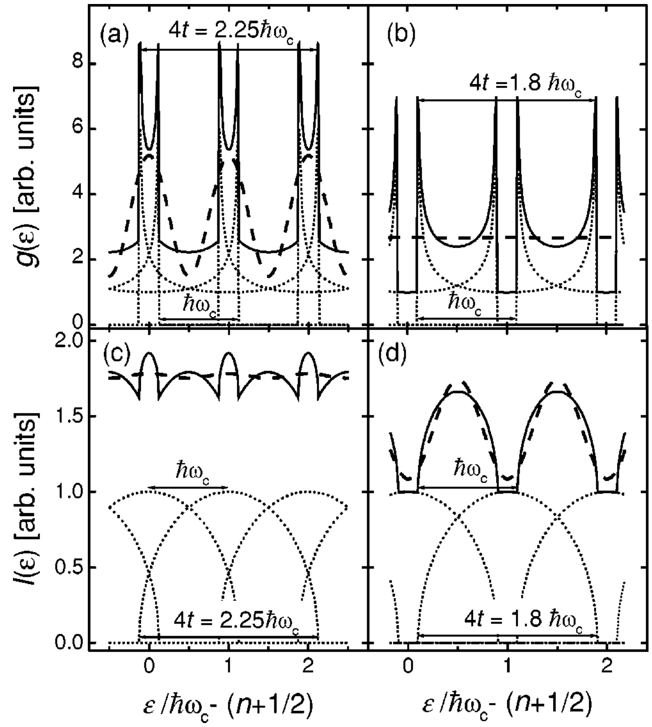

FIG. 1. (a) DOS (solid line) near the Fermi level and its first harmonic (dashed line) according to Eq. (3), at the ratio $4t/\hbar\omega_c = 2.25$; (b) the same at the ratio $4t/\hbar\omega_c = 1.8$; (c) the quantity $I(\epsilon)$ (solid line) and its first harmonic (dashed line) according to Eq. (7), at $4t/\hbar\omega_c = 2.25$; (d) the same at $4t/\hbar\omega_c = 1.8$. In all four panels: the dotted lines are the contributions from individual Landau levels.

been obtained by subtracting slowly varying backgrounds from the measured magnetic torque $\tau(B)$ and resistance $R(B)$ and (for the magnetization oscillations, $\tilde{M} \propto \tau/B$) subsequent dividing by B. The torque was measured using the capacitance cantilever torque magnetometer described in detail in Ref. 29. The oscillations of the capacitance due to the dHvA effect did not exceed 2×10^{-4} pF or $\approx 0.04\%$, corresponding to an angular displacement of $\leqslant 0.001°$. Thus, any artificial effects of changing the field orientation during the field sweep can be neglected. The fast Fourier transformation (FFT) spectra shown in the insets reveal the fundamental frequency of ≈ 3930 T in agreement with previous works.[23,25] The second harmonic contribution is about 1% of that from the fundamental one at the highest field.

Clear beats with four nodes are seen in both the dHvA and SdH curves. We have assured that the observed beats originate from the warping of the cylindrical FS by checking the angular dependence of their frequency.[25] The positions of the nodes determined as midpoints of narrow field intervals at which the oscillations inverse the phase are plotted in Fig. 3. The straight line is a linear fit of the magnetization data revealing the beat frequency $\Delta F = 40.9$ T that, according to Eq. (4), corresponds to $4t/\epsilon_F = \Delta F/F = 1/96$. The error bars in the node positions do not exceed $\pm 3\times10^{-4}$ T^{-1} for $N = 3$–5 and are somewhat bigger, $\simeq \pm 10^{-3}$ T^{-1}, for $N = 6$ due to a lower signal-to-noise ratio. We note that although the angle $\theta = 14.8°$ corresponds to a region in the vicinity of

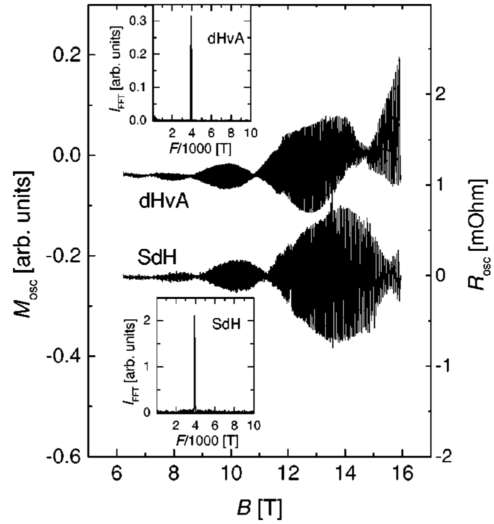

FIG. 2. dHvA (left scale) and SdH (right scale) oscillations in β-(BEDT-TTF)$_2$IBr$_2$ at $\theta \approx 14.8°$. Insets: corresponding FFT spectra.

the maximum beat frequency $[\max\{\Delta F(\theta)\} \approx 42.0$ T for the given field rotation plane], the sensitivity of the node positions to the field orientation is still quite high: the nodes shift by $\approx 2.3\times10^{-3}$ T^{-1} at changing θ by 1°. Thus, if one has to remount the sample between the torque and resistance measurements, even a slight misalignment may cause a substantial additional error. In our experiment both quantities were measured at the same field sweep, hence, such an error was eliminated.

From Figs. 2 and 3 one can see that the nodes of the SdH oscillations are considerably shifted to higher fields with re spect to those of the dHvA oscillations. The shift grows with increasing field. Both these observations are fully consistent with the above theoretical prediction.

In order to make a further comparison between the experiment and theory, we plot the quantity $\tan(\phi)$ (where ϕ is the

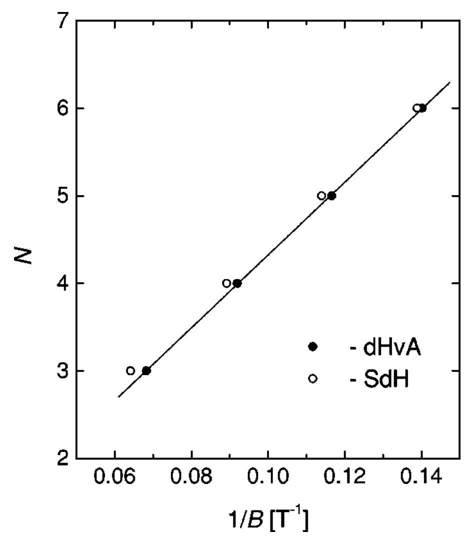

FIG. 3. The positions of the nodes in the oscillating magnetization (filled symbols) and resistance (open symbols) versus inverse field. The straight line is the linear fit to the magnetization data.

phase shift between the beating of the SdH and dHvA oscillations obtained from Fig. 3) as a function of magnetic field in Fig. 4. A linear fit to this plot (dashed line in Fig. 4) has a slope of 0.0371/T.[30] A substitution of this value and the cyclotron mass $m^*=4.2m_e$ into Eq. (9) yields an estimation for the interlayer bandwidth $4t\simeq 0.48$ meV or the ratio $4t/\epsilon_F=\Delta F/F\simeq 1/230$. This is somewhat smaller than the value 1/96 obtained directly from the ratio between the beating and fundamental frequencies. However, taking into account an approximate character of the presented theoretical model, the difference is not surprising. Further theoretical work is needed in order to provide a more explicit basis for the quantitative description of the phenomenon.

Summarizing, the beats of the SdH oscillations in β-(BEDT-TTF)$_2$IBr$_2$ are found to be shifted towards higher fields with respect to those of the dHvA signal. We attribute this effect to interfering contributions from oscillating DOS and Fermi velocity to the interlayer conductivity of this layered compound. The observed behavior appears to be a general feature of Q2D metals which should be taken into account whenever the cyclotron energy becomes comparable to the interlayer transfer energy. Of a particular importance is the dependence of the beat phase on the magnetic field strength. Ignorance of this fact may lead to considerable errors in estimations of the FS warping from the beat frequency when $2t\simeq\hbar\omega_c$.

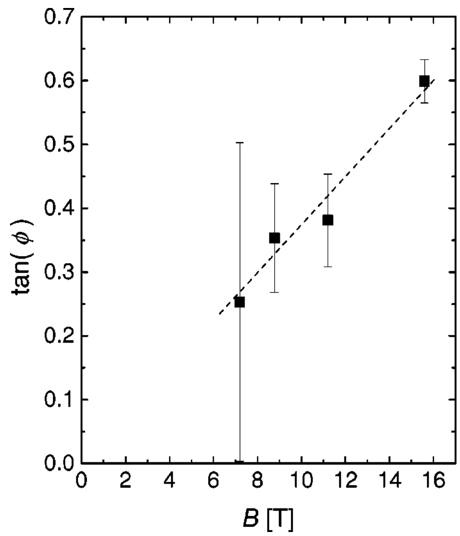

FIG. 4. Tangent of the phase shift between the node positions in the SdH and dHvA signals taken from the data on Fig. 3 as a function of magnetic field. The dashed line is a linear fit according to Eq. (9) (Ref. 30).

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[30] For the fit shown in Fig. 4 only the data corresponding to the nodes with $N=3-5$ were used. The phase shift for $N=6$ was disregarded due to the very large error bar.