New scheme for magnetotransport analysis in topological insulators

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The recent excitement about Dirac fermion systems has renewed interest in magnetotransport properties of multi-carrier systems. However, the complexity of their analysis, even in the simplest two-carrier case, has hampered a good understanding of the underlying phenomena. Here we propose a new analysis scheme that strongly reduces the numerical uncertainty of previous studies and therefore allows to draw robust conclusions. This is demonstrated explicitly for the example of three-dimensional topological insulators. Their temperature and gate voltage-dependent Hall coefficient and transverse magnetoresistance behavior, including the phenomenon of huge linear transverse magnetoresistance, is fully reproduced by the scheme, allowing for an unambiguous identification of the carrier numbers and mobilities. Also the Fermi level can be determined from the analysis.

We derive an upper limit for the transverse magnetoresistance as functions of mobility and field. Violation of this limit is a strong indication for field-dependences in the electronic band structure or scattering processes, that are not captured by our model, or for more than two effective carrier types. Remarkably, none of the three-dimensional topological insulators with particularly large transverse magnetoresistance violate the limit.

Keywords: Dirac fermion systems, topological insulators, Hall effect, magnetoresistance

Topological insulators continue to be of tremendous interest to condensed matter physicists [1-9], both to advance the fundamental understanding of topological matter and to pave the way for new applications. Topologically protected states with linear band dispersion have been observed for numerous systems, including (Bi,Sb)$_2$(Se,Te)$_3$, TlBi(Se,Te)$_3$, C$_3$As$_2$, and (Ta,Nb)(As,P), by angle-resolved photoemission spectroscopy (ARPES) [4-8]. However, (magneto)transport evidence for electrical conduction by Dirac fermions – relativistic quasiparticle in the solid associated with these bands – has remained more circumstantial [9-13]. This is largely due to finite conductivity contributions from topologically trivial (e.g., bulk) bands. To disentangle the different effects, transport experiments are usually being analyzed with two-carrier models [10-13,16]. Because of the large number of open parameters in these models, however, this has frequently lead to large uncertainties in the extracted information. As a consequence, unequivocal transport evidence for the expected ultrahigh mobilities of Dirac particles, a cornerstone for “topotronic” devices, remains to be found.

Here we propose a new scheme for such analyses, that largely eliminates previous problems. It clarifies the physical meaning of both the $R_{H}$ sign inversion and the huge linear transverse magnetoresistance (TrMR) [12,13], phenomena deemed characteristic of Dirac fermion systems. It also allows to determine the Fermi level as well as the upper limit for TrMR as functions of the Hall mobility and field.

We start by describing the differences between the common two-carrier analysis and our new scheme. In the former, the resistance $R_{xx}(B)$ and the Hall resistance $R_{xy}(B)$, where $B$ is the magnetic field, are characterized by four free parameters (the charge carrier numbers $n_1$ and $n_2$, and the mobilities $\mu_1$ and $\mu_2$) and two constant parameters (the charge carrier types $q_1$, $q_2 = \pm e$, where $e$ is the elementary charge) that need to be anticipated. Following the usual notation, $n_i$ is positive for both electrons and holes, whereas $\mu_i$ is negative for electrons and positive for holes. In the new scheme, only two free parameters are used, namely the relative charge carrier number difference

$$N \equiv n_1 - n_2$$

and the relative mobility difference

$$M \equiv \frac{\mu_1 - \mu_2}{\mu_1 + \mu_2}$$

Two further parameters, the effective Hall coefficient

$$R_{H} \equiv \lim_{B \to 0} \frac{R_{xy}}{B}$$

and the effective Hall mobility

$$\mu_{H} \equiv \lim_{B \to 0} \frac{R_{xy}B}{R_{xx}B}$$

can be directly read off the data. No ad hoc assumption has to be made on the charge carrier type. $N$ and $M$ can thus be determined with minimal ambiguity. $q_1$ and $q_2$ are determined as functions of $N$, $M$, and $\mu_{H}$ (see supplemental material for the complete description).

Next we show how to understand the sign inversion in $R_{H}$, observed in many Dirac fermion systems as a function of temperature $T$ or gate voltage $V_{G}$ [12,13]. Figure 1(a, left) depicts the typically observed signature in consecutive (1-4) $R_{xy}(B)$ traces: the slope varies continuously from 1 to 4. This is to be contrasted with the expectation for $R_{xy}(B)$ traces resulting from a single Dirac
FIG. 1. (Color online) (a, left) Sign inversion of \( R_{xy}(B) \) traces typically observed in a multi-carrier system as a function of temperature \( T \) or gate voltage \( V_G \) (1-4). The slope of \( R_{xy}(B) \) varies continuously from 1 to 4. (a, right) Electronic band dispersion of a single Dirac cone, where \( E \) is the energy and \( k \) is the wave number (left), and corresponding \( R_{xy}(B) \) traces at \( T = 0 \) (right) as \( E_F \) is varied across the Dirac point (5-8). The sign change of \( R_{xy} \) is accompanied by a discontinuity in the slope of \( R_{xy}(B) \). (b) Contour plots of the Hall factor \( \alpha \) for \( q_2/q_1 = -1 \) as functions of \( N \) and \( M \), for \( |\mu_1| > |\mu_2| \) (left) and \( |\mu_1| < |\mu_2| \) (right). The \( R_H \) sign inversion occurs only for \( |\mu_1| < |\mu_2| \), the situation of higher mobility majority carriers. The four situations (1-4) depicted in (a) could arise, for instance, along the arrow, with sign inversion between 2 and 3. (c) Contour plots of the transverse magnetoresistance \( \text{TrMR} \) for \( q_2/q_1 = -1 \) and \( |\mu_B| = 1 \) as functions of \( N \) and \( M \), for \( |\mu_1| > |\mu_2| \) (left) and \( |\mu_1| < |\mu_2| \) (right). The same arrow depicted in (c) indicates a monotonic increase between 2 and 3. The upper limits for the \( \text{TrMR} \) are 5.96 and 12.5 for \( |\mu_1| > |\mu_2| \) and \( |\mu_1| < |\mu_2| \), respectively.

The Hall factor. The values of the parameters \( N \) and \( M \) can assume in this case follow from their definitions (Eqns. 7 and 8) as \( 0 < N < 1 \) and \( 1 < |M| < \infty \). For any \((N, M)\) combination within these limits \( \alpha \) directly follows from Eqn. 7. Figure 1(b) shows contour plots of \( \alpha \) in the full \( N \) range and for \( 1 < M < 5 \) (left) and \( -5 < M < -1 \) (right). These describe the situations \( |\mu_1| > 1.5|\mu_2| \) and \( |\mu_1| < 0.667|\mu_2| \), respectively. Smaller mobility differences would be captured by plots to larger values of \( |M| \). These are, however, less relevant here because we aim at separating contributions of highly mobile Dirac fermions from those of topologically trivial fermions with much lower mobility.

Our first key result follows directly from these contour plots. A sign change of \( \alpha \) occurs only in the right panel. Therefore, within the two-carrier picture, also \( R_H \) can show sign inversion only for \( |\mu_1| < |\mu_2| \). In experiments on putative three-dimensional topological insulators (3D-TIs) where the observed sign inversion in \( R_H \) was taken as evidence for the presence of Dirac surface states [10, 12, 14, 15], Dirac fermions were thus the minority carriers, and transport was dominated by topologically trivial charge carriers of lower mobility, most likely associated with residual bulk states. To illustrate this further, two concrete examples of temperature and gate voltage tuning are given in what follows.

The experimentally observed \( R_H \) sign inversion as a function of temperature can be understood by taking the temperature dependence of the Fermi distribution function \( f(E, T) \) into account. Figure 2(a) shows a sketch of the electronic band structure of a 3D-TI, for the situation where \( E_F \) lies above the Dirac point. The Dirac fermions are thus electron like (n-type carriers). The temperature dependence of \( f(E, T) \) is also sketched, for different temperatures decreasing from 1 to 4. At high temperatures (1), the majority carriers \( (n_1) \) are thermally excited bulk holes (p-type carriers). With decreasing temperature \( (1 \rightarrow 4) \), \( n_1 \) decreases exponentially, resulting in a decrease of \( n_+ \) and \( N \). Minor variations are also expected for \( n_2, \mu_1, \) and \( \mu_2 \), but they are neglected here for simplicity. To extract information on the system at the sign inversion of \( R_H \), we replot a section of the \( \alpha(N, M) \) contour plot of Fig. 1(b, right) around \( |\mu_2/\mu_1| = 5 \) \((M = -1.5)\), a situation considered realistic for experimentally studied 3D-TIs, in Fig. 2(b). Upon lowering the temperature

\[
R_H = \frac{1}{n_+ q_1} \alpha, \tag{5}
\]

can be expressed as

\[
n_+ = n_1 + n_2 \tag{6}
\]
is the total charge carrier number and

\[
\alpha = \frac{N + 2M + NM^2}{(N + M)^2} \tag{7}
\]
(1 → 4), sign inversion (α = 0) occurs at N = 0.923, corresponding to only 4% of surface carriers (n_2 = 0.04n_1). Larger mobility differences (smaller negative M values, towards top of Fig. 2(b)) correspond to even smaller fractions of surface carriers.

Gate voltage tuning can be mimicked by a variation of E_F around the Dirac point (Fig. 3(a, left), E_F increases from 5 to 8). At not too low temperatures, the above situation with minority surface carriers (n_2) is still relevant here because of the very small density of states of Dirac particles near the Dirac point. The corresponding R_{xy}(B) traces (Fig. 3(a, center)) and α(N, M) contour plots for q_2/q_1 = 1 (E_F below Dirac point, 5 and 6 in Fig. 3(b, left)) and q_2/q_1 = -1 (E_F above Dirac point, 7 and 8 in Fig. 3(b, right)) reveal the situation at R_{xy}(B) sign inversion (details on α for q_2/q_1 = 1 are provided in the supplemental material): For the typical mobility ratio |μ_2/μ_1| = 5 (M = -0.667 for q_2/q_1 = 1 and M = -1.5 for q_2/q_1 = -1) also considered above, it occurs at N = 0.923, corresponding to 4% of surface carriers, similar to the temperature tuning case.

Interesting conclusions can, however, also be drawn if R_H(T) reveals no sign inversion. Such a situation may arise in a truly bulk-insulating 3D-TI where surface carriers are the majority carriers (n_1). Here, the exponential decrease of the number of minority bulk holes (n_2) with decreasing temperature (1 → 4) results in only a small decrease of n_+ (Fig. 2(c, center)) and an increase of the N (Fig. 2(c), arrow assumes again a mobility ratio |μ_1/μ_2| = 5 (M = 1.5)). Such a minor effect on the R_{xy}(B) isotherms can, on its own, hardly be taken as strong evidence for the detection of Dirac fermions. However, in conjunction with transverse magnetoresistance measurements, strong conclusions can be drawn, as detailed in what follows.

The transverse magnetoresistance TrMR = [R_{xx}(B) − R_{xx}(0)]/R_{xx}(0) of a two-carrier system with q_2/q_1 = -1

\[
\text{TrMR} = \frac{(N^2 - 1)M^2(1 - M^2)(\mu_HB)^2}{(2M + N + NM^2)^2 + N^2(1 - M^2)^2(\mu_HB)^2}
\]

depends on the parameters N, M, and \(\mu_H\) defined in Eqs. 4 and 5. As stated above, \(\mu_H\) can be directly read off from R_{xy}(B) and R_{xx}(B) data. Then, for a given \(\mu_HB\), TrMR follows for each (N, M) from Eqn. 8. Thus, as for α, contour plots of TrMR(N, M) can be determined and analyzed for the different situations of interest (Fig. 3(c), Fig. 5(c, f), Fig. 3(c)).

The sign inversion in R_H(T) in a system with surface minority carriers (Figs. 1 and 2(a-c)) is, for the exemplary case of |μ_HB| = 1, accompanied by a monotonic increase of TrMR(T) with decreasing temperature, in agreement with recent experiments [18]. By contrast, if surface carriers are the majority carriers (Fig. 2(d-f)), TrMR(T) should decrease with decreasing temperature. The sign inversion in R_H(V_G) is accompanied by a distinct feature in TrMR: a non-monotonic variation of TrMR(V_G) with a minimum of TrMR at the Dirac point (Fig. 3(c)). This insight establishes a new tech-
FIG. 3. (Color online) (a) Sketch of the electronic band dispersion of a 3D-TI around the Dirac point. An increase of $E_F$ by $V_G$ (5: lowest $E_F$; 8: highest $E_F$; left) and consecutive $R_{xy}(B)$ (center) and TrMR($B$) (right) traces are also presented. (b) Contour plots of $\alpha(N,M)$ ($q_2/q_1 = 1$, left) around $|\mu_2/\mu_1| = 5(M = -0.667)$ and $\alpha(N,M)$ ($q_2/q_1 = -1$, right) around $|\mu_2/\mu_1| = 5(M = -1.5)$. No sign inversion occurs at the Dirac point (6 $\rightarrow$ 7). (c) Contour plot of TrMR($N,M$) for $|\mu_B| = 1$, $q_2/q_1 = 1$ (left) and $q_2/q_1 = -1$ (right). The minimum TrMR is observed at the Dirac point (6 $\rightarrow$ 7).

Our two-carrier analysis scheme advances the understanding of this phenomenon by revealing that, for any given $|\mu_H B|$ value, there is an upper limit to the TrMR.

Figure 4 shows this TrMR limit for the two cases $q_2/q_1 = \pm 1$ and more mobile minority carriers ($|\mu_1| > |\mu_2|$). Interestingly, the TrMR limit increases linearly with $|\mu_H B|$ for $|\mu_H B| > 10^3$ and $|\mu_H B| > 10^{-1}$ for $q_2/q_1 = -1$ and $q_2/q_1 = 1$, respectively. The corresponding $N$ and $M$ values are also plotted (middle and lower panel). They suggest that linear increase of the TrMR limit with $|\mu_H B|$ occurs if $|\mu_1| \ll |\mu_2|$. The largest TrMR values for various Dirac fermion materials reported in the literature, together with the corresponding $\mu_H$ values at the same field, are also summarized in Fig. 4. None of them overshoots the limit. This shows that, surprisingly, all TrMR observed to date are consistent with a two-carrier model.

Let us not conclude without mentioning limitations of our considerations. The two-carrier model assumes that the charge carrier numbers and mobilities, and thus $N$ and $M$ are independent of $B$. Thus, systems with a strongly $B$-dependent electronic structure or scattering processes cannot be expected to be described. Whether or not a certain materials class obeys the TrMR limit discussed above is therefore an indication of the validity of these conditions. The fact that a large number of Dirac fermion systems all conform with the TrMR limit (Fig. 4) underpins the validity of this analysis for this materials class. Strongly field-dependent parameters have for instance been observed in strongly correlated electron systems [31, 32], which indeed break the limit [33].

Finally, we mention several possible applications of the new two-carrier analysis scheme. As the two-carrier model does not specify the origin of the carriers, the analysis is valid not only for intrinsic two-band transport, but can also describe extrinsic carriers arising from spatial inhomogeneity or by multi-layer films.

In summary, we have put forward a new two-carrier analysis scheme that clarifies the physical meaning of the $R_{xy}$ sign inversion and the huge linear TrMR. Transport behavior observed in 3D-TIs is discussed. Several features, such as the absence of a sign inversion of $R_{xy}$ in the case where surface carriers are the majority carriers and a minimum of the TrMR at the Dirac point are suggested. Upper limit of the TrMR is determined as a function of $|\mu_H B|$. All huge linear TrMR values observed to date in Dirac fermion systems are consistently explained within the analysis, implying the absence of $B$-dependent parameters in the two-carrier model.

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FIG. 4. (Color online) Upper limit of the TrMR for $q_2/q_1 = -1$ (colored in red) and for $q_2/q_1 = 1$ (colored in black) as a function of $|\mu B|$. The corresponding $N$ and $M$ values are also plotted. A linear increase of the TrMR limit with $|\mu B|$ occurs if $|\mu_1| \ll |\mu_2|$. The largest published TrMR values for various Dirac fermion systems (colored in red) and several pure elements (colored in black) are also plotted [11, 13, 14, 18, 23, 30], together with the corresponding $|\mu B|$ values given for the same magnetic field in the same reference, for the same sample batch. Remarkably, none of them overshoots the limit.

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