Angular dependence of magnetoresistance in strongly anisotropic quasi-two-dimensional metals for various Landau-level shapes

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We present the quantum-mechanical calculations of the angular dependence of interlayer conductivity \( \sigma_{zz}(\theta) \) in a tilted magnetic field in quasi-2D layered metals. Our calculation shows that the LL shape is important for this angular dependence. In particular, the amplitude of angular magnetoresistance oscillations (AMRO) is much stronger for the Gaussian LL shape than for the Lorentzian. The ratio \( \sigma_{zz}(\theta = 0)/\sigma_{zz}(\theta \to \pm 90^\circ) \) is also several times larger for the Gaussian LL shape. AMRO and Zeeman energy splitting lead to a spin current. For typical organic metals and for a medium magnetic field 10T this spin current is only a few percent of the charge current, but its value may almost reach the charge current for special tilt angles of magnetic field. The spin current has strong angular oscillations, which are phase-shifted as compared to the usual AMRO.

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I. INTRODUCTION

The angular magnetoresistance oscillations (AMRO) is a prominent feature of strongly anisotropic layered conductors, which gives an important information about their electronic properties (see, e.g.,12 for reviews). AMRO are actively used to investigate various layered compounds, including organic metals1–7 high-temperature cuprate superconductors,8–12 heterostructures13 etc.

AMRO were first observed14 in 1988 in a quasi-2D strongly anisotropic organic metal \( \beta \)-BEDT-TTF\(_2\)IBr\(_2\). The first explanation of AMRO appeared next year15 and used the geometrical arguments for the Fermi surface of the corrugated-cylinder shape, which corresponds to strongly anisotropic electron dispersion

\[
\epsilon_{3D}(k) \approx \epsilon_{2D}(k_x,k_y) - 2t_z \cos(k_z d),
\]

where \( h k_z \) is out-of-plane electron momentum, \( h \) is the Planck’s constant, \( d \) is the interlayer spacing, and the interlayer transfer integral \( t_z \) is much less than the Fermi energy \( E_F \). For the quadratic and isotropic in-plane electron dispersion \( \epsilon_{2D}(k_x,k_y) = h^2 (k_x^2 + k_y^2)/2m^* \) Yamaji obtained15 that the minima of interlayer conductivity \( \sigma_{zz}(\theta) \) correspond to the zeros of \( J_0(\kappa) \), where \( J_0 \) is the Bessel function of the zeroth order, \( \kappa \equiv k_F d \tan \theta \), \( k_F \) is the in-plane Fermi momentum, and \( \theta \) is the angle between the applied magnetic field \( \mathbf{B} \) and the normal to the conducting planes. The direct calculation of interlayer conductivity, using the electron dispersion in Eq. (1) and the Boltzmann transport equation in the \( \tau \)-approximation, gives16

\[
\sigma_{zz} = \sigma_{zz}^0 \left\{ J_0(\kappa) \right\}^2 + 2 \sum_{\nu = 1}^{\infty} \frac{\left\{ J_{2\nu}(\kappa) \right\}^2}{1 + (\nu \omega_c \tau)^2},
\]

where the cyclotron frequency \( \omega_c = eB_z/m^*c \), \( \tau \) is the mean free time, and the interlayer conductivity without magnetic field

\[
\sigma_{zz}^0 = e^2 \rho_F \langle v_z^2 \rangle \tau = 2e^2 t_z^2 m^* \tau d/\pi h^4,
\]

where \( \rho_F = m^*/\pi h^2 d \) is the 3D DoS at the Fermi level in the absence of magnetic field per two spin components, and the mean square interlayer electron velocity \( \langle v_z^2 \rangle = 2t_z^2 d^2/h^2 \). Here \( e \) is the electron charge, \( m^* \) is the effective electron mass, \( B_z \) is the component of magnetic field perpendicular to conducting layers, and \( c \) is the light velocity. Eq. (2) agrees with the result of Yamaji at \( \omega_c \tau \to \infty \). A microscopical calculation17 of quasi-2D AMRO also gives Eq. (2) when the number of filled Landau levels (LLs) \( n^F_{LL} \gg 1 \).

The calculations of AMRO in Refs.15–21 assume a well-defined 3D electron dispersion \( \langle 1 \rangle \), i.e. that the LL separation \( h\omega_c \) and broadening \( \Gamma = h/2\tau \) are much less than \( t_z \). The inverse “weakly incoherent” limit \( t_z \ll \Gamma \) with the momentum-conserving “coherent” interlayer hopping was also considered.18 The interlayer conductivity was calculated as a tunnelling conductivity between two adjacent conducting layers with short-range disorder in magnetic field, which again resulted to Eq. (2). The calculation in Refs.17,22 is performed under the assumption that the electron self-energy \( \text{Im} \Sigma = \Gamma_0 \) is independent of energy and magnetic field. This assumption, being almost equivalent to the \( \tau \)-approximation, is incorrect in 2D or strongly anisotropic quasi-2D layered compounds with \( t_z, \Gamma_0 \ll \hbar \omega_c \), i.e. in the presence of strong magnetic quantum oscillations (MQO).23–31 Even if MQO are suppressed by temperature or long-range disorder, that smear the Fermi distribution function \( \langle 1 \rangle \). The longitudinal interlayer magnetoresistance \( R_{zz}(B_z) = 1/\sigma_{zz} \) and of the LL broadening \( \Gamma = \Gamma(B_z) \), which changes the angular dependence \( R_{zz}(\theta) \) in Ref.22 a simple amendment to Eq. (2) in the limit \( t_z \ll \Gamma \) was proposed (see Eq. (36) of Ref.22), which includes the renormalization of the prefactor,

\[
\sigma_{zz}^0 \to \sigma_{zz}^0(B_z) \approx \sigma_{zz}^0 \left[ 1 + (2\omega_c \tau)^2 \right]^{-1/4},
\]

and the similar renormalization of the effective mean scat-
tering time in Eq. \(2\),
\[
\tau \rightarrow \tau (B_z) \approx \tau \left[1 + \left(2\omega_c \tau \right)^2 \right]^{-1/4}.
\] (5)

However, this modification of Eq. \(2\) has also several drawbacks. First, it disregards the additional "quantum" term, coming from MQO. This term was first obtained in Refs.\(^{23,24}\) for magnetic field perpendicular to the conducting layers. For tilted magnetic field this "quantum" term is given by the second term in the curly brackets in Eq. \(6\) below or in Eq. \(29\) of Ref.\(^{22}\). Second, Eq. \(2\) is derived for the Lorentzian Landau level (LL) shape while the actual LL shape in strong magnetic field for strongly anisotropic quasi-2D layered metals with \(t_z \ll \hbar \omega_c\) is closer to Gaussian.\(^{22}\) The aim of this paper is to perform a more rigorous calculation of the angular dependence of interlayer magnetoresistance in strongly anisotropic quasi-2D metals, which includes the contribution from MQO and is applicable for various LL shapes.

Eq. \(2\) gives the dependence only on the polar tilt angle \(\theta\) of magnetic field, because it assumes an isotropic in-plane dispersion \(\epsilon_{2D}(k_x, k_y)\). Its generalization for the anisotropic in-plane dispersion also within the \(\tau\)-approximation was considered analytically in Refs.\(^{16,22}\) and numerically in Refs.\(^{22,24}\), which also gives the azimuthal-angle dependence of MR.

II. CALCULATION

To calculate the interlayer conductivity \(\sigma_{zz}\), we use the same two-layer model as in Refs.\(^{22,29,30}\). We consider only two adjacent conducting layers with short-range impurities and with a coherent interlayer electron hopping, which conserves the in-plane electron momentum (see Eqs. \(8\)-(11) of Ref.\(^{22}\)). This model is applicable when the interlayer transfer integral \(t_z\) is less than the LL broadening.\(^{22}\) The interlayer conductivity can be calculated using the Kubo formula, which is valid when the interlayer transfer integral \(t_z\) is less than the LL broadening.

\[
\sigma_{zz} = \frac{e^2}{\hbar} \frac{2d}{2\zeta} \sum_{s=\pm 1} \int d\varepsilon \left[-n'_F(\varepsilon)\right] \times \left\{\left|G_{R}(r, \varepsilon)\right|^2 \cos(\theta) - \text{Re} \left[\left(G_{R}(r, \varepsilon)\right) \left(G_{R}(r, -\varepsilon)\right) e^{iqy}\right]\right\}.
\] (6)

Here \(n'_F(\varepsilon) = -1/\{4T \cosh^2 [(\varepsilon - \mu)/2T] \}\) is the derivative of the Fermi distribution function, \(\mu\) is the chemical potential, \(r \equiv (x, y)\), \(q \equiv e Bd \sin \theta/\hbar c\) and \(G_{R}(r, \varepsilon) = \langle G_{H}(r_1, r_1 + r, \varepsilon) \rangle\) is the retarded electron Green’s function as function of the coordinate and energy, averaged over impurity positions. The impurity averaging of each Green’s function in Eq. \(9\) can be performed separately, because the vertex corrections have the next order of smallness in the parameter \(t_z\) and because the impurities are short-range. For short-range impurities in the non-crossing approximation the averaged electron Green’s function is given by\(^{24,29}\)

\[
G(r_1, r_2, \varepsilon) = \sum_{n, k_y} \Psi^*_{n, k_y}(r_2) \Psi_{n, k_y}(r_1) G(\varepsilon, n),
\] (7)

where \(n\) is the LL number,

\[
G(\varepsilon, n) = \frac{1}{\varepsilon - \epsilon_{2D}(n) - \Sigma(\varepsilon)},
\] (8)

and \(\epsilon_{2D}(n) \equiv \epsilon_n = \hbar \omega_c (n + 1/2)\). In the noncrossing approximation the electron self-energy \(\Sigma(\varepsilon)\) depends only on energy \(\varepsilon\) (see Appendix of Ref.\(^{22}\)), being a periodic function with the period \(\hbar \omega_c\). Moreover, Eq. \(9\) contains the bare electron wave functions

\[
\Psi_{n, k_y}(r) = \Psi_n(x - l^2_{H} k_y) \exp(ik_y y),
\] (9)

where

\[
\Psi_n(x) = \frac{\exp \left(-x^2/2l^2_{H}\right)}{(\pi l^2_{H})^{1/4} 2n/\sqrt{\pi}}.
\]

\(H_n(x/l_H)\) is the Hermite polynomial and \(l_H = \sqrt{\hbar c/e B_z}\) is the magnetic length. This considerably simplifies the calculation. For the retarded and advanced Green’s functions the sign of \(\text{Im} \Sigma\) is fixed. In strong magnetic field \(\hbar \omega_c \gg \Gamma_0\) the electron Green’s function \(G(\varepsilon, n)\) can be calculated restricting to only one LL at \(\varepsilon \approx \epsilon_{2D}(n)\), which in the noncrossing approximation gives a dome-like rather than Lorentzian LL shape.\(^{22}\) The inclusion of diagrams with the intersection of impurity lines adds the exponential tails in the electron density of states (DoS) \(\rho(\varepsilon) = -\text{Im} G(\varepsilon, n)/\pi\) for each LL.\(^{22}\) As the angular dependence of MR depends on the LL shape, we first calculate Eq. \(9\) without restriction to any particular form of the electron Green’s function, and then compare the results for various LL shapes.

Now we substitute the electron Green’s function from Eq. \(9\) to Eq. \(9\). The first term in curly brackets, coinciding with Eq. \(50\) of Ref.\(^{22}\) and responsible for the so-called “classical” \(G_{R}G_{A}\) part of conductivity \(\sigma_{zz}\), rewrites as (see Appendix I)

\[
\mathcal{Cl} = \int d^2r |G(r, \varepsilon)|^2 \cos (qy) \times \sum_{n, p \in \mathbb{Z}} \text{[Re} G(\varepsilon, n) \text{Re} G(\varepsilon, n + p)] + \text{Im} G(\varepsilon, n) \text{Im} G(\varepsilon, n + p)] Z(n, p),
\]

where the LL degeneracy per unit area \(g_{LL} = 1/2\pi l^2_{H} = eB_z/2\pi \hbar c\),

\[
Z(n, p) = \exp \left(\frac{(ql_H)^2}{2}\right) \left(\frac{(ql_H)^2}{2}\right)^p \times L^p_n \left(\frac{(ql_H)^2}{2}\right)^n \left(n^2 + p^2\right)^{n/2},
\]

and \(L^p_n (x)\) is the Laguerre polynomial. The second term in curly brackets in Eq. \(9\), which is absent in Refs.\(^{10,22}\).
and responsible for the so-called "quantum" part of conductivity $\sigma_{zz}$, rewrites as (see Appendix II)

$$Q \equiv \text{Re} \left[ \int d^2r \left< G_R(r, \varepsilon) \right>^2 \exp (i g y) \right]$$

$$= g_{LL} \sum_{n,p \in Z} [\text{Re} G(\varepsilon, n) \text{Re} G(\varepsilon, n + p) - \text{Im} G(\varepsilon, n) \text{Im} G(\varepsilon, n + p)] Z(n, p).$$

Interlayer conductivity

$$\sigma_{zz}(T) = \frac{1}{2} \sum_{s=\pm 1} \int d\varepsilon \left[ -n'_F(\varepsilon) \right] \sigma_{zz}(\varepsilon),$$

where

$$\sigma_{zz}(\varepsilon) = (C I - Q) 2e^2 t_z^2 d/\pi \hbar \equiv \sigma_{zz}^0,$$

and the interlayer conductivity in the absence of magnetic field $\sigma_{zz}^0 = 2e^2 t_z^2 d/\pi \hbar \approx 2e^2 g_{LL} t_z^2 d/\hbar^2 \omega_c \Gamma_0$.

When many Landau levels (LL) are filled, i.e., at $n \sim n_{LL}^F = [\mu/\hbar \omega_c] \gg 1$, one can use the asymptotics of Laguerre polynomials,

$$L_n^a(z) \approx \frac{\Gamma(\alpha + n + 1)}{n!} \left( \frac{\alpha + 1}{2} \right)^n \left( \frac{\alpha + 1}{2} \right) z^{-\frac{n}{2}} \times \exp \left( \frac{z}{2} \right) J_\alpha \left( 2 \sqrt{\left( n + \frac{\alpha + 1}{2} \right) z} \right),$$

which gives at $0 \leq p \ll n$

$$Z(n, p) \approx J_p^2 (\sqrt{2n + 1} q_H).$$

Eq. (18) can be further simplified using $\sqrt{2n_{LL}^F + 1} q_H \approx k_F d \tan \theta$ and that $Z(n, p)$ has a weak dependence on $n$:

$$Z(n, p) \approx Z(n_{LL}^F, p) \approx J_p^2 (k_F d \tan \theta).$$

It is often convenient to cast the sum over LL number $n$ into a sum over harmonics using the Poisson summation formula:

$$\sum_{0}^{\infty} f(n) = \int_{0}^{\infty} f(n)dn + \sum_{k=-\infty}^{\infty} \int_{0}^{\infty} f(n) \exp(2\pi i k n) dn.$$

One can show that the zero-harmonic $k = 0$ of the quantum part $Q$ of interlayer conductivity $\sigma_{zz}$ in Eq. (13) is almost zero, provided the dependence on $n$ of $Z(n, p)$ is much weaker than the dependence of $G(\varepsilon_F, n)$, which is valid for $n \gg 1$. Then $Z(n, p) \approx J_p^2 (k_F d \tan \theta)$ can be factored out from the sum over $n$ in Eq. (13). Substituting Eq. (8) to Eq. (13) and applying the Poisson summation formula (20) we obtain for $k = 0$:

$$\bar{Q} \approx \frac{1}{2} \sum_{p \in Z} J_p^2 (k_F d \tan \theta) \left( \frac{\omega_c}{\hbar} \right)^2 \times$$

$$\int_{-\infty}^{\infty} dn \left[ (n - u)(n + p - u - v)^2 \right] \left( (n + p - u + v)^2 + v^2 \right) = 0,$$

where $u \equiv [\varepsilon - \text{Re} \Sigma(\varepsilon)]/\hbar \omega_c - 1/2$ and $v \equiv \text{Im} \Sigma(\varepsilon)/\hbar \omega_c$. The integral over $n$ in Eq. (21) is zero for each $p$, because the residues in the poles at $n = u + iv$ and $n = u - p + iv$ cancel each other for each $p \neq 0$, while at $p = 0$ the residue is zero, which can be checked by a direct calculation. Hence, $\bar{Q} \approx 0$. One can show, taking the dependence $Z(n, p)$ into account, that $Q$ is smaller than the classical part $Cl$ by a factor $\sim p d Z(n, p)/dn \sim p/n \ll 1$. Note that this statement does not depend on the LL shape, because Eq. (21) is valid for arbitrary $\Sigma(\varepsilon)$.

### III. RESULTS AND DISCUSSION

In a strong magnetic field, $\hbar \omega_c \gg \Gamma$, the details of AMRO essentially depend on the LL shape, determined by the Green’s function $G(\varepsilon, n)$. Therefore, below we consider the Lorentzian and Gaussian LL shapes separately and compare the results.

#### A. AMRO for different LL shapes

For Lorentzian LL shape $\text{Im} \Sigma(\varepsilon) = \Gamma = \Gamma(B)$ in Eq. (8) is independent of $\varepsilon$. This approximation is equivalent to that in Ref. 29, and for the monotonic part $\sigma_{zz}^L$ of interlayer conductivity one confirms Eq. (2) with the renormalized $\sigma_{zz}^0$ and $\tau = \hbar/2\Gamma$ according to Eqs. (1) and (5). The calculation of AMRO in the presence of MQO for the Lorentzian LL shape can be performed rather simply (see Appendix III). Combining Eqs. (14), (15) and (C2)-(C4) one obtains

$$\frac{\sigma_{zz}^L}{\sigma_{zz}^0} = \frac{\Gamma_0}{\Gamma_0} \sum_{p=\pm 1}^{\infty} \int d\varepsilon \left[ -n'_F(\varepsilon) \right] \sum_{p=\pm 1}^{\infty} A_p \left( \frac{J_p(\kappa)}{1 + (p \omega_c \tau)^2} \right)^2,$$

where $A_p$ and $A_p$ are given by Eqs. (C3) and (C4). Substituting Eq. (20) to Eq. (14), (15) and performing the standard integration over $\varepsilon$ one can express the result for the angular dependence of $\sigma_{zz}^L$ in the presence of MQO as a harmonic series:

$$\frac{\sigma_{zz}^L}{\sigma_{zz}^0} = \frac{\Gamma_0}{\Gamma_0} \sum_{k=-\infty}^{\infty} (-1)^k \exp \left( \frac{2\pi ikF}{\hbar \omega_c} \right) R_D(k) R_T(k) \times R_S(k) \left[ \frac{J_0(\kappa)}{\omega_c \tau} \right]^2 + \sum_{p=1}^{\infty} \frac{2 \left[ J_p(\kappa) \right]^2}{1 + (p \omega_c \tau)^2},$$

where the Dingle factor

$$R_D(k) = \exp \left( \frac{-\pi k}{\omega_c \tau} \right),$$

the temperature damping factor

$$R_T = \frac{2\pi^2 k_B T}{\sinh (2\pi^2 k_B T/\hbar \omega_c)}$$

and the spin factor is given by

$$R_S(k) = \cos \left( \frac{\pi g k}{2m_e \cos \theta} \right),$$
where $m^*$ and $m_e$ are the effective and free electron masses and the g-factor $g \approx 2$ unless the spin-orbit or e-e interaction is strong. The spin factor in Eq. (23) also has a strong angular dependence, giving the so-called spin-zeros angular dependence of MQO amplitudes. The angular dependence of MQO amplitudes is given by a product of two factors in the second line of Eq. (23): $R_S(k)$ and the AMRO factor in the curly brackets. Hence, the traditional fitting of the experimentally observed angular dependence of MQO amplitudes by the spin-zero factor only is not correct. The extra factor $(1 + \pi k/\omega_c \tau)$, multiplying $[J_0(\kappa)]^2$ in the second line of Eq. (23), enhances the AMRO amplitude of MQO as compared to the AMRO of monotonous part of MR. At $\omega_c \tau \lesssim 1$ this extra factor $(1 + \pi k/\omega_c \tau) \gg 1$, and even the ratio $\sigma_{zz}^f/\sigma_{zz}^L$ of the oscillating and monotonous parts of conductivity has more complicated angular dependence than just given by the spin factor $R_S$.

In 2D and strongly anisotropic quasi-2D layered compounds in strong magnetic field, when $\omega_c \tau \gg 1$, the LL shape is not Lorentzian. For a physically reasonable white-noise or Gaussian correlator of the disorder potential $U(r)$, $(U(0)U(r)) \propto \exp(-r^2/2d^2)$, theory predicts the Gaussian LL shape of the Landau levels (for reviews see, e.g., Refs. 42 and 43):

$$|\text{Im}G(\epsilon, n)| = \left(\frac{\sqrt{\pi}}{\Gamma}\right) \exp\left[-(\epsilon - \epsilon_n)^2/\Gamma^2\right].$$

(25)

At $\hbar\omega_c \gg \Gamma, T$, when the LLs have Gaussian shape, only few LLs at the Fermi level contribute to conductivity, because $|\text{Im}G(\epsilon = \mu, n)|$ is negligibly small for any $|n - n^F_{LL}|, |p| \geq 2$. Then, in the sum over $p$ and $n$ in Eq. (16) one may keep only three LLs: $n = n^F_{LL}$ or $n = n^F_{LL} \pm 1$, and $p = 0, \pm 1$:

$$\frac{I_1}{\Gamma_0 \hbar \omega_c} = \frac{2}{\pi} \sum_{n - n^F_{LL}, p = 0, \pm 1} Z(n, p) \text{Im}G(\epsilon, n) \text{Im}G(\epsilon, n + p).$$

(26)

The LL with $n = n^F_{LL} \pm 1$ also contain a small factor $|\text{Im}G(\mu, n \pm 1)|$ at $\hbar \omega_c \gg \Gamma, T$. However, they cannot be completely neglected. First, the terms $p \neq 0$ are responsible for the damping of AMRO. Without these terms the interlayer conductivity, given by Eqs. (13) - (16), would be strictly zero in the Yamaji angles. Second, when $\epsilon/\hbar \omega_c$ is integer, $\text{Im}G(\epsilon, n^F_{LL} + 1) = \text{Im}G(\epsilon, n^F_{LL})$, and the terms $n = n^F_{LL}, p = 1$ and $n = n^F_{LL} + 1, p = 0, -1$ give the same contribution as the term $n = n^F_{LL}, p = 0$. At higher temperature $T > \hbar \omega_c$ one has to keep several terms in the sum over $n$ but not over $p$ in Eq. (16), which at $\mu \gg T \gg \hbar \omega_c$ only very slightly affects AMRO.

In Figs. 1 and 2 we plot the calculated AMRO for the Lorentzian and Gaussian LL shapes for four different values of $\Gamma_0 = h/2\tau$: $0.5K, 1.5K, 3.0K$ and $5.0K$, corresponding to $\omega_0 \tau = 10, 3.3, 1.67$ and 1.0 respectively at $\theta = 0$. Comparison of Figs. 1 and 2 shows that the same value of $\Gamma_0$ suppresses AMRO much stronger for the Lorentzian LL shape than for the Gaussian. In particular, at finite $\Gamma \lesssim \hbar \omega_c$ the minima of conductivity at the Yamaji angles are much deeper for the Gaussian LL shape than for Lorentzian. Neglecting this may lead to the incorrect determination of $\omega_c \tau$ from the experimental data on AMRO amplitude.

At $\Gamma \ll \hbar \omega_c$, Eq. (26) gives exponentially small values of $\sigma_{zz}^G \sim \sigma_{zz}^L \exp\left[-(\hbar \omega_c/2\Gamma)^2\right]$ in the Yamaji angles. Besides finite LL broadening $\Gamma$, MR in the Yamaji maxima is limited by the additional “incoherent” mechanisms of interlayer transport, such as the interlayer hopping via resonance impurities and dislocations, or the boson-assisted tunneling. Approximately, the contribution of the incoherent channels to $\sigma_{zz}$ does not depend on the tilt angle $\theta$ of magnetic field and gives a constant upward shift of the curves in Figs. 1 and 2.

B. High tilt angle

From Figs. 1 and 2 one observes that not only the AMRO amplitude but also the ratio $\sigma_{zz}(\theta \rightarrow \pm 90^\circ)/\sigma_{zz}(\theta = 0)$ depend on the LL shape: the saturation value of $\sigma_{zz}$ at $\theta \rightarrow \pm 90^\circ$ looks considerably smaller for the Gaussian LL shape than for the Lorentzian.
Lorentzian. However, the calculated absolute values of \( \sigma_{zz}(\theta \to \pm 90^\circ) / \sigma_{zz}^0 \) depend only on \( \omega_0 \tau \cdot k_F d \) but not on the LL shape. These values agree well with Eq. (10) of Ref.\(^{21}\), which predicts

\[
\sigma_{zz}(\theta \to \pm 90^\circ) / \sigma_{zz}^0 = 1 / \left( 1 + (k_F \omega_0 \tau)^2 \right),
\]

where \( \omega_0 = eB_0 / m^* c \). In Ref.\(^{21}\) Eq. (27) was obtained in the \( \tau \)-approximation using the quasi-classical electron trajectories along the well-defined 3D Fermi surface. The \( \tau \)-approximation does not work in strong perpendicular-to-layers magnetic field, but it may work properly when the magnetic field is along the conducting layers so that \( B_z \to 0 \). One can also expect that the LL shape is not important in the limit \( \theta \to \pm 90^\circ \) and \( B_z \to 0 \). To check this, we now calculate \( \sigma_{zz}(\theta \to \pm 90^\circ) / \sigma_{zz}^0 \) for the Lorentzian and Gaussian LL shapes without the use of a 3D FS and of the \( \tau \)-approximation.

At high tilt angle the argument of the Bessel’s functions in Eq. (19) \( \kappa = k_F d \tan \theta \gg 1 \), and one can use its asymptotic expansion, which gives

\[
Z(n, p) \approx \frac{2}{\pi \kappa} \cos^2 \left( \pi - \frac{\pi}{2} - \pi / 4 \right) = \frac{1 + \cos(2\pi - \pi / 2)}{\pi \kappa}. \tag{28}
\]

The square brackets contain a sum of the monotonic and alternating terms as function of \( p \). At \( B_z \to 0 \), when the LL separation \( h\omega_c \ll \Gamma \), the factor \( \text{Im}G(\epsilon, n + p) \) in Eq. (10) depends very weakly on \( p \), and the alternating term gives a negligible contribution to Eq. (10). Substituting only a constant term from Eq. (28) to Eq. (10) gives at \( \theta \to \pm 90^\circ \)

\[
\frac{I_1}{\Gamma_0 h\omega_c} \approx \frac{2}{\pi^2 \kappa} \sum_{n, p \in Z} \text{Im}G(\epsilon, n)\text{Im}G(\epsilon, n + p). \tag{29}
\]

At \( h\omega_c = \hbar eB_0 / m^* c \ll \Gamma \) one can replace the summations over \( n \) and \( p \) by their integrations. For the Lorentzian LL shape this gives

\[
I_1 \approx \int_{-\infty}^{\infty} \frac{dp}{\pi\kappa} \Gamma_0 h\omega_c (2 \pi^2 \kappa) = 2 \Gamma_0 / \kappa h\omega_c = (\omega_0 \tau k_F d)^{-1} \tag{30}
\]

in agreement with Eq. (14) of Ref.\(^{22}\). For Gaussian LL shape at \( \theta \to \pm 90^\circ \) we obtain the same result:

\[
I_1 \approx \frac{2}{\pi \kappa} \int_{-\infty}^{\infty} \frac{dp}{\pi \kappa} \exp \left[ -\left( \frac{\epsilon - \epsilon_n}{\Gamma} \right)^2 \right] \times \int_{-\infty}^{\infty} \frac{dp}{\pi \kappa} \exp \left[ -\left( \frac{\epsilon - \epsilon_{n+p}}{\Gamma} \right)^2 \right] = 2 \Gamma_0 / \kappa h\omega_c. \tag{31}
\]

Thus, the ratio \( \sigma_{zz}(\theta \to \pm 90^\circ) / \sigma_{zz}^0 = (\omega_0 \tau k_F d)^{-1} \) is the same for Lorentzian and Gaussian LL shapes. This result is natural, because when \( \theta \to \pm 90^\circ \) and \( B_z \) is small, so that \( \Gamma \gg h\omega_c \), the LLs are smeared and their shape is not important. However, \( \sigma_{zz}(\theta = 0) \) depends on the LL shape. Substitution of Eqs. (C1) and (25) to Eq. (26), keeping only one term \( n = n_{LL}, p = 0 \), gives that at \( T = 0 \) in the maxima of MQO the value of \( \sigma_{zz}(\theta = 0) \) for the Gaussian LL shape is \( \pi \) times larger than for the Lorentzian for the same \( \Gamma \). Therefore, in Fig. 2 the ratio \( \sigma_{zz}(\theta \to \pm 90^\circ) / \sigma_{zz}(\theta = 0) \) is considerably smaller than in Fig. 1. Since \( k_F d \) is usually known from the AMRO period, and \( \omega_0 \) (determined by the effective mass \( m^* \)) is known from the MQO period, the experimentally obtained ratio \( \sigma_{zz}(\theta \to \pm 90^\circ) / \sigma_{zz}^0 \) provides a tool to determine \( \tau \) with high accuracy.

C. Spin current and the influence of spin on AMRO

The spin current, as a key object of spintronics, attracts a great attention for its present-day and potential applications (see, e.g., Refs.\(^{45,50}\) for reviews). In our system, the non-zero spin current conductivity \( s_{zz} = \sigma_{zz} - \sigma_{zz} \) appears because the electrons with opposite spin orientations give nonequal contributions to \( \sigma_{zz} \). The Fermi energy of spin up and down electrons differs by the Zeeman energy \( g \mu_B B \), giving different phase of MQO, which leads to the MQO of the spin current. The MQO amplitudes of the spin-current conductivity \( s_{zz} \) and of the usual charge conductivity \( \sigma_{zz} \) have completely different angular dependence. For the Lorentzian LL shape and neglecting the Zeeman splitting of the Fermi momentum \( k_F \) in the argument of the Bessel’s functions, the MQO amplitudes of the spin-current are given by the second line of Eq. (27) with the replacement \( R_S(k) \to R_S^{spin}(k) \), instead of Eq. (21) given by

\[
R_S^{spin}(k) = \sin(\pi g k m^*/2m^* \cos \theta). \tag{32}
\]

The spin-current MQO are damped by temperature and disorder, similar to the usual MQO. Hence, the measurement of the spin-current MQO is not simpler than the measurement of the usual MQO, and this measurement is useful only if it gives any additional information about a compound, which cannot be extracted from the MQO of \( \sigma_{zz} \). For example, if the \( g \)-factor cannot be reliably extracted from the spin-zero angles of the usual MQO in the available range of tilt angles, or if these spin-zero angles casually coincide with the Yamaj's angles. The non-sinusoidal shape of MQO and the interplay of MQO and AMRO make the angular dependence of the harmonic amplitudes of both \( \sigma_{zz} \) and \( s_{zz} \) even more complicated than just given by Eqs. (24) and (32). For example, one can observe only minima of the conductivity harmonic amplitudes instead of strict spin zeros, given by Eqs. (24) and (32).

The monotonic part of spin current appears mainly because of the slightly different angular dependence of the contributions to conductivity from electrons with different spin. The difference of Fermi momenta for spin up and down electrons, originating from the Zeeman energy splitting \( g \mu_B H \), leads to the difference \( \delta \kappa \) in the argument of the Bessel’s functions in Eq. (19):

\[
\delta \kappa = g \mu_B B_0 \tan \theta d / \hbar v_F \approx 2 \mu_B B_0 m^* d \tan(\theta) / (\hbar^2 k_F). \tag{33}
\]

The monotonic part \( s_{zz} \) of the spin-current conductivity, determined as the difference between the monotonic parts of conductivities with spin up and down as \( s_{zz} \approx \sigma_{zz}(\kappa + \delta \kappa) - \sigma_{zz}(\kappa) \), for the Lorentzian LL shape in the first
order in $\kappa \ll 1$ is given by

$$\frac{\bar{s}_{zz}}{\sigma_{zz}^0} \approx \left\{ 2J_0 (\kappa)J'_0 (\kappa) + 4 \sum_{\nu=1}^{\infty} \frac{[J_\nu (\kappa) J'_\nu (\kappa)]}{1 + (\nu \omega_c \tau)^2} \right\} \delta \kappa$$

$$= -\delta \kappa \sum_{\nu=-\infty}^{\infty} \frac{J_\nu (\kappa) [J_{\nu+1} (\kappa) - J_{\nu-1} (\kappa)]}{1 + (\nu \omega_c \tau)^2}, \quad (33)$$

where we have applied $2J'_\nu (\kappa) = J_{\nu-1} (\kappa) - J_{\nu+1} (\kappa)$. In a field $B_0 = 10T$ and for the parameters $d = 20A$, $k_F = 0.14 A^{-1}$ and $m^* \approx 2m_e$, corresponding to the organic metal $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$ (see Ref.24), $\delta \kappa \approx 0.1 \tan \theta$ is not negligible. For these parameters, in Fig. 4 we plot the angular dependence of $\bar{s}_{zz}/\sigma_{zz}^0$, calculated without expansion in $\delta \kappa$, i.e. from Eq. (23) for $k = 0$ (neglecting the MQO), for three different values of $\Gamma$, independent of $B_0$ and corresponding to $\omega_c \tau = 10$ (solid green line), 1 (dashed red line) and 0.5 (dotted blue line). We also checked that the first-order expansion in $\delta \kappa \approx 0.1 \tan \theta$, given by Eq. (33), works very well for $|\theta| < 86^\circ$.

In the Yamaji angles $\sigma_{zz} (\theta) / \sigma_{zz}^0 \ll 1$, and the spin current for these angles can be comparable to the charge current, being also considerably smaller than for other angles at $\omega_c \tau \gg 1$. Note, that at $\omega_c \tau \gg 1$ the monotonic part of spin current changes sign in the proximity of the Yamaji angles from $^\circ-$ to $^\circ+$, and it changes its sign back in the extrema of $\sigma_{zz} (\theta)$. In heterostructures the spin current can be considerably larger than shown in Fig. 8, because of a larger value of $\delta \kappa$, which is proportional to the interlayer distance $d$.

IV. CONCLUSIONS

We have presented the quantum-mechanical calculations of the angular dependence of interlayer magneetoresistance in quasi-2D layered metals. The previous calculations of AMRO usually neglected the magnetic quantum oscillation17,22 or even used the semiclassical Boltzmann transport equation in the constant-$\tau$ approximation16,18,20. However, even if MQO are not seen, being damped by temperature or long-range disorder, they strongly influence the interlayer conductivity and its angular dependence in a strong magnetic field, when $h \omega_c \gg t_z, \Gamma$. In the present study we take MQO into account from the beginning and consider the influence of MQO on AMRO. Our calculation is applicable for various shapes of the Landau levels, thus generalizing the calculation in Refs.17,22,29. This is important, because when the interlayer transfer integral $t_z$ is less than the LL separation $h \omega_c$, the LL shape is not Lorentzian24,33,37,42. In addition, we take into account the so-called “quantum term” in the magnetoresistance25,27 also originating from MQO and neglected in the previous studies17,22,29.

Our calculation shows that the LL shape is important for the angular dependence of magnetoresistance. In particular, the AMRO amplitude is much stronger for the Gaussian LL shape than for the Lorentzian (compare Figs. 1 and 2). The ratio $\sigma_{zz} (\theta = 0) / \sigma_{zz} (\theta \to \pm 90^\circ)$ is also several times larger for the Gaussian LL shape. For the Lorentzian LL shape the angular dependence of interlayer conductivity is given by Eqs. (22), (C3), (C4) or by Eqs. (23), (24) which combine MQO and AMRO. For arbitrary LL shape one can use Eqs. (11)–(18) by Eq. (22) or by Eqs. (18) or (19). In the high-field limit one can apply Eq. (20) instead of Eq. (10) to calculate $\sigma_{zz} (\theta)$.

We also estimated the spin current, which appears because of AMRO. For typical parameters of the organic metal $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$ and in the field $B \sim 10T$ the spin current is about 2% of the zero-field charge current (see Fig. 8), but it may almost reach the charge current for special tilt angles of magnetic field. In heterostructures the spin current can be considerably larger. The angular oscillations of the spin current are stronger and shifted by the phase $\sim \pi/2$ as compared to the usual charge-current AMRO.

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Appendix A: Classical part of conductivity

Substituting Eq. (7) to the first line of Eq. (11) one obtains

$$Cl = \int dy_2 dy_1 dx_2 dx_1 \cos [q(y_2 - y_1)]$$

$$\times \sum_{p,n,k_y,k'_y} \Psi^*_{n,k_y} (r_1) \Psi_{n,k_y} (r_2) G (\epsilon, n)$$

$$\times \Psi^*_{n+p,k_y} (r_2) \Psi_{n+p,k_y} (r_1) G^* (\epsilon, n + p),$$

where the wave functions are given by Eqs. (9) and (10). Integration over $y_2, y_1$ (in a unit square) gives:

$$Cl = 4\pi^2 Re \int dx_2 dx_1 \sum_{p,n,k_y,k'_y} \Psi^*_{n+p} (x_2 - l^Z_H k_y)$$

$$\times \Psi_{n+p} (x_1 - l^Z_H k_y) \Psi^*_{n} (x_1 - l^Z_H k'_y) \Psi_{n} (x_2 - l^Z_H k'_y)$$

$$\times G^* (\epsilon, n + p) G (\epsilon, n) \delta (k_y + q - k'_y),$$
Summation over $k_y'$ cancels $\delta$-function. Then we use the identity:

$$
\int_{-\infty}^{\infty} dx e^{-c^2x^2} H_n(a + cx) H_{n+p}(b + cx) = \frac{2\pi \sqrt{\pi} \Gamma(n+p)}{\Gamma(n)} L_n^p(-2ab), \quad 0 \leq p.
$$

(A2)

Using Eqs. (A2) and (10) one may get:

$$
\int_{-\infty}^{\infty} dx \Psi_{n+p}(x - l_H^2k_y)(x - l_H^2(k_y + q)) = (A3)

\exp \left( -\frac{(gl_H)^2}{4} \right) \left( \frac{gl_H}{\sqrt{2}} \right)^p L_n^p \left( \frac{(gl_H)^2}{2} \right) \frac{n!}{(n+p)!}.
$$

The integration over $x_1, x_2$ in Eq. (A1) is performed using Eq. (A3). Then, making the summation over $k_y$, which just gives the LL degeneracy $g_{LL} = 1/2\pi l_H^2 = eB_z/2\pi \hbar c,$ we obtain Eq. (11).

Appendix B: Quantum Part of conductivity

Substituting Eq. (7) to the first line of Eq. (13) gives

$$
Q = \int dy_2 dy_1 dx_2 dx_1 \exp \left[ i q(y_2 - y_1) \right]
$$

\times \sum_{p,n,k_y,k_y'} \Psi_{n,k_y'}(r_1) \Psi_{n,k_y'}(r_2) G(\epsilon, n)

\times \Psi_{n+p,k_y}(r_2) \Psi_{n+p,k_y}(r_1) G(\epsilon, n+p),
$$

which after the substitution of Eq. (9) and integration over $y_1, y_2$ becomes

$$
Q = 4\pi^2 \text{Re} \int dx_2 dx_1 \sum_{p,n,k_y,k_y'} \Psi_{n+p}(x_2 - l_H^2k_y)
$$

\times \Psi_{n+p}(x_1 - l_H^2k_y) \Psi_n(x_1 - l_H^2k_y') \Psi_n(x_2 - l_H^2k_y')

\times G(\epsilon, n+p) G(\epsilon, n) \delta(k_y + q - k_y').
$$

The integration over $x_1, x_2$ is similar to that in Eq. (A1) and can be easily done using Eq. (A3). Summation over $k_y$ gives the LL degeneracy. Performing these integrations we obtain Eq. (13).

Appendix C: Harmonic expansion of interlayer conductivity for the Lorentzian LL shape

For the Lorentzian LL shape one can put $\text{Im} \Sigma(\epsilon) = \Gamma = \text{const}$ in Eq. (8). Then the imaginary part of the electron Green’s function

$$
\text{Im} G(\epsilon, n) = \Gamma / [(\epsilon - \epsilon_n)^2 + \Gamma^2].
$$

(C1)

Substituting this and Eq. (10) to Eq. (10) one obtains

$$
I_1 = \sum_{n,p \in Z} \frac{(2/\pi) \hbar \omega_c \Gamma \Gamma_0 \Gamma^2 |J_p(\kappa)|^2}{[(\epsilon - \epsilon_n + p)^2 + \Gamma^2][(\epsilon - \epsilon_n)^2 + \Gamma^2]}.
$$

(C2)

The low limit of the summation over $n$ in Eq. (C2) can be extended to $-\infty$, because many LL are filled but only few LLs at the Fermi level $E_F$, i.e. with LL number $n \approx E_F/\hbar \omega_c \gg 1$, contribute considerably to conductivity. Eq. (19) and, hence, Eq. (C2) are valid at $n \gg 1$, and $|p| \ll n$. The summation over $n$ in Eq. (C2) can be easily performed using the identities:

$$
A_0 = \sum_{n \in \mathbb{Z}} \frac{(2/\pi) \hbar \omega_c \Gamma^3}{[(\epsilon - \hbar \omega_c(n + n/2))^2 + \Gamma^2]}^2
$$

$$
= \frac{\sinh (2\pi \Gamma / \hbar \omega_c)}{\cosh (2\pi \Gamma / \hbar \omega_c) + \cos (2\pi \epsilon / \hbar \omega_c)}
$$

(C3)

$$
A_p = \sum_{n \in \mathbb{Z}} \frac{(2/\pi) \hbar \omega_c \Gamma^3}{[(\epsilon - \hbar \omega_c(n + n + p/2))^2 + \Gamma^2]}^2
$$

$$
\times 1 \frac{1 + \cos \left( \frac{2\pi \epsilon}{\hbar \omega_c} \right) \cos \left( \frac{2\pi \Gamma}{\hbar \omega_c} \right)}{\cosh (2\pi \Gamma / \hbar \omega_c) + \cosh (2\pi \Gamma / \hbar \omega_c)}.
$$

(C4)

However, we are mainly interested in the monotonic part and in the harmonic expansion of MQO, which can be obtained using the Poisson summation formula (20). The monotonic part of Eq. (C2) is

$$
\tilde{I}_1 = \sum_{p \in \mathbb{Z}} \int_{-\infty}^{\infty} dn \frac{(2/\pi) \hbar \omega_c \Gamma \Gamma_0 \Gamma^2 |J_p(\kappa)|^2}{[(\epsilon - \epsilon_n + p)^2 + \Gamma^2][(\epsilon - \epsilon_n)^2 + \Gamma^2]}.
$$

(C5)

in agreement with Eq. (2) with the renormalized $\sigma_{xy}$ and $\tau = \hbar / 2\Gamma$ according to Eqs. (4) and (5). The harmonic expansion of $I_1$ is

$$
\tilde{I}_1 = \sum_{k,p \in \mathbb{Z}} \int_{-\infty}^{\infty} dn \frac{(2/\pi) \hbar \omega_c \Gamma \Gamma_0 \Gamma^2 |J_p(\kappa)|^2}{[(\epsilon - \epsilon_n + p)^2 + \Gamma^2][(\epsilon - \epsilon_n)^2 + \Gamma^2]} \exp(2\pi i kn).
$$

(C6)

For $p = 0$ the integration over $n$ reduces to

$$
\int_{-\infty}^{\pi/2} \frac{dn \hbar \omega_c \Gamma \exp(2\pi i kn)}{[(\epsilon - \epsilon_n - \epsilon)^2 + \Gamma^2]^2}
$$

$$
= (1 + \frac{2\pi \hbar \Gamma}{\hbar \omega_c}) \exp \left( \frac{2\pi \hbar \Gamma}{\hbar \omega_c} \right).
$$

(C7)

For $p \neq 0$ the integral over $n$ is

$$
\int_{-\infty}^{\infty} \frac{dn \hbar \omega_c \Gamma \exp(2\pi i kn)}{[(\epsilon - \epsilon_n - \epsilon)^2 + \Gamma^2]^2}
$$

$$
= \exp \left( \frac{2\pi \hbar \Gamma}{\hbar \omega_c} \right) \frac{1}{1 + \left( p \hbar \omega_c / 2\Gamma \right)^2}.
$$

(C8)
Substituting Eqs. (C7) and (C8) to Eq. (C6) we obtain

\[ I_1 = \frac{\Gamma_0}{\Gamma} \sum_{k=-\infty}^{\infty} (-1)^k \exp \left( \frac{2\pi i k \tau}{\hbar \omega_c} \right) \exp \left( -\frac{\pi k}{\omega_c \tau} \right) \times \]

\[ \left( |J_0(\kappa)|^2 \left( 1 + \frac{\pi k}{\omega_c \tau} \right) + \sum_{p \in Z} \frac{|J_p(\kappa)|^2}{1 + (p \omega_c \tau)^2} \right) \]

\[ (C9) \]

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