Incoherence, metal-to-insulator transition, and magnetic quantum oscillations of interlayer resistance in an organic conductor

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An analytic theory is developed for the conductivity across the layers σ_{zz} in a layered conductor in perpendicular magnetic field under the conditions of interlayer incoherence. The latter assumes a small hopping integral between the layers $t \ll \hbar/\tau$ and the presence of localized states in the tails of broadened Landau levels (LLs) (τ is the electron scattering time within the layers). In the incoherent regime, σ_{zz} strongly depends on the in-plane conductivity mechanisms because electrons spend most of their time within the weakly coupled layers. At high fields $\Omega\tau \gg 1$, an integer quantum Hall effect (IQHE) within the layers develops which changes dramatically magnetic quantum oscillations in the σ_{zz} compared to the standard Lifshitz-Kosevich theory (Ω is the cyclotron frequency). At low fields, σ_{zz} displays Shubnikov–de Haas (SdH) oscillations which in the limit $\Omega\tau \gg 1$ transforms into sharp peaks. The peaks reach their maximum values $\sigma_{zz} \propto \frac{\hbar\Omega}{T}$ when LLs cross the chemical potential μ. When μ falls into the tails between the LLs, the σ_{zz} displays first a thermal activation behavior $\sigma_{zz} \propto \exp[-(\hbar\Omega - \mu)/T]$ and, then at lower temperatures T, crosses over into a variable-range-hopping regime with $\sigma_{zz} \propto \exp(-\sqrt{T_0/T})$, where $T_0 \propto |B-B_0|^{\gamma}$. Above B_0, the in-plane electrons are in the quantum-Hall-insulator regime and the background interlayer magnetoresistance R_b has an insulatorlike temperature dependence. Below B_0, the in-plane electrons are in the conventional SdH oscillation regime and R_b has a metal-like temperature dependence. On the insulating side, R_b displays a universal dependence on the scaling variable $(B-B_0)/T^{\kappa}$. Scaling is destroyed in tilted magnetic fields at angles corresponding to the spin zeros. All the above features in the σ_{zz} have been observed in the β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$, in which the critical exponent is equal to $\kappa = 1/\gamma = 0.65$. The IQHE regime at high fields in this quasi-two-dimensional organic conductor is favored by the fixed value of the chemical potential. It is shown that at low temperatures ($T \ll \hbar/\tau$), oscillations of the conductivity and magnetization are related by the condition $\sigma_{zz} \propto B^2 \partial\widetilde{M}/\partial B$, in agreement with observations in β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$. The analysis shows that the above features in the conductivity cannot be explained within the model with a narrow-band dispersive electron transport across the layers because the model is incompatible with the incoherence condition $t \ll \hbar/\tau$. Moreover, in the self-consistent Born approximation, this model yields a nonphysical negative conductivity $\sigma_{zz} < 0$.

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I. INTRODUCTION

Magnetic quantum oscillations in layered organic conductors [ET salts, i.e., the ones which contain a molecule BEDT-TTF (bisethylenedithio-tetrathiafulvalene] often strongly deviate from the Lifshitz-Kosevich (LK) theory developed for conventional metals with arbitrary shape of the Fermi surface (FS).[1–5] The most striking example is the highly anisotropic layered organic superconductor β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ (β'' salt),[1,2] which displays an incoherent (nondispersive) electron transport across the layers and presumably has a FS corresponding to a two-dimensional (2D) rather than a three-dimensional (3D) conductor because the momentum across the layers is not preserved. Experimentally, the interlayer incoherence in the β'' salt has numerous manifestations in anomalous magnetic quantum oscillations.[1,2,6] Some of these anomalies will be discussed later.

The prime evidence of the incoherence is the absence of beats in magnetic quantum oscillations as well as the fact that a resistive peak in parallel to the layer's magnetic field never has been observed in the β'' salt.[1–4] The absence of beats means a very small value of the hopping integral between the layers t. In organic salt β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$, an estimate yields $t < 10^{-6}$ eV,[6] which is caused by large anion molecules SF$_5$CH$_2$CF$_2$SO$_3$ between the layers. The dispersion across the layers $\varepsilon(p_z)$ and related 3D FS have no meaning in that case for a very simple reason: the warping of the cylinder FS caused by the electron interlayer hopping, $\sim t$, is much less than its smearing by the disorder potential, $\sim \hbar/\tau$. In other terms, the inequality $\hbar/t \gg \tau$ means that interlayer hopping time is large and electrons scatter many times within the layer before hopping to a neighboring layer, so that there is no coherent wave function perpendicular to the layers.

Under such conditions, the conductivity across the layers σ_{zz} is carried out through uncorrelated electron tunneling between impurity-broadened 2D Landau levels belonging to the neighboring layers.[7] The same concept of interlayer incoherence is well established in anisotropic high-T_c cuprates.[8–13]

Although both Subnikov–de Haas (SdH) and de Haas–van Alphen (dHvA) oscillations are strongly influenced by the interlayer incoherence, to treat this effect in thermodynamic dHvA oscillations is much easier. A theory of dHvA oscillations in intercalated layered conductor with disorder in layer stacking was developed in Ref. 14. The disorder across the layers means nothing but incoherence since p_z, the momentum component perpendicular to the layers, is not preserved and the related dispersion $\varepsilon(p_z)$, as well as the corresponding warped cylinder FS, has no meaning in that case. It was established in Ref. 14 that local defects in layer stacking result in additional frequencies in the spectrum of dHvA oscillations. As will be shown in this paper, the influence of the interlayer incoherence on magnetic oscillations of the conductivity σ_{zz} is more complex.

In less anisotropic (than the β'' salt) organic conductors of the ET family, electrons have a dispersion across the layers. In that case, the accepted standard is to take the electron interlayer kinetic energy in the form $\varepsilon(p_z)=t\cos(p_z a/\hbar)$. If $t \gg \hbar/\tau$, the corresponding 3D FS is a warped cylinder producing beats in the SdH and dHvA oscillations. The beats in the dHvA oscillations in layered conductors have been studied in Ref. 14. In the SdH conductivity across the layers, these beats are known as the "slow oscillations." They are well established experimentally and explained theoretically in Ref. 15 within the standard cosine-dispersion model for the interlayer electron hopping. It was also demonstrated in this paper that under the conditions $t \gg \hbar\Omega \gg \hbar/\tau$, the magnetic oscillations of conductivity in layered conductors such as ET salts cannot be described correctly without taking into account a quantum correction to the quasiclassical Boltzmann conductivity σ_{zz}.

A formal application of the standard model with the cosine interlayer dispersion $\varepsilon(p_z)=t\cos(p_z a/\hbar)$ to calculations of the SdH oscillations in the highly anisotropic β'' salt was done in Refs. 16 and 17. In this connection, a remark is in order. It is clear that any approach to the problem in question based on a dispersive interlayer electron transport cannot explain neither metal-to-insulator transition nor scaling in the background magnetoresistance observed in Ref. 1. As will be shown later, the model and approximations used in Refs. 16 and 17 are in principal conflict with the concept of the interlayer incoherence both mathematically and physically. Moreover, they result in a negative nonoscillating conductivity across the layers $\sigma_{zz}<0$.

On the other hand, the incoherence condition $\hbar/t \gg \tau$, as was proved in Ref. 7, does not change the angular-dependent magnetoresistance oscillations (AMROs), which is not a surprise since AMRO hold even in bilayers in tilted magnetic fields.[18] To the contrary, the SdH and dHvA oscillations in the incoherent layered conductor deviate dramatically from the standard LK picture and display very unusual behavior below the upper critical magnetic field B_{c2}. The experiments of Ref. 19 show that both SdH and dHvA amplitudes are enhanced in the β'' salt in the superconducting state in contrast to all previous observations and all theories so far, which predict a decrease of magnetic oscillations in the vortex state of a 2D superconductor due to several specific mechanisms of damping.[20] This anomalous enhancement of the dHvA and SdH oscillations in the β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ salt below the B_{c2} is most likely related to the restoration of the interlayer coherence in the superconducting state.[21] The latter enhances the layer-stacking factors, modulating the amplitudes of magnetic oscillations. These factors are absent in purely 2D models of magnetic quantum oscillations in the superconducting state.[20]

The spin factor in the LK theory oscillates with the angle between the magnetic field and the conducting plane so that the amplitudes of harmonics became zero at some tilt angles. This spin-splitting zero effect is anomalous in the β'' salt as well.[6] The spin zeros are shifted compared to the LK theory in the dHvA oscillations, but in the SdH measurements, they exactly follow the LK predictions.[22] A theory of this unusual phenomenon based on a combined effect of the small chemical potential oscillations and interlayer incoherence will be considered elsewhere.

Here, we will concentrate on another puzzling manifestation of the interlayer incoherence in the β'' salt. It was found in Refs. 1 and 6 that the SdH oscillations in this layered conductor in perpendicular magnetic field B display a metal-to-insulator transition at the critical field $B_0=3.5$ T. Below B_0, the background magnetoresistance R_b has a metal-like temperature dependence, i.e., it decreases with the decrease of temperature. Above B_0, the temperature dependence of the R_b is insulating. Moreover, a normalized background magnetoresistance R_b displays a universal monotonously increasing dependence on the scaling variable $(B-B_0)/T^{\kappa}$, with the critical exponent $\kappa=0.65$.

The purposes of this paper are (i) to develop a theory of the SdH oscillations in the conductivity across the layers in a regime of incoherent electron hopping between the conducting layers, and (ii) to explain the metal-to-insulator transition induced by a perpendicular magnetic field in the β'' salt and the scaling behavior of the background magnetoresistance in this compound based on the assumption that the integer quantum Hall effect (IQHE) holds in this compound at high magnetic fields.

The paper is organized as follows. In Sec. II, basic equations are derived and the analogy with the IQHE is discussed. The SdH oscillations in the interlayer conductivity is considered in Sec. III. Metal-to-insulator transition and scaling in the background magnetoresistance as well as the role of the variable-range-hopping mechanism in the total conductivity are explained in Sec. IV. Results and conclusions of the paper are summarized in Sec. V. In Appendix A, the effective quantum hopping amplitude across the layers is considered. The incompatibility of the interlayer incoherence with the models based on the dispersion across the layers and the self-consistent Born approximation (SCBA) is discussed in Appendixes B and C.

II. BASIC EQUATIONS AND ANALOGY WITH THE INTEGER QUANTUM HALL EFFECT

A theory of the SdH oscillations for the case of incoherent electron transport across the layers was developed in Ref. 23. Equations for the conductivity obtained in this paper are written in terms of the density of states (DOS) $g(\varepsilon)$ and velocity $v_z^2(\varepsilon)$ of electrons within the broadened Landau levels (LLs). They can be applied both to the dispersive and nondispersive electron transport, including the case when some fractions of states within the broadened LLs are localized as in the IQHE.[24] The latter means that $v_z^2(\varepsilon)=0$ for energies belonging to the localized states at the tails of Landau levels, and $v_z^2(\varepsilon)\neq 0$ for the stripes of delocalized states at the center of LLs.

The SdH conductivity σ_{zz}, obtained in Ref. 23, is a sum of the Boltzmann (σ_B) and quantum (σ_Q) terms: $\sigma_{zz}=\sigma_B+\sigma_Q$, where

$$\sigma_B=\sigma_0\int d\varepsilon\frac{dE}{\pi}g(\varepsilon)v_z^2(\varepsilon)\left(-\frac{\partial f}{\partial E}\right)\tau S[\lambda,\delta(E,\varepsilon)], \quad (1)$$

$$\sigma_Q=\sigma_0\int d\varepsilon\frac{dE}{\pi}g(\varepsilon)v_z^2(\varepsilon)\left(\frac{\partial f}{\partial E}\right)\frac{2\pi}{\Omega}\frac{\partial}{\partial\lambda}S[\lambda,\delta(E,\varepsilon)]. \quad (2)$$

Here, $\lambda(E)=2\pi/\Omega\tau$, $\delta(E,\varepsilon)=2\pi(E+\varepsilon)/\hbar\Omega$, $\sigma_0=e^2N_L/\hbar\Omega$, $\Omega=eB/mc$, B is the magnetic field, m is the electron mass, c is the speed of light, $N_L=\Phi/S\Phi_0$ is the electron density at the LL, Φ is the flux through a sample, $\Phi_0=2\pi\hbar c/e$ is the flux quantum, $f=f(E-\mu)$ is the Fermi function, μ stands for the chemical potential (CP), and

$$S(\lambda,\delta)=\sum_{p=-\infty}^{\infty}(-1)^p e^{-|p|\lambda}\cos p\delta=\frac{\sinh\lambda}{\cosh\lambda+\cos\delta}. \quad (3)$$

The Landau levels in the problem in question are broadened by the intralayer disorder and by the interlayer incoherence. The variable ε describes the LLs' broadening. The shape of this broadening is given by the DOS $g(\varepsilon)$:

$$E_n(\varepsilon)=\hbar\Omega(n+1/2)+\varepsilon. \quad (4)$$

The electron velocity in the direction perpendicular to the layers, v_z, is related to the tunneling matrix elements between the layers by

$$v_z(\varepsilon)=\frac{|t_{\varepsilon,\varepsilon}|a}{\hbar\sqrt{2}}, \quad (5)$$

where a and $\hbar\sqrt{2}/|t_{\varepsilon,\varepsilon}|$ are correspondingly the distance between the layers and the time of tunneling.

Experiment[2] shows a nonmetallic temperature behavior of the resistance across the layers $\rho_{zz}(T)=1/\sigma_{zz}(T)$ in the β'' salt even at $B=0$, which displays a broad maximum at 35 K. This insulating resistance testifies in favor of the hopping mechanism of electronic transport across the layers in this organic compound.

Although little is known about the mechanism of incoherence, a strong point of the above equations is that we can learn much about the $\sigma_{zz}(B,T)$ from Eqs. (1) and (2) under rather general assumptions on this mechanism and yet without resorting to any specific shape for the DOS $g(\varepsilon)$. In this connection, one can argue first that the interlayer electron hopping in the incoherent regime is a random one-dimensional (1D) walk yielding nonzero conductivity in the direction perpendicular to the layers.

As is well known, the 1D walk is a special case, in which for any type of disorder the DOS $g(\varepsilon)$ has mobility edges separating a stripe of delocalized states from the energy tails at which velocity $v_z(\varepsilon)=0$. In principle, all states can be localized for strong enough disorder, but this is not the case for the β'' salt since the conductivity $\sigma_{zz}(B,T)\neq 0$ in experiments. Thus, there is at least a narrow stripe of delocalized states in this ET salt, where the $v_z(\varepsilon)\neq 0$, which provides the nonzero conductivity across the layers. We can assume then in a most general way that as in the IQHE the delocalized states are centered at $\varepsilon=0$, where $g(\varepsilon)$ has a narrow peak (of the order of t) and wide tails spreading between the LLs.[25–27]

The analogy with the integer quantum Hall effect is relevant and important for the problem in question because under the condition of incoherence, $\hbar/t\gg\tau$, electrons moving across the layers spend most of their time within the 2D layers. By scattering many times within the layers, electrons then hop to the neighboring layer, providing thereby a conductivity across the layers which bears a strong impact of the 2D conductivity mechanism within the layers. On the other hand, under the condition $\Omega\tau\gg 1$, for which Eqs. (1) and (2) have been obtained in Ref. 23, the IQHE regime develops within the weakly coupled conducting layers. This regime in the β'' salt is favored by the fixed value of the chemical potential at the Fermi level $\mu\approx E_F$ and relatively small Landau level occupation index [see also Sec. IV C]. Small oscillations of the chemical potential with amplitudes much less than the separation between LLs can be neglected in this problem. The Landau levels in the IQHE regime are broadened by disorder and have a narrow stripe of delocalized states in their center.[24–26] The localization is a crucial ingredient of the IQHE. The idea that the IQHE within the layers can hold in some organic ET salts has been discussed in the literature,[3] although a direct experimental proof of this is absent so far.

Thus, assuming the presence of a narrow stripe of delocalized states in the DOS $g(\varepsilon)$, we can simplify Eqs. (1) and (2), and rewrite them as follows:

$$\sigma_{zz}=\int\frac{dE}{\pi}\left(-\frac{\partial f}{\partial E}\right)\sigma_\tau G_{zz}(\lambda,E), \quad (6)$$

$$G_{zz}(\lambda,E)=G_B(\lambda,E)+G_Q(\lambda,E), \quad (7)$$

$$G_B(\lambda,E)=S[\lambda,\Delta(E)],\quad G_Q(\lambda,E)=-\lambda\frac{\partial}{\partial\lambda}S[\lambda,\Delta(E)], \quad (8)$$

where $\Delta(E)=2\pi E/\hbar\Omega$ and

$$\sigma_\tau=\frac{e^2N_L\tau\langle v_z^2\rangle}{\hbar\Omega}. \quad (9)$$

The average of the velocity squared is given by

$$\langle v_z^2\rangle=\frac{a^2}{2\hbar^2}\int_{\varepsilon_{min}}^{\varepsilon_{max}}d\varepsilon g(\varepsilon)|t_{\varepsilon,\varepsilon}|^2. \quad (10)$$

The integral in Eq. (10) is taken within the narrow stripe of delocalized states at the center of the LLs. The tails in the DOS $g(\varepsilon)$ can be wide, but the width of the delocalized states stripe is small, so that $(\varepsilon_{max}-\varepsilon_{min})<t\ll\hbar/\tau$. This important point simplifies equations for the conductivity and yet stresses a similarity of the physics behind the σ_{zz} to that in the 2D IQHE systems (see Secs. IV and V for more details).

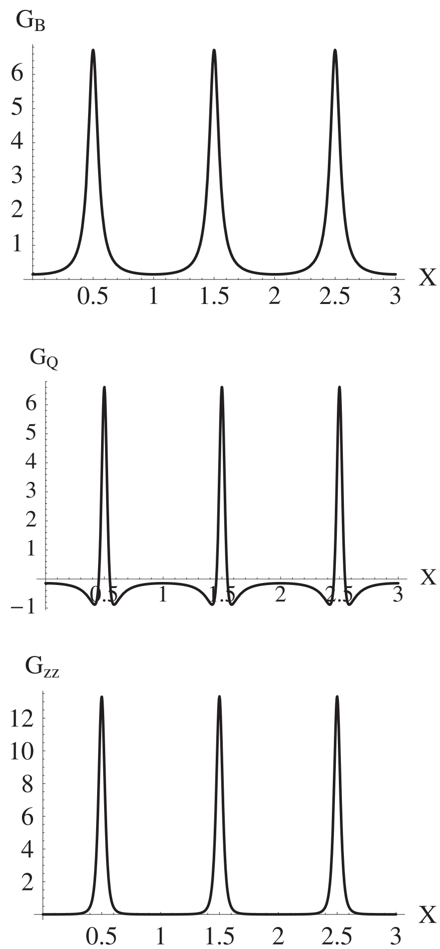

FIG. 1. (a) The Boltzmann, $G_B=S(\lambda,2\pi X)$ and (b) the quantum, $G_Q=-\lambda\frac{\partial}{\partial\lambda}S(\lambda,2\pi X)$ contributions into the conductivity σ_{zz} in Eq. (6), and (c) their sum $G_{zz}=G_B+G_Q$. $X=E/\hbar\Omega$, $\lambda=0.3$.

III. SHUBNIKOV–de HAAS OSCILLATIONS OF THE CONDUCTIVITY

The scattering time $\tau(E)$, in general, is a model-dependent function of the energy, which is inversely proportional to the scattering probability of electrons by impurities. The latter depends on the DOS of the system, which oscillates in external magnetic field and makes, in general, $\tau(E)$ an oscillating function of the inverse magnetic field, too. However, in our model, these oscillations are small because conducting (delocalized) electrons belong to a narrow stripe in the DOS, while the rest of the electrons are localized and produce a reservoir of states, stabilizing oscillations in $\tau(E)$ which can be approximately set $\tau=\text{const}$ (see Appendix A for more details).

Under the conditions $\tau=\text{const}$ and $\lambda\ll 1$, the functions $G_B(\lambda,E)$ and $G_Q(\lambda,E)$ are sharply peaked at $E=E_n$, and between the LLs they nearly compensate each other, as shown in Fig. 1. Correspondingly, the conductivity σ_{zz} tends to zero between the peaks like in the IQHE. The amplitude of the peaks in Fig. 1(c) is twice larger than in Figs. 1(a) and 1(b). Therefore, one can approximate the function $G_{zz}(\lambda,E)=G_B(\lambda,E)+G_Q(\lambda,E)$ by a periodic set of δ-like Lorentzians [see Fig. 1(c)]

$$G_{zz}(\lambda,E)\approx\frac{2}{\pi}\sum_{n=-\infty}^{\infty}\frac{\eta}{(n+1/2-E/\hbar\Omega)^2+\eta^2},\qquad(11)$$

where $\eta=\lambda/2\pi$. The sum in Eq. (11) can be written in a simple analytic form with the help of the identity

$$\frac{1}{\pi}\sum_{p=-\infty}^{\infty}\frac{\eta}{(n+a)^2+\eta^2}=\frac{\sinh 2\pi\eta}{\cosh 2\pi\eta-\cos 2\pi a}.\qquad(12)$$

The width of peaks in Fig. 1 in the energy scale is of the order of $\hbar/\tau$. If $T\gg\hbar/\tau$, the peaked function $-(\partial f/\partial E)$ is broader than peaks in the function $G_{zz}(\lambda,E)$. Correspondingly, Eq. (6) for $T\gg\hbar/\tau$ yields

$$\sigma_{zz}(B)\approx\sigma_\tau\frac{\hbar\Omega}{2\pi T}\sum_n\cosh^{-2}\left(\frac{E_n-\mu}{2T}\right).\qquad(13)$$

The oscillations of the conductivity as a function of magnetic field are completely determined by Eq. (13) provided that the shape of the chemical potential $\mu(B)$ is known. Experiments tell that the chemical potential in the β'' salt vary very little with magnetic field and is approximately equal to the Fermi energy E_F.[1] Although reasons behind such behavior of the CP are not completely known, it is clear that the function $\mu(B)$ in layered organic ET salts differs from that in a 2D electron gas because of the electronic miniband structure attached to each Landau level. This miniband structure in the occupation below the Fermi energy E_F, as was shown in Ref. 28, strongly reduces the amplitude and changes the shape of the chemical potential oscillations through a specific integral factor depending on the DOS within the minibands. In a 2D electron gas, the LLs are flat and, at zero temperature, $\mu(B)$ jumps between them with the amplitude $\hbar\Omega$.[29–32] In the 3D case, the amplitude of the CP oscillations is dramatically reduced to the value $\hbar\omega_c\sqrt{\hbar\omega_c/E_F}$. Because of that, $\mu\approx E_F$ in the LK theory. In superlattices and layered conductors, the amplitude is between the 2D and 3D values. It depends on the layer-stacking parameters through the DOS of the occupied miniband structure below the E_F (Ref. 28). (More precisely, through the layer-stacking factor introduced in Ref. 14.) Although the miniband structure in the β'' organic salt is unknown, one can assume that the electron population of these states below the E_F is the physical reason that stabilizes the chemical potential at the E_F in this layered conductor.[1]

In view of the above reasoning, and in accordance with the experiment,[1] we can put $\mu\approx E_F$ in Eq. (13) for the conductivity σ_{zz}. This equation then yields σ_{zz} as an oscillating function of the $\hbar\Omega$ (i.e., magnetic field), which is shown in Fig. 2. Under the condition $\hbar\Omega/T\gg 1$, the conductivity σ_{zz} becomes a sharply peaked function which has maxima when one of the LLs exactly coincides with the chemical potential, $E_n=\mu$. The conductivity at maxima is equal to $\sigma_{zz}=\sigma_\tau\frac{\hbar\Omega}{2\pi T}$. At minima, as well as at all other magnetic fields at which the chemical potential μ falls between the LLs, the conductivity σ_{zz} becomes an exponentially small function of temperature and $\hbar\Omega$:

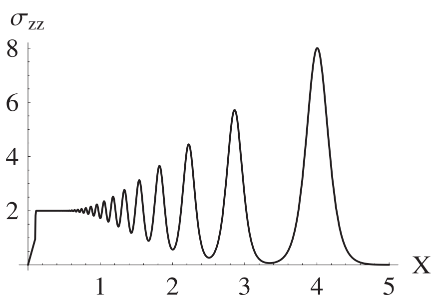

FIG. 2. The conductivity σ_{zz} [see Eq. (13)] in units of σ_τ as a function of $X=\hbar\Omega$. The conventional energy units are accepted, in which $T=0.5$ and $E_F=10$.

$$\sigma_{zz}=\sigma_\tau\frac{\hbar\Omega}{2\pi T}\exp\left(-\frac{\hbar\Omega-\delta\mu}{T}\right), \quad (14)$$

where $\delta\mu$ is a separation between the CP and the center of the nearest partially filled broadened LL. Such thermally activated dependence of the conductivity across the layers $\sigma_{zz}(T)$ was established experimentally in the β'' organic salt.[1,2]

At lower temperatures, $T\ll\hbar/\tau$, the conductivity regime changes. In that case, one can approximate the derivative $(-\partial f/\partial E)$ in Eq. (6) by the delta function $\delta(E-\mu)$, which yields

$$\sigma_{zz}\approx\sigma_\tau(G_B[\lambda,\Delta(\mu)]+[G_Q\lambda,\Delta(\mu)]). \quad (15)$$

As a function of magnetic field, the conductivity σ_{zz} (15) in the low-temperature regime, $T\ll\hbar/\tau$, is given by the sharply peaked function $G_{zz}[\lambda,\Delta(\mu)]$ of the variable $\Delta(\mu)=2\pi\mu/\hbar\Omega$ shown in Fig. 1. The Boltzmann and quantum terms in Eq. (15) nearly compensate each other between the peaks, which in the limit $\eta\to 0$ become narrow Lorentzians of Eq. (11). The same type of behavior is also typical for the diagonal conductivity of conventional 2D semiconductor quantum Hall effect (QHE) systems,[25–27] which makes the analogy with the QHE important for further consideration based on the approach developed in Ref. 24.

Using Eqs. (11) and (12), we can rewrite Eq. (15) in the following simple analytic form:

$$\sigma_{zz}\approx 2\sigma_\tau\frac{\sinh(2\pi\nu)}{\cosh(2\pi\nu)+\cos(2\pi\mu/\hbar\Omega)}, \quad (16)$$

which permits establishing a relationship between the SdH and dHvA oscillations in the considered system. Indeed, the magnetization oscillations in a layered quasi-2D conductor with the fixed value of the chemical potential $\mu\approx E_F$ and a weak coupling between the layers can be described at zero temperature by the sum[33]

$$\tilde{M}=M_0\sum_{p=1}^{\infty}\frac{(-1)^p}{p}\exp(-2\pi\nu p)\sin\left(\frac{2\pi\mu p}{\hbar\Omega}\right). \quad (17)$$

Neglecting small corrections of the order of $\hbar/\tau\mu\ll 1$, we can establish a relation between the SdH conductivity of Eq. (16) and the oscillating part of magnetization $\tilde{M}$:

$$\frac{\sigma_{zz}}{\sigma_\tau}\approx AB^2\frac{\partial}{\partial B}\frac{\tilde{M}}{M_0}, \quad (18)$$

where $A=2e\hbar/\pi mc\mu$. Since $AB\simeq\hbar\Omega/\pi\mu\ll 1$, one may conclude that the relative amplitude of the SdH oscillations σ_{zz}/σ_τ is much less than the relative amplitude of the magnetization oscillations $\tilde{M}/M_0$. Equation (18) was established experimentally in the β'' salt.[1] It is worthy to note here that a proportionality of the SdH conductivity oscillations to the $B^2\partial\tilde{M}/\partial B$ in the LK theory is a consequence of a simplifying assumption, fulfilled in many isotropic conventional 3D metals, that both SdH and dHvA oscillations are determined completely by the DOS oscillations. This is not the case in general. In particular, in some anisotropic and quasi-2D conductors, electron velocities are oscillating functions of the magnetic field because of the Landau bandwidth oscillations. The most prominent example is the Weiss oscillations observed in periodically modulated 2D electron gas.[34–36] In our case, the average velocity within the delocalized stripe Eq. (10) is determined by the variable-range-hopping mechanism and does not oscillate. We consider the conductivity due to this mechanism in the next section.

IV. VARIABLE RANGE HOPPING, MAGNETORESISTANCE OSCILLATIONS, AND METAL-TO-INSULATOR TRANSITION

A. Integer quantum Hall effect regime and variable range hopping

The background conductivity σ_τ in Eq. (9) depends on the hopping amplitude $|t_{\varepsilon,\varepsilon}|$. Below we argue that in β'' salt, this amplitude is a function of temperature and magnetic field. The point is that under the condition of incoherence, $t\ll\hbar/\tau$, which holds in the β'' salt, electrons scatter many times within the layers before hopping to the neighboring layer. In other words, by moving from the top layer to the bottom layer, electrons spend a major fraction of time within the layers. The corresponding quantum amplitude related to the electron path across the layers is a product of the hopping amplitudes within and between the layers. We argue below that because of the weak coupling between the layers and the fixed value of the chemical potential, the β'' salt is a good candidate for establishing the IQHE regime within the layers at high fields. If this is the case, then the amplitude $|t_{\varepsilon,\varepsilon}|$ in the β'' salt is proportional to the average hopping amplitude within the layers, which is the Mott exponent.

Although, so far, the quantum Hall effect has not been observed directly in layered organic ET salts, numerous indirect experimental evidences testify that at fields $\hbar\Omega\gg t$, the IQHE regime is achieved within the layers in these conductors (see Ref. 3 for a review). Among the whole family of ET salts, the β'' salt has the smallest hopping integral t and a fixed value of the chemical potential, which make conditions for the IQHE regime within the layers the most favorable. Also, that compound has a much smaller Fermi surface than other ET salts, which is equal only to 1/5 of the value given by the band structure calculations. As a result, the β'' salt has the smallest Landau orbit occupation index (for example, n

=11 at 18 T, while in the other ET salts, it is usually three times larger[37]). The small Landau index in 2D conductors favors the localization effects necessary for the IQHE.

Therefore, taking into account all the above reasoning, we will assume that the IQHE regime holds in the β'' salt under the conditions $\hbar\Omega \gg \hbar/\tau \gg t$. In the IQHE regime, the diagonal conductivity evolves on the background of localization which plays a crucial role in the IQHE physics. The hopping amplitude between localized states within the layers in the IQHE regime is determined by the variable-range-hopping (VRH) mechanism, with the amplitude proportional to the Mott exponent $\exp(-\sqrt{T_0/T})$.[38,39] The latter means that the amplitude $|t_{\varepsilon,\varepsilon}|^2$ at low temperatures $T \ll \hbar/\tau$ is proportional to this factor, too. The factor $\exp(-\sqrt{T_0/T})$ depends not only on temperature but through a characteristic temperature $T_0 = T_0(B)$ on the magnetic field B, too. That point will be discussed later. (More details on the Mott VRH mechanism can be found also in Appendix A.)

Thus, the temperature and field dependencies of the conductivity σ_{zz} at low temperatures are governed by the IQHE physics within the weakly coupled layers. It is believed that in 2D conductors in perpendicular magnetic field, the extended states are at the center of the broadened Landau levels and all the other states are localized.[25–27] The picture of localization, in general, is rather complex. Qualitatively, at high enough fields, the localization within the layers means that Landau orbits drift along the closed equipotential contours of the random impurity potential.[26,27] At places where contours come close, electrons can tunnel from one contour to another, providing thereby a conductivity mechanism through the extended states. In the β'' salt, this process includes as well contours lying in the neighboring layers, which yield nonzero conductivity across the layers.

The energies of the equipotential contours depend on the random potential relief, and have some dispersion in values. Therefore, the hopping between contours includes a thermal activation and tunneling, i.e., it goes through the Mott VRH mechanism. Because of that, the effective interlayer hopping amplitude $|t_{\varepsilon,\varepsilon}(T)|$ can be written as follows: $|t_{\varepsilon,\varepsilon}(T)|^2 = |t^0_{\varepsilon,\varepsilon}|^2 \exp(-\sqrt{T_0/T})$. Equations (9) and (10) then yield a well-known Mott law for the background hopping conductivity

$$\sigma_\tau(T_0/T) = \sigma_\tau(0)\exp(-\sqrt{T_0/T}). \quad (19)$$

Here, $\sigma_\tau(0)$ is given by Eqs. (9) and (10), in which $|t^0_{\varepsilon,\varepsilon}|^2$ substitutes the amplitude $|t_{\varepsilon,\varepsilon}(T)|^2$. The characteristic temperature T_0 in the IQHE conductors is inversely proportional to the correlation length ξ, which has a singular dependence on the magnetic field so that $T_0 \propto 1/\xi \propto |B-B_0|^\gamma$. Such type of dependence of the correlation length on a magnetic field manifests a transition to the quantum-Hall-insulator state.[40–42]

In the GaAs/AlGaAs heterostructures, the critical exponent index $\gamma \approx 2.35$ determines the scaling properties of the conductivity near the plateau-to-plateau or plateau-to-Hall-insulator phase transitions.[38–41] The numerical values of the critical index γ and critical field B_0 in the β'' salt should be taken from the experiment. To this end, we will first derive in the next section the conductivity σ_{zz} as a function of temperature.

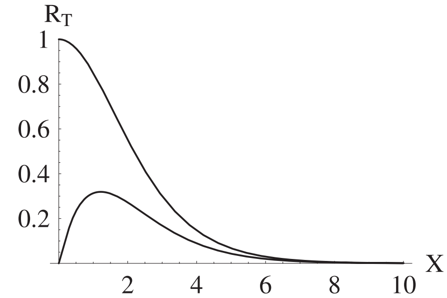

FIG. 3. Temperature damping factor $R_T(1)$ of the fundamental ($p=1$) harmonic as a function of the variable $X=2\pi^2 T/\hbar\Omega$ for the SdH (lower curve) and dHvA (upper curve) oscillations in the case of incoherent electron hopping across the layers [see Eq. (22)]. The upper curve displays a standard LK temperature factor [$R_T(1)$ with $T_0=0$]. The most pronounced difference between the SdH and dHvA factors is at low temperatures $T \ll T_0$, where the VRH conductivity mechanism gives an additional Mott exponent $\exp(-\sqrt{T_0/T})$ in the SdH case. Temperature is measured in conventional units in which $T_0=3$.

B. Temperature behavior of the Shubnikov–de Haas conductivity

Consider the temperature behavior of the conductivity σ_{zz}. Using Eq. (3) for the series expansion of the function $S[\lambda,\Delta(E)]$, Eq. (19), and the integral relation

$$\int_0^\infty dE\left(-\frac{\partial f}{\partial E}\right)\cos\left(\frac{2\pi pE}{\hbar\Omega}\right) \approx R_T(p)\cos\left(\frac{2\pi p\mu}{\hbar\Omega}\right), \quad (20)$$

one can write Eq. (6) for the conductivity σ_{zz} in the following form:

$$\sigma_{zz} = \sigma_\tau(0)\sum_{p=-\infty}^{\infty}(-1)^p R_D(p) R_T(p)\cos\left(\frac{2\pi p\mu}{\hbar\Omega}\right). \quad (21)$$

Here, $R_D(p)=\exp(-2\pi|p|\Omega\tau)$ is the Dingle factor,

$$R_T(p) = \frac{Xp}{\sinh(Xp)}\exp(-\sqrt{T_0/T}) \quad (22)$$

is the temperature factor, and $X=2\pi^2 T/\hbar\Omega$.

The temperature factor $R_T(p)$ (22) differs from the standard LK temperature factor by the Mott VRH exponent. In the LK theory, the temperature dependence of the harmonics is the same for the SdH and dHvA oscillations. This is not the case in the quasi-2D layered conductors with the incoherent transport across the layers. The temperature factors $R_T(p)$ for the dHvA oscillations (upper curve) and SdH oscillations (lower curve) are shown in Fig. 3. Remarkably, the same shape deviation from the LK theory in the $R_T(p)$ (bent down at low temperatures) has been observed in the SdH experiments on the β'' salt (see Fig. 4 in Ref. 37). We will discuss this in more detail in Sec. V.

A similar anomalous behavior of the $R_T(p)$ in SdH experiments with some ET salts has been reported before.[4] Interestingly, the above anomaly was observed only in those ET salts which display an indirect evidence of the IQHE within the layers and show no beats in the magnetoresistance. Direct measurements of the in-plane diagonal and Hall conductivities have not been done so far in ET salts because it is not easy to do experimentally as explained in a review paper.[4]

C. Scaling and the metal-to-insulator transition

The appearance of the Mott VRH exponent in the SdH temperature factor $R_T(p)$ caused by the incoherence has very important consequences. One of these is a scaling of the background conductivity. This scaling means a universal dependence of the background conductivity on the variable $|(B-B_0)|/T^{0.65}$, which has been observed in the SdH experiments on the β'' salt in Ref. 1.

In our theoretical approach, the scaling follows from the fact that the characteristic temperature T_0 is inversely proportional to the correlation length, which has a singularity at the magnetic field B_0. At this field, a transition to the Hall-insulator state holds within the layers,[40–42] and conductivity across the layers is provided by the electron tunneling between equipotential contours belonging to the neighboring layers. Putting then $T_0 \propto 1/\xi \propto |B-B_0|^\gamma$ into the VRH factor, we can write it in the form

$$\exp(-\sqrt{T_0/T}) = \exp[-A(|B-B_0|/T^{0.65})^{0.77}]. \tag{23}$$

Correspondingly, for the hopping conductivity Eq. (19), we have

$$\sigma_\tau(B,T) = \sigma_\tau(0)\exp[-A(|B-B_0|/T^{0.65})^{0.77}]. \tag{24}$$

Here, we take into account the following relations:

$$\sqrt{\frac{T_0}{T}} \propto \sqrt{\frac{|B-B_0|^\gamma}{T}} = \left(\frac{|B-B_0|}{T^{1/\gamma}}\right)^{\gamma/2} \tag{25}$$

and set $\gamma^{-1}=0.65$ in accordance with experiments.[1]

The magnetoresistance $R_{zz}(B,T)=1/\sigma_{zz}(B,T)$ is inversely proportional to the conductivity given by Eqs. (21) and (23). The SdH oscillations of the magnetoresistance $R_{zz}(B,T)$ calculated with the help of Eqs. (21) and (23) at different temperatures are plotted in Fig. 4. This plot displays correctly all specific features observed in the SdH experiments on the β'' salt in Refs. 1 and 2: the upturn of the background magnetoresistance, the insulating type of magnetoresistance (the background magnetoresistance is higher for lower temperatures), and the scaling. The scaling in the background magnetoresistance is illustrated in Fig. 5, in which the VRH exponent $R_b/R(0)=\exp[1.7(X)^{0.77}]$ is plotted as a function of the universal scaling variable $X=|(B-B_0)|/T^{0.65}$. The constant A is taken equal to $A=1.7$ in these conventional units.

The logarithmic scaling plot of the ratio $R_b/R(0)$ in the β'' salt is shown in Fig. 4 of Ref. 1. It approximately consists of two straight lines crossing smoothly at $X=10$. Therefore, at two-thirds of the plot, from $X\approx 10$ to $X\approx 30$, the scaling plot is approximately a VRH exponent of Eq. (24) with the fixed value of the coefficient A. Within the interval $X\approx[0-10]$, the line in the plot is more inclined toward the X axis, which means a smaller coefficient A. Although a nature of this crossover is unclear because the compound in question is too complex, we may relate two crossing straight lines in the logarithmic plot with the two types of the VRH conductivity mechanisms of electrons caused by the intra- and interlayer hoppings. Indeed, the VRH conductivity (19) within the planes is well documented in the semiconductor 2D IQHE systems (see Refs. 38 and 39, and references therein). The electron tunneling across the layers is a 1D VRH process which yields the same exponent Eq. (19) in the conductivity, but with the different coefficient A in Eq. (24). In more details, the VRH is discussed in Appendix A.

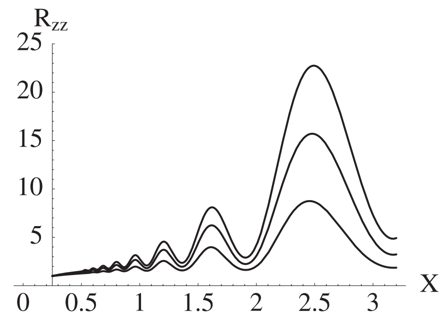

FIG. 4. Resistivity $R_{zz}=1/\sigma_{zz}$ in units of the $\sigma_\tau(0)$ [see Eqs. (13) and (19)] as a function of the variable $X=\hbar\Omega$ at different temperatures (the lower temperature corresponds to the higher curve). In accepted conventional units, these temperatures and other parameters are $T=0.7$, 1, and 2; $T_D=2$; $T_0=0.25$; and $E_F=30$.

The β'' salt is a complex and not completely understood compound. Even at $B=0$, a broad peak in the temperature behavior of the interlayer resistance $R_{zz}(T)$ is not explained so far.[2] Like in high-T_c cuprates, a sharp drop in the $R_{zz}(T)$ to the superconducting value $R_{zz}(T)=0$ at $T=T_c\approx 5$ K follows after a wide semiconducting region where $R_{zz}(T)$ increases with the decrease of temperature. The magnetic field decreases T_c and the upset of a superconducting mixed state at low temperatures below the upper critical field changes the physics of the interlayer transport. It is beyond the scope of the present consideration and has not been taken into account in Fig. 5, which is plotted for $B>B_0$.

Certainly, the VRH and superconductivity compete in the β'' salt at low temperatures, resulting in the saturation of the

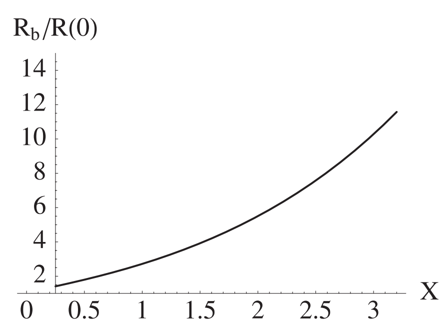

FIG. 5. The VRH exponent $R_b/R(0)=\exp[A(X)^{0.77}]$ plotted as a function of the universal scaling variable $X=|(B-B_0)|/T^{0.65}$. The constant A is taken equal to $A=1.7$ in adopted conventional units.

$R_{zz}(T)$ at T of about a tenth of millikelvin, shown in Fig. 2 and Fig. 4 of Ref. 37. Because of this competition, a deviation from the LK theory in the temperature dependence of the Fourier amplitudes demonstrated in Fig. 4 of Ref. 37 differs at very low temperatures from that in Fig. 3. On the other hand, at higher temperatures, where the superconducting effects are absent, the curves in Fig. 4 of Ref. 37, in agreement with Eq. (24) of our approach, go higher and start to bend down off the standard LK theory lines at higher temperatures in stronger magnetic fields $B > B_0$.

The experimental value of the critical field at which metal-to-insulator transition in the background magnetoresistance holds is $B_0 = 3.5$ T. This value is very close to the upper critical field at zero temperature, which in the β'' salt is $B_{c2} = 3.6$ T. Below the $B_{c2}(T)$, the magnetoresistance $R_{zz}(B,T)$ declines gradually toward zero and oscillations fade away. The magnetoresistance curves go down more rapidly toward zero resistance at lower temperatures since the critical field $B_{c2}(T)$ is higher for lower temperatures.[1,6] This results in the change of the type of magnetoresistance behavior from insulatinglike to metalliclike at fields below the B_{c2}. Moreover, the IQHE regime within the weakly coupled conducting layers also vanishes at low fields, where it crosses over into the conventional SdH oscillation regime as shown in Fig. 2. Correspondingly, at low fields, the temperature factor $R_T(p)$ (22) regains its standard form without the Mott VRH exponent (see the upper curve in Fig. 3) and the temperature behavior of the σ_{zz} (21) became metallic. Experimentally, this transition from the insulator quantum Hall regime to the SdH regime within the conducting layers in the β'' salt looks very much like a phase transition from the insulatinglike to metallike type of the magnetoresistance across the layers.[1] It is worthy to note here that in conventional semiconductor-based Hall systems, a transition from the IQHE regime to the SdH regime in diagonal conductivity with the decrease of magnetic field is a well-known phenomenon.

V. RESULTS AND CONCLUSION

According to the results of this paper, we arrived at the following picture of the quantum magnetic oscillations of the conductivity in the β'' salt. The interlayer incoherence in ET salt β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ is caused by a small hopping integral between the layers $t \ll \hbar/\tau$. It means that the hopping time $\hbar/t \gg \tau$ is large and electrons scatter many times within the layers before hopping to the neighboring layer. The momentum across the layers is not preserved in this case, and there is no conventional Fermi surface for 3D metals. Nonetheless, the quantization of the closed 2D FS (Fermi contour) in perpendicular magnetic field gives rise to the SdH and dHvA oscillations, which deviate from the predictions of the standard LK theory valid for wideband conventional 3D metals with arbitrary FS.

The conductivity σ_{zz} under conditions of the incoherence is due to uncorrelated random electron hopping across the layers. Between the hopping, electrons spend a lot of time within the layers where they experience multiple scattering on random impurity potential. Therefore, the conductivity across the layers, to much extent, is determined by the electron dynamics within the layers. On the other hand, if $\Omega\tau \gg 1$, the conditions for the IQHE regime are satisfied within the layers in view of the small value of the hopping integral and because the chemical potential μ is fixed at the Fermi energy E_F in the β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ organic salt. The physical reason behind this behavior of the chemical potential is unknown completely so far. In 2D electron gas at zero temperature, the CP μ jumps between the Landau levels. This is not the case in layered conductors and in 2D conventional quantum Hall heterojunctions, where the localized states fix the chemical potential. In the β'' salt, the electron population of the miniband structure attached to each Landau level as well as the electrons populating the 1D sheets at the 2D FS and electrons localized by disorder, which do not take part in the quantum oscillations directly, fix the CP μ. In a varying magnetic field, the Landau levels broadened by incoherence and disorder sweep through the fixed chemical potential $\mu = E_F$, giving rise to periodic quantum oscillations of the conductivity σ_{zz} in inverse magnetic field.

The localization play a crucial role in the IQHE because nonzero conductivity in this phenomenon is due to a narrow stripe of the delocalized states at the center of each impurity-broadened Landau level. The mechanism of the conductivity in the IQHE regime is the variable range hopping of electrons within the narrow stripe of the delocalized states,[38,39] which yields the Mott type of the background conductivity $\sigma_{zz} \propto \exp(-\sqrt{T_0/T})$ with $T_0 \propto 1/\xi$.

In conventional 2D semiconducting quantum Hall conductors, the coherence length has a singularity $\xi \propto |B - B_c|^{-\gamma}$ near each plateau-to-plateau transition in the Hall conductivity. In increasing magnetic field, this series of transitions terminates at B_0, where a transition to the Hall-insulator state holds.[41,42] Plateaus in the Hall conductivity never have been observed in the β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$, and only transition to the Hall insulator survives in this layered conductor at B_0. Above B_0, the in-plane electrons are in the quantum-Hall-insulator regime, the coherence length $\xi \propto |B - B_0|^{-\gamma}$, and the background magnetoresistance R_b has an insulatorlike temperature dependence, which is clearly seen in Fig. 4. The background magnetoresistance R_b displays a universal monotonously increasing dependence on the scaling variable $(B - B_0)/T^{0.65}$ as shown in Fig. 5. Such type of behavior was observed experimentally in the β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ (Ref. 1) (for more details, see discussion in Sec. IV C). The Hall-insulator regime means that percolation of 2D Landau orbits throughout the whole planes is impossible, but electron tunneling between equipotential contours from neighboring layers makes the conductivity across the layers, σ_{zz}, nonzero. At low fields, the IQHE regime is destroyed and the conductivity enters the SdH regime as one can see in Fig. 2. In the SdH oscillation regime, the background magnetoresistance $R_b(T)$ has a metal-like temperature dependence. At yet lower fields, the β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ salt becomes a superconductor and R_b goes to zero. The upper critical field at zero temperature in the β'' salt is $B_{c2} = 3.6$ T. Below this field, a wide transition region to the superconducting state with zero resistance $R_b(T) = 0$ is observed.[1,6] The wide width of this transition is caused by the fact that the irreversibility line goes well below the upper critical field $B_{c2}(T)$ in β'' salt.[19]

This physical picture is well supported by the calculations of the conductivity on the basis of Eqs. (1) and (2) within the model, which assumes a narrow stripe of delocalized states at the center of Landau levels and long tails of localized states between them caused by the intra- and interlayer disorders. The adopted model leaves some freedom in the choice of the electron scattering time $\tau(E)$ and DOS $g(\varepsilon)$ describing the Landau level broadening. The principal simplification of equations comes from the fact that a stripe of delocalized states is very narrow (much less than $\hbar/\tau$) so that σ_{zz} can be written in a general form of Eq. (6). The analysis of this equation is summarized in Figs. 1–5. It shows full qualitative agreement of the theory and experiments of Refs. 1 and 2. In particular, the following unusual features are found: the magnetoresistance oscillations hold on the insulating background (Fig. 4), the background magnetoresistance R_b displays a metal-to-insulator transition and the scaling properties (Fig. 5), the magnetoresistance oscillations display a crossover from the SdH to IQHE regime (Fig. 2), and the temperature behavior of harmonics in the SdH and dHvA oscillations differs. The latter is because the extra Mott hopping exponent in the temperature factor of the SdH conductivity oscillations (22) is absent in the thermodynamic dHvA oscillations.

At $T \gg \hbar/\tau$, the conductivity σ_{zz} exhibits a thermally activated behavior which was observed in Ref. 2 [see Eq. (14)]. At lower temperatures, $T \ll \hbar/\tau$, the conductivity crosses over gradually to the Mott VRH regime (19) with the universal scaling variable $(B-B_0)/T^{0.65}$ in the exponent. This regime holds for $B > B_0$ ($B_0 \approx 3.5$ K is very close to the upper critical field, which in β'' salt is equal to $B_{c2} \approx 3.6$ K at $T=0$). At fields $B < B_{c2}(T)$, the superconductivity sets up and decreases the background magnetoresistance, the more the lower the temperature, for $B_{c2}(T)$ is smaller at higher temperatures. Because of that, the R_b below B_0 restores a metal-like behavior. Therefore, the field B_0 separates approximately the metalliclike part of the plot $R_b(T)$ from the VRH insulatorlike one at $B > B_0$.

The above reasoning explains qualitatively the metal-to-insulator-like transition and scaling behavior found in the background magnetoresistance in Ref. 1. Equation (19) shows that at low temperatures $T \ll \hbar/\tau$, oscillations of the conductivity and magnetization are related as in the LK theory by $\sigma_{zz} \propto B^2 \partial \tilde{M}/\partial B$. This relation between the SdH and dHvA oscillations was experimentally established in the β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ salt.[1] A crossover from one VRH regime to another in the logarithmic plot of the background magnetoresistance in Fig. 2 of Ref. 1 is explained qualitatively in Sec. IV C by the two types (intra- and interlayer) of the VRH hopping mechanisms. The Coulomb effects in the VRH conductivity are discussed in Appendix A.

Thus, the approach developed in this paper picks up correctly all unusual experimental observations found in the quantum magnetic oscillations of the highly anisotropic layered organic β'' salt.

A few concluding remarks are in order. First, it is clear that the model with dispersive electron transport across the layers cannot describe either metal-to-insulator transition or scaling in the background magnetoresistance because these phenomena as well as the incoherence are incompatible with conventional LK approach. The latter is valid only for 3D metals with well defined 3D FS and electron dispersion. This point is discussed in detail in Appendixes B and C in the context of the model, with the cosine dispersion adopted in Refs. 16 and 17 to explain the SdH oscillations in the β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$.

Second, I would like to note here the following. Although equations for the conductivity (1)–(5) have been obtained in Ref. 23, the physical picture of the quantum magnetoresistance oscillations in the β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ salt discussed in this paper differs from that adopted in Ref. 23. In this connection, I give an analysis of the drawbacks of a model used in Ref. 23 in Appendixes B and C. In Appendix B, I analyze the applicability of the SCBA approximation to the problem in question and show the principal irrelevance of a narrow cosine-dispersion model for calculations of the σ_{zz} in a highly anisotropic layered conductor under the condition $t \ll \hbar/\tau$.

The scattering time calculation in quasi-2D organic conductors in quantizing magnetic field is a very complex and unsolved problem so far. In the model approach of the present paper, it is taken as a constant. In the SCBA, this can be justified by Eq. (C1), in which tails in the DOS $g(\varepsilon)$ and the large value of the reservoir states $R \gg 1$ smooth oscillations in τ. The use of the SCBA raises problems only in the model with a narrow-band dispersion across the layers adopted in Refs. 16 and 17, and results in the nonphysical negative and nonoscillating conductivity $\sigma_{zz} < 0$ following Eqs. (C2)–(C5).

In conclusion, the approach developed in this paper is rather general and open for further research of the problem in question, in particular, within the frame of more specific models of the incoherence in layered quasi-2D conductors.

ACKNOWLEDGMENTS

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APPENDIX A: VARIABLE-RANGE-HOPPING MECHANISM AND THE EFFECTIVE QUANTUM HOPPING AMPLITUDE BETWEEN THE LAYERS

At high fields and low temperatures, the localization within the layers means qualitatively that Landau orbits drift along the closed equipotential contours of the impurity potential. At places where contours (including contours from the neighboring layers) come close, electrons can tunnel from one contour to another, providing thereby the delocalization and nonzero conductivity across the layers. The hopping between contours includes a thermal activation and tunneling, i.e., it goes through the Mott VRH mechanism.

In the spirit of the VRH concept, we can estimate the amplitude $|t_{\varepsilon,\varepsilon}|^2$ as a quantity proportional to the electron hopping probability between the two 1D closed equipotential contours at which Landau orbits are localized. If R is a distance of hopping, then

$$|t_{\varepsilon,\varepsilon}|^2 \propto \exp\left[-\left(\frac{1}{RN(0)T}+\frac{2R}{\xi}\right)\right]. \quad \text{(A1)}$$

In this equation, we take account of the thermal activation which helps the tunneling if the initial and final levels are separated in energy by a stripe of the order of $1/RN(0)$. Here, $N(0)$ is a one-dimensional DOS at the closed equipotential contours averaged over a random potential and taken near the Fermi level. The optimal hopping distance is $R=\sqrt{\xi/2N(0)T}$ and the corresponding average amplitude is $|t_{\varepsilon,\varepsilon}|^2 \propto \exp(-\sqrt{T_0/T})$, where $T_0=8/N(0)\xi$. In fact, the quantity T_0 is a fitting parameter which can be found only from the experiment. The quantity $N(0)$ determines the average energy separation (or the gap) between the initial and final states in the electron hopping. This gap, in the case when all states are localized and the Coulomb repulsion between electrons plays a dominant role, can be estimated as $C\frac{e^2}{4\pi\epsilon R}$. The appropriate Efros-Shklovskii VRH mechanism yields $T_0=C\frac{e^2}{4\pi\epsilon\xi}$ as was shown in Ref. 38. Here, C is of the order of unity and ϵ is the dielectric constant.

Thus, both 1D Mott hopping mechanism and the Efros-Shklovskii mechanism, which take account of the Coulomb electron-electron effect, result in the same "square-root" exponent factor $\exp(-\sqrt{T_0/T})$ in the VRH conductivity. This factor is well established experimentally in the IQHE conductivity site.[38,39] The quantity T_0 is a fitting parameter of the experiment. This fitting does not answer the question in which proportion 1D Mott and Coulomb-gap mechanisms do contribute to the square-root exponent $\exp(-\sqrt{T_0/T})$. Therefore, by taking effective DOS in the form $1/N_{eff}=1/N(0)+C\frac{e^2}{4\pi\epsilon}$, one can qualitatively take account of the Coulomb effect in the 1D VRH mechanism.

The VRH concept was originally applied to the problem of the conductivity peak broadening $\Delta\nu$ in the IQHE in semiconducting heterojunctions.[38] It was shown that the temperature, current, and frequency dependencies of the $\Delta\nu$ in the quantum Hall conductors can be well described within this paradigm. The square root $\sqrt{T_0/T}$ in the hopping conductivity exponent is well established in conventional IQHE systems. According to our analysis, the same is true for the β'' salt (see discussion in Sec. IV C). This means that the conductivity in these systems is governed by the one-dimensional VRH with plausible assistance of the Coulomb effects. For two- and three-dimensional systems, there should be another exponent $(T_0/T)^{\alpha}$, with $\alpha=1/3$ and $\alpha=1/4$, respectively. In our model, the square-root exponent is because electrons tunnel from one 1D equipotential contour to another. The contours may lie within the same layer as well as in the neighboring layers, and the Coulomb correlations may assist the tunneling as we discussed above.

In a tilted magnetic field, the configuration of the closed equipotential contours at the Fermi level would change (the chemical potential is fixed at $\mu \approx E_F$ in the β'' salt). The reason is that the separation between the LLs depends on the perpendicular component of the magnetic field, while the spin splitting depends on its absolute value. At special values of the tilt angle, the ratio of the separation between LLs to the spin splitting is an integer. At this angles, called the spin-zero angles, the completely filled spin-polarized Landau levels appear at the Fermi level. Correspondingly, the configuration of the equipotential contours and their population at the Fermi energy will change also. At the spin-zero angles, additional completely populated spin-polarized contours appear between the partially populated equipotential contours responsible for the VRH conductivity, which may destroy a percolation in the Mott VRH mechanism as well as related metal-to-insulator transition. Actually, this unusual phenomenon was really observed recently. According to Ref. 6, at the tilt angle corresponding to the spin zero of the fundamental harmonic, the metal-to-insulator transition and the scaling have been destroyed completely in the β'' salt.

APPENDIX B: INCOHERENCE VERSUS COSINE DISPERSION ACROSS THE LAYERS

In this section and in Appendix C, we discuss a crucial role of the incoherence in understanding the unusual magnetoresistance oscillations observed experimentally in β'' salt. We will show here that the narrow cosine-dispersion model adopted in Refs. 16 and 17 is completely inconsistent with the interlayer incoherence and yields nonphysical negative and nonoscillating values for the conductivity across the layers $\sigma_{zz}<0$.

We first note here that Eqs. (1) and (2) result from the summation of the harmonic series which, besides the standard Dingle and temperature factors, contains a new one, specific only for the SdH oscillations:[23]

$$N_{zz}(p) = \int d\varepsilon g(\varepsilon) v_z^2(\varepsilon) \exp\left(\frac{2\pi i p \varepsilon}{\hbar\Omega}\right). \quad \text{(B1)}$$

This kinetic factor depends on the DOS $g(\varepsilon)$ and is valid both for coherent and incoherent interlayer electron hoppings across the layers. If, following Ref. 16, we assume a coherent electron dispersion across the layers $\varepsilon(p_z)=t\cos(ap_z/\hbar)$, then a simple calculation yields $N_{zz}(p)=\frac{a^2 t\Omega}{2\pi\hbar p}J_1\left(\frac{2\pi p t}{\hbar\Omega}\right)$. Here, $J_1(x)$ is a Bessel function. In Ref. 16, the authors, in view of the smallness of the hopping integral $t \ll \hbar\Omega$, used the following approximation: $J_1(x) \approx x/2$. Then $N_{zz}(p) \approx \frac{a^2 t^2}{2}$ becomes independent of the harmonics index p, which, at first glance, makes possible a formal summation on p. This was done in Ref. 16, yielding an expression for the conductivity similar to Eq. (6). However, there is a problem.

We must note in this connection that the above approximation assumes actually a stronger inequality $2\pi p t \ll \hbar\Omega$, which becomes invalid for large p even if $t \ll \hbar\Omega$. To make summation on p with the approximate value $N_{zz}(p) \approx \frac{a^2 t^2}{2}$ correct, the Dingle factor $R_D(p)=\exp(-2\pi|p|/\Omega\tau)$ must cut off contributions from terms with larger p. This gives an additional condition, $t \ll \hbar/\tau$, which means an interlayer incoherence incompatible with the simple dispersion relation $\varepsilon(p_z)=t\cos(ap_z/\hbar)$ adopted in Ref. 16.

Thus, contrary to the statement given in Ref. 43, Eq. (6) was obtained in Ref. 16 incorrectly because the summation on p in a narrow-band dispersive model can be done mathematically correct only with the help of an exact value for the factor $N_{zz}(p)=\frac{a^2t\Omega}{2\pi\hbar p}J_1\left(\frac{2\pi pt}{\hbar\Omega}\right)$. Otherwise, the approach is in conflict with the incoherence. Unfortunately, this fact was not recognized in Ref. 16, which contains yet another serious mistake caused by misuse of the SCBA, as will be shown in Appendix C.

As was pointed out in Ref. 23 and above in this paper, the incoherence plays a crucial role in the derivation of Eq. (6). In that case, only a narrow stripe of delocalized states at the center of the broadened Landau levels contributes to the factor $N_{zz}(p)$, which, therefore, can be approximated by $N_{zz}(p)\approx\langle v_z^2\rangle$. The average velocity squared is independent of p and given by Eq. (10). The corresponding condition necessary for the summation on p reads as $(\varepsilon_{max}-\varepsilon_{min})<t\ll\hbar/\tau$, and is compatible with the incoherence condition.

APPENDIX C: SELF-CONSISTENT BORN APPROXIMATION AND THE SHUBNIKOV–de HAAS OSCILLATIONS

We discuss in this section the applicability of the self-consistent Born approximation to the quantum magnetic oscillations in our problem, and show that in case of dispersive electron transport across the layers, it yields either negative or zero conductivity depending on the strength of the reservoir of nonconducting electronic states. It will be shown below that this drawback of the SCBA is related only to the narrow-band dispersive model and recovers in the case of incoherent interlayer conductivity mechanism.

In the self-consistent Born approximation, the equation for $\tau(E)$ in the presence of the reservoir of states is

$$\frac{\tau_0}{\tau(E)}=\frac{1}{1+R}\left[R+\int d\varepsilon g(\varepsilon)S[\lambda,\delta(E,\varepsilon)]\right]. \qquad (C1)$$

This equation comes from the assumption that the inverse scattering time in the SCBA is proportional to the total DOS, $N(E)=N_R(E)+N_B(E)$, i.e., $N(E)/N(0)=\tau_0/\tau(E)$, where $N(0)$ is the DOS for the 2D electrons at $B=0$, $N_R(E)$ is the DOS of the reservoir, $N_B(E)$ is the oscillating DOS in a quantizing magnetic field B, $R=N_R(E)/N(0)$, and τ_0 is the intralayer scattering time. The reservoir DOS $N_R(E)$ is a smooth function of E due to the states localized at impurities within the layers and electrons on 1D sheets of the FS which do not take part in magnetic oscillations directly. Usually, the quantity $R\approx R(E_F)$ is taken as a constant at the Fermi energy.[30–32] In general, Eq. (C1) tells us that at large $R\gg 1$, oscillations in $\tau(E)$ are suppressed and $\tau\approx$const. This is really the case for the incoherent electron hopping between the layers. The DOS $g(\varepsilon)$ in that case has a narrow peak and wide tails that smooth oscillations in the integral term of Eq. (C1).

In the dispersive model adopted in Ref. 16, the DOS $g(\varepsilon)$ has sharp boundaries at $\varepsilon=\pm t$ and, under the condition $t\ll\hbar/\tau$, Eq. (C1) takes the form

$$\frac{\lambda}{\lambda_0}\approx\frac{1}{1+R}\{R+S[\lambda,\delta(E,0)]\}. \qquad (C2)$$

This is exactly Eq. (30) of Ref. 16, where it was derived incorrectly since a transition from Eq. (C1) to Eq. (C2) assumes the incoherence condition $t\ll\hbar/\tau$, which is at odds with the adopted narrow-band dispersion model. The consequences of Eq. (C2) are the following:

$$\tau G_B=\tau S[\lambda,\delta(E,0)]=\tau_0\left[(1+R)-R\frac{\lambda_0}{\lambda}\right], \qquad (C3)$$

$$\tau G_Q=-\tau_0\lambda_0\frac{\partial}{\partial\lambda}S[\lambda,\delta(E,0)]=-\tau_0(1+R). \qquad (C4)$$

Note that the Boltzmann term cancels partially with the quantum term and both terms do not oscillate in this approximation. Substitution of these equations into Eq. (6) [Eq. (19) in Ref. 16] yields a nonphysical negative conductivity

$$\sigma_{zz}=\int\frac{dE}{\pi}\left(\frac{\partial f}{\partial E}\right)\sigma_\tau R<0. \qquad (C5)$$

In the absence of a reservoir, $R=0$ so that the Boltzmann and quantum terms cancel each other and $\sigma_{xx}=0$.[43]

We see that the use of a narrow-band cosine-dispersion model in the self-consistent Born approximation has serious drawbacks. Such approach under the condition $t\ll\hbar/\tau\ll\hbar\Omega$ is mathematically incorrect and yields a negative nonoscillating conductivity Eq. (C5). On the contrary, Eqs. (1) and (2) are free of the above problems. They provide a basis for more general research including the incoherence and localization effects in the σ_{zz} within the SCBA as well as in other approximations.

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