For the innovation of spintronic technologies, Dirac materials, in which low-energy excitation is described as relativistic Dirac fermions, are one of the most promising systems because of the fascinating magnetotransport associated with extremely high mobility. To incorporate Dirac fermions into spintronic applications, their quantum transport phenomena are desired to be manipulated to a large extent by magnetic order in a solid. We report a bulk half-integer quantum Hall effect in a layered antiferromagnet EuMnBi₂, in which field-controllable Eu magnetic order significantly suppresses the interlayer coupling between the Bi layers with Dirac fermions. In addition to the high mobility of more than 10,000 cm²/V s, Landau level splittings presumably due to the lifting of spin and valley degeneracy are noticeable even in a bulk magnet. These results will pave a route to the engineering of magnetically functionalized Dirac materials.

INTRODUCTION

A conductive material with magnetic order is an integral component for spintronic devices, such as spin valves and spin transistors (1). There, charge transport correlated with magnetism, such as giant magnetoresistance effect, enables high-speed and/or nonvolatile device operations. Dirac fermions with linear energy dispersion with momentum space have been of current interest for spintronic applications because a variety of quantum transport phenomena manifest themselves in an external magnetic field due to extremely high mobility. A typical example is an unusual half-integer quantum Hall effect (QHE) (2, 3) that is observable even at room temperature in graphene (4). More recently, the quantum anomalous Hall effect was observed for the surface Dirac state in magnetic topological insulator thin films (5, 6). To expand potential application of such a distinct quantum transport enriched by magnetic order, it is highly desirable to explore bulk systems that host various magnetism and dimensionality.

Despite recent discovery of a number of new bulk Dirac materials, novel quantum transport features have been elucidated mostly in non-magnetic materials, as exemplified by so-called Dirac/Weyl semimetals, such as Cd₃As₂ (7–9), Na₃Bi (10), and TaAs (11). Since the emergence of the three-dimensional (3D) Dirac-like dispersion stems from specific lattice symmetry for the above materials, it would be, in principle, impossible to substitute the constituent elements with magnetic ones while keeping the crystal structure. As strongly correlated magnetic systems, on the other hand, certain heavy transition metal oxides, such as pyrochlore iridates, have been predicted to have Weyl semimetallic states (12). At present, however, quantum transport phenomena associated with Dirac-like quasi-particles remain experimentally elusive.

In this context, layered compound AMnBi₂ [A = Sr²⁺ (13, 14) and Eu²⁺ (15)] would provide an ideal arena to reveal the interplay between Dirac fermions and ordered magnetic moments. This is because the conducting layers of Bi square net hosting quasi-2D Dirac fermions and the insulating magnetic layers consisting of the Mn-Bi and A layers are spatially separated (Fig. 1E), where we can develop a variety of magnetic layers while keeping the Dirac-like band structure. For EuMnBi₂, a signature of coupling between charge transport and magnetism was recently discerned upon the magnetic order of Eu moments (spin S = 7/2 for Eu²⁺) adjacent to the Bi layer (15). By applying fields up to 55 T that enable complete control of the magnetic order of Eu sublattice, we here demonstrate its strong impact on interlayer hopping of quasi-2D Dirac fermions on the Bi layer, which gives rise to the multilayer quantum Hall state.

RESULTS AND DISCUSSION

As shown in Fig. 1A, the magnetic susceptibility M/H parallel to the c axis for EuMnBi₂ steeply decreases below the antiferromagnetic (AFM) transition temperature T_N ~22 K, indicating that the Eu moments are aligned parallel to the c axis (15). To reveal the AFM arrangement of the Eu sublattice, we have measured the resonant x-ray scattering spectra near the Eu L₃ absorption edge. At 5 K, we found the (0 0 1 1) reflection at $E = 6.975$ keV that is forbidden in the present space group (I4/mmm) (inset to Fig. 1B). Considering the evolution of the reflection intensity below T_N (Fig. 1B) and the observation of polarization rotation as well as a sharp resonance at the Eu L₃ edge (fig. S1), it can be assigned to resonant magnetic scattering from the Eu sublattice. On the basis of the analyses on several magnetic reflections (see figs. S2 and S3), we derive the most probable magnetic structure as shown in Fig. 1E. The Eu moments order ferromagnetically in the
The resistivity data for the field perpendicular to the c axis (\(H_{||}c\)) at 0.1 T (blue) and 7 T (red). Open symbols are the data for the field perpendicular to the c axis (\(H_{\perp}c\)) at 0.1 T. (B) Intensity of resonant magnetic reflection (0 0 11) at 6.975 keV at 0 T. The inset shows the profile of the (0 0 11) reflection along [001] at 6.975 keV (resonant) and 7.00 keV (nonresonant). In-plane resistivity \(\rho_{xx}\) (C) and interlayer resistivity \(\rho_{zz}\) (D) at 0 and 9 T (\(H_{||}c\)). Schematic sample configuration for the resistivity measurement is shown in each panel, emu/mol, electromagnetic unit per mole; a.u., arbitrary unit. (E) Schematic illustration of the plausible magnetic structure for EuMnBi\(_2\) at zero field, together with the formal valence of each ion. The arrangement of the Mn sublattice is assumed to be the same as in SrMnBi\(_2\) (45). (F) Magnetic phase diagram for the Eu sublattice as functions of field (\(H_{||}c\)) and temperature. PM and AFM denote the paramagnetic and antiferromagnetic states, respectively. \(H_H\) and \(H_F\) correspond to the transition fields to the spin-flop AFM and PM (forced ferromagnetic) phases, respectively. Black arrows are schematic illustration of the Eu moments sandwiching the Bi\(^{3-}\) layer. Note the Mn sublattice orders at ~315 K (>\(T_N\)).

At 9 T, \(\rho_{xx}(T)\) exhibits marked positive magnetoresistance effects that evolve with decreasing temperature down to ~40 K, followed by a steep drop at \(T_N\). On the other hand, \(\rho_{zz}(T)\) at 9 T shows minimal (longitudinal) magnetoresistance effects above \(T_N\), but shows a much larger jump on cooling at \(T_N\) than that at 0 T. These suggest that the increase of anisotropy in resistivity below \(T_N\) is further enhanced at 9 T; the increase in \(\rho_{zz}/\rho_{xx}\) with decreasing temperature from 25 K (just above \(T_N\)) to 2 K exceeds 1000% at 9 T, whereas it is approximately 180% at 0 T. Judging from the temperature profile of \(M/H\) at 7 T for \(H_{||}c\) in Fig. 1A (and also magnetic phase diagram in Fig. 1F), the Eu moments are oriented perpendicular to the c axis in the AFM phase at 9 T, which appears to strongly suppress the interlayer conduction between the Bi layers. We will again discuss the effect of the Eu spin flop on the resistivity in terms of its field profile (vide infra).

The magnetotransport properties enriched by the Eu magnetic order are further highlighted by the magnetization and resistivity measured in the magnetic field up to 55 T applied along the c axis (Fig. 2). The magnetization at 1.4 K exhibits a clear magnetomagnetic (spin-flop) transition at \(H = H_{||}c\) (~5.3 T), corresponding to the reorientation of the Eu moments to be perpendicular to the field (Fig. 2A). In the forced ferromagnetic state above \(H_{||}c\) (~22 T), the magnetization is saturated close to 7 \(\mu_B\), reflecting the full moment of localized Eu 4f electrons. The temperature variation of \(H_{||}c\) and \(H_{\perp}c\) is plotted in Fig. 1F (see fig. S4 for details), which forms a typical phase diagram for an anisotropic antiferromagnet in the field parallel to the magnetization-easy axis.

The interlayer resistivity is markedly dependent on the AFM states of the Eu sublattice (Fig. 2B). Above \(T_N\) (at 27 and 50 K), \(\rho_{zz}\) is almost independent of field, except for clear Shubnikov–de Haas (SdH) oscillations at 27 K. At 1.4 K, on the other hand, \(\rho_{zz}\) exhibits a large jump at \(H_{||}c\) followed by giant SdH oscillations that reach \(\Delta \rho_{zz}/\rho ~50\%\). This high-\(\rho_{zz}\) state is terminated at \(H_{\perp}c\), above which the \(\rho_{zz}\) value is substantially reduced. The origin of such \(\rho_{zz}\) enhancement (that is, suppression of interlayer coupling) in the spin-flop phase remains as an open question at present: the interlayer charge transfer caused by the electron’s hopping on the local Eu moments would not change, if Eu moments were simply reoriented perpendicular to the c axis while keeping the same AFM pattern. We should note here that the Mn sublattice that antiferromagnetically orders at ~315 K (15) as well as the Eu one play a vital role in achieving the high-\(\rho_{zz}\) state. As shown in the inset in Fig. 2B, the \(\rho_{zz}\) value at 0 T for EuZnBi\(_2\) is analogous to that in the spin-flop phase for EuMnBi\(_2\) (that is, the Eu moments are aligned in the ab plane with staggered stacking along the c axis; see fig. S5E). For SrMnBi\(_2\), on the other hand, the \(\rho_{zz}\) value at 0 T is comparable to that for EuMnBi\(_2\) but shows a minimal magnetoresistance effect up to 9 T. The magnetic order in both the Eu and Mn sublattices is thus essential for enhancing \(\rho_{zz}\). As a possible model based on these facts, the magnetic order of the Mn sublattice might be significantly modulated upon the Eu spin flop due to the f-d coupling. It is also likely that we need to take into consideration the anisotropy of Eu\(^{3+}\) 4f orbitals induced by the crystal field splitting (16), which might reduce wave function overlap with the Mn sites along the c axis when the Eu moment and orbital rotate. Revealing the detailed mechanism would, however, be an issue for future experimental and theoretical works.

Another important feature is that the \(\rho_{zz}\) peak around 20 T shows a sizable hysteresis between the field-increasing and field-decreasing runs. [Correspondingly, a hysteretic anomaly also manifests itself in \(\rho_{xx}\) (Fig. 2C).] Because no clear anomaly is discerned in the magnetization curve around 20 T (fig. S6), the Eu moments play a minor role;
instead, a possible transition between the Landau levels with different spin orientation might be responsible for this hysteresis, as discussed below.

The in-plane resistivity exhibits a large positive (transverse) magnetoresistance effect, irrespective of the Eu magnetic order (Fig. 2C). At 50 K, the $\rho_{xx}(H)$ profile is strikingly $H$-linear without saturation up to 35 T, resulting in the magnetoresistance ratio of $\rho(H = 35 \text{ T})/\rho(0) \sim 2000\%$. Such large $H$-linear magnetoresistance is occasionally observed in Dirac semimetals (7–10, 17). At lower temperatures, the SdH oscillations are superimposed; at 1.4 K, in particular, the magnitude of oscillation is largely enhanced in the spin-flop AFM phase between $H_r$ and $H_c$, similarly to $\rho_{zz}$. The enhanced SdH oscillations in the spin-flop phase are also noticeable for the Hall resistivity $\rho_{yx}$ (Fig. 2D), which show plateau-like structures at 1.4 K. In the following, we will analyze the details of $\rho_{yx}$ plateaus in terms of the multilayer QHE in the stacking 2D Bi layers.

In Fig. 3A, we plot the inverse of $\rho_{yx}$ at 1.4 K (spin-flop phase) as a function of $B_r/B$, where $B_r$ is the frequency of SdH oscillation and $B$ is the magnetic induction. $B_r/B$ is the normalized filling factor [corresponding to $(n + \frac{1}{2})\gamma$ in Eq. 1] (18), which is used to compare the samples with different $B_r$ (Table 1). The inverse of $\rho_{yx}$ also exhibits clear plateaus at regular intervals of $B_r/B$, the positions of which nicely correspond to deep minima in $\rho_{xx}$ (Fig. 3B) and pronounced peaks in $\rho_{zz}$ (Fig. 3C). All these features signify the multilayer QHE, as previously observed for the GaAs/AlGaAs superlattice (19, 20). Although the $\rho_{xx}$ minima do not reach zero, $\omega_c\tau$ estimated from $\rho_{xx}/\rho_{zz}$ is much larger than unity (for example, $\sim 5$ at around $B_r/B = 1.5$; see fig. S7A), where $\omega_c$ is the cyclotron frequency and $\tau$ is the scattering time. What is prominent in the present compound is that the values of $1/\rho_{yx}$ are quantized to half-integer multiples, when scaled by $1/\rho_{xx}^0$, the step size of successive plateaus (see fig. S7B for definition). Given the conventional view of QHE, this quantization of $\rho_{xx}^0/\rho_{yx}$ leads to the normalized filling factor of $n + \frac{1}{2}$, where $n$ is a nonnegative integer. This is consistent with the plateaus occurring at half-integer multiples of $B_r/B$ (vertical dotted lines in Fig. 3A, where a small shift corresponds to the phase factor as explained below). Such a half-integer (normalized) filling factor is known to stem from the nontrivial $\pi$ Berry’s
Table 1. Parameters related to the SdH oscillations and quantized Hall plateaus in the spin-flop phase (at 1.4 K and 5.3 to 22 T). $B_F$ and $\gamma$ are the results of linear fit to the Landau fan plot.

<table>
<thead>
<tr>
<th>Sample no.</th>
<th>$p_{yy}$</th>
<th>$B_F$ (T)</th>
<th>$\gamma$ (phase factor)</th>
<th>Sample thickness ($\mu$m)</th>
<th>$p_{yy}^0$ ($\mu\Omega$cm)</th>
<th>$s$ (degeneracy factor)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$\rho_{xx}$ $\rho_{yx}$</td>
<td>26.1(2)</td>
<td>$-0.12(4)$</td>
<td>130</td>
<td>525</td>
<td>5.5</td>
</tr>
<tr>
<td>2</td>
<td>$\rho_{xx}$ $\rho_{yx}$</td>
<td>23.1(2)</td>
<td>$-0.12(2)$</td>
<td>78</td>
<td>578</td>
<td>5.0</td>
</tr>
<tr>
<td>3</td>
<td>$\rho_{xx}$</td>
<td>19.5(1)</td>
<td>$-0.08(2)$</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
</tbody>
</table>

The results of linear fit to the Landau fan plot.

The phase of Dirac fermions (2, 3), which in two dimensions leads to the Hall resistance quantized as follows (21, 22)

$$\frac{1}{R_{\text{yx}}} = \pm s \left( n + \frac{1}{2} - \gamma \right) \frac{e^2}{h}$$

where $e$ is the electronic charge, $h$ is the Planck’s constant, $s$ is the spin and valley degeneracy factor, and $\gamma$ is the phase factor expressed as $\gamma = \frac{\varphi}{\sqrt{\varphi^2 + \pi}}$, with $\varphi$ as the Berry’s phase (23). The observed half-integer filling factor thus corresponds to $\gamma \approx 0$, that is, nontrivial $\pi$ Berry’s phase in the present QHE.

Furthermore, following standard analyses on the SdH oscillations using fan diagram, we plot the values of 1/$\rho_{xx}$ vs $1/B$ (Fig. 3A, inset). On the basis of a semiclassical expression of oscillating part in $\rho_{xx}$ ($\propto$ $B$ $\delta$ sin $n \pi$, $\delta$ |$\Delta$ $\varphi_{\text{sc}}$| $\cos$ ($\pi$ $B$ $\delta$ $+$ $\varphi_{\text{sc}}$)), a linear fitting yields $\gamma - \delta$ close to 0 ($-0.1$) for all the samples (Table 1), where a phase shift $\delta$ is determined by the dimensionality of the Fermi surface, varying from 0 (for 2D) to $1/8$ (for 3D). Because the value of $\delta$ tends to be negligibly small for quasi-2D Fermi surfaces even in bulk materials (18, 24), the fitted results indicate $\gamma \approx 0.1$, which again verifies the nonzero Berry’s phase in this compound.

The quantization of $\rho_{yx}^0$ to half-integer multiples is well reproduced for two samples (#1 and #2 in Fig. 3A). The thickness of sample #2 is ~60% of that of #1. Nevertheless, their difference in $\rho_{xx}^0$ is only ~10%. This fact ensures that the observed Hall plateaus are of bulk origin, which should be attributed to the parallel transport of the 2D Bi layers stacking along the $c$ axis, as is the case for multilayer quantum Hall systems, including semiconductor superlattice (19, 25), Bechgaard salts (26, 27), Mo$_x$O$_{11}$ (28, 29), and Bi$_x$Se$_y$ (30). The inverse Hall resistivity is hence expressed as $1/\rho_{yx} = Z^0/\rho_{yx}$, where $Z^0 = 1/(e/2) - 8.9 \times 10^6$ (cm$^{-1}$) is the number of the Bi layers per unit thickness and $c$ is the $c$ axis length. This gives the step size between the successive $1/\rho_{yx}^0$ plateaus as $1/\rho_{yx}^0 = Z^0(c^2/h)$, from which we have estimated the degeneracy factor $s$ to be ~5 to 6, as shown in Table 1 (see also fig. S7B and the related discussions).

Provided that there exist four valleys in EuMnBi$_2$ (31) as is the case of SrMnBi$_2$ (13, 32), $s$ should be 8 (including double spin degeneracy). Even having taken into account errors in sample thickness ($\pm 10$ to 20%), the $s$ value of 8 is somewhat larger than the estimated one, which may be attributable to the inhomogeneous transport arising from dead layers and/or the imperfect contacts.

From the SdH frequencies in the spin-flop phase, we are capable of estimating the 2D carrier density per Bi layer at 1.4 K to be $n_{2D} = se_{\text{Bi}}h/4.9 \times 10^{12}$ assuming $s = 8$, which results in 3D density $n_{3D} = n_{2D}Z^0 \times 4.4 \times 10^{19}$ cm$^{-3}$ (sample #1). This is comparable to that estimated from $\rho_{xx}$ at ~20 T: $n_{\text{Bi}} = B/\rho_{xx} \approx 2.9 \pm 0.2 \times 10^{19}$ cm$^{-3}$, where errors arise from the oscillatory component. From the value of residual resistivity $\rho_0$, we have obtained the mobility $\mu = n_{3D}/\rho_0 \sim 14,000$ cm$^2$/V s at ~2 K, which attains a markedly high value despite the transport coupled with the Eu magnetic order.

As shown in Fig. 3B, the $N = 2$ Landau level clearly splits into two peaks in the second derivative of resistivity $-d^2\rho_{xx}/dB^2$, whereas the splitting for $N = 3$ is barely discernible. This Landau level splitting is likely to be more pronounced for $N = 1$ (at higher fields), supposedly forming a dip structure in $\rho_{xx}$ as well as $-d^2\rho_{xx}/dB^2$. Unfortunately, only one of the split Landau levels is accessible for $N = 1$, because the spin-flop phase is terminated at $H_c$ (a spiky peak in $-d^2\rho_{xx}/dB^2$; see also Fig. 4). With further decreasing temperature down to 50 mK, another Landau level splitting appears to evolve (thick arrow in Fig. 4). Although the origin of these splittings is unclear at present, it should be relevant to the spin and valley degrees of freedom, as is often the case in the conventional QHE in semiconductor heterostructures (33). It is surprising that such lifting of spin and valley degeneracy is clearly observed at moderately high fields (~20 T) even in the bulk system. This may be indicative of a large Landé $g$ factor and/or strong electron correlations, characteristic of Dirac fermions formed on the Bi layer (34–36).

Finally, we mention the hysteretic anomalies in $\rho_{xx}$ and $\rho_{yx}$ around 20 T (Fig. 2). It should be noted here that similar hysteretic phenomena of resistivity have been discovered in many 2D electron gas systems in both the regimes of the integer (37, 38) and fractional QHE (39, 40). Their physical origin is considered to be relevant to the crossing of Landau levels for electrons (or for composite fermions in the fractional QHE) with different spin polarization (41), where magnetic domains
are likely to form. In the present compound, because the resistivity shows substantial hysteresis near the transition between the split Landau levels (in the $N = 1$ state as shown in Fig. 4), it might originate from the dissipative conduction along such domain walls. Although detailed discussions about its mechanism are beyond the scope of the present study, the observed distinct hysteresis may suggest the possible importance of the spin polarization of Landau level for Dirac fermions.

Here, we have presented a marked tuning of magnitude in interlayer conduction of quasi-2D Dirac fermions, utilizing the AFM order of Eu moments. In addition to the staggered moment alignment along the $c$ axis, the field-induced flop of the Eu moment direction appears to further reduce the interlayer coupling and hence confine the Dirac fermions within the constituent 2D Bi layer well enough to quantize the Hall conductivity in a bulk form (42). Such a magnetically active Dirac fermion system would form a promising class of spintronic materials with very high mobility.

**MATERIALS AND METHODS**

Single crystals of EuMnBi$_2$, SrMnBi$_2$, and EuZnBi$_2$ were grown by a Bi self-flux method. For EuMnBi$_2$, high-purity ingots of Eu (99.9%), Mn (99.9%), and Bi (99.999%) were mixed in the ratio of Eu/Mn/Bi = 1:1:9 and put into an alumina crucible in an argon-filled glove box. From Le Bail fitting of the measured profiles, the lattice constants are estimated to be $a = 4.5416(4)$ Å and $c = 22.526(2)$ Å, $a = 4.5609(4)$ Å and $c = 23.104(2)$ Å, and $a = 4.6170(3)$ Å and $c = 21.354(2)$ Å for EuMnBi$_2$, SrMnBi$_2$, and EuZnBi$_2$, respectively.

At low fields, magnetization (up to 7 T) and resistivity (up to 14 T) were measured down to 1.9 K using Magnetic Property Measurement System (Quantum Design) and Physical Properties Measurement System (Quantum Design), respectively. In-plane resistivity $\rho_{xx}$ and Hall resistivity $\rho_{yx}$ were measured by a conventional five-terminal method with electrodes formed by room temperature curing silver paste (fig. S8D). The typical sample dimension is ~2.20 mm (length) ×0.5 mm (width) × 0.1 mm (thickness). The voltage terminals were needed to cover the whole thickness of the sample side to avoid the admixture of the interlayer resistance. Interlayer resistivity $\rho_{zz}$ was measured by a four-terminal method on bar-shaped samples with a typical dimension of ~1.5 mm in length along the $c$ axis and ~0.4 × 0.4 mm$^2$ in cross section (fig. S8E). Current terminals were formed so as to completely short out the in-plane current. The magnetization and resistivity up to 55 T were measured using the nondestructive pulsed magnet with a pulse duration of 36 ms at the International MegaGauss Science Laboratory at the Institute for Solid State Physics. The measurement temperature range was 1.9 to 150 K. The magnetization was measured by the induction method, using coaxial pickup coils. The resistivity ($\rho_{xx}$, $\rho_{y}$, and $\rho_{zz}$) was measured by a lock-in technique at 100 kHz with ac excitation of 1 to 10 mV. The resistivity measurement up to 28 T at ~50 mK was performed with a lock-in amplifier at 17 Hz with ac excitation of 100 μA by using a dilution refrigerator embedded in the cryogen-free hybrid magnet at High Field Laboratory for Superconducting Materials in the Institute of Materials Research, Tohoku University (44).

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at https://advances.sciencemag.org/cgi/content/full/2/1/e1501117/DC1

**REFERENCES AND NOTES**

Quantum Hall effect in a bulk antiferromagnet EuMnBi$_2$ with magnetically confined two-dimensional Dirac fermions

Hidetoshi Masuda, Hideaki Sakai, Masashi Tokunaga, Yuichi Yamasaki, Atsushi Miyake, Junichi Shiogai, Shintaro Nakamura, Satoshi Awaji, Atsushi Tsukazaki, Hironori Nakao, Youichi Murakami, Taka-hisa Arima, Yoshinori Tokura and Shintaro Ishiwata

Sci Adv 2 (1), e1501117.
DOI: 10.1126/sciadv.1501117