Comment on "Magnetic quantum oscillations of the conductivity in layered conductors"

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We discuss the recent theory of Gvozdikov [Phys. Rev. B **70**, 085113 (2004)] which aims at explaining the Shubnikov-de Haas oscillations of the longitudinal resistivity ρ_{zz} observed in the quasi-two-dimensional organic compound $\beta'' - (\text{BEDT} - \text{TTF})_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$. We point out that the self-consistent equations of the theory yielding the longitudinal resistivity and the magnetic field dependence of the chemical potential have been incorrectly solved. We show that the consideration of the self-consistent Born approximation (which determines the relaxation rate in Gvozdikov's paper) leads in fact to the complete absence of the longitudinal conductivity σ_{zz} at leading order in high magnetic fields.

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Gvozdikov[1] recently derived a theory of the Shubnikov-de Haas oscillations of the longitudinal conductivity σ_{zz} in quasi-two-dimensional (2D) metals in high perpendicular magnetic fields. He found that σ_{zz} minima display a thermally activated behavior and that σ_{zz} peaks are split in presence of small oscillations of the chemical potential μ. He also claimed that his theory can explain the giant oscillations of the magnetoresistance $\rho_{zz} = \sigma_{zz}^{-1}$ observed[2] in the layered organic compound $\beta'' - (\text{BEDT} - \text{TTF})_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$.

In this Comment, we point out that the theoretical results have been incorrectly derived and that the theoretical model adopted by Gvozdikov is inconsistent with existing experiments. In the following, we first address the framework of the theory and the problems with the calculation of σ_{zz}. Then, we shall focus on the thermodynamic quantities such as the density of states and the chemical potential. Finally, we shall discuss possible theoretical explanations of experimental observations.

I. THE LONGITUDINAL CONDUCTIVITY σ_{zz}

A. Model and framework

The system under consideration is a quasi-two-dimensional metal with an interlayer hopping integral t. In the paper,[1] the interlayer dispersion is modelled by an unknown density of states with a characteristic width of the order of t. Gvozdikov's paper[1] was aimed at deriving the oscillations of σ_{zz} as a function of magnetic field perpendicular to the layers under the regime of a weakly incoherent interlayer transport.

The incoherent transport was discussed in literature (see, e.g., Ref. 3) in the cases of the so-called weakly and strongly incoherent regimes. The former occurs when the intralayer momentum is conserved in the process of

transfer of the electron from one layer to another. The incoherence is relevant when a large number of in-plane scatterings takes place before an interlayer tunneling occurs, i.e. when $t \ll \hbar/\tau$, where τ is the mean free time. Interlayer transport is then incoherent because the successive tunneling events get uncorrelated. This situation was first discussed in the paper[4] in relationship with the conductivity in cuprates and it gives a Drude-type formula for the interlayer conductivity where the scattering time is the time of inelastic intralayer scattering. The strongly incoherent transport occurs when the intralayer momentum is not conserved by tunneling and there is no interference between the wavefunctions on adjacent layers.

In Gvozdikov's theory[1] intralayer inelastic processes are apparently[5] not considered since the width $\Gamma = \hbar/2\tau$ of the spectral function in the layer is determined by the elastic scattering on impurities (note that in our definition there is a difference of a factor 2 with Gvozdikov's notation). The additional complicacy considered in Ref. 1 consists of the inelastic processes at tunneling. The Landau levels and (implicitly) the centers of the Landau orbits are assumed to be conserved. This is valid provided that the characteristic value of the energy change at tunneling, γ, is much less than the intralayer spectral width Γ. Under this regime of so-called weak incoherence by Gvozdivov (that does not correspond to the definition of weak incoherence given above), the inelastic processes at tunneling do not play a role for the interlayer transport and we return to a theory where only elastic scattering on impurities is relevant. It should be noted that there is no clear distinction between t and γ in Ref. 1.

The derivation of the longitudinal conductivity σ_{zz} for elastic impurity scattering and a cosinelike interlayer energy dispersion was already reported in the Refs 6,7,8. The paper[6] was interested in the regime $\hbar\Omega \leq t$ (here Ω is the cyclotron frequency), while the papers[7,8] were

concerned with the 2D limit $\hbar\Omega \gg t$ in connection with the experiment.[2]

B. Derivation of σ_{zz}

The longitudinal conductivity σ_{zz} is derived within the Kubo formula written in the formalism of the Green functions. Vertex corrections have been neglected in Ref. 1 which means that a point-like impurity model has been implicitly assumed. Within this model the self-energy part entering into the expression of the disorder averaged Green functions is independent of the Landau levels index n. This fact allows to compute analytically the sum over n using the Poisson summation formula.

Gvozdikov has derived an expression for σ_{zz}, Eqs. (21)-(22). In addition to the condition $t \ll \Gamma$, the 2D limit $\hbar\Omega \gg t$ is required for its validity. Under the same inequalities, one can rewrite these equations (21)-(22) as

$$\sigma_{zz} = \int dE \, (-f'(E)) \, [\sigma_B + \sigma_Q], \qquad (1)$$

where

$$\sigma_B = \sigma_\tau \frac{\lambda_0}{\lambda(E)} S(\lambda(E), E), \qquad (2)$$

$$\sigma_Q = -\sigma_\tau \lambda_0 \frac{\partial S(\lambda(E), E)}{\partial \lambda}. \qquad (3)$$

Here $f(E)$ is the Fermi-Dirac distribution function which depends on the chemical potential μ, σ_τ is the conductivity at zero magnetic field, $\lambda(E) = \pi/\Omega\tau(E)$, $\lambda_0 = \pi/\Omega\tau_0$ [$\tau(E)$ and τ_0 are respectively the *elastic* quasi-particle lifetimes at energy E in the presence and absence of magnetic field] and the function S is defined by

$$S(\lambda(E), E) = \frac{\sinh \lambda(E)}{\cosh \lambda(E) + \cos(2\pi E/\hbar\Omega)}. \qquad (4)$$

Formulas (1)-(4) are incomplete without a prescription for calculating the function $\lambda(E)$. To proceed further, Gvozdikov applied the self-consistent Born approximation (SCBA) to the quasi-2D spectrum. This approximation stipulates the proportionality between the density of states $N(E)$ and $\tau^{-1}(E)$, i.e.,

$$\frac{\tau_0}{\tau(E)} = \frac{N(E)}{N_0}. \qquad (5)$$

Here N_0 is the density of states of the quasi-2D system at zero magnetic field. The density of states $N(E)$ under magnetic field is expressed as a Fourier series and is a function of the lifetime $\tau(E)$. In the 2D limit ($\hbar\Omega \gg t$) and for $t \ll \Gamma$, it is possible to sum up this series and the relation (5) leads to the following self-consistent equation for $\lambda(E)$:

$$\lambda(E) = \lambda_0 \, S\left(\lambda(E), E\right). \qquad (6)$$

Equation (6) yields straightforwardly the fact that the term $\sigma_B = \sigma_\tau$ [see Eq. (2)], i.e., this latter does not oscillate with the magnetic field. Gvozdikov got then that the oscillations in the strong 2D regime arise from the term σ_Q only. Here we want to stress that this result[1] has been incorrectly derived from the self-consistent Eq. (6). All the subsequent results based on the formula (28) for σ_{zz} in Ref. 1 are accordingly incorrect. Indeed, differentiating the formula (6) with respect to λ, we obtain that $\lambda_0 \partial S/\partial \lambda = 1$. Substituting this result into Eq. (3) we straightforwardly have $\sigma_Q = -\sigma_\tau$, which means that in frame of SCBA the total conductivity

$$\sigma_{zz} = 0 \qquad (7)$$

in the limit $\hbar\Omega \gg t$. A non-zero expression for σ_{zz} may be obtained by considering next order corrections in the parameters $t/\hbar\Omega$ and t/Γ.

It is worth noting that the Eqs. (1)-(4) are exactly the same as Eqs. (19)-(21) given in the work[7] considering a cosinelike dispersion relation with respect to the interlayer momentum. The reason for this is that the specific form of the interlayer density of states is unimportant under the conditions $t \ll \hbar\Omega$ and $t \ll \hbar/\tau$. The important difference between the two theories is that in Ref. 7 the total (not just the intralayer) spectral width due to elastic scattering on impurities $\Gamma = \hbar/2\tau$ is postulated as being constant, i.e. energy independent: the SCBA leading to Eq. (6) and Eq. (7) is not considered as an inherent property of the transport theory.

We would like to stress that Eq. (7) is a consequence of the application of SCBA in high magnetic fields only. In smaller magnetic fields $\hbar\Omega \leq t$, the same SCBA yields[6] small oscillations of σ_{zz} around the zero field value which is nonzero due to 3D elastic scattering on impurities.

II. THERMODYNAMIC QUANTITIES

A. Density of states

It is important to note that Eq. (6) determines as well the function $\lambda(E)$ (or the lifetime $\tau(E) = \pi/\Omega\lambda(E)$) as the density of states $N(E) = N_0\lambda(E)/\lambda_0$. It has been recognized in the Section IV of Ref. 7 that the self-consistent equation (6) for the density of states is exactly the same equation as one would obtain using the SCBA with a strict 2D spectrum, which has has been well known for a long time.[9,10] The approximation consisting in replacing $\lambda(E)$ everywhere by the peak value $\lambda(E_n) \approx \sqrt{2\lambda_0}$ as done in Ref. 1 is inconsistent with Eq. (6). In fact, the self-consistent Eq. (6) yields a function $\lambda(E)$ which strongly oscillates with E, especially in high magnetic fields. When $\lambda_0 \leq 2$, $\lambda(E)$ even vanishes between Landau levels energy positions $E_n = (n+1/2)\hbar\Omega$, which means that the density of states splits into separate bands centered on the energies E_n (see Refs. 9,10). Going beyond SCBA, it is possible to calculate more accu-

rately the tails of the bands to avoid the unphysical cut-off produced by Eq. (6). Moreover, next corrections due to the interlayer hopping, of the order of $t/\hbar\Omega$ and t/Γ, may yield finite but small values for $N(E)$ between Landau levels. However, in any cases, the model adopted by Gvozdikov automatically implies sharp peaks for $N(E)$ when $E \approx E_n$ and the quasiabsence of states between Landau levels for $\Omega\tau_0 > \pi/2$.

B. Chemical potential

Eq. (23) of Ref. 1 [reproduced here with Eq. (6)] completely determines the density of states and thus allows to compute the chemical potential μ as a function of magnetic field. Therefore, there is no freedom for the choice of some phenomenological model describing the chemical potential oscillations in accordance with the experiments, as surprisingly done in the Section 4 of Ref. 1.

If the density of states consists of sharp bands centered on the Landau levels, one expects that μ is pinned to a value close to $(n + 1/2)\hbar\Omega$ for most of the range in magnetic fields and drops suddenly to another Landau band once a Landau band becomes filled or empty. This process gives rise to strong magnetic quantum oscillations of the chemical potential. This expectation is physically inconsistent with the experimental observation of an inverse sawtooth profile of the magnetization oscillations in the compound $\beta'' - (\text{BEDT} - \text{TTF})_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$,[11] that implies[12] the presence of negligibly small oscillations of μ. Furthermore, the experiments on thermodynamic and transport quantities[2,11] rather suggest a significant amount of states between the Landau levels for $\Omega\tau_0 \sim 1$ in contradiction with the result obtained within the self-consistent Born approximation.

Now, we discuss the different forms possible for the chemical potential oscillations under the assumption of a constant lifetime (as in Section 4 of Ref. 1), because they have been incorrectly presented and used in Ref. 1. For an energy independent lifetime $\bar{\tau}$, the equation obeyed at zero temperature by the chemical potential μ in the 2D limit can be explicitly calculated[12,13]

$$\mu = E_F + \frac{\hbar\Omega}{\pi} \arctan\left(\frac{\sin(2\pi\mu/\hbar\Omega)}{e^\nu + \cos(2\pi\mu/\hbar\Omega)}\right) \quad (8)$$

where $\nu = \pi/\Omega\bar{\tau}$ and E_F is the Fermi energy at zero magnetic field. As pointed out by Grigoriev,[14] it is possible to invert this self-consistent formula (8) to obtain

$$\mu = E_F + \frac{\hbar\Omega}{\pi} \arctan\left(\frac{\sin(2\pi E_F/\hbar\Omega)}{e^\nu - \cos(2\pi E_F/\hbar\Omega)}\right). \quad (9)$$

When $\nu \ll 1$, the density of states within this model consists of sharp peaks centered around the Landau levels (as in the SCBA), and this Eq. (9) yields a staircase dependence for $\mu/\hbar\Omega$ as a function of magnetic field. Correspondingly, the oscillating part $\tilde{\mu}/\hbar\Omega = (\mu - E_F)/\hbar\Omega$

exhibits a sawtooth dependence (with the so-called direct shape).[12]

Within the phenomenological model assuming a reservoir of states and a constant lifetime, the equation for μ now takes the form[12]

$$\mu = E_F + \frac{\hbar\Omega}{\pi(1 + R)} \arctan\left(\frac{\sin(2\pi\mu/\hbar\Omega)}{e^\nu + \cos(2\pi\mu/\hbar\Omega)}\right) \quad (10)$$

where R is a dimensionless parameter measuring the strength of the reservoir. Equation (10) embraces naturally the former model with $R = 0$. For an important reservoir, $R \gg 1$, the oscillating part $\tilde{\mu}$ is extremely small ($\tilde{\mu} \ll \hbar\Omega$), implying that μ is almost fixed to the zero field value E_F. In this particular case, it is then justified to replace μ by E_F in the right-hand side of this Eq. (10) to obtain

$$\mu \approx E_F + \frac{\hbar\Omega}{\pi R} \arctan\left(\frac{\sin(2\pi E_F/\hbar\Omega)}{e^\nu + \cos(2\pi E_F/\hbar\Omega)}\right). \quad (11)$$

Then, $\tilde{\mu}/\hbar\Omega$ exhibits the (so-called) inverse sawtooth waveform when $\nu \ll 1$, but with a negligibly small amplitude (because $R \gg 1$). On the contrary, the magnetization oscillations which are proportional to $R\tilde{\mu}/\hbar\Omega$ are not limited in amplitude by the reservoir parameter R (see Ref. 12).

In his discussion of the effects of the chemical potential oscillations on σ_{zz}, Gvozdikov[1] has used Eq. (8) (i.e. no reservoir) with μ replaced by E_F in the right-hand side of the equation. We want to stress that it is generally mathematically incorrect to replace μ by E_F[12,13] in the right-hand side of Eq. (8) especially when $\nu \ll 1$ [e.g., compare the resulting equation and Eq. (9)]. An additional remark is that Eq. (31) for μ considered in Ref. 1 [Eq. (8) with a minus instead of a plus between the two terms in the right-hand side] to describe the direct sawtooth case has no basis at all.

III. THEORETICAL APPROACHES

As already mentioned above, the theory in Ref. 7, also aimed at explaining the oscillations of σ_{zz} reported in Ref. 2, is based on the same Eqs. (1)-(4) for σ_{zz}. The difference with Gvozdikov's theory rests on the function $\lambda(E)$ [or equivalently on the lifetime $\tau(E)$], which is (phenomenologically) assumed to be constant. Within this model both contributions σ_B and σ_Q oscillate with magnetic fields. The thermal activation of σ_{zz} minima stems from a cancellation at zero order in $t/\hbar\Omega$ of these two contributions between the Landau levels.[7,8] Contrary to SCBA, the cancellation does not occur at the Landau levels.

Now, we would like to address one alternative explanation to the experiment.[2] Owing to some similitudes, it is tempting to bring together the present problem in a layered conductor and the problem encountered in the

theory of the quantum Hall effect[15]. At the experimental side, in this quasi-2D conductor and in the 2D electron gases under the regime of the integer quantum Hall effect, a thermal activation is observed: in the first system it concerns the minima of the interlayer conductivity σ_{zz},[2] and in the second system the minima of the longitudinal conductivity σ_{xx}.[16] The puzzling point is that the density of states extracted from different measurements of the thermodynamic quantities points out a rather large amount of states between the Landau peaks in the layered organic conductor[2,11] and in the 2D electron gases under the regime of the integer quantum Hall effect[17,18] which is typically observed when $\Omega\tau_0 \geq 1$. As a result, we have to face the same difficulty, i.e., to capture transport and thermodynamic properties within a complete and consistent microscopic theory. In the regime of the integer quantum Hall effect, it is believed that most of the states of the 2D layer are localized by the disorder and act as a reservoir which almost fixes the chemical potential.[16] The core of the extended states is located at the energies E_n. At finite but not too low temperatures, the main means of conduction when the chemical potential sits between the Landau levels is the thermal excitation of quasiparticles at the edges of the mobility gap.[16] We could presume the presence of similar localized states in the layers responsible for the thermal activation of σ_{zz} minima at high magnetic fields in the compound $\beta'' - (\mathrm{BEDT-TTF})_2\mathrm{SF}_5\mathrm{CH}_2\mathrm{CF}_2\mathrm{SO}_3$.

For the moment there exists no proper quantum calculations for σ_{xx} in the quantum Hall effect regime,[15] so that this scenario for σ_{zz} only rests on pure qualitative considerations. In fact, the principal theoretical difficulty is the microscopic treatment of the impurity scattering effects on the Landau levels quantization. For this purpose, it is worth noting that the consideration of a model of impurity potential with zero-range is unphysi-cal in high magnetic fields B since in the limit $B \to \infty$ the magnetic length $l_B = (\hbar c/eB)^{1/2}$ always becomes smaller than the correlation radius of the potential. It has been shown[19] for the 2D systems that the SCBA is no more valid when the range of the potential exceeds l_B so that the investigation of impurity effects within the usual perturbation theory becomes very complicated. However, it seems that even this ingredient which consists to consider a finite range for the impurity potential is not sufficient to explain the presence of a background reservoir of states.[20] Apparently, new ideas are needed.

IV. CONCLUSION

The results reported in the paper[1] are based on the inconsequent use of SCBA. The proper SCBA application at leading order in high magnetic field leads to the zero conductivity $\sigma_{zz} = 0$ and to strong oscillations of the density of states and of the chemical potential. Both latter effects can be suppressed in the presence of a significant number of states between Landau levels. An explanation for the thermally activated behavior of σ_{zz} minima could be the presence of localized states as in the integer quantum Hall effect. However, a concrete self-consistent magnetotransport theory going beyond SCBA and including these localized states is still missing.

[1] V.M. Gvozdikov, Phys. Rev. B **70**, 085113 (2004).

[2] M.-S. Nam *et al.*, Phys. Rev. Lett. **87**, 117001 (2001).

[3] P. Moses and R.H. McKenzie, Phys. Rev. B **60**, 7998 (1999).

[4] N. Kumar and A.M. Jayannavar, Phys. Rev. B **45**, 5001 (1992).

[5] This fact is deduced from the use of the self-consistent Born approximation [mentioned in the paragraph containing Eq. (17) in Ref. 1] determining microscopically the scattering time τ.

[6] P.D. Grigoriev, Phys. Rev. B **67**, 144401 (2003).

[7] T. Champel and V.P. Mineev, Phys. Rev. B **66**, 195111 (2002) ; *ibid* **67**, 089901(E) (2003).

[8] T. Champel and V.P. Mineev, Physica B **346-347**, 392 (2004).

[9] T. Ando, J. Phys. Soc. Jpn. **37**, 1233 (1974).

[10] T. Ando, A.B. Fowler, and F. Stern, Rev. Mod. Phys. **54**, 437 (1982).

[11] J. Wosnitza *et al.*, Phys. Rev. B **61**, 7383 (2000).

[12] T. Champel, Phys. Rev. B **64**, 054407 (2001).

[13] T. Champel and V.P. Mineev, Philos. Mag. B **81**, 55 (2001).

[14] P.D. Grigoriev, Zh. Éksp. Teor. Fiz. **119**, 1257 (2001) [Sov. Phys. JETP **92**, 1090 (2001)].

[15] M. Janssen, O. Viehweger, U. Fastenrath, and J. Hajdu, *Introduction to the Theory of the Integer Quantum Hall Effect* (Wiley-VCH, Weinheim, 1994).

[16] *The Quantum Hall Effect*, edited by R.E. Prange and S.M. Girvin (Springer-verlag, new-York, 1990).

[17] E. Gornik *et al.*, Phys. Rev. Lett. **54**, 1820 (1985).

[18] J.P. Eisenstein *et al.*, Phys. Rev. Lett. **55**, 875 (1985).

[19] M.E. Raikh and T.V. Shahbazyan, Phys. Rev. B **47**, 1522 (1993).

[20] L. Spies, W. Apel, and B. Kramer, Phys. Rev. B **55**, 4057 (1997).