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Monotonic growth of interlayer magnetoresistance in strong magnetic field in very anisotropic layered metals

P. D. Grigoriev[1)]

L.D. Landau Institute for Theoretical Physics, 142432 Chernogolovka, Russia

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It is shown, that the monotonic part of interlayer electronic conductivity strongly decreases in high magnetic field perpendicular to the conducting layers. We consider only the coherent interlayer tunnelling, and the obtained result strongly contradicts the standard theory. This effect appears in very anisotropic layered quasi-two-dimensional metals, when the interlayer transfer integral is less than the Landau level separation.

Introduction. The investigation of the angular and magnetic field dependence of magnetoresistance provides a powerful tool of studying the electronic properties of various metals. The Fermi surface geometry of the most metals has been measured using the magnetic quantum oscillations (MQO) of magnetoresistance [1–3]. The angular dependence of magnetoresistance also gives the important information about the electronic structure and is widely used to investigate the electronic properties of layered compounds: organic metals (see, e.g., Refs. [4–7] for reviews), cuprate high-temperature superconductors [8–11], heterostructures [12] etc.

In layered quasi-2D metals, where the interlayer transfer integral t_z is considerably smaller than the in-plane electron Fermi energy, the electron dispersion is given in the tight-binding approximation by

$$\epsilon_{3D}\left(\mathbf{k}\right) \approx \epsilon\left(k_x, k_y\right) - 2t_z \cos(k_z d), \qquad (1)$$

where $\epsilon\left(k_x, k_y\right)$ is the in-plane electron dispersion, k_z is out-of-plane electron momentum, and d is the interlayer spacing. If t_z is much larger than the Landau level (LL) separation $\hbar\omega_c = \hbar eB/m^*c$, the standard theory of galvanomagnetic properties [1–3] works well. This theory predicts several special features of magnetoresistance in quasi-2D metals: the angular magnetoresistance oscillations [13, 14] and the beats of the amplitude of MQO [1].

In strongly anisotropic layered quasi-2D metals, when $t_z \sim \hbar\omega_c$, many new qualitative effects emerge. For example, the slow oscillations of magnetoresistance appear [15, 16] and the beats of MQO of transport quantities become shifted [17, 16]. These effects are not described by the standard theory [1–3], because it is valid only in the lowest order in the parameter $\hbar\omega_c/t_z$. When this parameter becomes of the order of unity, the standard theory is no longer applicable.

The monotonic part of magnetoresistance also changes when $t_z \lesssim \hbar\omega_c$. According to the standard theory [2], external magnetic field along the electric current leads only to MQO but does not influence the monotonic (background) part of this current. However, the monotonic increase of interlayer magnetoresistance R_{zz} with the increase the magnetic field $\mathbf{B}$ perpendicular to the conducting layers has been observed in various strongly anisotropic layered metals [18–24]. This monotonic growth of magnetoresistance was attributed to the "strongly incoherent" regime, where the interlayer tunnelling described by the usual Hamiltonian term in Eq. (5) is not effective, and the new mechanisms of interlayer electron transport play the major role. For example, the variable-range electron hopping between the localized states in strong magnetic field leads to the insulating behavior and to the exponential dependence of interlayer conductivity on temperature and magnetic field [25]. In another model, where the in-plane electron motion is fully metallic but the interlayer electron transport goes via rare local crystal defects (e.g., resonance impurities), the interlayer conductivity σ_{zz} also has metallic-type temperature dependence but decreases strongly with the increase of the out-of-plane component of magnetic field [23]. The boson-assisted interlayer tunnelling can describe only the unusual temperature dependence of interlayer conductivity at $T \sim 10{-}150\,\mathrm{K}$ [26–28], but it does not explain its magnetic field dependence.

In Ref. [29] it was shown, that the monotonic growth of magnetoresistance $R_{zz} \propto \sqrt{B_z}$ appears also in the standard model, described by the Hamiltonian in Eqs. (2)–(6), in strong magnetic field at very weak interlayer coupling: $\hbar\omega_c \gg \Gamma_0 > t_z$, where $\Gamma_0 = \hbar/2\tau_0$ is the electron level broadening due to impurity scattering in the absence of magnetic field and τ_0 is the electron mean free time. This contradicts the common opinion [30] that in the "weakly incoherent" regime, i.e. at $\Gamma_0 > t_z$,

[1)] e-mail: grigorev@itp.ac.ru

the interlayer magnetoresistance does not differ from the coherent almost 3D limit $t_z \gg \Gamma_0$. This increase of magnetoresistance was also missed in Refs. [31–33], where the Born approximation has been incorrectly applied to describe almost the 2D-electron system. However, in Ref. [29] the approximate and phenomenological electron Green's function has been used, given by Eq. (22) of Ref. [29]. Below we rederive the main result of Ref. [29] more rigorously, which gives the prefactor $4/\pi \approx 1.27$ times greater for the interlayer conductivity than in Ref. [29].

The model. The electron Hamiltonian in layered compounds with small interlayer coupling contains 3 main terms:

$$\hat{H} = \hat{H}_0 + \hat{H}_t + \hat{H}_I. \tag{2}$$

The first term $\hat{H}_0$ is the noninteracting 2D-electron Hamiltonian summed over all layers:

$$\hat{H}_0 = \sum_{m,j} \varepsilon_{2D}(m)\, c^+_{m,j} c_{m,j}, \tag{3}$$

where $\{m\} = \{n, k_y\}$ is the set of quantum numbers of electrons in magnetic field on a 2D conducting layer, $c^+_{m,j}(c_{m,j})$ are the electron creation (annihilation) operators in the state $\{m\}$ on the layer j, and $\varepsilon_{2D}(m)$ is the corresponding free electron dispersion given by

$$\varepsilon_{2D}(n, k_y) = \hbar\omega_c\,(n + 1/2)\,. \tag{4}$$

The second term in Eq. (2) gives the coherent electron tunnelling between two adjacent layers:

$$\hat{H}_t = 2t_z \sum_j \int d^2\mathbf{r}[\Psi^\dagger_j(\mathbf{r})\Psi_{j-1}(\mathbf{r}) + \Psi^\dagger_{j-1}(\mathbf{r})\Psi_j(\mathbf{r})], \tag{5}$$

where $\Psi_j(\mathbf{r})$ and $\Psi^\dagger_j(\mathbf{r})$ are the creation (annihilation) operators of an electron on the layer j at the point $\mathbf{r}$. This interlayer tunnelling Hamiltonian is called "coherent" because it conserves the in-layer coordinate dependence of the electron wave function (in other words, it conserves the in-plane electron momentum) after the interlayer tunnelling. The last term

$$\hat{H}_I = \sum_i \int d^3\mathbf{r} V_i(\mathbf{r})\, \Psi^\dagger(\mathbf{r})\Psi(\mathbf{r}) \tag{6}$$

gives the electron interaction with impurity potential. The impurities are taken to be point-like and randomly distributed on conducting layers with volume concentration n_i and areal concentration $N_i = n_i d$ on each layer. The impurity distributions on any two adjacent layers are uncorrelated. The potential $V_i(\mathbf{r})$ of any impurity located at point $\mathbf{r}_i$ is given by

$$V_i(\mathbf{r}) = U\delta^3(\mathbf{r} - \mathbf{r}_i)\,. \tag{7}$$

We also introduce the 2D point-like impurity potential with the strength $V_0 = U\,|\psi(z_i)|^2 \approx U/d$ of each impurity:

$$V_i(x, y) = V_0\delta(x - x_i)\,\delta(y - y_i)\,. \tag{8}$$

In the limit, $t_z \ll \Gamma_0, \hbar\omega_c$, the interlayer hopping t_z must be considered as a perturbation for the disordered uncoupled stack of 2D metallic layers. The 2D metallic electron system in magnetic field in the point-like impurity potential has been extensively studied [34–40]. In the self-consistent single-site approximation the coordinate electron Green's function, averaged over impurity configurations, is given by

$$G(\mathbf{r}_1, \mathbf{r}_2, \varepsilon) = \sum_{n,k_y} \Psi^{0*}_{n,k_y}(r_2)\Psi^0_{n,k_y}(r_1) G(\varepsilon, n)\,, \tag{9}$$

where $\Psi^0_{n,k_y}(r_1)$ are the 2D electron wave functions in perpendicular magnetic field [41], and the Green's function $G(\varepsilon, n)$ does not depend on k_y:

$$G(\varepsilon, n) = \frac{1}{\varepsilon - \hbar\omega_c\,(n + 1/2) - \Sigma(\varepsilon)}, \tag{10}$$

where $\Sigma(\varepsilon)$ is the electron self-energy part due to scattering by impurities.

The interlayer conductivity σ_{zz}, associated with the Hamiltonian (5), can be calculated using the Kubo formula and the formalism, developed for the metal-insulator-metal junctions [42]. In analogy to Eq. (44) of Ref. [30],

$$\sigma_{zz} = \frac{4e^2t_z^2 d}{\hbar L_x L_y} \int d^2\mathbf{r} d^2\mathbf{r}' \int \frac{d\varepsilon}{2\pi}\,[-n'_F(\varepsilon)] \times \\ \times \langle \mathrm{Im}G_R(\mathbf{r}, \mathbf{r}', j, \varepsilon)\mathrm{Im}G_R(\mathbf{r}', \mathbf{r}, j+1, \varepsilon)\rangle\,. \tag{11}$$

The angular brackets in Eq. (11) mean averaging over impurity configurations. Since the impurity distributions on adjacent layers are uncorrelated, one can perform this averaging separately for each layer. The averaged Green's functions are translational invariant: $\langle G_R(\mathbf{r}, \mathbf{r}', j, \varepsilon)\rangle = \langle G_R(\mathbf{r} - \mathbf{r}', j, \varepsilon)\rangle$. Therefore, one can perform the integration over $\mathbf{r}'$, which removes the sample size $L_x L_y$:

$$\sigma_{zz} = \frac{2\sigma_0\Gamma_0}{\pi\nu_{2D}} \int d^2\mathbf{r} \int d\varepsilon\,[-n'_F(\varepsilon)] \times \\ \times \langle \mathrm{Im}G_R(\boldsymbol{r}, j, \varepsilon)\rangle\,\langle \mathrm{Im}G_R(\boldsymbol{r}, j+1, \varepsilon)\rangle\,, \tag{12}$$

where we introduced the interlayer conductivity without magnetic field

$$\sigma_0 = e^2t_z^2\nu_{2D}d/\hbar\Gamma_0, \tag{13}$$

$\nu_{2D} = 2N_{LL}/\hbar\omega_c = m^*/\pi\hbar^2$ is the 2D density of states (DoS) at the Fermi level in the absence of magnetic field per two spin components, and N_{LL} is the LL-degeneracy per unit area.

When the magnetic field is perpendicular to the conducting layers, the coordinate dependence of the electron Green's function on the adjacent layers is the same. Then the integration over $\boldsymbol{r}$ for the Green's function of the form (9) is very simple and gives the factor N_{LL}:

$$\sigma_{zz} = \frac{\sigma_0 \Gamma_0 \hbar\omega_c}{\pi} \int d\varepsilon \left[-n'_{\mathrm{F}}(\varepsilon)\right] \sum_n \left|\mathrm{Im} G_R(\varepsilon, n)\right|^2. \quad (14)$$

In the zero-temperature limit, where $-n'_{\mathrm{F}}(\varepsilon) = \delta(\varepsilon - \mu)$, and in weak magnetic field, where the summation over n can be replaced by the integration over n, Eq. (14) gives

$$\sigma_{zz}(B) = \sigma_0 \Gamma_0 / \left|\mathrm{Im}\Sigma(\mu, B)\right| \quad (15)$$

in agreement with the standard theory.

Calculation. In strong magnetic field, when $\hbar\omega_c \gg \Gamma_0 = \pi n_i U^2 \rho_{3D} = \pi N_i V_0^2 N_{LL}/\hbar\omega_c = \hbar/2\tau_0$, one can consider each Landau level separately. In the self-consistent single-site approximation [34] the electron Green's function on each LL is given by

$$G(E, n) = \frac{E + E_g(1 - c_i) - \sqrt{(E - E_1)(E - E_2)}}{2EE_g}, \quad (16)$$

and the DoS on each LL is described by the well-known dome-like function [34]

$$\frac{-\mathrm{Im} G_R(E, n)}{\pi} = D(E) = \frac{\sqrt{(E - E_1)(E_2 - E)}}{2\pi |E| E_g}, \quad (17)$$

where the electron energy E is counted from the last occupied LL: $E \equiv \varepsilon - \varepsilon_{2D}(n_{\mathrm{F}}, k_y)$, and $E_g = N_{LL} V_0$, where the LL-degeneracy per unit area is $N_{LL} = 1/2\pi l_{Hz}^2 = eB/2\pi\hbar c$. The boundaries of the DoS dome in Eq. (17) are

$$E_1 = E_g\left(\sqrt{c_i} - 1\right)^2, \; E_2 = E_g\left(\sqrt{c_i} + 1\right)^2, \quad (18)$$

where c_i is the dimensionless ratio of the impurity concentration to the electron concentration on one LL:

$$c_i = N_i/N_{LL} = 2\pi l_{Hz}^2 n_i d. \quad (19)$$

The function $D(E)$ in Eq. (17) is nonzero in the interval $0 < E_1 < E < E_2$ and normalized to unity: $\int D(E)\, dE = 1$. The LL half-width

$$\Gamma_B \equiv (E_2 - E_1)/2 = 2E_g\sqrt{c_i} \propto \sqrt{B}. \quad (20)$$

The LL broadening Γ_B in Eq. (17) is much larger than Γ_0 and depends on magnetic field, which is emphasized by the subscript "B". The ratio

$$\Gamma_B/\Gamma_0 \approx \sqrt{4\hbar\omega_c/\pi\Gamma_0} \gg 1 \quad (21)$$

grows as $\sqrt{B}$ in high magnetic field.

Taking zero temperature and substituting Eq. (17) into Eq. (14) we obtain

$$\sigma_{zz}(E) = \frac{\sigma_0 \Gamma_0 \hbar\omega_c}{\pi} \sum_n \left(\frac{\sqrt{(E - E_1)(E_2 - E)}}{2|E| E_g}\right)^2, \quad (22)$$

where $E \equiv \mu - \varepsilon_{2D}(n_{\mathrm{F}}, k_y)$ and the real part of the square root must be taken, which is nonzero only in the interval $E_1 < E < E_2$. The monotonic part $\bar{\sigma}_{zz}$ of conductivity can be obtained by the averaging of Eq. (22) over the oscillation period $\hbar\omega_c$:

$$\bar{\sigma}_{zz} = \int_{E_1}^{E_2} \sigma_{zz}(E)\, dE/\hbar\omega_c =$$
$$= \frac{\sigma_0\Gamma_0}{2\pi E_g^2}\left[\frac{E_2 + E_1}{2}\ln\left(\frac{E_2}{E_1}\right) + E_1 - E_2\right] =$$
$$= \frac{2\sigma_0\Gamma_0}{\pi E_g}\left[\frac{1 + c_i}{2}\ln\left(\frac{\sqrt{c_i} + 1}{\sqrt{c_i} - 1}\right) - \sqrt{c_i}\right]. \quad (23)$$

When $c_i \gg 1$, this simplifies to

$$\bar{\sigma}_{zz} \approx \frac{2\sigma_0\Gamma_0}{\pi E_g\sqrt{c_i}} = \sigma_0\sqrt{\frac{4\Gamma_0}{\pi\hbar\omega_c}}. \quad (24)$$

The interlayer conductivity in Eq. (24) decreases with the increase of magnetic field: $\bar{\sigma}_{zz} \propto B^{-1/2}$. Qualitatively, this dependence is obtained by substituting $\left|\mathrm{Im}\Sigma(\mu, B)\right| \approx \Gamma_B$ and Eq. (21) into Eq. (15):

$$\bar{\sigma}_{zz} \approx \frac{\sigma_0\Gamma_0}{\left|\mathrm{Im}\Sigma\right|} \approx \sigma_0\frac{\Gamma_0}{\Gamma_B} = \sigma_0\sqrt{\frac{\pi\Gamma_0}{4\hbar\omega_c}}. \quad (25)$$

In Ref. [29] the qualitative arguments, similar to those in the derivation of Eq. (25), have been applied to show the monotonic growth and the change in the angular dependence of interlayer magnetoresistance. However, the arguments in Eq. (25) are not strict, because $\left|\mathrm{Im}\Sigma(\mu, B)\right| \neq \Gamma_B$, being a strongly oscillating function of magnetic field B and of Fermi level μ. Therefore, the calculated value of $\bar{\sigma}_{zz}$ in Eq. (24) is $4/\pi \approx 1.27$ times greater than the qualitative estimate in Eq. (25), and the above calculation of interlayer conductivity, resulting in Eq. (24), is more strict than in Ref. [29].

The obtained correction by a factor of $4/\pi \approx 1.27$ to the result of Ref. [29] is not very important compared to the large field-dependent factor $\sqrt{\hbar\omega_c/\Gamma_0} \propto \sqrt{B_z}$. The Green's function given by Eq. (16) is also approximate, and the factor $4/\pi \approx 1.27$ gives the estimate of the possible inaccuracy of the calculation. However, the inclusion of the neglected diagrams with the intersections of the impurity lines will not increase the accuracy, because the initial model given by Eqs. (3)–(8) does not include many relevant minor factors. For example, this model considers only one type of impurities, while in real compounds there are always several types, which

results in the averaging over the impurity strength V_0 in Eq. (8). Nevertheless, the above derivation of Eq. (24) is more rigorous and self-consistent than in Ref. [29], which gives a stricter proof of the main result: the increase of the monotonic part of interlayer magnetoresistance in strong magnetic field $\propto \sqrt{B_z}$.

Discussion. The physical origin of the decrease of the mean interlayer conductivity $\bar{\sigma}_{zz}$ can be understood as follows. The 2D-electrons in magnetic field are much stronger affected by the impurity potential than in 3D: they become localized, and the energy of each localized electron state m is shifted by the energy $W(m) \sim N_i V_0$. This energy shift depends on the electron state m and on the conducting layer j. Therefore, when the electron tunnels between two conducting layers, the energy of the initial and final states are different, which decreases the interlayer conductivity.

The large increase of the effective imaginary part of the electron self energy $|\mathrm{Im}\Sigma(\mu, B)|$ as compared to Γ_0 in the limit $\hbar\omega_c \gg \Gamma_0, t_z$, resulting to the decrease of the interlayer conductivity according to Eq. (25), can also be obtained by the following qualitative arguments. The average difference $\Delta W(m)$ of the energy shifts of two localized electron states is determined by the fluctuation of the number of impurities effectively interacting with the localized electron. This number is approximately $N_i/N_{LL} = c_i > 1$, and the typical fluctuation of this number is $\sim \sqrt{c_i}$. The average difference of the energy shift between two localized states is $\Gamma_W \approx \sqrt{\left\langle (\Delta W)^2 \right\rangle} \sim W/\sqrt{c_i} \sim \sqrt{\Gamma_0 \hbar\omega_c}$ serves as an effective $|\mathrm{Im}\Sigma|$ in Eq. (25). Indeed, the fluctuating shift of the electron energy is equivalent to the coordinate dependent $\mathrm{Re}\Sigma(\boldsymbol{r})$ in the electron Green's function in Eq. (10). The averaging of the electron Green's function over impurity configurations is then similar to the integration over $\mathrm{Re}\Sigma(\boldsymbol{r})$ with distribution of the width Γ_W. For the Lorentzian distribution of the energy shift W

$$D(W) = \Gamma_W/\pi \left[(W - \langle W \rangle)^2 + \Gamma_W^2 \right] \qquad (26)$$

this immediately gives the imaginary part $\Gamma_W \sim \sim \sqrt{\Gamma_0 \hbar\omega_c}$ of the electron Green's function:

$$\langle G_R(\varepsilon, n) \rangle = \int \frac{dW\, D(W)}{E - W - \varepsilon_{2D}(n) - i\Gamma_0} = \\ = \frac{1}{E - \langle W \rangle - \varepsilon_{2D}(n) - i(\Gamma_0 + \Gamma_W)}. \qquad (27)$$

In the Green's function in Eq. (16), obtained in the self-consistent single-site approximation [34], this averaging over the energy shifts of localized electron states is hidden, and the resulting value of $|\mathrm{Im}\Sigma| \sim \Gamma_W \gg \Gamma_0$ in the interval $E_1 < E < E_2$ where the DoS is nonzero.

Eq. (24) gives the decrease of the monotonic part of conductivity $\bar{\sigma}_{zz} \propto B_z^{-1/2}$. It has a singularity at $B = 0$ because it is derived in the limit of strong magnetic field when $\hbar\omega_c \gg \Gamma_0$. In the crossover region $\hbar\omega_c \sim \Gamma_0 > t_z$ the above arguments remain qualitatively valid, but the quantitative dependence $\bar{\sigma}_{zz}(B)$ requires additional calculation.

In the calculation we assumed the normalized impurity concentration $c_i > 1$, because the numerous weak defects and the impurities, situated far from the conducting layers, are important for the lifting of LL-degeneracy in all layered materials [39]. Therefore, $c_i > 1$ even in the strongest pulsed magnets with $B \sim 100T$.

We do not go beyond the self-consistent single-site approximation [34] in studying the influence of the impurity potential, because further corrections give only the small tails to the DoS distribution [35–37, 40]. Hence, these corrections do not change the main result. We also disregard the electron-electron interactions, which restricts our study to the limit when the Fermi energy is much greater than the cyclotron energy, so that many Landau levels are occupied. The chemical potential oscillations [43, 44] are also neglected for two reasons: (i) they do not considerably affect the nonoscillating part of conductivity and (ii) they are strongly damped (almost cancelled) by the MQO of the sample volume [45]. This magnetostriction was directly observed in beryllium [45]. No chemical potential oscillations are observed also in very anisotropic layered organic metals β''-$(\mathrm{BEDT\text{-}TTF})_2\mathrm{SF}_5\mathrm{CH}_2\mathrm{CF}_2\mathrm{SO}_3$ [46].

To summarize, we calculate the interlayer conductivity in strong magnetic field in very anisotropic quasi-two-dimensional metals. The calculation is performed in the framework of the coherent tunnelling model, given by the Hamiltonian in Eqs. (2)–(7). In this calculation the impurity scattering is considered in the self-consistent single-site approximation, which is much more accurate for layered almost 2D-metals than the traditionally used Born approximation. This allows to obtain the new qualitative effect: the strong growth of interlayer magnetoresistance with the increase of magnetic field along conductivity and perpendicular to the conducting layers (see Eq. (24)). This result may explain the numerous experimental observations in strongly anisotropic layered organic metals [18–24], where the interlayer conductivity strongly decreases with the increase of magnetic field along conductivity in contrast to the standard theory [2, 3, 14, 30].

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