Anomalous conductivity tensor in the Dirac semimetal Na₃Bi

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Abstract – Na₃Bi is a Dirac semimetal with protected nodes that may be sensitive to the breaking of time-reversal invariance in a magnetic field B. We report experiments which reveal that both the conductivity and resistivity tensors exhibit robust anomalies in B. The resistivity ρₓₓ is B-linear up to 35 T, while the Hall angle exhibits an unusual profile approaching a step function. The conductivities σₓₓ and σₓᵧ share identical power-law dependences at large B. We propose that these significant deviations from conventional transport result from an unusual sensitivity of the transport lifetime to B. The transport features are compared with those in Cd₃As₂.

In Dirac semimetals – the analogs of graphene in three-dimensional material – the bulk Dirac node is protected against gap formation via hybridization. The iridate pyrochlores were initially predicted [1] to have protected nodes, but crystal growth has been problematical. Recently, Young et al. [2] identified a class of materials in which time-reversal invariance (TRI) leads to node protection when the nodes occur at high-symmetry points (the time-reversal invariant momenta or TRIM). Subsequently, Wang et al. proposed that crystalline symmetry can protect Dirac nodes even when they occur away from TRIM. From band calculations, they identified Na₃Bi [3] and Cd₃As₂ [4] as Dirac semimetals. Photoemission [5–9] and scanning tunneling microscopy [10] recently confirmed that bulk Dirac nodes exist in both Na₃Bi and Cd₃As₂. Many groups [11–14] predict that, in Dirac and Weyl semimetals, charge pumping associated with the chiral anomaly can be observed in an intense magnetic field B. Recently, the chiral anomaly was detected in Na₃Bi as an enhanced conductivity “plume” that is locked to the direction of the applied magnetic field B [15]. The chiral anomaly experiments were performed on crystals in which the Fermi energy E_F is only 30 meV above the node. Here we report detailed transport experiments on Na₃Bi crystals in which E_F is ~10× higher. We show that, even at low B, the breaking of TRI leads to robust anomalies in the conductivity tensor which appears to originate from a strongly B-dependent transport lifetime τ_tr(B).

Among the anomalies are a robust B-linear magnetoresistance and an unusual step-like field profile of the Hall angle tan θ. The quantum oscillations also suggest that the Fermi surface (FS) has two frequency components. These unusual features show that even in crystals with large E_F the transport properties are highly unusual.

Na₃Bi single crystals were crystallized from the Na-rich compositions (90 and 95%) tuned to preclude the formation of the superconductor NaBi as an impurity phase [16,17] (growth details are published in ref. [18]). The crystal structure was confirmed by X-ray diffraction (the lattice structure is sketched in fig. 1(C)). The deep-purple crystals grow with the largest facets normal to the c-axis (001). To avoid deterioration of the crystals (which fully oxidize within 30 s of exposure to air), we attached contacts with Ag epoxy to the crystals inside an Ar glove-box and then covered them with oil before transferring to the cryostat (see ref. [15] for the detailed sample mounting process). The resistivity profile ρ vs. T is metallic (fig. 1(A)) with residual values ranging from 1.72 to 87 µΩcm. The Hall resistivity ρₓᵧ is n-type and strictly B-linear (fig. 1), with a nearly T-independent Hall coefficient R_H = ρₓᵧ/B (ẑ|I|ẑ and ẑ|c), where I is the current. Batch B and C samples were measured without post annealing, while G1 was post-annealed for 1 month. Table 1 lists the transport quantities measured in 8 samples. The samples B5, ···, B12, and C1 were measured without post annealing. Samples F1 and G1 were post-annealed.
I. Introduction

Table 1: Transport parameters in 8 samples of Na\textsubscript{3}Bi. \(n_H\) is the Hall density inferred from \(R_H\). \(k_F\) is the FS wave vector inferred from the period of the quantum oscillations and \(n_F = g_\nu k_F^2/3\pi^2\) with \(g_\nu = 2\). \(\mu'\) is the transport mobility derived from \(\rho(4\,\text{K})\) and \(n_F\), while \(\mu\) is directly read from the profile of \(\sigma_{xy}(B)\) in fig. 3(B) (main text). MR(9\,T) is the MR ratio measured at 9\,T. \(\tau_F\) is calculated from \(\mu = e v_F \tau_F/k_F\). \(f_1\) and \(f_2\) are the periods inferred from the SdH oscillations. The quantities \(\rho, n_H\) and \(\mu\) are subject to the large uncertainty in estimating \(t\) (\(\pm 50\,\mu\text{m}\)), but \(k_F, n_F\) and \(\mu'\) are unaffected. F1 and G1 were post-annealed for 2 weeks and 1 month, respectively. The quantities \(\rho(4\,\text{K})\) and \(n_H\) are strongly affected by the large uncertainty in \(t\), but \(k_F, n_F, \mu\) and \(\tau_F\) are not.

| Sample | \(\rho(4\,\text{K})\) (\(\mu\Omega\text{cm}\)) | \(n_H\) (\(10^{19}\text{cm}^{-3}\)) | \(k_F\) \(A^{-1}\) | \(n_F\) (\(10^{19}\text{cm}^{-3}\)) | \(\mu'\) | \(\mu\) (cm\(^2\)/Vs) | MR(9\,T) | \(\tau_F\) (ps) | \(f_1\) | \(f_2\) |
|--------|-----------------|-----------------|-------------|-----------------|-------------|-----------------|--------|-------------|--------|--------|
| B5     | 34              | 0.083           | 3.8         | --              | --          | 5.69            | --     | --          | --     | --     |
| B6     | 6.2             | 0.079           | 3.4         | 35000           | 17          | 2.55            | --     | --          | --     | --     |
| B10    | 7.5             | 0.081           | 3.6         | 13000           | 9.62        | 1.49            | --     | --          | --     | --     |
| B11    | 87              | 0.073           | 2.6         | 5500            | --          | 10.5            | --     | --          | --     | --     |
| B12    | 7.4             | 0.082           | 3.7         | 23000           | 10.3        | 1.94            | 247.3  | 225.3       | --     | --     |
| C1     | 5.1             | 0.084           | 4.0         | 13600           | 16.2        | 1.93            | 223    | 202.8       | --     | --     |
| F1     | 6.6             | 0.082           | 3.7         | 14600           | 32.8        | 2.11            | 252.8  | 203.3       | --     | --     |
| G1     | 1.72            | 0.085           | 4.1         | 78900           | 97.1        | 6.71            | 241.8  | 225.3       | --     | --     |
| E1     | --              | --              | --          | --              | --          | --              | --     | --          | --     | --     |

II. Results and Discussion

Fig. 1: (Color online) Magnetotransport in Na\textsubscript{3}Bi. Panel A: the zero-field resistivity \(\rho\) and the Hall coefficient \(R_H\) vs. \(T\) (measured with \(\mathbf{H}\parallel c\)). The inset shows the crystals sealed in a vial. The largest facet is normal to \(\hat{e}\). Panel (B) shows the Hall resistivity \(\rho_{xy}\) vs. \(B\) measured at 2\,K in B6. Panel (C): the \(H\)-linear magnetoresistance in sample B6 measured at 2\,K at selected tilt angles \(\theta\) to \(\hat{e}\). The MR ratio is largest at \(\theta = 0^\circ\) (and 180\(^\circ\)). \(B = \mu_0 \mathbf{H}\) with \(\mu_0\) the vacuum permeability. The crystal structure of Na\textsubscript{3}Bi is sketched in the inset (adapted from ref. [5]).

Fig. 2: (Color online) Torque measurements of the de Haas-van Alven (dHvA) oscillations in Na\textsubscript{3}Bi. The dHvA oscillations (solid curve in panel (A)) can be fit well to the LK expression with one period. Because of difficulties related to prevention of oxidation, measurements of the crystal thickness \(t\) have a large uncertainty (\(\pm 50\,\mu\text{m}\)). This affects the estimates of \(\rho\) and \(n_H\). However, the quantities inferred from Shubnikov-de Haas (SdH) oscillations (fig. 2) as we discuss below, as well as those from the field profile of \(\sigma_{xy}(B)\) (fig. 3(B)), namely \(k_F, n_F, \mu\) and \(\tau_F\), are unaffected by the large uncertainty in \(t\).

Figure 1(B) plots the MR curves in sample B6 for selected \(\theta\) (the tilt angle between \(\hat{e}\) and the field \(\mathbf{H} = B/\mu_0\), for 2 weeks and 1 month, respectively, before measuring.
with \( \mu_0 \) the vacuum permeability). As shown by the fan pattern, the MR ratio \( \rho_{xx}(B)/\rho_{xx}(0) \) decreases rapidly as \( \mathbf{H} \) is tilted into the \( a-b \) plane (\( \theta \to 90^\circ \)).

All the samples display prominent Shubnikov-de Haas oscillations in \( \rho_{xx}(B) \), from which the Fermi surface cross-section \( S_F \) and the Fermi wave vector \( k_F \) are determined. In addition, we have measured the de Haas-van Alven (dHvA) oscillations using torque magnetometry. Figure 2(A) shows the oscillatory component of the magnetization together with a fit to the Lifshitz-Kosevich (LK) expression. From fits to the dHvA amplitudes vs. \( 1/H \) and \( T \) (panels (B) and (C)), we determine the effective mass \( m^* \), the Fermi velocity \( v_F \) and the quantum lifetime \( \tau_Q \). The carrier density \( n = g_v k_F^2/3\pi^2 \) (with the valley and spin degeneracies \( g_v \) and \( g_s \) both equal to 2) ranges from 2.6-4.1 \( \times 10^{19} \text{ cm}^{-3} \), consistent with the Hall effect (table 1). In addition, by varying \( \theta \) in the MR, we verify that the FS cross-section \( S_F \) is nearly spherical (fig. 2(D)).

Because of unintentional doping from vacancies, the Fermi energy \( E_F \) is high in the conduction band, as implied by the \( n \)-type sign of the Hall resistivity \( \rho_{yx} \). The band calculations [3] predicted the existence of two Dirac nodes centered at \((0,0, \pm k_D)\) caused by gap inversion (sketch in fig. 2(D)). As \( E_F \) rises in the conduction band, the two independent Dirac cones merge into a band with when \( E_F \) exceeds the Lifshitz-transition energy \( E_L \). Recent ARPES experiments have confirmed the predicted dispersion and measured \( k_D \) to be 0.095 Å\(^{-1} \) [5] and 0.10 Å\(^{-1} \) [9] (see also [19]). However, because the ARPES spectra do not access states high above \( E_F \), we cannot determine at present the actual sign of \( E_F - E_L \) in the conduction band.

We further uncover an interesting and persistent feature in the SdH oscillations, \textit{i.e.} a weak beating pattern in the SdH oscillations. Figure 3(A) shows the SdH traces from 5 samples. The Fourier spectra of the oscillations reveal two frequencies \( f_1 \) and \( f_2 \) corresponding to two values of \( S_F \) differing by \( \sim 16\% \) (values reported in table 1). The beating suggests that the orbits may reflect quantum interference between the two orbits. A systematic trend in the splitting \( f_1 - f_2 \) has not been found, but we hope to explore them further in high-mobility crystals.

The mobility \( \mu \) of each sample is directly measured from the field profile of the Hall conductivity \( \sigma_{xy} \). As shown in fig. 3(B), \( \sigma_{xy}(B) \) has the characteristic dispersion-resonance profile produced by cyclotron motion of the carriers. By the Bloch-Boltzmann theory, the extrema in \( \sigma_{xy}(B) \) occur at the peak fields \( \pm B_\mu \), with \( 1/B_\mu = \mu \). With increasing \( \mu \), from sample B10 (\( \mu = 21640 \text{ cm}^2/\text{Vs} \)) to B6 (39250 cm\(^2\)/Vs) and G1 (91000 cm\(^2\)/Vs), the peak field \( B_\mu \) systematically decreases. The variation in \( \mu \) strongly influences the Hall-angle profile (see below).

In the relaxation-time \textit{ansatz}, the Boltzmann equation describing changes to the distribution function \( f_k \) caused by an electric field \( \mathbf{E} \) is expressed as [21]

\[
e\mathbf{E} \cdot \nabla f_k + e\mathbf{v} \times \mathbf{B} \frac{\partial g_k}{\partial k} = -\frac{g_k}{\tau_r},
\]

where \( e \) is the elemental charge and \( g_k = f_k - f_k^{(0)} \) with \( f_k^{(0)} \) the Fermi-Dirac function. \( E_k \) and velocity \( \mathbf{v} \) are, respectively, the energy and velocity at state \( k \). The \textit{ansatz} yields the conductivity tensor \( \sigma_{ij} \), with

\[
\sigma_{xx} = ne\mu/D, \quad \sigma_{xy} = ne\mu^2B/D.
\]

where \( D = 1 + (\mu B)^2 \). From eq. (2), the ratio \( \sigma_{xy}/\sigma_{xx} = \mu/B \), which is the Hall angle \( \tan \theta \), is linear in \( B \). By contrast, the resistivity \( \rho_{xx} \) is \( B \)-independent because the Hall electric field \( E_y \) exactly balances the Lorentz force. Significantly, \( \sigma_{xx} \sim 1/B^2 \) decreases much faster than \( \sigma_{xy} \sim 1/B \) when \( \mu B \gg 1 \). These standard predictions assume that \( \tau_r \) (hence \( \mu \)) is a constant independent of \( B \). In conventional metals and semimetals in the impurity-scattering regime (elastic scattering), this assumption is firmly established; the predicted trends are a cornerstone of semiclassical transport.

In Na\(_3\)Bi, however, the observed field dependencies of the diagonal elements \( \sigma_{xx} \) and \( \rho_{xx} \) disagree in an essential way from the standard predictions (only \( \rho_{yx}(B) \) appears conventional). As we noted in fig. 1, the resistivity

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The MR in G1 is plotted in (a) vs. \( B \) at 10, to \( \mu \)A. A general trend is that the MR increases with \( B \). In addition to the present report and ref. [15], the unusual comments [22]; we exclude metals with open orbits [21].

MR is rare in conventional conductors (see Abrikosov’s change occurs at 0.5 T. To persuade ourselves that the topological insulator Bi to persuade ourselves that the \( \mu \)-linear profile, the field profile of \( \tan \theta = \rho_{yy}/\rho_{xx} \) is compared in 4 samples. As \( H \) increases, \( \tan \theta \) rapidly saturates to an \( H \)-independent value, which implies the anomalous relationship \( \tau_{tr} \sim 1/H \). In G1, the change occurs at 0.5 T.

increases linearly with \( B \) instead of saturating. A \( B \)-linear MR is rare in conventional conductors (see Abrikosov’s comments [22]; we exclude metals with open orbits [21]). In addition to the present report and ref. [15], the unusual \( B \)-linear transverse MR has been observed in (type-B crystals) of the Dirac semimetal Cd3As2 [20], as well as in the topological insulator Bi2Te3 [23]. These results have stimulated renewed theoretical interest in linear MR. In ref. [24], it is predicted that diffusion of the guiding centers in weak disorder potentials varying on length scales much longer than the cyclotron radius leads to linear transverse MR even up to 300 K.

To persuade ourselves that the \( B \)-linear MR is pervasive in Na3Bi, we have investigated 8 samples (table 1). Figure 4(A) shows that the \( B \)-linear MR is a very robust feature in Na3Bi. Across the samples, the MR ratio (measured at 15 T) increases from \( \sim 14 \) in B10, to 163 in G1 (the sample with the highest \( \mu \)). In B11, we show that the \( B \)-linear profile extends to 35 T with no evidence of deviation.

A second dramatic anomaly is seen in the Hall-angle profile. In fig. 4(B), we compare \( \tan \theta \) measured in four samples with increasing \( \mu \); B10, B12, B6 and G1. As shown, \( \tan \theta \) initially rises very rapidly in weak \( B \) at a rate dictated by the mobility, but saturates to a plateau value at large \( B \). Whereas the saturation is gradual in the samples with low mobility (B10 and B12), the rise becomes abrupt in higher-mobility samples (B6 and G1). In G1, especially, the profile resembles a step-function profile. \( \tan \theta \) assumes a virtually \( B \)-independent value from \( B = 0.5 \) to 15 T instead of increasing linearly with \( B \). Since \( \tan \theta = \mu B \), the simplest interpretation of the step-function profile is that, starting in weak \( B \), the transport lifetime varies with \( B \) as

\[
\tau_{tr} \sim 1/B. \tag{3}
\]

The merit of eq. (3) is that it also accounts for the \( B \)-linear MR profile, i.e. \( \rho_{xx} = 1/\mu \tau_{tr} \sim B \). Interestingly, with eq. (3), the high-field \( B \)-dependence of \( \sigma_{xx} \) is reduced by one power of \( B \) to \( \sigma_{xx}(B) \sim 1/B \) (eq. (2)), but leaves that of \( \sigma_{xy} \sim 1/B \) unchanged because \( \mu \) cancels out at large \( B \). Hence both \( \sigma_{xx} \) and \( \sigma_{xy} \) vary as \( 1/B \) at large \( B \), consistent with the step profile of \( \tan \theta \).

To verify this, we plot the \( B \)-dependences of \( \sigma_{xx} \) and \( \sigma_{xy} \) in log-log scale for the two high-mobility samples B6 and G1 (fig. 5). In both samples, the two conductivities have the same power-law dependence \( B^{-\beta} \) above a relatively low \( B \). Consistent with the behavior of \( \tan \theta \), this occurs at \( B = 2 \)T and 0.3 T in B6 and G1, respectively. The measured value of \( \beta \) is 1.0 in B6, but is slightly larger (1.15) in G1.

Fig. 4: (Color online) Robust \( H \)-linear magnetoresistance in Na3Bi (panel (A)). In the 8 samples shown, \( \rho_{xx}(B) \) is measured with \( H || \hat{e} \) at 2 K in all cases except in B11 (at 1.6 K). In B11, the MR persists without observable deviation to 35 T.

Fig. 5: (Color online) Log-log plots of \( \sigma_{xx} \) and \( \sigma_{xy} \) vs. \( B \) in G1 and B6. Consistent with eq. (3), both quantities approach the same power law \( B^{-\beta} \) when \( B \) exceeds 0.3 and 2 T in G1 and B6, respectively. The measured \( \beta \) is 1.15 and 1.0 in G1 and B6, respectively. Curves for B6 are shifted vertically.
We also notice the strong similarities (and some differences) between the magnetoresistance (MR) results in Na$_3$Bi and those in Cd$_3$As$_2$. In ref. [20], the Cd$_3$As$_2$ samples investigated fall into two groups. Set-A samples are needle-shaped single crystals, while Set-B samples are polycrystals cut from the boule. A $B$-linear MR very similar to the MR observed in Na$_3$Bi is observed in all Set-B samples as well as in the Set-A crystals with “lower mobility” (100000 to 150000 cm$^2$/Vs). In Set-A crystals displaying ultrahigh mobility (150000 to 10$^7$ cm$^2$/Vs), the MR evolves to a $B^2$ profile.

The ratio of the transport to quantum lifetimes, $R_t = \tau_r/\tau_q$, reaches very large values (10$^4$) in the ultrahigh mobility Cd$_3$As$_2$ crystals. Liang et al. [20] infer that the ultrahigh mobilities likely arise from a protection mechanism that strongly suppresses back-scattering for currents flowing along the needle axis when time-reversal invariance (TRI) prevails. Application of a magnetic field $B$ breaks TRI and results in the lifting of the protection, hence a giant MR. The MR ratios attain values significantly larger than those seen in Na$_3$Bi.

It seems to us that a crucial difference between the two Dirac semimetals is that the scattering rate $1/\tau_r$ in Cd$_3$As$_2$ is highly anisotropic especially in crystals with the highest mobilities (as determined using the Montgomery technique). The anisotropy notwithstanding, the main finding is the strong effect of $B$ on $1/\tau_r$, similar to eq. (3). Because its scattering rate $1/\tau_r$ is nearly isotropic, Na$_3$Bi provides a simpler platform to unravel the mechanism underlying deviations from conventional transport.

Theoretically, in the Weyl semimetal, the Weyl nodes which come in pairs act as sources and sinks of Berry curvature (Chern flux) [11–14]. To realize a finite Berry curvature $\tilde{\Omega}(k)$, TRI must be broken in the Weyl semimetal. The 3D Dirac semimetal may be regarded as the limiting case when TRI is restored. In this limit, the Weyl nodes coincide in $k$ space but are prevented from hybridizing by crystalline symmetry. Conversely [3], one expects that, in a Dirac semimetal, the breaking of TRI by an applied $B$ will render $\Omega(k)$ finite, and lead to essential changes in its FS best detected by transport experiments.

The large negative longitudinal MR associated with the chiral anomaly is most pronounced in Na$_3$Bi crystals with $E_F$ close to the node [15]. However, as shown by the present results, even in crystals with large $E_F$, the finite Berry curvature seems to lead to major anomalous features in the conductivity tensor elements $\sigma_{ij}$. The results in both Na$_3$Bi and Cd$_3$As$_2$ [20] invite a re-examination of transport in Dirac semimetals.

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