Angular dependence of magnetoresistance and planar Hall effect in semimetals in strong magnetic fields

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The semiclassical transport theory is especially powerful for investigating galvanomagnetic effects. Generally, the semiclassical theory is applicable only in weak fields because it does not consider Landau quantization. Herein, we extend the conventional semiclassical theory by considering Landau quantization through the field dependence of carrier density in semimetals. Using this approach, we simultaneously explain the qualitative change in the angular dependence of transverse magnetoresistance (TMR), anisotropic magnetoresistance, and planar Hall effect in bismuth with an increase in the magnetic field. We also considered the quantitative applicability of our theory. We found that the field dependence of mobility can result in a qualitative agreement in TMR far beyond the quantum limit ($\gtrsim$10 T).

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I. INTRODUCTION

The galvanomagnetic effect, in which the direction and amplitude of current are changed by a magnetic field, is one of the oldest and most fundamental subjects in solid-state physics. Every textbook of solid-state physics provides its introductory explanation, and some books discuss it in detail [1,2]. Despite it being discovered centuries ago, the galvanomagnetic effect continues to drive novel physics even today. Recently, the galvanomagnetic effect has attracted significant attention in connection with topological condensed-matter physics [3,4]. For example, the possibilities of observing the chiral anomaly in solids are being actively discussed, such as in negative magnetoresistance [3,5–7] and in the planar Hall effect (PHE) [7–16].

The semiclassical transport theory has proved to be very useful in explaining the galvanomagnetic effect [1,4,17]. For example, it could successfully explain the complex galvanomagnetic effect in bismuth, a multivalley semimetal with Dirac electrons [18–22]. The angular dependence of transverse magnetoresistance (TMR), anisotropic magnetoresistance (AMR), and PHE in Bi have been accurately explained by the semiclassical theory for weak fields ($\lesssim$ 1 T) [16,22,23]. However, the angular dependence exhibits an essential transformation by the magnetic field from weak to strong. (The experimental details are provided later in the paper.) The semiclassical theory has failed to explain these field-induced transformations in the angular dependencies [16,23,24]. This leads to a simple question. Does the apparent failure of the semiclassical theory indicate the appearance of novel physics, or imply the limitation of the semiclassical theory?

Regarding the possibility of novel physics, Yang *et al.* argued the possibility that the field-induced transformation in Bi originates from the effect of the chiral anomaly in Weyl and Dirac electrons [16]. However, the same study also pointed out that a phase shift of $\pi/2$ in the bisectrix channel of PHE did not agree with the chiral anomaly model. Typically, the semiclassical theory is not applicable in strong fields, because it does not consider Landau quantization. If this difficulty is overcome, it can reveal the origin of the field-induced transformation in the galvanomagnetic effect in Bi.

This study aims to point out the critical effect of Landau quantization in magnetoresistance (MR); it also aims to clarify the origin of the nontrivial field-induced transformation in the angular dependence of TMR, AMR, and PHE in Bi. We extend the semiclassical transport theory by considering Landau quantization through the field dependence of carrier density, which is crucial for semimetals in strong fields. Using this approach, we provide a critical point of view for MR in the strong-field region. The results of our modified theoretical approach exhibit a field-induced transformation in the angular dependence of TMR, AMR, and PHE, all of which agree well with the experiments.

The electronic structure and Landau quantization of Bi are accurately known owing to a number of previous studies [18,21,22]. There is no ambiguity in the electric structure. Therefore, Bi provides an ideal environment for our purpose.

Before we discuss the theory, let us summarize the experimental results on the angular dependence of TMR, AMR, and PHE in Bi. TMR in Bi exhibits a large angular dependence, where the magnetic field is rotated in the binary-bisectrix plane with the current along the *trigonal* axis [Fig. 1(a)]. The angular dependence is sufficiently large to be observed at 300 K [25]. This highly anisotropic TMR originates from the extremely high mobility of Bi ($\mu \sim 10^8$ cm^2/V s) [26]. The TMR is anisotropic only in weak fields ($\lesssim$1 T) and the anisotropy disappears in strong fields ($\gtrsim$10 T), i.e., the TMR becomes nearly isotropic [23,24]. This is rather surprising because the anisotropy should be enhanced by the magnetic field. (Note that the loss of threefold symmetry in TMR has been observed in strong fields [23,25]; however, here we

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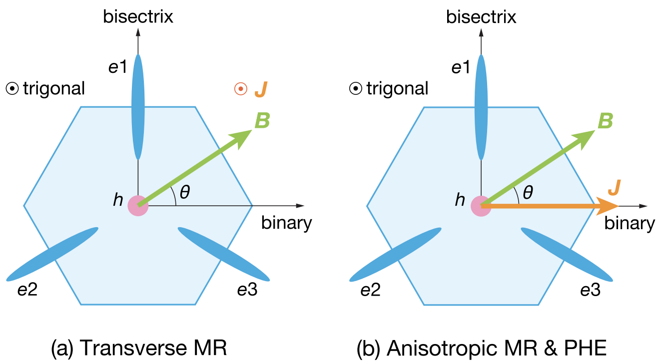

FIG. 1. Configuration of the current $\boldsymbol{J}$ and magnetic field $\boldsymbol{B}$ for (a) TMR and (b) AMR and PHE. One circle and three ellipsoids express the hole and the electron pockets of the Fermi surface in Bi, respectively.

concentrate on the behavior of a large component that holds the threefold symmetry.) Remarkable amplitudes of AMR and PHE are observed with the current along the *binary* axis by rotating the magnetic field in the same plane as that of TMR [Fig. 1(b)] [11,16]. In weak fields ($\lesssim$1 T), both AMR and PHE have two components of angular oscillation with period $\pi/2$ and π. In strong fields ($\gtrsim$10 T), however, the angular oscillation with period $\pi/2$ disappears. We primarily address these nontrivial field-induced phenomena in this study.

II. THEORY

Each part of the theory—the Landau levels and carrier density of Bi [27–29], and the semiclassical theory for multivalley systems [30–32]—has been already well established in previous studies. By combining these parts, we can examine the effect of field-dependent carrier density on TMR, AMR, and PHE simultaneously. TMR, AMR, and PHE are all calculated from the resistivity tensor $\hat{\rho}$, which is given by the inverse of the conductivity tensor, $\hat{\rho} = \hat{\sigma}^{-1}$. For multivalley systems, the total conductivity tensor is given by the summation of each valley conductivity, $\hat{\sigma} = \sum_i \hat{\sigma}^{(i)}$ [30]. Each conductivity tensor in an ellipsoidal valley is written as

$$\hat{\sigma}^{(i)} = en_i\left(\hat{\mu}_i^{-1} \pm \hat{B}\right)^{-1}, \tag{1}$$

where $e > 0$ is the elementary charge and $\hat{B}$ is the magnetic field tensor [30,33]. n_i and $\hat{\mu}_i$ are the carrier density and the mobility tensor of each valley, respectively. The sign $\pm$ corresponds to the sign of charge. It is naively expected that Eq. (1) is not applicable in the strong-field region, because it was obtained without Landau quantization. A possible approximation to overcome this difficulty is the extended semiclassical approach, where the field-independent carrier density n_i is replaced by the field-dependent one $n_i(B)$ in Eq. (1) [17]. In this extended semiclassical approach, it is assumed that the carrier density can be decoupled from the other parts of the conductivity tensor. The validity of this decoupling was verified by a fully quantum approach based on the Kubo formula with the Landau quantization [34]. This allows us to calculate the carrier density separately from the other parts at each magnetic field. In addition, this extended semiclassical approach agrees significantly well with the fully quantum approach by the Kubo formula even in the case of Dirac electrons except for the quantum oscillation [17].

This approach has not yet been applied to specific materials. In the present work, we employ the extended semiclassical theory with the Landau levels and the field and angular dependencies of carrier density in Bi, which have been accurately determined by several studies [22,27–29]. The field-induced angular dependence of carrier density plays a crucial role in solving the anomalies in strong fields.

One hole pocket is located at the T point along the trigonal axis, and three electron pockets are located at three equivalent L points (Fig. 1). The electrons around the L point can be well described as the Dirac electrons with an additional g factor, which originates from the multiband effect of spin-orbit coupling [35–38]. Its Landau quantized energy is given as [29]

$$\begin{aligned} \epsilon_{n,\sigma}(k_z) &= \sqrt{\Delta^2 + 2\Delta\xi_{n,\sigma}(k_z)} + \frac{\sigma g' \mu_B B}{2}, \\ \xi_{n,\sigma}(k_z) &= \left(n + \frac{1}{2} + \frac{\sigma}{2}\right)\hbar\omega_c + \frac{\hbar^2 k_z^2}{2m_z}, \end{aligned} \tag{2}$$

where n is the Landau level index, and $\sigma = \pm$ corresponds to the degree of freedom of the Kramers doublet. Δ is the half of the band gap and ω_c is the cyclotron frequency. g' is the additional g factor, and μ_B is the Bohr magneton. k_z and m_z are the wavenumber and effective mass along the magnetic field direction, respectively. The holes around the T point can be well approximated as a nearly free particle with a modified g factor [27,37] (for details see Ref. [39]). The Fermi energy is determined in order to satisfy the charge neutrality condition: $n_h(B) = \sum_{i=1\text{-}3} n_{ei}(B)$, where $n_h(B)$ and $n_{ei}(B)$ are the carrier densities of holes and electrons, respectively. Finally, we calculate the magnetoconductivity tensors by substituting n_i in Eq. (1) with the computed field-dependent carrier density. This theoretical approach enables the calculation of the galvanomagnetic effect even at strong fields, where Landau quantization is noticeable [17].

III. RESULTS

The left panels of Fig. 2 are the polar plots of the inverse of TMR $\rho_{33}^{-1}(\theta)$, where the field is rotated in the binary-bisectrix plane with the current along the trigonal axis [Fig. 1(a)]. Subscripts 1, 2, and 3 correspond to binary, bisectrix, and trigonal directions, respectively. (Here, we plot the inverse of TMR to make the comparison with the experiment easier [23]. This plot makes the comparison with the theoretical carrier density easier as well.) We used the electron mobilities $\mu_1 = 11000$, $\mu_2 = 300$, $\mu_3 = 6700$, and $\mu_4 = -710$, and the hole mobilities $\nu_1 = 2200$ and $\nu_3 = 350$ (in units of $\mathrm{T}^{-1} = 10^4\ \mathrm{cm}^2/\mathrm{V\,s}$), which were obtained by Hartman for bulk Bi at 4.23 K [26]. In weak fields, $\rho_{33}^{-1}(\theta)$ takes the maxima (minima) for $B \parallel$ binary (bisectrix). The highly anisotropic $\rho_{33}^{-1}(\theta)$ in weak fields originates from the anisotropy of mobilities. In particular, ρ_{33}^{-1} is proportional to the effective mass *perpendicular* to the magnetic field, because the conductivity of the electron pocket along the bisectrix axis ($e1$ in Fig. 1) is given as $\sigma_{33}^{e1} \simeq en_{e1}/(\mu_2 \cos^2\theta + \mu_1 \sin^2\theta)B^2$, and the mobilities are inversely proportional to the effective mass. As the field

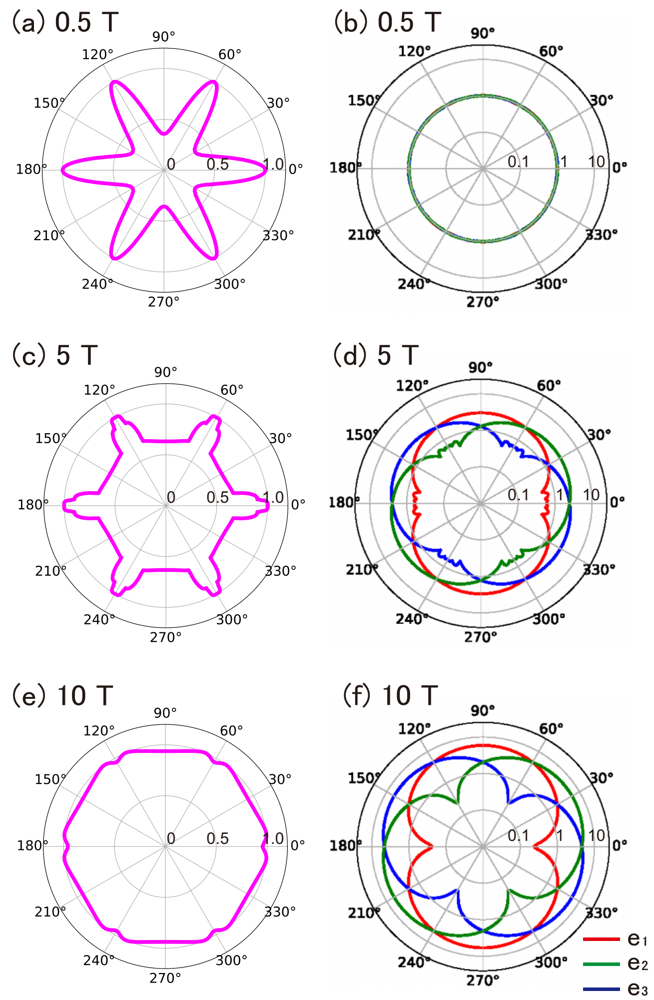

FIG. 2. Left: Angle dependence of inverse TMR with the current along the trigonal axis, $\rho_{33}^{-1}(\boldsymbol{B})$, at 0.5, 5, and 10 T. For easy comparison with the experiment [23], the values of ρ_{33}^{-1} are normalized by its maximum values. Right: Angle dependence of electron carrier densities, $n_{e1,e2,e3}(\boldsymbol{B})$, which are normalized by those in the weak-field limit, $n_{ei}(0.1\ \mathrm{T}) = 9.7 \times 10^{16}\ \mathrm{cm}^{-3}$. 0° and 90° correspond to the binary and bisectrix axes, respectively.

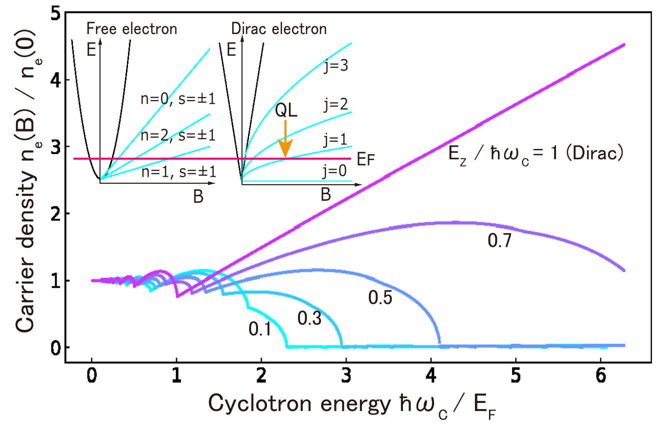

FIG. 3. Magnetic field dependence of carrier density for different Zeeman energies, which characterizes the amplitude of the spin-orbit coupling. The insets show the field dependence of the Landau levels for the free and Dirac electrons.

increases, the star-shaped peaks at the binary become less prominent and vanish at 10 T [Fig. 2(e)]. Here, the angular dependence of TMR is almost isotropic. This regression of anisotropy is perfectly consistent with the experiment, especially at low temperatures [23,24].

The qualitative change in the anisotropy can be easily understood by considering the angular dependence of the carrier density of electrons, $n_{ei}(\theta)$, shown in the right panels in Fig. 2. In weak fields, $n_{ei}(\theta)$ is isotropic, so that the anisotropy of TMR originates solely from the anisotropy of mobilities ($\mu_1 \gg \mu_2$). In strong fields, on the other hand, $n_{ei}(\theta)$ becomes anisotropic. $n_{ei}(\theta)$ takes its maximum (minimum) values for $B \parallel$ bisectrix (binary). The anisotropy of $n_{ei}(\theta)$ is orthogonal to that of $\rho_{33}^{-1}(\theta)$, where they compensate for each other. This is the reason why the TMR becomes isotropic at high fields.

The origin of the anisotropy of $n_{ei}(\theta)$ is related to (i) the characteristics of Dirac electrons in semimetals and (ii) the anisotropy of effective mass. Let us discuss the first possible origin. In semimetals, where electrons and holes coexist even at zero temperature, each carrier density can change to a great extent as long as it maintains charge neutrality [22,27,29]. To understand this origin as clearly as possible, here we consider a semimetal with one electron and one hole carrier as an example. As shown in Fig. 3, the electron's carrier density $n_e(B)$ begins to have remarkable field dependence when the field reaches the quantum limit ($\hbar\omega_c \sim E_F$), where all electrons are confined to the lowest Landau level. (The details of the calculation are given in Ref. [39].) In the case of Dirac electrons, where the Zeeman energy E_z becomes equivalent to the cyclotron energy $\hbar\omega_c$ due to the large spin-orbit coupling [21,35], the lowest Landau level is barely affected by the magnetic field (cf. the inset in Fig. 3). Thus, $n_e(B)$ increases linearly in B due to Landau degeneracy. On the other hand, in the case of nearly free electrons ($E_z/\hbar\omega_c \ll 1$), $n_e(B)$ decreases with the field, because the energy of the lowest Landau level increases with the magnetic field (see the inset in Fig. 3). Therefore, the large increase in the carrier density is a characteristic of Dirac electrons in semimetals with large spin-orbit coupling. Now, we discuss the second probable origin. The increase in $n_{ei}(B)$ depends on the inverse of cyclotron mass m_c, because the field value in the quantum limit is roughly given by $E_F \simeq \hbar\omega_c \propto 1/m_c$. Here, m_c is given by the effective mass *perpendicular* to the field. The anisotropy of $\rho_{33}^{-1}(\theta)$ in weak fields, which is proportional to m_c, is canceled out by the anisotropy of n_{ei} in strong fields. Consequently, the isotropic TMR in strong fields is the inherent characteristic of semimetals with Dirac electrons.

We calculate ρ_{11} (AMR) and ρ_{12} (PHE) with the same extended semiclassical theory and the same model of Bi as in Fig. 2. The arrangement of $\boldsymbol{J}$ and $\boldsymbol{B}$ is shown in Fig. 1(b). We used the mobility for a thin film obtained by Yang *et al.*: $\mu_1 = 142.8$, $\mu_2 = 1.99$, $\mu_3 = 32.7$, $\mu_4 = -3.38$, $\nu_1 = 18.8$, and $\nu_3 = 1.57$ (in T^{-1}) [16]. In Fig. 4, we assumed that the charge neutrality is violated as $[n(B) - p(B)]/n(0) = 0.265$ according to the experimental report [16]. Note that the following results are essentially unchanged even if we change

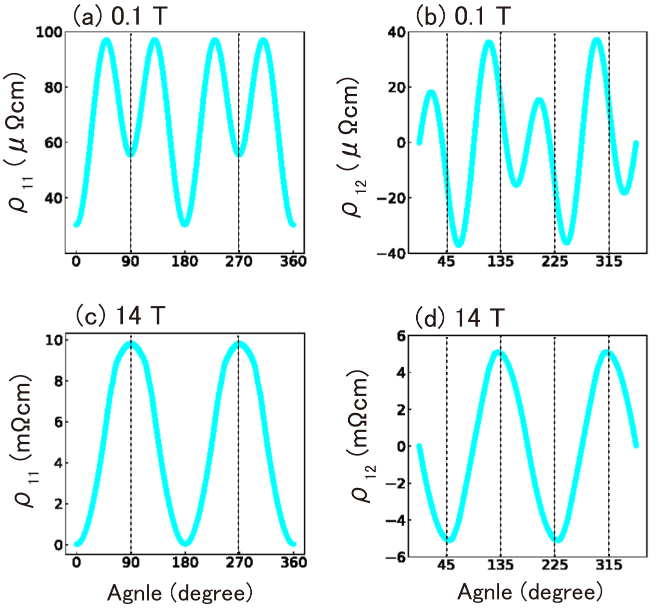

FIG. 4. Angular dependence of ρ_{11} (AMR) and ρ_{12} (PHE) with the current along the binary axis, assuming the mobilities for a thin film.

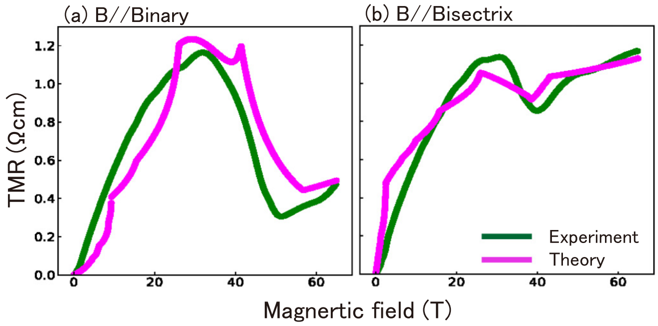

FIG. 5. TMR as a function of magnetic field along the (a) binary axis and (b) bisectrix axis. The theoretical results are obtained by the extended semiclassical theory with the field dependence of mobility. The experimental data are from Ref. [24].

the degrees of the violation [39]. In weak fields [Figs. 4(a) and 4(b)], both $\rho_{11}(\theta)$ and $\rho_{12}(\theta)$ show four maxima; i.e., the angular oscillation has two components with periods π and $\pi/2$. These properties have been already reported by Yang *et al.* based on the conventional semiclassical theory [16]. They also pointed out that the experimental results in strong fields—the peak of period $\pi/2$ disappears—are impossible to be fitted by the conventional semiclassical theory. That is why they argued the possible scenario of chiral anomaly. In contrast, our results for strong fields [Figs. 4(c) and 4(d)] show that the angular oscillation of the period $\pi/2$ disappears both in $\rho_{11}(\theta)$ and $\rho_{12}(\theta)$, which can explain the experimentally observed field-induced transformation. It is clear that the field and angular dependencies of n_{ei} play a crucial role for the field-induced transformation. Even in AMR and PHE, the angular dependence of $n_{ei}(\theta)$ in strong fields is the same as that in TMR (Figs. 1 and 2). Therefore, the field-induced transformation in AMR and PHE shares the same origin as in TMR, which has never been previously pointed out. We revealed that the nontrivial field-induced transformations of angular-dependent TMR, AMR, and PHE are all well explained qualitatively by our extended semiclassical theory in a unified manner. However, this approach is not sufficient to obtain quantitative agreement with the measured amplitude of galvanomagnetic effects far beyond the quantum limit ($\gtrsim$10 T). For example, the theoretical value of TMR was estimated to be two orders larger than the experimental value at 10 T even when we considered the field-dependent carrier density [39]. This discrepancy can be corrected by considering the field dependence of mobilities. We hypothesized several forms of the field-dependent mobility and found that the functional form of $\hat{\mu} = \hat{\mu}_0/(1 + \gamma_i B)$ can well fit the TMR from weak fields to strong fields, as shown in Fig. 5. (The details are provided in Ref. [39].) The theoretical results of TMR both for $B \parallel$ binary and bisectrix axes agree well quantitatively with those of experiments including the sudden drop at around 40 T, which is due to the evaporation of Dirac electrons [24]. Although the microscopic derivation of this functional form is still missing, such field dependence may be derived by the guiding center diffusion [40] or quantum correction to the relaxation time [41,42].

IV. CONCLUSIONS

In summary, there are three aspects to our conclusions. First, we indicated that the nontrivial field-induced transformation observed in the angular-dependent TMR, AMR, and PHE in Bi can be naturally explained via the angular dependence of carrier density in strong fields using the extended semiclassical approach. We demonstrated that nontrivial behavior does not indicate the appearance of novel topological physics, such as the chiral anomaly. The key components of the modified theory are Landau quantization and the field dependence of carrier density, which have not been considered by the conventional semiclassical theory. Second, we showed the crucial role of the field and angular dependence of carrier density in strong fields. This property originates from the characteristics of semimetals with Dirac electrons. These aspects are not only valid for Bi, but can also be applied to various semimetals. Using this approach, we can further improve the accuracy of the analysis of galvanomagnetic effects and identify new phenomena, especially in topological semimetals. Finally, we found that the field-dependent carrier mobility, which is inversely proportional to the field, is essential for the quantitative explanation of the TMR far beyond the quantum limit ($\gtrsim$10 T). Although the cause of the field dependence is an open question, the functional form has been proposed in several theoretical studies.

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