Transfer and conservation of angular momentum is at the heart of spintronics [1–5]: spin transfer torque works to convert spin-polarized current to and from magnons, thus enabling the electrical control of magnetism. For example, current induced magnetization reversal and spin torque ferromagnetic resonance through spin injection have been realized in various heterostructures based on a heavy metal element (with large spin-orbit coupling) and ferromagnet, such as Pt/Pt, Ta/CoFeB and Pt/Co [2–5]. Recently, it has been reported that unidirectional magnetoresistance (UMR) emerges in such heterostructures under in-plane magnetization [6–8]; the resistance value is different depending on the sign of the outer product of current J and magnetization M vectors. There, the spin accumulation direction, either parallel or antiparallel to M, at the interface by the spin Hall effect has been proposed to be a major origin of UMR [6–8], in analogy to the giant magnetoresistance (GMR) effect [9,10], which depends on the relationship of magnetizations, parallel or antiparallel, in stacked ferromagnetic metal layers. In a broader context, the UMR is expected to be further enhanced in a topological insulator (TI) with a large spin Hall angle [11–14] via spin-momentum locking [Fig. 1(a)], because the spin polarization at the surface would govern the amplitude of this effect.

In this study, we investigate the UMR of TI heterostructures [12,15,16] composed of nonmagnetic TI (Bi1−xSbx)2Te3 (BST) [17,18] and magnetic TI Crx(Bi1−xSbx)2–xTe3 (CBST) [19] on insulating InP substrate. By tuning the composition y, we could control the Fermi energy EF of the surface state inside the bulk band gap, which is confirmed by the Hall effect measurement [15–18]. Here, the main players of conduction are top and bottom surfaces with single Dirac cones around the Γ point [15,16,20,21]. In addition, in the heterostructure, only one surface involved in the Cr-doped layer effectively interacts with magnetism [15,16]. Thus, in terms of the symmetry consideration of spin-momentum locking [Fig. 1(a)], we can expect that magnetoresistance depends on the relative configuration between surface electron spin and M directions, parallel [Fig. 1(b)] or antiparallel [Fig. 1(c)].

Thin films of TI heterostructures were grown with molecular beam epitaxy in the same procedures as described in Refs. [15] and [16]. The nominal compositions of TI heterostructure Crx(Bi1−xSbx)2–xTe3/(Bi1−ySby)2Te3 are x ∼ 0.2 and y ∼ 0.86. Using photolithography and Ar ion milling, thin films were patterned into the shape of a Hall bar, 10 μm in width and 36 μm in length. After that, the electrodes Au(45 nm)/Ti(5 nm) were formed by electron beam deposition [22]. The transport measurements were performed mainly at 2 K in the Physical Property Measurement System (Quantum Design) using a dc current source and a voltmeter.

Figure 1(e) shows the measured magnetoresistance of the heterostructure CBST(3 nm)/BST(5 nm) [Fig. 1(d)]. First, we notice that resistance decreases with increasing in-plane magnetic field B. Because of the out-of-plane anisotropy of M in CBST, M initially points along the z direction forming the exchange gap in the surface Dirac state. As magnetic field is applied up to 0.7 T, the magnetization direction gradually changes to the in-plane so that the eventual gap closing of the Dirac surface state causes negative magnetoresistance [22]. Also, we note that the resistance measured under +1 μA (red) and −1 μA (blue) at 2 K shows a noticeable deviation as shown in Fig. 1(e); the difference ΔRxx between the two current directions is plotted in Fig. 1(f). Here, ΔRxx is antisymmetrized as a function of B and M. ΔRxx is initially almost zero at 0 T where M is pointing out of plane, and then increases as the field increases up to 0.7 T. At higher...
magnetic field above 0.7 T, $\Delta R_{xx}$ becomes almost constant, whose sign is reversed in accordance with $M$ reversal in CBST. Furthermore, $\Delta R_{xx}$ is also reversed in sign, as shown in Figs. 1(h) and 1(i), for the inverted heterostructure BST(3 nm)/CBST(5 nm) [Fig. 1(g)], while showing the similar absolute magnitude of UMR. This is most likely because the manner of the spin-momentum locking is opposite between the top and bottom surfaces as depicted in Figs. 1(d) and 1(g). This leads to the cancellation of UMR in the case of the single-layer CBST film [22]. Figures 1(j) and 1(k) show the current amplitude dependence of UMR. While $\Delta R_{xx}$ shows a negligibly small difference with current amplitude of 0.1 $\mu$A, it is progressively enhanced with increasing current. The current $J$ dependence of $\Delta R_{xx}$ at 0.7 T is summarized in Fig. 1(k), which shows a linear relationship in a low current region, typically $J < 0.5 \mu$A. Therefore, the relationship between electric field $E_x$ and current density $j_x$ should be expressed in a nonlinear form in such a low current region,

$$E_x = R_{xx}j_x + R_{xx}^{(2)} j_x^2. \tag{1}$$

Here, $\Delta R_{xx} = 2R_{xx}^{(2)} j_x$ is linearly proportional to current density. The derivation from the linear relationship in Fig. 1(k) at high current (> 0.5 $\mu$A) is attributed to the heating effect by fairly large current excitation, up to $\Delta T = 2.3$ K at $J = 3 \mu$A as estimated from the change of $R_{xx}$ [22]. Hereafter, we applied $\pm 1 \mu$A for the measurements to get enough $S/N$ ratio but to make the heating effect as small as possible.

In Table I, we compare the magnitude of UMR in the present device with those of previously reported

| Material                  | $j$ (A/cm$^2$) | $R_{xx}$ (Ω) | $\Delta R_{xx}$ (Ω) | $\Delta R_{xx}/R_{xx}$ (%) | $\Delta R_{xx}/j_{xx}$ (arb.units) |
|--------------------------|----------------|--------------|---------------------|-----------------------------|---------------------------------|
| Ta/Co [7]                | $10^7$         | 574          | 0.011               | 0.0019                      | 1.3                             |
| Pt/Co [7]                | $10^7$         | 176          | 0.0025              | 0.0014                      | 1                               |
| GaMnAs heterostructure [6]| $7.5 \times 10^5$ | 1720         | 2                   | 0.12                        | $1.1 \times 10^3$              |
| CBST/BST (this study)    | $5.0 \times 10^3$ * | 14000        | 57                  | 0.41                        | $5.7 \times 10^5$ *             |

TABLE I. Comparison of UMR magnitude for various heterostructures. Note that the values marked with an asterisk * for CBST/BST are calculated with assuming the thickness of conductive region ~2 nm of the top and bottom surface states [25]. Even if the total thickness of the whole film (~8 nm) were taken, the values would be changed only by a factor of 4.
heterostructures [6–8]. Since it is linear in current, we adopt the quantity $(\Delta R_{xx} = R_{xx} - \hat{R}_{0})$ as a measure of UMR for a fair comparison. In the TI heterostructure, we define the current density by considering each surface conduction thickness of $\sim 1$ nm [25] (see also the legend of Table 1). Even though the current density is much smaller than other systems, $\Delta R_{xx}$ is comparable or larger. Therefore, the amplitude of $(\Delta R_{xx} = R_{xx} - \hat{R}_{0})$ is quite large, $10^2$–$10^6$ times larger than other bilayer systems, e.g., GaMnAs heterostructure or Pt/Co [6–8].

To elucidate a possible origin of such a large UMR, we investigated angular dependence of the signal. Figures 2(b) and 2(c) show the in-plane magnetic-field directional dependence of normalized $\Delta R_{xx}$ and $M_y (\propto \cos \phi)$; here definition of azimuth angle $\phi$ is shown in Fig. 2(a). The $|\Delta R_{xx}|$ is largest at $B||y$ axis (\(\phi = 0^\circ, 180^\circ\), and $360^\circ$), scaling well with the $\cos \phi$ dependence of $M_y$. Figures 2(e) and 2(f) show the out-of-plane magnetic-field directional dependence of normalized $\Delta R_{xx}$ [see Fig. 2(d)]. Here, $M_x$ and $M_z$ are estimated from the variation of anomalous Hall effect. In accord with the in-plane case, the $|\Delta R_{xx}|$ is largest at $B||y$ axis ($\theta = -90^\circ, 90^\circ$). It is noticeable, however, that the $\Delta R_{xx}$ does not simply scale with $M_y$. This is perhaps because the finite $M_z$ component makes the Dirac dispersion massive, which effectively weakens the spin-momentum locking [26]. To summarize, UMR emerges only when the $M_y$ component is finite.

One possible origin of such nonlinear magnetoresistance might be an additional voltage caused by heat gradient along the $z$ direction such as the anomalous Nernst effect and spin Seebeck effect [27–29]. In both processes, induced voltage would be expressed as $V_{thermal} \propto M \times (\nabla T)_z$ [27–29], so that the finite $M_y$ component might cause an additional voltage along the $x$ direction. However, we can safely exclude this possibility since the additional voltage should exhibit the same sign for both heterostructures of CBST = BST = InP [Fig. 1(d)] and BST = CBST = InP [Fig. 1(g)] when InP works as a heat bath; this is inconsistent with the experimentally observed opposite sign shown in Figs. 1(f) and 1(i). Therefore, the origin of UMR should be explored in intrinsic scattering mechanisms related to electron spins. To clarify the microscopic origin, we studied the temperature dependence under higher magnetic field [Fig. 3(a)]. UMR at low magnetic field decreases with increasing temperature until it almost vanishes at around Curie temperature $T_C \sim 24$ K [22], confirming its close relevance to the ferromagnetic magnetization. This is also evident from the absence of UMR within the present experimental error in the single-layer

FIG. 2. (a) Schematic sample configuration for the measurement of in-plane magnetic field $\phi$ dependence of $\Delta R_{xx}$. $\phi$ is measured from the $y$ axis. (b), (c) Magnetic field and $\phi$ dependence (by 30° step) of normalized $\Delta R_{xx}$, $\Delta R_{xx}/\Delta R_{xx}^0$. Here, $\Delta R_{xx}^0$ is $\Delta R_{xx}$ at $\phi = 0^\circ$ and 1.0 T. (d)–(f) The same as (a)–(c) for the out-of-plane magnetic field $\theta$ dependence. $\theta$ is measured from the $z$ axis. Here, $\Delta R_{xx}^0$ is $\Delta R_{xx}$ at $\theta = 90^\circ$ and 1.0 T.

FIG. 3. (a) Magnetic field dependence of $\Delta R_{xx}$ at various temperatures. (b) Schematic concepts of asymmetric scattering of spin-polarized surface Dirac electrons by magnons. (c) Temperature dependence of $\Delta R_{xx}$ under various magnetic fields. (d) Numerical calculation results of temperature dependent $\Delta R_{xx}$ under various magnetic fields.
BST film [22]. On the other hand, UMR is strongly suppressed at high magnetic field, meaning that it does not simply scale with $M_\text{c}$. This indicates that the UMR in TI cannot be explained in terms of the GMR mechanism that was proposed for the case of ferromagnet–normal-metal bilayers [6–8]. Rather, such a field induced suppression of $|\Delta R_{xx}|$ is reminiscent of the cases of the spin Seebeck effect [28,29] and magnon Hall effect [30], in which the magnon population and hence the signal magnitude are suppressed by gap opening of spin wave (magnon) at higher field. This leads us to consider the scattering of surface Dirac electrons by magnons as a microscopic origin of UMR.

With spin-momentum locking, conservation of angular momentum leads to the one-way scattering by magnon: Taking the quantization direction along $M|y$ axis, the angular momentum of the magnon is $+1$ (note that spin angular momentum points opposite to $M$). Thus, as shown in Fig. 3(b), when an electron with $s_y = -1/2$ spin (left branch) is backscattered to $s_y = 1/2$ (right branch), the electron absorbs the magnon because of the conservation of angular momentum. On the other hand, when it goes from $s_y = 1/2$ to $s_y = -1/2$, it emits magnon as a reverse process. Phenomena related to such a transfer of angular momentum between electron spin and magnetization have been widely recognized in the field of spintronics [1], for example, the spin Seebeck effect [27–29], spin Peltier effect [31], spin pumping [32,33], and spin torque ferromagnetic resonance [2,3,13,14]. In such a scattering process by a magnon, we can derive the formula of UMR by the Boltzmann transport equation with the relaxation time approximation [24] as follows (see Supplemental Material [22]):

$$\Delta R_{xx} \propto j_x \int k_x \left( \frac{1}{\tau^+} + \frac{1}{\tau^-} \right) \left( \frac{\partial^2 f}{\partial E^2} \right),$$

$$\frac{1}{\tau^+} \propto \frac{1}{e^{\hbar \omega / k T} - 1} \left( 1 - \frac{1}{e^{\hbar (\omega + \hbar v v k_x - E_F)} - 1} \right),$$

$$\frac{1}{\tau^-} \propto \left( \frac{1}{e^{\hbar \omega / k T} - 1} + 1 \right) \left( 1 - \frac{1}{e^{\hbar (\omega - \hbar v v k_x - E_F)} - 1} \right).$$

Here, $f$ is the Fermi distribution function and $\hbar \omega$ is the magnon energy with wave number $\sim 2k_F$ ($k_F$: Fermi wave number). $\tau^+$ ($\tau^-$) is relaxation time of magnon scattering from left (right) branch to right (left) one. The first factors of Eqs. (3) and (4) are the probability of magnon absorption and emission, respectively, and the second ones are the probability that the final state of the electron is unoccupied. Since $1/\tau^+$ and $1/\tau^-$ are not equal in general, equation (2) gives finite UMR in TI. Note that Eq. (2) is derived for the one-dimensional (1D) Dirac dispersion but this can be readily extended to the actual 2D case without essential change of the scheme [22].

Figures 3(c) and 3(d) display the comparison of experimental results of the temperature and magnetic-field dependence of UMR with the calculated ones based on the above model assuming the magnon energy of $g \mu_B B$ [34] with $g \sim 2$ for the localized Cr moment in CBST. Both results give qualitative consistency; the UMR monotonically increases with decreasing temperature at low magnetic field (1.1 and 5.0 T), while the magnon gap as large as $\sim 20$ K opens at 13.9 T so that the UMR takes a peak structure around the temperature comparable with the magnon gap. Here, the deviation of numerical calculation from the experimental result above 10 K originates from the breakdown of the adopted spin wave approximation [22] at temperatures close to $T_c \sim 24$ K. This microscopic model helps us to understand why the UMR in TI is so large: One reason is the spin-momentum locking inherent in TI. Unlike the Rashba interface with two bands having opposite spin helicity, TI with single spin-momentum locking can accumulate spin efficiently without cancellation. Another factor is that TI with tuned $E_F$ around the Dirac point can have a small Fermi momentum $k_F$ lower than $\sim 500 \mu m^{-1}$ [17,18]. Therefore, magnons with small wave number and low energy can dominantly contribute to electron scattering, which is easily populated even at low temperatures.

Finally, we discuss the $E_F$ dependence of UMR in the field-effect transistor of TI heterostructure. Here, the AlO$_x$ layer with a thickness of 30 nm was deposited as a top gate dielectric. Figures 4(a) and 4(b) show the gate voltage $V_G$ dependence of $R_{yx}$ and $R_{xx}$ under $B||z$ axis at 0.01 and 14 T. Anomalous Hall effect [$R_{yx}$ measured at 0.01 T, shown by a black line in Fig. 4(a)] and $R_{xx}$ [Fig. 4(b)] take maxima at around $-4$ V, indicating that $E_F$ of the top surface state is tuned close to the Dirac point [15]. The $V_G$ dependence of UMR measured under the $B||y$ axis is summarized in Figs. 4(c) and 4(d). To exclude the $V_G$ dependence of relaxation time (denoted as $\tau^{(0)}$ in the Supplemental Material [22]) we plot $\Delta R_{xx}$, not $\Delta R_{xx}/R_{xx}$ in Fig. 4(d) [22]. First, the sign of $\Delta R_{xx}$ does not change with $V_G$, i.e.,
irrespective of $E_F$ position in hole ($V_G = -19$ V) and electron ($V_G = 0$ and 18 V) doping regions. This can be understood by considering the scattering process for the hole side in the same way as shown in Fig. 3(b) for the electron side [22]. Moreover, the UMR is maximized at $E_F$ being close to the Dirac point ($V_G = -6$ V). As $k_F$ decreases with $E_F$ approaching the Dirac point, the wave number and energy of magnon contribute to the scattering process decrease so that the related magnon population increases. This, in combination with the decrease of $k_F$, results in the maximum $\Delta R_{xx}$ and UMR with $E_F$ around the Dirac point.

To summarize, we observed UMR in magnetic TI, which is shown to be several orders of magnitude larger than in other reported systems [6–8]. The origin of UMR is identified to be the asymmetric scattering of electrons by magnons. Improvement of theoretical calculation [22] and understanding of the relationship with spin-orbit torque remain as future issues [12–14].

We thank T. Yokouchi, T. Ideue, Y. Okamura, N. Ogawa, S. Seki, K. Hamamoto, and N. Nagaosa for fruitful discussions, and S. Shimizu for experimental support. This research was supported by the Japan Society for the Promotion of Science through the Funding Program for World-Leading Innovative R & D on Science and Technology (FIRST Program) on “Quantum Science on Strong Correlation” initiated by the Council for Science and Technology Policy and by JSPS Grant-in-Aid for Scientific Research(S) No. 24224009 and No. 24226002 from MEXT, Japan.

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