Slow oscillations of magnetoresistance in quasi-two-dimensional metals

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Abstract

Slow oscillations of the interlayer magnetoresistance observed in the layered organic metal $\beta$-(BEDT-TTF)$_2$IBr$_2$ are shown to originate from the slight warping of its Fermi surface rather than from independent small cyclotron orbits. Unlike the usual Shubnikov-de Haas effect, these oscillations are not affected by the temperature smearing of the Fermi distribution and can therefore become dominant at high enough temperatures. We suggest that the slow oscillations are a general feature of clean quasi-two-dimensional metals and discuss possible applications of the phenomenon.
Quantum oscillations of magnetization (de Haas-van Alphen, dHvA, effect) and magnetoresistance (Shubnikov-de Haas, SdH, effect) in strong magnetic fields have been successfully used for many years in order to investigate Fermi surfaces and other electronic properties of metals [1]. In three-dimensional (3D) metals the oscillations are determined by only a small fraction of conduction electrons near extremal cyclotron orbits on the Fermi surface (FS). Therefore they are very weak and can perfectly be described in terms of the so-called Lifshitz-Kosevich (LK) formalism [1]. Recently, however, materials of lower dimensionality such as two-dimensional electron gas systems, layered metal oxides and organic conductors have been of high interest. In these materials the relative number of electrons contributing to the oscillations is much higher than in 3D metals and the main assumptions of the standard theory, i.e. a weak oscillation amplitude and constant chemical potential, are often no longer fulfilled.

Some crystalline organic metals, being very clean and having extremely large anisotropy of electronic properties, are excellent objects for studying specific features of the magnetic quantum oscillations in the low-dimensional limit [2, 3, 4, 5, 6]. Various deviations of the oscillatory magnetization from the LK theory observed in these compounds can be fairly well explained by a semi-phenomenological description of the quasi-two-dimensional (Q2D) dHvA effect developed in a number of theoretical works [3, 7, 8, 9]. The situation with the SdH effect is less satisfactory. Despite some progress in understanding several anomalies of the oscillatory magnetoresistance [3, 10, 11] a consistent theory is still lacking. Moreover, there are still open qualitative questions. One of these questions concerns the origin of slow oscillations of magnetoresistance which were first found in the Q2D organic metal $\beta$-(BEDT-TTF)$_2$IBr$_2$ [12, 13]. Similar oscillations have been observed in other layered organic conductors, e.g. $\beta$-(BEDT-TTF)$_2$I$_3$ [14, 15], $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ [16], and $\kappa$-(BEDT-TSF)$_2$C(CN)$_3$ [17]. Since the behavior of these oscillations strongly resembles that of the SdH effect, they have been supposed to originate from additional very small pockets of the FS. However, the band structure calculations which are basically believed to correctly reproduce the FS topology of organic metals (see e.g. [18] for a review) do not explain such small groups of carriers in any of the cited compounds.

In this Letter, we report on detailed studies of the oscillating interlayer magnetoresistance and magnetization of $\beta$-(BEDT-TTF)$_2$IBr$_2$ at various orientations of magnetic field. Our results provide an unequivocal evidence that the slow oscillations of the magnetoresistance
do not reveal any new carriers but are ultimately caused by a weak warping of the single cylindrical FS in this Q2D metal. We propose a theoretical explanation of the phenomenon which appears to be in good agreement with the experiment.

The experiment was performed on a high-quality \(R(290\,\text{K})/R(2\,\text{K}) \geq 3000\) single crystal with the dimensions \(0.6 \times 0.3 \times 0.12\,\text{mm}^3\). The sample was mounted into a measuring cell in a \(^3\text{He}-\text{cryostat}\) allowing simultaneous registration of the resistance and magnetic torque \([15]\) at different orientations of the magnetic field produced by a 14 T superconducting magnet. The field orientation was defined by the angle \(\theta\) between the field direction and the normal to the highly conducting \(ab\)-plane.

The FS of \(\beta-(\text{BEDT-TTF})_2\text{IBr}_2\) is a slightly warped cylinder with the axis along the least conducting direction \([13, 20, 21]\). The general behavior of the oscillating interlayer magnetoresistance and magnetization is shown in Fig. 1 and fully agrees with the previous reports \([13, 20]\). Cyclotron orbits on the cylindrical FS give rise to rapid SdH (Fig. 1a) and dHvA (Fig. 1b) oscillations with the frequency \(F = 3930\,\text{T}\) that corresponds to the FS cross-sectional area \(S \approx 0.53S_{\text{BZ}}\) (\(S_{\text{BZ}}\) is the Brillouin zone area). The amplitudes of both the SdH and dHvA oscillations are clearly modulated due to a slight warping of the FS. A comparison of the beat and fundamental frequencies yields the evaluation of the warping: \(\Delta S/S = 2F_{\text{beat}}/F \approx 10^{-2}\).

In addition to the rapid SdH oscillations, the magnetoresistance exhibits prominent slow oscillations with the frequency \(F_{\text{slow}} \approx 42\,\text{T}\). Due to a large cyclotron mass, \(m \approx 4.2m_e\) (\(m_e\) is the free electron mass), the fundamental oscillations rapidly diminish with increasing temperature and can barely be resolved at the highest field at \(T = 1.4\,\text{K}\). By contrast, the amplitude of the slow oscillations remains almost the same as at 0.6 K. Noteworthy, no trace of slow oscillations has been found in our magnetization measurements.

A clue for understanding the origin of the slow oscillations is the dependence of their frequency on the tilt angle \(\theta\) displayed in Fig. 2a \([22]\). Unlike the rapid oscillations having the \(1/\cos\theta\)-dependence typical of Q2D metals, the slow oscillations show a strong non-monotonic change of their frequency with \(\theta\). Such a behavior immediately reminds that of the beat frequency \(F_{\text{beat}}(\theta)\). At certain magnetic field orientations, repeated periodically with \(\tan\theta\), the areas of cyclotron orbits in a Q2D metal become independent of the orbit positions in \(k\)-space \([23]\). This obviously results in periodic drops of \(F_{\text{beat}}\) to zero \([20]\). At the same angles the semiclassical part of the magnetoresistance turns out to sharply increase.
Fig. 2b shows the angular dependence of the background magnetoresistance of our sample at $B = 14$ T, revealing two prominent AMRO peaks at $\theta \approx 33^\circ$ and $-20^\circ$. It clearly correlates with the angular dependence of $F_{\text{slow}}$: the latter rapidly decreases as the magnetoresistance approaches the AMRO peaks.

It is of course very tempting to directly compare $F_{\text{slow}}$ and $F_{\text{beat}}$. Unfortunately, the rapid oscillations were generally observed in a relatively small field interval in our experiment, so that the beat period could not be reliably measured in a sufficiently wide angular range. Nevertheless, estimations made at a few angles (see e.g. the data in Fig. 1) reveal the relationship $F_{\text{slow}}(\theta) \approx 2F_{\text{beat}}(\theta)$ within the experimental error bar. Thus, one can conclude that the slow oscillations and beats of the rapid oscillations have the same physical origin, i.e. both are directly related to the warping of the cylindrical FS.

In order to clarify the mechanism responsible for the slow oscillations, we first note that the interlayer conductivity contains several factors which, in general, oscillate in magnetic field with the frequency $F$ determined by the cross-sectional area of the FS cylinder. The amplitudes of the oscillations are modulated, due to the warping of the cylinder, with the frequency $F_{\text{beat}} = (2t_\perp/\epsilon_F)F \ll F$ ($t_\perp$ is the interlayer transfer integral and $\epsilon_F$ the Fermi energy). The product of two oscillating quantities with modulated amplitudes $\tilde{\alpha}$ and $\tilde{\beta}$ yields a slowly oscillating term, e.g. $(1 + \tilde{\alpha} \cos x)(1 + \tilde{\beta} \cos x) = 1 + (\tilde{\alpha} + \tilde{\beta}) \cos x + (\tilde{\alpha}\tilde{\beta}/2) \cos 2x + \tilde{\alpha}\tilde{\beta}/2$. Here, the last term describes slow oscillations.

To be more explicit, we consider the interlayer conductivity as determined from the Boltzmann transport equation [24]:

$$\sigma_{zz} = e^2 \int d\epsilon (-n'_F(\epsilon)) I(\epsilon) \tau(\epsilon),$$

(1)

where $n'_F(\epsilon) = -1/\{4T\cosh^2[(\epsilon - \mu(B))/2T]\}$ is the derivative of the Fermi distribution function, $I(\epsilon) \equiv \sum |v_z(\epsilon)|^2$ the square of the electron interlayer velocity $v_z$ summed over all states at the energy $\epsilon$, and $\tau(\epsilon)$ is the momentum relaxation time at the energy $\epsilon$. In Born approximation, $\tau(\epsilon)$ is inversely proportional to the density of states (DoS): $\tau(\epsilon) \propto \rho^{-1}(\epsilon)$ and oscillates in a magnetic field [24]:

$$\tau(\epsilon) \propto \left[1 + 2 \sum_{p=1}^{\infty} (-1)^p \cos \left(\frac{2\pi p \epsilon}{\hbar \omega_c} \right) J_0 \left(\frac{4\pi p t_\perp}{\hbar \omega_c} \right) R_D \right]^{-1},$$

(2)

where $\omega_c = eB/m$ is the cyclotron frequency, and $R_D = \exp(-2\pi^2 pk_BT_D/\hbar \omega_c)$ is the usual
scattering damping factor \( I(\epsilon) \) (\( T_D \) is the Dingle temperature). If the cyclotron energy is comparable to the interlayer transfer integral, the oscillations of the quantity \( I(\epsilon) \) become also important. They are given by

\[
I(\epsilon) \propto 1 + \frac{\hbar \omega_c}{\pi t_\perp} \sum_{p=1}^{\infty} \frac{(-1)^p}{p} \cos \left( \frac{2\pi p \epsilon}{\hbar \omega_c} \right) J_1 \left( \frac{4\pi p t_\perp}{\hbar \omega_c} \right) R_D.
\]

(3)

The beats of the oscillations of \( \tau \) and \( I \) are given by the 0-th and 1-st order Bessel functions \( J_0 \) and \( J_1 \) in Eqs. (2) and (3), respectively. Their product eventually gives rise to a slowly oscillating term in the conductivity. Since the FS warping is large enough in our case, \( 4\pi t_\perp > \hbar \omega_c \), one can approximate the Bessel functions as

\[
J_0(z) = \frac{\sqrt{2}}{\pi z} \cos(z - \pi/4) \quad \text{and} \quad J_1(z) = \frac{\sqrt{2}}{\pi z} \sin(z - \pi/4).
\]

Further, taking into account the weak amplitude of the SdH effect (see Fig. 1) and strong harmonic damping (the second harmonic never exceeded 1% of the fundamental one in our experiment), we neglect oscillations of the chemical potential \( \mu \) and restrict our consideration to the lowest order in the damping factors. Then, substituting Eqs. (2) and (3) into Eq. (1) and performing the integration over energy at finite temperature, we obtain:

\[
\sigma_{zz} = \sigma_0 \left\{ 1 + 2 \sqrt{\frac{\hbar \omega_c}{2\pi^2 t_\perp}} \left[ 1 + a^2 \right] \times \right.
\]

\[
\times \cos \left( \frac{2\pi \mu}{\hbar \omega_c} \right) \cos \left( \frac{4\pi t_\perp}{\hbar \omega_c} - \frac{\pi}{4} + \phi \right) R_D R_T +
\]

\[
+ \frac{\hbar \omega_c}{2\pi^2 t_\perp} \left[ 1 + \sqrt{1 + a^2} \cos \left( 2 \left[ \frac{4\pi t_\perp}{\hbar \omega_c} - \frac{\pi}{4} + \frac{\phi}{2} \right] \right) \right] R_D^2 \right\}
\]

where \( \phi = \arctan(a) \) and \( a = \hbar \omega_c/2\pi t_\perp \). \( R_T = (2\pi^2 k_B T/\hbar \omega_c)/\sinh(2\pi^2 k_B T/\hbar \omega_c) \) is the temperature damping factor which is equal to that in the LK theory and comes from the integration with the Fermi distribution function. The scattering factor \( R_D^* \) has the same form as \( R_D \) but includes a different Dingle temperature \( T_D^* \) instead of \( T_D \) as will be discussed below. The coefficient \( \sigma_0 \) in Eq. (4) can be estimated as \( \sigma_0 = e^2 N_{LL} 2t_\perp^2 d^2 / \pi \hbar^2 \omega_c k_B T_D \) where \( N_{LL} \) is the Landau level degeneracy, \( N_{LL}/\hbar \omega_c = m^*/2\pi \hbar^2 \) is the DoS at the Fermi level.

The second term in the curly brackets of Eq. (4) describes the fundamental SdH oscillations modulated with the frequency \( F_{\text{beat}} = 2t_\perp m/e \hbar \). It is the last term in Eq. (4) that gives the slow oscillations of \( \sigma_{zz} \) (hence of \( R_\perp \propto 1/\sigma_{zz} \)) with the frequency equal to the double beat frequency, in agreement with the experiment. The oscillations of \( F_{\text{beat}} \) as a function of the angle \( \theta \), in accordance with the AMRO effect [20, 23] lead to identical oscillations of \( F_{\text{slow}} \) that explains the angular dependence plotted in Fig. 2. It is interesting that, unlike
many anomalies which were studied earlier and associated with a strong enhancement of the oscillation amplitude and harmonic contents, the present phenomenon exists even in the limit of a weak amplitude and constant chemical potential, i.e. when no substantial deviations from the standard LK description were expected.

From Eq. (4) it becomes clear why the slow oscillations are virtually independent of temperature (Fig. 1). Indeed, they are determined by the interlayer transfer integral (i.e. warping of the FS) but not by the electron energy. Therefore, they are not affected by the temperature smearing of the Fermi distribution function and do not contain the factor \( R_T \). This is why the slow oscillations, though originating from the cyclotron motion on the same large orbits on the FS cylinder as the fundamental SdH oscillations, persist up to relatively high temperatures at which the latter are completely suppressed.

Another notable feature of the slow oscillations is that their Dingle temperature \( T^*_D \) is different from \( T_D \) entering the Dingle factor of the usual SdH oscillations. The usual Dingle temperature includes all mechanisms of the smearing of DoS oscillations. These are not only microscopical scattering events but also macroscopic spatial inhomogeneities of the sample. These inhomogeneities lead to macroscopic spatial variations of the electron energy in Eq. (4). Their effect is equivalent to the local shift of the chemical potential \( \mu \). The total signal is an average over the entire sample and such macroscopic inhomogeneities lead to the damping of the magnetic quantum oscillations similar to that caused by temperature. Since the slow oscillations do not depend on \( \mu \), they are not affected by this type of smearing and the corresponding Dingle temperature \( T^*_D \) is determined by only short-range scatterers. One can therefore estimate relative contributions from macroscopic inhomogeneities and from local defects to the scattering rate by comparing \( T_D \) and \( T^*_D \). In Fig. 3 we plot the normalized amplitudes of the fundamental and slow oscillations as functions of inverse magnetic field. For the fundamental oscillations an angle \( \theta = 19.3^\circ \) has been chosen. This angle corresponds to the AMRO peak, thus complications in the determination of the amplitude due to the beats could be avoided. Fits of the data according to Eq. (4) (solid lines in Fig. 3) yield the Dingle temperatures \( T_D = (0.8 \pm 0.02) \) K and \( T^*_D = (0.15 \pm 0.02) \) K. It is unlikely that the big difference between these values is only caused by the slightly different orientations. Therefore, we conclude that inhomogeneities play an important role in damping the SdH oscillations in the present sample.

Although the scattering is dominated by crystal imperfections at low temperature,
electron-electron and electron-phonon interactions must also be taken into account in both $R_D$ and $R_D^*$. This leads to a finite temperature dependence of the amplitude of the slow oscillations. A correct evaluation of this effect based on exact calculations of the imaginary part of the electron self-energy would allow to obtain additional information about the many-body interactions from the slow oscillations.

Finally, we note that the interference between the oscillating relaxation time and inter-layer Fermi velocity considered above is not the only possible source of the slow oscillations. There are other oscillating quantities which do not enter Eq. (1) directly but are contained in the relaxation time $\tau$ and can, in principle, also lead to a similar effect. Taking into account these additional factors will lead to a change in the magnitude and phase of the slow oscillations, leaving, however, their dependence on temperature and magnetic field essentially the same.

In conclusion, we have shown that the slow oscillations in the Q2D metal $\beta$-(BEDT-TTF)$_2$IBr$_2$ originate from the warping of its cylindrical FS. We propose a model explaining this phenomenon as a general feature of clean Q2D metals developing when the cyclotron energy becomes comparable with the interlayer transfer energy. In particular, slow oscillations observed in some other layered organic metals [14, 15, 16, 17] may have the same origin. Since the slow oscillations are not affected by the temperature smearing of the Fermi distribution function, one can use them for estimating the warping of the FS even at relatively high temperatures at which the fundamental SdH oscillations are suppressed. On the other hand, the temperature and field dependencies of the slow oscillations can give a valuable information on scattering processes.

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to Fig. 1a does not exactly fit to the curve in Fig. 2a.

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Figure captions

Fig. 1. Interlayer resistance (a) and oscillating part of magnetic torque (b) of $\beta$-(BEDT-TTF)$_2$IBr$_2$ versus magnetic field at $\theta \approx -15^\circ$. The curves at different temperatures are offset for clarity.

Fig. 2. Angular dependencies of the frequency of the slow oscillations (a) and background resistance at $B = 14$ T, $T = 0.44$ K (b). Lines are guides for the eye.

Fig. 3. Amplitudes of the fast (a) and slow (b) oscillations normalized to the background resistance plotted versus inverse magnetic field at $T = 0.44$ K. The lines are fits to Eq. (4).
Fig. 1 of "Slow oscillations..." by M. Kartsovnik et al.
Fig. 2 of "Slow oscillations..." by M. Kartsovnik et al.
Fig. 3 of "Slow oscillations..." by M. Kartsovnik et al.