Quantum contribution to magnetotransport in weak magnetic fields and negative longitudinal magnetoresistance

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Longitudinal magnetoresistance (LMR) refers to the change in resistance due to a magnetic field when the current and the magnetic field are parallel to each other. For this to be nonzero in weak magnetic fields, kinetic theory stipulates that the electronic dispersion must satisfy certain conditions: it should either be sufficiently anisotropic or have topological features. The former results in a positive LMR, while the latter results in a negative LMR. Here, I propose a different mechanism that leads to LMR in *any* dispersion without a need to satisfy the above requirements. The mechanism is quantum in origin but is applicable in the said regime. It arises due to the change in the density of states with the magnetic field and is not kinetic in origin. Remarkably, LMR is found to be negative even if the dispersion is nontopological, provided it is nonparabolic. An analytical expression is derived for this contribution to LMR. It is found to depend on the orbital magnetic susceptibility. The analytical findings are confirmed by numerical calculations.

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I. INTRODUCTION

Magnetotransport—the motion of charge carriers in the presence of both an electric and a magnetic field—is one of the most commonly studied phenomena in solids. As a charge accelerates under the electric field, it suffers repeated collisions with scatterers, giving rise to resistance. With the introduction of the magnetic field, the charge now experiences an additional Lorentz force and bends away from its linear path. It spends more time in traversing the direction of the electric field and suffers more collisions, resulting in an increased resistance. Thus, magnetoresistance is expected to be positive and arise only when the current and the magnetic field have components perpendicular to each other [1].

Based on the above kinetic picture which is essentially classical, one does not expect longitudinal magnetoresistance (LMR) to exist because it requires the current to be parallel to the magnetic field; and even if it exists, it should not be negative (N). Nevertheless, in solids where electrons do not have a free-particle dispersion, LMR—and, in some cases, NLMR—can arise within the same basic mechanism at weak fields where kinetic theory is valid. It has been shown that if the dispersion possesses a certain kind of anisotropy such that the velocity of electrons parallel and perpendicular to the magnetic field cannot be decoupled, LMR can be nonzero and is necessarily positive [2–4]. On the other hand, if the dispersion features topological properties, along with the Lorentz force the kinetics is influenced by an additional contribution that arises from the Berry curvature. Then anisotropy is not a necessity and a nonzero LMR can arise. In this case, however, LMR is negative [5–9]. Apart from these two mechanisms, others that lead to LMR—and, in some cases, NLMR—are known to exist. However, these are either extrinsic in origin, such as scenarios that require specific models of scattering [10,11] and inhomogeneities [12–14], or are beyond the semiclassical regime, requiring, for example, a very high magnetic field which forces electrons to occupy only the lowest Landau level [15] or low enough temperatures such that quantum interference effects lead to weak (anti)localization [16].

In this work, I show that there exists another mechanism, intrinsic in origin and applicable in weak fields but not kinetic in nature, that contributes to magnetotransport. This gives rise to a nonzero value of LMR in cases where kinetic theory predicts a zero value, which can even become negative. The mechanism derives from the change of density of states due to the magnetic field and is quantum in origin in spite of appearing in a classical regime. A simple understanding can be obtained by considering a familiar context in which the same mechanism is at play: Landau diamagnetism. It is well known that classically Landau diamagnetism cannot arise since the magnetic field through its kinetic contribution cannot affect the total energy of a system. Quantum mechanically, however, it is allowed since the density of states becomes a function of the magnetic field through the formation of discrete Landau levels. Note that in spite of being quantum in origin, the effect manifests at weak fields such that $\omega_c \ll E_F$, where ω_c is the cyclotron frequency and E_F is the Fermi energy ($\hbar = 1$). Extending this mechanism to transport, magnetoresistance should also inherit a similar contribution, irrespective of the orientation of the current and the magnetic field. Importantly, the orbital magnetic susceptibility, while being diamagnetic for a parabolic dispersion (Landau diamagnetism), becomes paramagnetic when nonparabolicity is introduced in the dispersion [17]. The same can arise in the context of magnetotransport, with magnetoresistance switching sign from positive to negative as the dispersion acquires nonparabolicity. The different physical origins of the

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two contributions to magnetoresistance, kinetic and quantum, are expected to show up in their functional dependence on the magnetic field: the former is expected to be a function of $\omega_c\tau$, where τ is the relevant scattering time, whereas the latter should be a function of $\frac{\omega_c}{E_F}$. Because $\omega_c\tau$ can reach values much larger than one while satisfying $\omega_c \ll E_F$, in general the kinetic contribution will dominate over the quantum contribution. However, if the former is identically zero, the latter can become the leading contribution. As discussed earlier, this could happen with LMR, making the new quantum contribution relevant in this context.

Below I substantiate the above claims with analytical and numerical calculations. LMR is calculated for a dispersion that is separable in directions parallel and perpendicular to the magnetic field using the Kubo formula. The choice of such a dispersion is not necessary, but is done for two reasons: first, it greatly simplifies the calculation, and second, it is known that the kinetic contribution to LMR for such a choice is identically zero [2]; therefore, any LMR found is necessarily of quantum origin. A general expression for the quantum contribution is derived which is found to be intimately related to the orbital magnetic susceptibility. It is explicitly shown that in contrast to conventional wisdom, even a parabolic dispersion exhibits LMR, which becomes negative as nonparabolicity is introduced in the dispersion.

II. MODEL

Consider a metallic system with a dispersion

$$E(\mathbf{k}) = \varepsilon_{xy}(k_x, k_y) + \varepsilon_z(k_z). \tag{1}$$

Without any loss of generality, it is assumed that the minimum value of each term is zero. A magnetic field **B**, described by the vector potential $\mathbf{A} = (0, Bx, 0)$, is applied in the z direction. The dispersion becomes (spin is ignored for simplicity)

$$E(n, k_z) = \varepsilon_{xy}(n) + \varepsilon_z(k_z), \tag{2}$$

where $\varepsilon_{xy}(n)$ denotes the Landau levels in two dimensions. The eigenfunctions are given by

$$\psi_{k_z,k_y,n} = e^{ik_z z + ik_y y}\phi_n\left(x - k_y l_B^2\right), \tag{3}$$

where ϕ_n are the Landau level eigenfunctions corresponding to $\varepsilon_{xy}(n)$ and $l_B^2 = \frac{1}{eB}$. The corresponding single-particle Green's function is

$$G(k_z, k_y, x, x', \omega) = \sum_n \frac{\phi_n^*\left(x - k_y l_B^2\right)\phi_n\left(x' - k_y l_B^2\right)}{\omega - \xi_n(k_z) + \frac{i}{2\tau}sgn(\omega)}, \tag{4}$$

where $\xi_n(k_z) = E(n, k_z) - E_F$. Here, I have included a phenomenological scattering time τ without worrying about its microscopic origin and assumed it to be field independent. This will be revisited later. Throughout this work, it will be assumed that scattering is weak so that $1/\tau \to 0$.

III. LONGITUDINAL MAGNETOCONDUCTIVITY

In calculating the longitudinal magnetoconductivity σ_{zz}, I closely follow Abrikosov [18], who first calculated the same for a parabolic dispersion. Using the Kubo formula,

$$\sigma_{zz}(B) = \mathrm{Re}\frac{e^2}{(2\pi)^3}\int d\omega \frac{n_F(\omega) - n_F(\omega + \Omega)}{\Omega}\int dk_z dk_y dx' v_z(k_z) G^R(k_z, k_y, x, x', \omega) v_z(k_z) G^A(k_z, k_y, x', x, \omega + \Omega). \tag{5}$$

Here, n_F is the Fermi function, $v_z = \frac{\partial E}{\partial k_z} = \frac{\partial \varepsilon_z}{\partial k_z}$, $G^{R(A)}$ is the retarded (advanced) Green's function corresponding to Eq. (4), and Ω is the external frequency. At $T = 0$, $\Omega = 0$, the frequency integral pins all energies on the Fermi surface. Using Eq. (4) in Eq. (5), I have

$$\sigma_{zz}(B) = \frac{e^2}{(2\pi)^3}\sum_{n,n'}\int dk_z dk_y dx' \frac{v_z^2(k_z)}{\xi_n^2(k_z) + \frac{1}{4\tau^2}}\phi_n^*\left(x - k_y l_B^2\right)\phi_n\left(x' - k_y l_B^2\right)\phi_{n'}^*\left(x' - k_y l_B^2\right)\phi_{n'}\left(x - k_y l_B^2\right).$$

Using the fact that the Landau level eigenfunctions form an orthonormal complete basis, the integral over x' gives $\delta_{n,n'}$. Using this, and completing the integral over k_y, I have

$$\sigma_{zz}(B) = \frac{e^2 eB}{(2\pi)^3}\sum_n \int dk_z \frac{v_z^2(k_z)}{\xi_n^2(k_z) + \frac{1}{4\tau^2}}. \tag{6}$$

Next, I make a change of variable: $\int dk_z \to \int \frac{2}{|v_z|} d\xi_n$, where the factor 2 is included since E is an even function of k_z (see comment [19]). In the limit $\frac{1}{\tau} \to 0$, $\frac{1}{\xi_n^2(k_z) + \frac{1}{4\tau^2}} \to 2\tau\pi\delta(\xi_n)$. Thus,

$$\sigma_{zz}(B) = \frac{e^2\tau eB}{2\pi^2}\sum_n |v_{zn}|, \tag{7}$$

where

$$v_{zn} = \left.\frac{\partial \varepsilon_z(k_z)}{\partial k_z}\right|_{k_z = \varepsilon_z^{-1}(E_F - \varepsilon_{xy}(n)) \geqslant 0}. \tag{8}$$

The summation over n runs from 0 to N, the maximum value of n for which $\varepsilon_{xy}(n) \leqslant E_F$. Equation (7) has a simple interpretation. The magnetic field has reduced the three-dimensional spectrum into a set of one-dimensional bands dispersing along k_z, each with a degeneracy proportional to B. The total conductivity is the sum of the velocity in the z direction at the Fermi energy contributed by all the partially occupied bands. As shown in Fig. 1, the number of such bands is given simply by the number of bands E_F crosses—this corresponds to N. At very high magnetic fields, only the lowest band is occupied ($N = 1$), which contributes to transport. This is a purely quantum regime. As the magnetic field decreases, more bands get populated by going below the Fermi level.

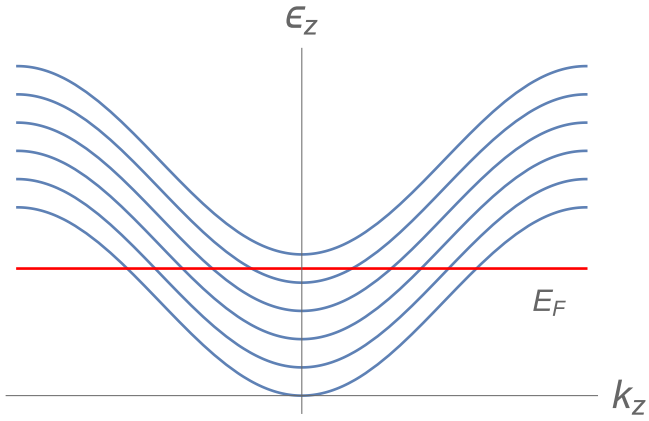

FIG. 1. In a magnetic field, a three-dimensional spectrum is reduced to a set of one-dimensional bands dispersing in the direction of the field. The sum in Eq. (7) is over those bands that intersect the Fermi energy E_F.

When the number of occupied levels is large ($N \gg 1$), one enters the semiclassical regime. In this regime, with change in B, the sum in Eq. (7) changes in two ways: a part that evolves smoothly and another that changes abruptly due to a sudden change from N to $N+1$ each time an extra band gets populated. Together, they give rise to LMR, the former appearing as a smooth background while the latter manifesting as quantum oscillations. Quantum oscillations are vestigial signatures of quantum effects in the semiclassical regime. The fully quantum regime along with quantum oscillations in the semiclassical regime have been extensively studied before by Arbrikosov [18] and others [15,21] as manifestations of quantum effects. However, the smooth background is considered to be purely classical, described by the kinetic theory, devoid of any quantum effects. Below, through explicit calculations, I show that this is not correct: the smooth background contribution to LMR also inherits an intrinsic quantum contribution, hitherto unexplored, with interesting consequences.

As a simple example, consider first a parabolic spectrum: $\varepsilon_{xy}(k_x,k_y)=\frac{k_x^2+k_y^2}{2m}$, giving $\varepsilon_{xy}(n)=(n+\frac{1}{2})\omega_c$, where $\omega_c=\frac{eB}{m}$, and $\varepsilon_z(k_z)=\frac{k_z^2}{2m}$. Then, $v_{zn}=\sqrt{\frac{2}{m}}\sqrt{E_F-(n+\frac{1}{2})\omega_c}$. The summation over n in Eq. (7) can be converted into an integral by using the Euler-MacLaurin formula (see Supplemental Material [22]). Ignoring the oscillating part and keeping only the smooth part up to $O(B^2)$, I find

$$\sigma_{zz}(B)\approx\sigma_{zz}(0)\left[1-\frac{1}{32}\frac{\omega_c^2}{E_F^2}\right], \tag{9}$$

where $\sigma_{zz}(0)=\frac{n_0e^2\tau}{m}$, with $n_0=\frac{(2mE_F)^{3/2}}{6\pi^2}$ being the zero-field charge density. Thus, even for a parabolic spectrum, the longitudinal conductivity is magnetic field dependent. This should be contrasted with the kinetic theory result, which predicts the absence of any field dependence. The field-dependent part scales with $\frac{\omega_c}{E_F}$ instead of $\omega_c\tau$, confirming its quantum origin. The negative sign implies that LMR, obtained by taking the inverse, is positive.

I now generalize the above idea to a general spectrum. The Landau levels $\varepsilon_{xy}(n)$ no longer have a simple analytical form. They are, instead, derived from the semiclassical quantization condition,

$$Sl_B^2=2\pi(n+\gamma), \tag{10}$$

where $S(\varepsilon)$ is the area of the surface enclosed by the isoenergy contour $\varepsilon_{xy}=\varepsilon$ in the two-dimensional k space and γ is the semiclassical phase. It is easy to check that for a parabolic dispersion, $S(\varepsilon)=\pi(k_x^2+k_y^2)|_{\varepsilon_{xy}=\varepsilon}=2\pi m\varepsilon$ and $\gamma=\frac{1}{2}$ reproduce the correct Landau level spectrum $\varepsilon(n)=(n+\frac{1}{2})\omega_c$. When the dispersion is nonparabolic, two changes arise: $S(\varepsilon)$ is no longer the area of a circle and, more importantly, γ is no longer a constant but a function of ε itself. While $S(\varepsilon)$ is a simple geometrical quantity, calculation of $\gamma(\varepsilon)$ requires more care. In the simplest case where singularities in the isoenergy contours and interband effects can be ignored, it was shown by Roth that [20,21]

$$\gamma(\varepsilon)-\frac{1}{2}=\frac{eB}{48\pi}\frac{\partial}{\partial\varepsilon}\int\delta(\varepsilon_{xy}-\varepsilon)\left[m_{xx}^{-1}m_{yy}^{-1}-\left(m_{xy}^{-1}\right)^2\right]d^2k, \tag{11}$$

where $m_{\alpha\beta}^{-1}=\frac{\partial^2\varepsilon_{xy}}{\partial k_\alpha\partial k_\beta}$. This can be written in terms of the two-dimensional orbital magnetic susceptibility χ. According to the Landau-Peierl's formula [20],

$$\chi(\varepsilon)=-\frac{e^2}{24\pi^2}\int\delta(\varepsilon_{xy}-\varepsilon)\left[m_{xx}^{-1}m_{yy}^{-1}-\left(m_{xy}^{-1}\right)^2\right]d^2k. \tag{12}$$

Combining the two,

$$\gamma(\varepsilon)-\frac{1}{2}=-\frac{\pi B}{2e}\frac{\partial\chi}{\partial\varepsilon}. \tag{13}$$

Going back to Eq. (7), the sum is once again computed using the Euler-Maclaurin formula, but keeping in mind that now a change in n is accompanied by changes in both S and γ [21]. Ignoring the oscillating part and keeping only the smooth part up to $O(B^2)$ as before, I find

$$\begin{aligned}\sigma_{zz}(B)&\approx\sigma_{zz}(0)-\left[\frac{\partial|v_z|}{\partial\varepsilon}\chi\Big|_{\varepsilon=0}+\int_0^{E_F}\frac{\partial|v_z|}{\partial\varepsilon}\frac{\partial\chi}{\partial\varepsilon}d\varepsilon\right]\frac{e^2\tau}{4\pi}B^2\\&=\sigma_{zz}(0)-\alpha B^2.\end{aligned} \tag{14}$$

Here, $\frac{\partial|v_z|}{\partial\varepsilon}\equiv\frac{\partial|v_z|}{\partial\varepsilon_{xy}}\Big|_{\varepsilon_{xy}=\varepsilon}$, where v_z is evaluated from Eq. (2) and expressed in terms of ε_{xy} [similar to Eq. (8) but now in (k_x,k_y) space]. The expression for LMR is obtained by inverting Eq. (14): $\rho_{zz}(B)\approx\rho_{zz}(0)+\alpha B^2$, where $\rho_{zz}=\frac{1}{\sigma_{zz}}$. Equation (14) clearly shows that the quantum contribution to LMR in a three-dimensional system is intimately related to the orbital magnetic susceptibility of the corresponding two-dimensional spectrum, confirming their common origin.

A remarkable feature of Eq. (14) is that the two terms constituting the coefficient α need not be of the same sign; therefore, α can pick a sign depending on which term wins. In the parabolic case, $v_z=\frac{k_z}{m}=\sqrt{\frac{2}{m}}\sqrt{E_F-\varepsilon}$ and $\chi=-\frac{e^2}{12\pi m}$ [from Eq. (12)]. The latter is independent of energy, so the second term constituting α drops out and the expression in Eq. (9) is recovered with α positive. However, once the dispersion becomes nonparabolic, the second term becomes nonzero and opposite in sign to the first term. For a sufficiently nonparabolic spectrum, α becomes negative, resulting in NLMR.

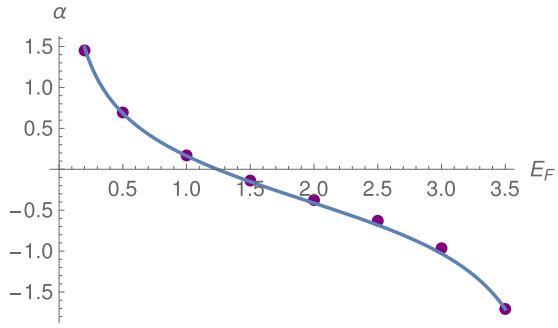

FIG. 2. Dependence of α (in arbitrary units) on E_F (in units of t) in longitudinal conductivity, $\sigma_{zz}(B) = \sigma_{zz}(0) - \alpha B^2$, for the separable spectrum in Eq. (1) with $\varepsilon_{xy} = 4t - 2t[\cos(k_x a) + \cos(k_y a)]$ and $\varepsilon_z = \frac{k_z^2}{2m}$. The solid line is according to the analytical expression in Eq. (14). The solid circles represent numerically calculated values obtained by fitting the curves in Fig. 3 to the quadratic equation above. At small E_F, the spectrum is close to parabolic and α is positive. However, as E_F is increased, the spectrum becomes nonparabolic and α becomes negative—in this regime, LMR is negative.

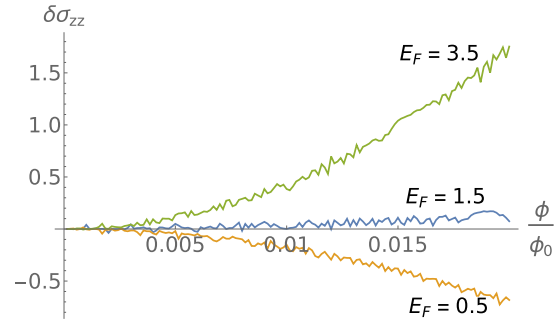

FIG. 3. Dependence of $\delta\sigma_{zz} = \sigma_{zz}(B) - \sigma_{zz}(0)$ (in arbitrary units) on B (expressed in terms of flux over flux quantum) at different values of E_F (in units of t) obtained by evaluating Eq. (7) exactly—see text. The spectrum is the same as in Fig. 2: $\varepsilon_{xy} = 4t - 2t[\cos(k_x a) + \cos(k_y a)]$ and $\varepsilon_z = \frac{k_z^2}{2m}$. A nonzero temperature $T = 0.1t$ has been used to suppress the quantum oscillations. At small values of E_F, σ_{zz} decreases with B, whereas at larger values of E_F, it increases—in this regime, LMR is negative.

Note that for this to happen, it is sufficient to have only the two-dimensional spectrum ε_{xy} nonparabolic; the dispersion along the magnetic field, ε_z, can still be parabolic. To illustrate this, consider the spectrum $\varepsilon_{xy} = 4t - 2t[\cos(k_x a) + \cos(k_y a)]$ and $\varepsilon_z = \frac{k_z^2}{2m}$, where t is the nearest-neighbor hopping parameter on a square lattice of lattice constant a. Using Eq. (12), one finds [17] $\chi(\varepsilon) = \frac{e^2}{12ta^2\pi^2} Q_{1/2}[1 - \frac{(\varepsilon-4)^2}{8}]$, where $Q_n[x]$ is the Legendre function of the second kind and ε is in units of t. Using this in Eq. (14), the integral is calculated to compute α. In Fig. 2, the dependence of α on E_F is plotted. At small E_F, the spectrum is close to parabolic, and α is positive. With increase in E_F, nonparabolicity becomes more pronounced and, at some value, α switches sign and becomes negative, resulting in NLMR. Equation (14) along with its consequences form the main result of this paper.

IV. NUMERICAL CALCULATION

As further proof, I now present an exact numerical evaluation of Eq. (7), which is then compared with the analytical result in Eq. (14). The Landau level spectrum $\varepsilon_{xy}(n)$ corresponding to $\varepsilon_{xy}(k_x, k_y) = 4t - 2t[\cos(k_x a) + \cos(k_y a)]$ is calculated numerically on a lattice model (see Supplemental Material [22] for details). Using Eq. (8), $v_{zn} = \sqrt{\frac{2}{m}}\sqrt{E_F - \varepsilon_{xy}(n)}$ is computed. This is inserted in Eq. (7) and the sum is evaluated numerically as a function of the field. This yields the total $\sigma_{zz}(B)$, which includes both the smooth as well as the oscillating parts. To remove the oscillating part, a small temperature is introduced. Temperature influences the two contributions differently: it introduces a negligible correction $\sim\left(\frac{T}{E_F}\right)^2$ (Sommerfeld correction) in the smooth part, but reduces the oscillating part exponentially as $\sim e^{-T/\omega_c}$ for $T \gg \omega_c$. This is exploited to suppress the oscillating part and reveal the smooth part of $\sigma_{zz}(B)$. Note that this is not just a theoretical trick, but also has experimental relevance: to observe the predicted behavior in the smooth part of LMR, one needs to be in the regime $\omega_c \lesssim T \ll E_F$. The effect of temperature is included by using the formula $\sigma_{zz}(E_F, T) = \int(-\frac{\partial n_F(E-E_F)}{\partial E})\sigma_{zz}(E, 0)dE$. The results are presented in Fig. 3. As expected, $\sigma_{zz}(B)$ varies quadratically with the field. At small values of E_F, it decreases with the field, leading to positive LMR, while at larger values of E_F, it becomes an increasing function of the field, leading to NLMR. The curves are fitted and the coefficient α is extracted. The extracted values of α are plotted in Fig. 2 alongside the analytical curve. It is seen that they are in excellent agreement.

V. EFFECT OF FIELD ON SCATTERING TIME

In arriving at Eq. (7), the scattering time τ was assumed to be a phenomenological constant. In a microscopic theory, τ depends on the density of states and, therefore, should change with the field. More importantly, since τ is inversely proportional to the density of states, one can wonder whether it will kill all the field dependence in σ_{zz} found so far. It turns out that this is not the case. This can be shown explicitly by considering a simple model where δ-function impurities are scattered randomly in a system with a parabolic spectrum. Assuming weak and dilute impurities, within the first Born approximation one finds (see Supplemental Material [22]) $\tau^{-1} = n_i U_0^2 \frac{eB}{\pi} \sum_{n=0}^{N} \frac{1}{|v_{zn}|}$, where U_0 is the Born scattering amplitude and n_i is the density of impurities. Inserting this in Eq. (7), it is clear that a cancellation does not occur. Carrying out the summation over the Landau levels as before (see Supplemental Material [22]), I find $\tau \approx \tau_0\left[1 - \frac{1}{96}\frac{\omega_c^2}{E_F^2}\right]$, where τ_0 is the scattering time in the absence of the field. Using this in Eq. (9), I get $\sigma_{zz}(B) \approx \sigma_{zz}(0)\left[1 - \frac{1}{24}\frac{\omega_c^2}{E_F^2}\right]$. The field dependence in τ, instead of destroying LMR, accentuates it.

VI. CONCLUDING REMARKS

To summarize, I have shown that a nonzero LMR can arise in *any* dispersion in weak magnetic fields, in contrast to the prediction of kinetic theory which states that LMR is nonzero only for dispersions of certain kinds. This arises because a magnetic field affects electronic transport not only kinetically, but also by modifying the density of states. The mechanism is inherently quantum in spite of manifesting in the classically weak-field regime. Importantly, the quantum contribution to LMR can become negative if the dispersion is sufficiently nonparabolic, even if the latter has no topological features. It is found that it is related to the orbital magnetic susceptibility. While the theory presented here considered the simplest case of a single isolated band, it can be extended to include coupled bands. Such extensions are important in the context of topological systems and will be investigated in the future.

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