Enhancing Thermopower and Nernst Signal of High-Mobility Dirac Carriers by Fermi Level Tuning in the Layered Magnet EuMnBi₂

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Dirac/Weyl semimetals hosting linearly dispersing bands have received recent attention for potential thermoelectric applications, since their ultrahigh-mobility carriers could generate large thermoelectric and Nernst power factors. To optimize these efficiencies, the Fermi energy needs to be chemically controlled in a wide range, which is generally difficult in bulk materials because of disorder effects from the substituted ions. Here it is shown that the Fermi energy is tunable across the Dirac point for layered magnet EuMnBi₂ by partially substituting Cd³⁺ for Eu²⁺ in the insulating block layer, which dopes electrons into the Dirac fermion layer without degrading the mobility. Clear quantum oscillation observed even in the doped samples allows to quantitatively estimate the Fermi energy shift and optimize the power factor (exceeding 100 μW K⁻² cm⁻¹ at low temperatures) in combination with the first-principles calculation. Furthermore, it is shown that Nernst signal steeply increases with decreasing carrier density beyond a simple theoretical prediction, which likely originates from the field-induced gap reduction of the Dirac band due to the exchange interaction with the Eu moments. Thus, the magnetic block layer provides high controllability for the Dirac fermions in EuMnBi₂, which would make this series of materials an appealing platform for novel transport phenomena.

1. Introduction

In the last 15 years, conducting materials, whose low energy excitation exhibits linear energy-momentum dispersion of relativistic Dirac fermions, have attracted much attention due to their extremely high mobility and unconventional quantum transport properties.[1] While early researches mainly focused on 2D systems, such as graphene[2] and surface states of topological insulators,[3] more recently the 3D bulk analogues called Dirac/Weyl semimetals have been intensively explored to discover various exotic physical properties.[4,5] These include chiral anomaly,[6–9] giant magnetoresistance,[10,11] and large anomalous Hall effect,[12–16] drawing attention for potential applications to advanced spin-electronic devices. Taking advantage of the bulk form, the Dirac/Weyl semimetals should also be promising for thermoelectric application,[17] since the high mobility carriers are able to achieve an excellent power factor (PF) as is the case for most of the conventional thermoelectric materials. For Dirac semimetal Cd₃As₂, for instance, a dimensionless figure of merit (ZT) reaches ~0.2 around 350 K, which is further enhanced to ZT ≈ 1 by applying magnetic fields up to 7 T owing to the strong magneto-thermopower effect.[18] Furthermore, a recent theoretical study suggested that the low-dimensional (e.g., 1D or anisotropic 2D) gapped Dirac dispersion is even more ideal, because a large density of states and a high group velocity can coexist.[19] In experiments, one-dimensional Dirac semimetal (Ta,Nb)₅Si₄Te₄ was found to show a large PF at low temperatures.[20–23]

Besides the thermoelectricity, the Dirac linear dispersion could also show high-efficient transverse thermoelectricity, that is, a large Nernst effect.[22,23] which was so far intensively investigated mainly for Bi[24–26] and graphite.[27,28] In these materials, the high carrier mobility as well as low carrier density plays a vital role, since the conventional Nernst coefficient roughly tracks \( \alpha_k E_F \tau \) where \( \alpha_k \) is the cyclotron frequency, \( \tau \) the relaxation time, and \( E_F \) the Fermi energy. Many of the Dirac/Weyl semimetals, such a condition is well satisfied and hence the large Nernst effects were recently observed (e.g., in Pb₁₋ₓSnₓSe,[29] Cd₁₋ₓAsₓ,[31,32] Ta(As,P),[33] and (Zr,Hf)Te[34,35]). In addition to the aforementioned conventional term, the anomalous term arising from the nonzero Berry curvature associated with the Dirac/Weyl points could also contribute to the Nernst signal (Sₜₜ), when \( E_F \) is located close to the Dirac/Weyl
point. The rapid increase in $S_{xx}$ at low fields was observed in some materials, which is likely relevant to the anomalous term.

For the optimization of the thermoelectric and Nernst power factors, wide-range as well as precise tuning of $E_F$ by chemical substitution is indispensable. In 3D bulk materials, however, the substituted ions tend to degrade the carrier mobility and/or modify the peculiar band structure, resulting in a significant reduction in PF. To overcome this issue, the layer structure consisting of the insulating and conducting layers that are spatially separated is of advantage, since the insulating layer works as a charge reservoir without disturbing the conducting layer (known as the block-layer concept or nano-block integration). We here propose layered material $\text{AMn}X_2$ ($A$: alkaline and rare earth ions, $X$: Sb, Bi) can realize this concept in the Dirac/Weyl semimetals, which consists of the alternative stack of the $X^-$ square net layer hosting quasi 2D Dirac fermions and the (Mott) insulating $\text{A}^2\text{+}-\text{Mn}^2\text{+}-\text{X}^3\text{–}$ block layer (Figure 1a). Among them, we focus on $\text{EuMnBi}_2$ as a parent material, since the quasi-2D Dirac bands, hosting an energy gap ($\approx 50$ meV) due to the strong spin-orbit coupling of Bi, cross $E_F$ without contamination of other trivial bands (Figure 1c). This is confirmed by the previous experiments, such as the transport measurement and the photoemission spectroscopy. In this study, we have demonstrated the chemical tuning of $E_F$ across the Dirac point for $\text{EuMnBi}_2$ by partially substituting Eu$^{2\text{+}}$ with Gd$^{3\text{+}}$ to dope electrons with keeping the same $4f^7$ configuration. Reflecting the validity of the block layer concept, the high mobility is retained even for the doped samples, allowing us to quantitatively estimate the $E_F$ shift by observing the quantum oscillation. Based on this, we have experimentally revealed the overall $E_F$ dependence of thermopower ($S_{xx}$) and Nernst signal ($S_{yx}$), which is compared with the theoretical calculation.

2. Results and Discussion

2.1. Determination of Fermi Energy Shift

We synthesized Gd-doped $\text{EuMnBi}_2$ single crystals by changing the nominal Gd concentration of the starting materials (see Methods). Although the energy dispersive x-ray analysis

![Figure 1. Fermi level control by Gd substitution in EuMnBi2.](image-url)
indicated the presence of Gd in the crystals, it was impossible to quantitatively determine the Gd concentration for the low-doped samples by this technique (see Supporting Information). Therefore, we have specified the variation in $E_F$ among the samples based on their transport properties, such as the Shubnikov-de-Haas (SdH) oscillations and Hall effects, as detailed below. We thereby label the Gd-doped samples as Gd#1–#6 in order of increasing $E_F$ (from low to high).

Figure 1d–i shows the field dependence of in-plane Hall resistivity $\rho_{yx}$ at 2 K for (Eu,Gd)MnBi$_2$ single crystals. For undoped EuMnBi$_2$, $\rho_{yx}$ has a positive and slightly non-linear slope with respect to field (Figure 1d), indicating the hole-type carriers are doped probably owing to the chemical defects and vacancies. Reflecting the decrease in hole-type carriers (i.e., the increase in $E_F$) with Gd substitution, the slope of $\rho_{yx}$ progressively increases for Gd#1–#3, while it suddenly decreases for Gd#4. It is likely that $E_F$ for Gd#4 is so close to the valence band top that the negative contribution from the electron-type carriers may compensate the positive $\rho_{yx}$ slope. For Gd#5, which is the sample from the same batch as Gd#4, the slope of $\rho_{yx}$ changes to negative, indicating the sign of Hall coefficient is sensitive to a small change in Gd concentration for these samples. For Gd#6, which was grown with the highest nominal Gd concentration, the magnitude of negative $\rho_{yx}$ slope is reduced to $\sim$1/100 of that for Gd#5 (inset to Figure 1i), consistent with the further increase in electron-type carriers. Thus, the measurements of $\rho_{yx}$ have qualitatively revealed the variation of the density and polarity of carriers upon Gd substitution.

It is important that the high mobility for the Dirac fermion on the Bi$^-$ layer remains essentially intact against the Gd substitution in the block layer, leading to the clear SdH oscillations for all the hole-doped samples (Figure 1d–i and insets therein). The increase in $E_F$ with Gd substitution is quantitatively estimated from the variation of the SdH frequency $B_F$, which is proportional to the extremal Dirac surface area. For the undoped and Gd#1–#3 samples, the SdH oscillations are extracted from $-d\rho_{yx}/d(1/B)^2$ at low fields (2.5–5 T) below the spin-flop transition of the Eu layer$^{[46,54]}$ (insets to Figure 1d–g), whereas for Gd#4 is discernible only at high fields (6.5–23 T) (inset to Figure 1h. See also Supporting Information). The resultant Landau fan plot is shown in Figure 1b, where the $B_F$ value deduced from the slope systematically decreases from $B_F = 21$ T (undoped) to $B_F = 9.2$ T (Gd#4). In inset to Figure 1b, the calculated $E_F$ versus $B_F$ is denoted by a solid curve, which is roughly expressed by $E_F \propto \sqrt{B_F}$ reflecting the linear dispersion. By plotting the experimental $B_F$ values on this curve, $E_F$ is estimated to shift from $\sim$39 meV (undoped) to $\sim$25 meV (Gd#4), where $E_F$ is measured from the valence-band top (inset to Figure 1c).

### 2.2. Thermoelectric Performance

Having demonstrated the systematic tuning of $E_F$, we show the corresponding variation of thermoelectric properties for (Eu,Gd)MnBi$_2$ single crystals. Figure 2 presents the $E_F$ dependence of the temperature profile of in-plane resistivity $\rho_{xx}$ (top panels), $S_{xx}$ (middle), and $S_{yx}$ (bottom) for (Eu,Gd)MnBi$_2$, where $E_F$ increases from Figure 2a (undoped) to Figure 2g (Gd#6). For undoped EuMnBi$_2$, in spite of the nice metallic behavior ($\rho_{xx} \approx 90 \mu\Omega \cdot cm$ at 300 K), the $S_{xx}$ value is fairly large, reaching the maximum of 70 $\mu$V K$^{-1}$ at 100 K (middle panel in Figure 2a). Note here that the temperature profile of $S_{xx}$ for normal metals monotonically increases with increasing temperature, which is not the case for (Eu,Gd)MnBi$_2$. The reduction in $S_{xx}$ at high temperatures likely arises from the negative contribution from the thermally-excited carriers in the conduction band, as is reproduced by the first-principles calculations (vide infra). When $E_F$ increases for the hole-doped samples, $S_{xx}$ shows an initial increase (Gd#1) followed by a gradual decrease (Gd#2–#4), while leaving the temperature dependence almost unchanged. For Gd#5, $S_{xx}$ changes to negative, while the temperature dependence of the absolute value still remains similar to that for the hole-doped samples. For heavily electron-doped Gd#6, however, $|S_{xx}|$ is much reduced ($\approx 10$ $\mu$V K$^{-1}$ at 300 K), showing a monotonic increase with temperature as is the case for normal metals.

To discuss the detail of the $E_F$ dependence of $S_{xx}$, we shall compare the experimental results with the calculated ones. We here focus on the hole-doped samples, where the carrier density $n$ as well as $E_F$ is quantitatively estimated from the SdH oscillation. Note that $n$ is defined as the summation of the electron density (positive sign) and hole density (negative sign), which are calculated based on the $E_F$ value (Figure 1c). Figure 3a shows the $n$ dependence of $S_{xx}$ in the hole-doped region at selected temperatures. At 300 and 150 K, the experimental data (open squares) are semi-quantitatively reproducible by the first-principles calculation (solid curves), indicating the optimum $n$ value is theoretically predictable. At 75 K, on the other hand, the calculation significantly overestimates the experimental results, especially when $n$ decreases down to $\approx 2.0 \times 10^{20}$ cm$^{-3}$. The decrease in positive $S_{xx}$ value with $E_F$ approaching the Dirac point arises from the negative contribution from the thermally-excited electron-type carriers in the conduction bands, which include not only the Dirac cone but also the parabolic band at the M point (Figure 1c). However, the contribution from the latter does not strongly affect the $n$ dependence of total $S_{xx}$, as was revealed by the additional calculation taking account of the shift of the parabolic band relative to the Dirac point (see Supporting Information). Therefore, the disagreement in $S_{xx}$ can be attributed to the contribution from the conduction band of the Dirac cone, the position of which may not be precisely reproduced by the present calculation.

In addition to this, the $k$-dependence of $\tau$, which is neglected in the constant-$\tau$ approximation, may not be negligible when $E_F$ approaches the Dirac point. To reveal the impact of $\tau$ on $S_{xx}$, the first-principles calculation of $\tau$ is necessary. However, it remains challenging owing to the huge computational cost$^{[55]}$ since the present material requires the spin-orbit coupling and the magnetism to be included in the calculation, otherwise the band gap is closed and the transport calculation fails.

Taking advantage of chemically tunable $S_{xx}$ presented above, we are able to optimize the thermoelectric PF (= $S_{xx}^2/\rho_{xx}$). Figure 3b shows the temperature dependence of PF for the hole-doped samples, where PF exhibits a peak around 50–75 K reflecting the temperature dependence of $S_{xx}$. $\rho_{xx}$ persistently shows nice metallic behavior regardless of the Gd concentration (top panels in Figure 2). For Gd#1–#3, in particular, the residual...
resistivity ratio $[\text{RRR} = \rho_{300 \text{K}}/\rho_{2 \text{K}}]$, which is a measure of the mean free path ($\propto \tau$), is almost unchanged from the undoped value (top panels in Figure 2a–d) and Table S1, demonstrating that the block layer works as an ideal charge reservoir for the Dirac fermion layer. Consequently, the $n$ dependence of PF is similar to that of $S_{xx}$, resulting in the highest PF for Gd#1 at all temperatures. The peak value of PF reaches 105 $\mu$W K$^{-2}$ cm$^{-1}$ at 60 K, which is comparable to that for the best $p$-type thermoelectric materials, such as Na$_2$CoO$_2$ ($\approx 160$ $\mu$W K$^{-2}$ cm$^{-1}$ at 75 K$^{[29]}$ and Ti-doped Nb$_4$SiTe$_4$ hosting 1D Dirac-like band ($\approx 60$ $\mu$W K$^{-2}$ cm$^{-1}$ at 210 K$^{[22]}$).

2.3. Nernst Signal Enhancement toward the Dirac Point

In addition to $S_{xx}$, $S_{yx}$ is also tunable with Gd substitution, although the $E_F$ dependence is totally different from that of $S_{xx}$ (bottom panels in Figure 2). For the hole-doped samples (Figure 2a–e), the temperature profile of $S_{yx}$ at 9 T exhibits the maximum at some temperature below 100 K, besides a weak anomaly at the antiferromagnetic transition temperature of the Eu layer ($T_N \approx 22$ K). (See also the inset to Figure 4 for the magnification of the low-temperature profiles) Noteworthy is that the peak value of $S_{yx}$ ($S_{yx}^{\text{max}}$) progressively increases as $E_F$ approaches the Dirac point; $S_{yx}^{\text{max}}$ reaches 50 $\mu$V K$^{-1}$ for Gd#3, which is approximately five times as large as $S_{yx}^{\text{max}}$ for the undoped sample. For Gd#4 located closest to the Dirac point, however, $S_{yx}^{\text{max}}$ suddenly decreases to 24 $\mu$V K$^{-1}$, followed by the negligibly small $S_{yx}$ values ($<1.5$ $\mu$V K$^{-1}$) in the entire temperature range for the electron-doped samples (Figure 2f,g). For a simple 2D Dirac band, the calculation predicts that $S_{yx}$ is an even function of $E_F$, showing a large positive peak at the Dirac point$^{[58,59]}$ which cannot reproduce the observed electron-hole asymmetric $S_{yx}$. Considering that there are some less-dispersive parabolic conduction bands just above $E_F = 0$ (e.g., at the M point shown in Figure 1c$^{[18]}$), the vanishingly small $S_{yx}$ for $E_F > 0$ may be caused by the multi-band effect. To explain the $E_F$ dependence of $S_{yx}$ for electron doped samples, therefore, the calculation taking account of the multi-band nature would be necessary, which remains as a future work.

We now discuss the $E_F$ dependence of $S_{yx}$ for the hole-doped samples in details, based on a theoretical model considering a massless Dirac band$^{[29,60]}$. It is known that this model reproduces the temperature and field dependence of $S_{yx}$ for Weyl semimetal NbP$^{[28]}$ specifically, it reproduces the low-temperature peak in $S_{yx}$ as was also observed for the present materials (inset to Figure 4). From the calculation, the peak value and the peak temperature ($T^{\text{max}}$) are given by $S_{yx}^{\text{max}} \propto v_F^2 / E_F$ and by $T^{\text{max}} \propto E_F$, respectively, where $v_F$ is the Fermi velocity. In Figure 4, we plot the experimental $S_{yx}^{\text{max}}$ and $T^{\text{max}}$ values versus $E_F$, together with the theoretical curves of the above relations. Note here the proportional constants of the theoretical curves are determined so as to reproduce the data for the undoped sample (with the largest $E_F$). Although the experimental $T^{\text{max}}$ data deviate downward from the theoretical line, its overall $E_F$ dependence roughly tracks $T^{\text{max}} \propto E_F$ (Figure 4b). On the other hand, the $S_{yx}^{\text{max}}$ data largely deviate upward from the curve of $S_{yx}^{\text{max}} \propto 1/E_F$ (Figure 4a). For the linear Dirac band, since $v_F$ is independent of $E_F$, this suggests $\tau$ progressively increases with $E_F$ approaching the Dirac point for Gd#1-#3.
followed by a sudden drop for Gd#4. In fact, a marked decrease in $\tau$ for Gd#4 is also suggested in the $\rho_{xx}$ data; the RRR value (a measure of $v_F \tau$) as well as the low-temperature mobility is much reduced for Gd#4 in comparison to the undoped and Gd#1–#3 samples (top panel in Figure 2e and Table S1, Supporting Information). Therefore, the decrease in $S_{yx}$ for Gd#4 is likely to arise mainly from the decrease in $\tau$. On the other hand, the RRR value and low-temperature mobility are almost unchanged from the undoped sample to Gd#3 (Table S1, Supporting Information). The anomalous enhancement of $S_{yx}$ max for these samples thus cannot be well explained by the simple model adopted here.

2.4. Impacts of Eu Magnetic Moments

As a plausible origin for this, we discuss the impact of the Eu local moments on $S_{yx}$. Above $T_N$ ($\approx 22$ K), the Eu moments exhibit isotropic paramagnetic behavior following the Curie-Weiss law.[46] Near $T_{max}$ ($\approx 80$ K), appreciable net magnetization is induced by the field of 9 T (e.g., 1.7 $\mu_B$/Eu at 50 K), which generates large spin splitting in the gapped Dirac bands owing to the exchange interaction between the Dirac fermions and Eu local moments. From the first-principles calculation,[53] the magnitude of spin splitting is roughly estimated to be $\approx 20$ meV for 1.7 $\mu_B$/Eu, significantly reducing the energy gap ($\approx 50$ meV). For a 2D Dirac band, it is theoretically revealed that the non-zero energy gap suppresses the steep increase in $S_{yx}$ toward the Dirac point.[58] Therefore, the magnetization-induced gap reduction may weaken such suppression, leading to the additional enhancement in $S_{yx}$ max observed for Gd#1–#3.

We have also observed a marked impact of the Eu local moments on $S_{xx}$ at 9 T (open circles in the middle panels of Figure 2a–e). The positive $S_{xx}$ values for the hole-doped samples are significantly reduced by the field of 9 T at mid-to-high temperatures. The magnitude as well as temperature range of such negative magneto-thermopower evolves with decreasing $|E_F|$ (from undoped to Gd#4). These facts are again consistent with the band gap reduction at 9 T, since it promotes the negative thermopower from the conduction band. Note here that the negative magneto-thermopower changes to the positive one at low temperatures. The latter can be explained by the Mott’s relations within the conventional semiclassical Drude-Boltzmann theory,[30] as is often observed in other Dirac/Weyl semimetals with high carrier mobility.[30,31,34] For (Eu,Gd)MnBi 2, the mobility is enhanced at low temperatures (especially below $T_N$), where the magnitude of the positive magneto-thermopower could overcome the negative one. Thus, the magnetic block layer in the
present materials plays a role not only as a source of charge carriers to tune $E_F$, but also as a source of exchange interaction to tune the band gap, which would enable us to improve the thermoelectric and Nernst efficiencies in various ways.

3. Conclusion

In conclusion, we have demonstrated the systematic control of Fermi energy for layered Dirac material EuMnBi$_2$ by partially substituting Gd$^{3+}$ for Eu$^{2+}$ in the block layer. Since the high carrier mobility is retained even for the doped samples, we observed the clear quantum oscillation and thereby quantitatively estimate the carrier density and the Fermi energy. Wide-range chemical control of the Fermi energy across the Dirac point enables us to experimentally clarify the overall variation of the transport and thermoelectric properties. The Fermi energy dependence of thermopower is roughly reproduced by the first-principles calculation, achieving the optimized power factor of more than 100 $\mu$W K$^{-2}$ cm$^{-1}$ around 50 K. On the other hand, the Fermi energy dependence of Nernst signal shows an anomalous increase toward the Dirac point, which cannot be explained within a simple rigid-band scheme. The calculation suggests that the field-induced Eu magnetization reduces the gap of the Dirac band via the exchange interaction, which may strongly affect the Fermi energy dependence. Therefore, the seven occupied 4$d$-orbitals so that the calculated band structure exhibits good consistency with those calculated with the hybrid functionals. For the transport calculation using the PBE+U method shown in Supporting Information, the band structure was calculated including the spin–orbit coupling as shown in Figure S4f, Supporting Information, the simplified rotationally-invariant formulation introduced for the PBE+U model were performed using very fine k-meshes from 600 $\times$ 600 to 960 $\times$ 960 $\times$ 40.

4. Experimental Section

Experimental Details: Single crystals of (Eu,Gd)MnBi$_2$ were grown by a Bi self-flux method. High purity ingots of Eu (99.9%), Gd (99.9%), Mn (99.9%), Bi (99.999%) were mixed in the ratio of Eu : Gd : Mn : Bi = 1 : x : 1 : 9 and put into an alumina crucible in an argon-filled glove box, where $x$ is 0, 0.005, 0.0075, 0.01, 0.06, and 0.1 for the undoped, Gd#1, Gd#2, Gd#3, Gd#4–#5, and Gd#6 samples, respectively. The crucible was sealed in an evacuated quartz tube and heated at 1000 °C for 10 h, followed by slow cooling to 350 °C at the rate of $\approx$ 2 °C h$^{-1}$, where the excess Bi flux was decanted using a centrifuge. The powder x-ray diffraction profile at room temperature indicates that the crystal structure of the obtained single crystals is tetragonal ([14]/mmm) and the lattice constants are almost unchanged irrespective of the nominal Gd concentration.

$\rho_{xx}$ and $\rho_{yx}$ were measured by a conventional 5-terminal method with electrodes formed by room-temperature curing silver paste. $S_x$ and $S_y$ were simultaneously measured by a steady-state method with a temperature difference ($\Delta T$) of $<$ 1 K (typically 2–4%) of the measurement temperature below 50 K) between the longitudinal voltage contacts. Note here $S_x = (V_x/\Delta T)(L_y/L_x)$, where $V_x$ is the field-induced transverse voltage, $L_x$ the distance between the longitudinal contacts, and $L_y$ the distance between the transverse contacts. The measurements were performed from 2 to 300 K at 9 T (B||c), using Physical Properties Measurement System (Quantum Design). For Gd#4, the resistivity up to 23 T at 1.4 K was measured using the non-destructive pulsed magnet with a pulse duration of 36 msec at the International Mega-Gauss Science Laboratory at the Institute for Solid State Physics. The voltage signal was measured by a lock-in technique at 100 kHz with the ac excitation of 1–10 mA.

Computational Details: The authors performed first-principles band structure calculations using the density functional theory with the Perdew–Burke–Ernzerhof parameterization of the generalized gradient approximation (PBE-GGA) and the projector augmented wave (PAW) method as implemented in the Vienna ab initio simulation package. The open-core treatment was applied for the Eu$^{2+}$ ions, to say, the seven occupied 4$f$ orbitals (4$f^7$) for Eu$^{2+}$ ion were included into the core of the PAW potential. The spin–orbit coupling was included. The G-type antiferromagnetic order of Mn atoms as observed in experiment was assumed. The plane-wave cutoff energy of 350 eV and a 16 $\times$ 16 $\times$ 16 k-mesh were used. Lattice parameters and atomic coordinates were taken from experiment.

After the band structure calculation, the $M_n$d + Bi-p Wannier functions extracted from the calculated band structure using the WANNIER90 code. For a technical reason, the conventional (tetragonal) unit cell with a 16 $\times$ 16 $\times$ 4 k-mesh was used for this purpose. Then, a tight-binding model was constructed with the obtained hopping parameters among the Wannier functions. Using this model, the chemical potential was determined so as to reproduce the experimental values of $V_y$. Here, $B_x$ on constant-k planes was evaluated and then an average was taken with respect to $k_x$.

The carrier density and the Seebeck coefficient were also evaluated using this model and the estimated chemical potential. The Seebeck coefficient was calculated on the basis of the Boltzmann transport theory with a constant $\tau$ approximation. Note that the Seebeck coefficient does not depend on $\tau$ if $\tau$ is assumed to be constant. Here an upward shift was applied for the conduction bands by 30 meV, because PBE-GGA is known to underestimate the band gap. This shift value was determined so that the calculated Seebeck coefficients agree well with experimental ones. These calculations using the tight-binding model were performed using very fine k-meshes from 600 $\times$ 600 $\times$ 40 to 960 $\times$ 960 $\times$ 40.

For further calculations shown in Supporting Information, other exchange-correlation functionals were also used. For calculating the band structure shown in Figure S4a–d, Supporting Information, an 8 $\times$ 8 $\times$ 8 k-mesh was used without including the spin–orbit coupling, because of the high computational cost for hybrid-functional calculations. Here the B3LYP and HSE06 functionals were used. For the PBE+U calculation shown in Figures S4 and S5, Supporting Information, the simplified rotationally-invariant formulation introduced by Dudarev et al. was adopted with $U_{eff} = U - J = 5$ eV for the Mn d-orbitals so that the calculated band structure exhibits good consistency with those calculated with the hybrid functionals. For the transport calculation using the PBE+U method shown in Figure S5, Supporting Information, the band structure was calculated including the spin–orbit coupling as shown in Figure S4f, Supporting Information, extracted the Wannier orbitals, constructed a tight-binding model using these Wannier orbitals, applied the same energy shift as PBE (30 meV) to the conduction bands calculated using this model, and calculated the Seebeck coefficient based on the Boltzmann transport theory. All of these procedures for transport calculation were done using the same computational conditions as the PBE calculation.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Fermi level tuning, magnetic Dirac semimetal, Nernst signal, quantum oscillation, thermopower

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