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Absence of Dirac states in BaZnBi₂ induced by spin-orbit coupling

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We report magnetotransport properties of BaZnBi₂ single crystals. Whereas electronic structure features Dirac states, such states are removed from the Fermi level by spin-orbit coupling (SOC) and consequently electronic transport is dominated by the small hole and electron pockets. Our results are consistent with three dimensional (3D) but also with quasi two dimensional (2D) portions of the Fermi surface. The spin-orbit coupling-induced gap in Dirac states is much larger when compared to isostructural SrMnBi₂. This suggests that not only long range magnetic order but also mass of the alkaline earth atoms A in ABX₂ (A = alkaline earth, B = transition metal and X=Bi/Sb) are important for the presence of low-energy states obeying the relativistic Dirac equation at the Fermi surface.

I. INTRODUCTION

Rapid development of graphene and topological matter could induce transformational changes in both information and energy science.1–4 Main physical observables are Dirac states with Berry phases and high mobility due to suppressed backscattering.5 This generates large linear magnetoresistance (MR), quantum Hall effect and is of high interest for electronic and optical device fabrication.6–14

Dirac states could be found not only at the surface of topological insulators or graphene but also in multiband bulk crystals, giving rise to magnetoresistant mobility up to μMR∼3400 cm²/Vs, comparable to that in graphene and topological insulators.15–19 In contrast to Na₃Bi and Cd₃As₂, ABX₂ materials feature quasi-two-dimensional (quasi-2D) electronic transport of Dirac states.15,16,20 Moreover, ABX₂ incorporate strongly correlated alternating Mn-Bi magnetic layers and sometimes rare earth atoms on A site.21–23 Localized magnetic moments with sufficient hybridization could provide a possible route to the Kondo effect and to transforming Dirac into a Weyl semimetal, separating the degenerate Dirac cones.24 This could result in spin-dependent band splitting and quasi-2D spin-polarized transport.

Coupling of magnons to Dirac states is indicated by Raman measurements in CaMnBi₂, where the enhanced interlayer exchange coupling drives a charge gap opening.25 In contrast, magnetic dynamics can be described by the Heisenberg model and is not influenced by Dirac fermions.26 It is of interest to establish conditions for Dirac states at the Fermi surface in ABX₂ materials since the momentum-anisotropy of Dirac states might tailor Dirac current for applications.17,27–30 In this study we have synthesized BaZnBi₂ single crystals that feature crystal structure identical to SrMnBi₂ and BaMnBi₂. Our first-principle calculations show presence of Dirac states near Fermi level and portions of Fermi surface with quasi-2D character, similar to other AMnBi₂ materials. However, SOC opens a larger gap in Dirac states when compared to SrMnBi₂, leaving small hole and electron pockets that dominate the electronic transport.

II. EXPERIMENTAL DETAILS

Single crystals of BaZnBi₂ were grown from an excess Bi flux.31 Ba, Zn and Bi granules were mixed in the ratio of Ba:Zn:Bi = 1:1:10, put into an alumina crucible and then sealed in a quartz tube. The quartz tube was heated to 1073 K, held there for 6 h, then cooled to 593 K at a rate of 2.4 K/h where the excess Bi flux was decanted using a centrifuge. Shiny needle-like single crystals with typical size 5 mm × 1 mm × 1 mm were obtained. The element analysis was performed using an energy-dispersive x-ray spectroscopy (EDX) in a JEOL LSM-6500 scanning electron microscope, confirming 1:1:2 stoichiometry. X-ray diffraction (XRD) data were obtained by using Cu Kα (λ = 0.15418 nm) radiation of a Rigaku Miniflex powder diffractometer on crushed crystals. Heat capacity and magnetotransport measurement up to 9 T were conducted in a Quantum Design PPMS-9 on cleaved and polished single crystals in order to remove residual Bi droplets on the crystal surface [Fig. 1(a)]. Thin Pt wires were attached to electrical contacts made with Epotek H20E silver epoxy, producing contact resistance of about 10 Ω. Sample dimensions were measured with an optical microscope Nikon SMZ-800 with 10 μm resolution. Magnetotransport in high magnetic field up to 18 T was conducted at the National High Magnetic Field Laboratory (NHMFL) in Tallahassee. Resistivity was measured using a standard four-probe configuration for both ρ₂ and ρ₁. Both measurements were conducted with 0.3 mA 16 Hz ac current excitation. All magnetotransport measurements were performed in transverse configuration with current always perpendicular to magnetic field.
The de Haas van Alphen (dHvA) oscillation experiments were performed at NHMFL Tallahassee. The crystals were mounted onto miniature Seiko piezoresistive cantilevers which were installed on a rotating platform. The field direction can be changed continuously between parallel ($\theta = 0^\circ$) and perpendicular ($\theta = 90^\circ$) to the c-axis of the crystal. For first-principles band structure calculations, we applied the WIEN2K\textsuperscript{32} implementation of the full potential linearized augmented plane wave method in the generalized gradient approximation\textsuperscript{33} of density-functional theory with SOC treated in a second variational method. The basis size was determined by $R_{\text{mt}} K_{\text{max}} = 7$ and the Brillouin zone was sampled with a regular $13 \times 13 \times 13$ mesh containing 196 irreducible $k$ points to achieve energy convergence of 1 meV. The Fermi surface was plotted in a 10 000 k-point mesh.

**III. RESULTS AND DISCUSSION**

The unit cell of BaZnBi\textsubscript{2} crystals can be indexed in the $I4/mmm$ space group by RIETICA software [Fig. 1(a)].\textsuperscript{34} The lattice parameters $a = b = 0.4855(2)$ nm and $c = 2.1983(3)$ nm agree with the previously reported values.\textsuperscript{35} Hence, the BaZnBi\textsubscript{2} [Fig. 1(a) inset] features an identical structure, but a somewhat compressed c-axis when compared to SrMnBi\textsubscript{2} and BaMnBi\textsubscript{2}. The heat capacity of BaZnBi\textsubscript{2} [Fig. 1(b)] is approaching the Einstein value of $4\cdot3\cdot R = 99$ J mol$^{-1}$ K$^{-1}$, where $R$ is the universal gas constant 8.31 J mol$^{-1}$ K$^{-1}$. From $C/T$ as a function of $T^2$ at low temperature [Fig. 1(b) inset] we obtain an electronic heat capacity $\gamma = 0.48(1)\text{ mJ mol}^{-1}\text{ K}^{-2}$ and a slope of 2.61 mJ mol$^{-1}$ K$^{-4}$. From the latter, the Debye temperature $\Theta_D = 155(1)$ K is
SOC reveals Dirac-like bands derived from the 5p
magnetotransport properties below we present results of
not apply in BaZnBi
5p
level where all components of the resistivity tensor should
consequence of magnetotransport of the lowest Landau
fields. Linear unsaturated magnetoresistance could be a
tures up to 50 K. The linear unnsaturated magnetoresis-
Corresponding calculated Fermi surfaces [Fig. 2(e,f)] are
states near the X points remain quasi-two-dimensional.
more small hole and electron pockets, while the electron
gaps into the Dirac states possibly [Fig. 2(c,d)] creating
indicating that the Dirac points are close to the Fermi
square lattices and displays a pseudogap feature, again
[Fig. 2(b)] is dominated by partial contribution of Bi
Brillouin zone, which are fairly close to the Fermi level
τ
frequency and
ity in a compensated semimetal.
MR = [ρ(B) - ρ(0)]/ρ(0)×100% at different tempera-
tures up to 50 K. The linear unsaturated magnetoresis-
tance is evident at low temperatures and in high magnetic
fields. Linear unsaturated magnetoresistence could be a
consequence of magnetotransport of the lowest Landau
level where all components of the resistivity tensor should
be linear in temperature.\textsuperscript{36,37} Other possible mechanisms
of the large linear magnetoresistance include the mobility
fluctuations in a strongly inhomogeneous system, ω_cτ ~ 1 limit in thin films (where ω_c = eB/m^* is cyclotron
frequency and τ^{-1} is scattering rate) and charge neutrality
in a compensated semiemetal.\textsuperscript{38,40} The former should
d not apply in BaZnBi_2 since our sample is stoichiometric
crystal without doping/disorder. To get more insight into
magnetotransport properties below we present results of
first principle calculations.

The first-principles band structure in the absence of
SOC reveals Dirac-like bands derived from the 5p_x and
5p_y orbitals of Bi square lattices near the X points in the
Brillouin zone, which are fairly close to the Fermi level
[Fig. 2(a)]. The density of states near the Fermi level
[Fig. 2(b)] is dominated by partial contribution of Bi
square lattices and displays a pseudogap feature, again
indicating that the Dirac points are close to the Fermi
level. However, spin-orbit coupling introduces sizeable
gaps into the Dirac states possibly [Fig. 2(c,d)] creating
more small hole and electron pockets, while the electron
states near the X points remain quasi-two-dimensional.
Corresponding calculated Fermi surfaces [Fig. 2(e,f)] are
quite different. This result suggests that linear MR [Fig.
1(c)] is unlikely to arise from the lowest Landau level
magnetotransport.

It appears that the SOC effect on the A atoms predomi-
nates the steric effect due to different sizes of A atoms,
since the same structural data were used in the calcula-
tions with and without SOC. To further distinguish
the two A-site effects, we performed non-SOC calcula-
tions for BaZnBi_2 with Ba atoms moved 5% closer to the Bi2 planes and all the other parameters remaining
unchanged. In this case, the steric effect was enhanced
considerably but it has little impact on the Bi2 bands
showing Dirac points [Fig. 3(a)]. In addition, we carried
out calculations for the experimental structure with SOC
on the Bi atoms only. We found [Fig. 3(b)] that the gapping
out of the Dirac state along the Γ - X line is nearly
vanishing. This shows that the large SOC on the A-site
atoms do facilitate the gapping out of the Dirac state in
BaZnBi_2.

Figure 4(a) shows the temperature dependences of in-
plane (ρ_a) and out-of-plane (ρ_c) resistivity at 0 and 9 T
for BaZnBi_2 single crystal. Temperature dependence of
resistivity is very anisotropic(ρ_c/ρ_a ~ 55 at 2 K and ~
30 at 300 K). The broad hump at ~ 150 K at ρ_c indicates
the crossover from high-T incoherent to low-T co-
herent conduction in a quasi-2D system.\textsuperscript{28} The tempera-
ture dependence of magnetoresistance (MR) for electric
current along a- and c-axes in magnetic fields up to 9
T are shown in the Fig. 4(b). The linear transverse
MR is established for both current directions, however
at much lower field for ρ_c. The temperature-dependent
cantilever signal [Fig. 4(c)] shows clear oscillations below
40 K. The oscillatory component obtained by subtracting
a smooth background is periodic in 1/B [Fig. 4(d)]. The
fast Fourier transform (FFT) analysis yields four frequen-
cies are estimated to be m^∗_A^∗/e, m^∗_B^∗/e, and m^∗_C^∗/e,
corresponding to about 1.0%, 1.2%, 1.4%, and 1.6% of its total Brillouin zone (BZ) in the (001) plane, respectively. These are likely
to arise from the small pockets in spin-orbit induced com-
penated Fermi surface [Fig. 2(f)]. The cyclotron mass can be obtained from fitting the temperature dependence of the oscillation amplitude at different frequencies using
Lifshitz-Kosevich formula: 41 A ~ sinh(α m^∗(T/B)/m^∗_e) where α = 2π^2 k_B/e^\hbar \approx 14.69 T/K, m^∗ = m/m^∗_e is
the cyclotron mass ratio (m^∗_e is the mass of free electron).
The fitting results are presented in Fig. 4(f), the rather small cyclotron masses corresponding to different
frequencies are estimated to be m^∗_A = 0.14 m^∗_e, m^∗_B = 0.12
m^∗_e, m^∗_C = 0.11 m^∗_e, and m^∗_D = 0.11 m^∗_e, in agreement with heat capacity result [Fig. 1(b)].

The response of the carriers to the applied magnetic
field and the magnitude of MR is determined by the mo-
tility in the plane orthogonal to the applied magnetic

\begin{figure}[!h]
\centering
\includegraphics[width=0.8\textwidth]{fig3.png}
\caption{(Color online). (a)) Band structure calculations for hypothetical BaZnBi_2 where Ba is 5% closer to Bi2 plane, all
other parameters remaining unchanged. This leaves mass of
A the same, while mimicking steric effects (lattice parameter change) due to different A size. (b) Band structure calculation for
BaZnBi_2 in its true unit cell but where spin-orbit coupling is selectively switched off on the Ba atoms only.}
\end{figure}
FIG. 4. (Color online). (a) Temperature dependence of the in-plane resistivity $\rho_a(T)$ and c-axis resistivity $\rho_c(T)$ of the BaZnBi$_2$ single crystal in B = 0 and 9 T magnetic fields, respectively. Large resistivity anisotropy indicates that the c-axis interlayer coupling of conductive Bi crystallographic square layers is small. (b) Field dependence of transverse magnetoresistance at 2 K for $\rho_a$ (filled circles) and $\rho_c$ (filled squares), respectively. (c) Cantilever oscillation as a function of magnetic field at different temperatures and its oscillatory component as a function of 1/B (d). (e) FFT spectra at various temperatures, data in (c), (d), and (e) use same legend. (f) Temperature dependence of the FFT amplitudes at different frequencies, the solid line is the fitting curve using Lifshitz-Kosevich formula.

FIG. 5. (Color online). (a) Angular-dependent MR for in-plane resistivity $\rho_a(T)$ at different tilt angles. Inset cartoon shows the configuration of the measurement. (b) Tilt angle $\theta$ dependence of $\rho_a(T)$ at $B = 18$ T and $T = 0.91$ K. The red line is the fitting curve using $|\cos(\theta)|$. dHvA oscillatory components at various tilt angles $\theta$ (c), and corresponding FFT spectra (d), the dashed lines are guides to the eyes. Data in (c) are using same scale and shifted vertically for clarity, while FFT spectra are normalized and shifted vertically (c,d) Cantilever oscillation as a function of magnetic field at different tilt angles $\theta$ and corresponding FFT amplitude. (e) The angular dependence of the oscillation frequencies $F_\alpha$, $F_\beta$, and $F_\gamma$. $F_\beta$ and $F_\gamma$ can be fitted very well by $|\cos(\theta)|$, indicating the quasi-2D feature of $\beta$ and $\gamma$ Fermi surface.

There should be no significant angle-dependent MR in the case of isotropic three-dimensional Fermi surface. For quasi-2D electronic systems, the 2D states respond only to the perpendicular component of the magnetic field $B|\cos(\theta)|$; hence MR oscillations should be observed in quasi-2D conducting states. Figures 5(a,b) show the angular dependence of the $\rho_a$. The rotating magnetic field [Fig. 5(a) inset] is parallel (perpendicular) to the crystallographic c-axis when $\theta = 0^\circ$ ($90^\circ$), respectively. The $MR(\theta)$ at 18 T is consistent with the $|\cos(\theta)|$ angular dependence [solid red line in Fig. 5(b)], commonly observed in AMnBi$_2$ (A = Ca, Sr, Ba). This reflects the contribution of quasi-2D cylinder-like states away from the Γ point in the Brillouin zone [Fig. 2(f)].

Angular-dependent quantum oscillation offer further insight into the geometry of the Fermi surface. Cantilever oscillatory components are shown in Fig. 5(c), corresponding FFT spectra are shown in Fig. 5(d). The small oscillation amplitude at $8^\circ$ reflects the angle error bar in measurement in which amplitude of oscillation should be zero at strict $0^\circ$ and $90^\circ$. As shown in Fig. 5(e), $F_\beta$ and $F_\gamma$ can be fitted very well by $|\cos(\theta)|$, while $F_\alpha$ is nearly constant at different angles.
$F_3$ and $F_4$ with similar angular dependence might come from the cylindrical states near A point, and $F_n$ can be attributed to small pockets near the center of BZ. On the other hand, it is difficult to trace the weak angular dependence of $F_n$ which could come from the large hole pockets around $\Gamma$ point.

It is instructive to compare mechanism of Dirac states formation with isostructural materials AMnBi$_2$ ($A = $ Sr, Ba). We note that nearly-linear Dirac-like energy dispersion is evident in first-principle calculation in all materials. However, a small SOC-induced gap of about 0.05 eV appears near the Dirac point when SOC is included in SrMnBi$_2$. Over lapping of the cylindrical states near A point, and small electron and hole-like pockets near Brillouen zone center. Our results suggest that the mass of the alkaline earth atoms A near Bi$_2$ square network, in addition to magnetic ordering is essential for formation of Dirac cones in ABX$_2$ materials.

**IV. CONCLUSIONS**

In summary, we show that spin–orbit coupling induces gap in Dirac states, causing their removal from the Fermi surface. Resulting electronic transport is governed by cylindrical states away from the $\Gamma$ point and small electron and hole-like pockets near Brillouen zone center. Our results suggest that the mass of the alkaline earth atom A near Bi$_2$ square network, in addition to magnetic ordering is essential for formation of Dirac cones in ABX$_2$ materials.

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