Effects of Crystalline Disorder on Interfacial and Magnetic Properties of Sputtered Topological Insulator/Ferromagnet Heterostructures

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ABSTRACT: Thin films of topological insulators (TIs) coupled with ferromagnets (FMs) are excellent candidates for energy-efficient spintronics devices. Here, the effect of the crystalline structural disorder of TI on the interfacial and magnetic properties of sputter-deposited TI/FM, Bi$_2$Te$_3$/Ni$_{80}$Fe$_{20}$, heterostructures is reported. Ni and a smaller amount of Fe from Py were found to diffuse across the interface and react with Bi$_2$Te$_3$. For improved crystalline c-axis-oriented Bi$_2$Te$_3$ films, a significant enhancement in Gilbert damping is observed, accompanied by an effective out-of-plane magnetic anisotropy and enhanced damping-like spin–orbit torque (DL-SOT), possibly due to the topological surface states (TSS) of Bi$_2$Te$_3$. Furthermore, a spontaneous exchange bias is observed in hysteresis loop measurements at low temperatures. This is caused by a topological antiferromagnetic interfacial layer formed due to a solid-state reaction between the diffused Ni with Bi$_2$Te$_3$ that couples with the FM, Ni$_{80}$Fe$_{20}$. For the increasing disorder of Bi$_2$Te$_3$, a significant weakening of the exchange interaction in the AFM interfacial layer is observed. These experimental results open the pathway for further exploration of crystalline-disordered TIs and their interfaces.

KEYWORDS: Topological Insulator, Ferromagnet, Interface, Spin Pumping, Spin Orbit Torque, Antiferromagnet

INTRODUCTION

Topological insulators (TIs) of the (Bi$_2$Sb)$_3$(Te$_3$Se)$_4$, family of compounds are van der Waals (vdW) chalcogenide materials with tetradymite structures. TIs possess large spin–orbit coupling (SOC) resulting in dissipation-less surface conducting states—topological surface states (TSS). 1–3 Introducing magnetic order in TIs leads to gap opening in the TSS bands and the possibility of dissipation-less quantum anomalous Hall (QAH) and axion insulator states. 4–21 Stimulated by these remarkable material properties, TIs are regarded as promising candidates for realization of energy efficient spintronic devices. TIs possess highly reactive surfaces, thus making them susceptible to the formation of interfacial phases when coupled with metallic films. 22–26 Because of their composition, these interfacial layers have the potential for hosting topologically nontrivial magnetic phases. 27 The majority of the reported experiments have studied TIs grown from molecular beam epitaxy (MBE) 22–24 which is a standard technique for growing high-quality, crystalline-ordered thin films. However, MBE suffers from low throughput and is constrained by sample dimensions, making it incompatible for integration in industrial CMOS processes. Magnetron sputtering, on the other hand, is the semiconductor industry’s accepted thin film deposition technique because of its potential for high throughput and large area film growth. Sputtering also allows easy deposition of TIs with varying crystalline disorder. 28–30 This opens up the possibility of exploration of their disorder-dependent electronic and magnetic properties.

Recently, the topological antiferromagnetic (AFM) compound Ni$_6$Bi$_4$Te$_6$ was observed in the interface of highly c-axis-oriented sputtered Bi$_2$Te$_3$/Ni$_{80}$Fe$_{20}$ heterostructures. 30 Ni from the Ni$_{80}$Fe$_{20}$ (Py) layer diffuses and reacts with Bi$_2$Te$_3$, and the reaction is promoted by the delocalized TSS electrons. 23–25 In addition, recent experiments have shown the presence of Dirac-like surface states even in amorphous Bi$_2$SbTe$_3$. 30 In this work, the effects of crystalline structural disorder on the interface and magnetic properties of Bi$_2$Te$_3$/Py heterostructures are investigated. The magnetic species, largely Ni and smaller amounts of Fe, are found to diffuse across the interface into Bi$_2$Te$_3$, resulting in a magnetic interfacial layer. For increasing c-axis-oriented texture of Bi$_2$Te$_3$, increasing amounts of diffused magnetic species were found to react with Bi$_2$Te$_3$, which also leads to enhanced magnetic interactions at low...
temperatures. This phenomenon was identified in hysteresis loop measurements of the magnetic moment versus applied magnetic field, \( m(H) \), for the Bi\(_2\)Te\(_3\)/Py samples. As a result of the interfacially diffused magnetic species (Ni, Fe) reacting with Bi\(_2\)Te\(_3\), the magnetic moment, \( m \), at saturation field is reduced by \( \Delta m \) in the Bi\(_2\)Te\(_3\)/Py compared to control Py samples suggesting a change in valence state of the magnetic species. The values of \( \Delta m \) become smaller for increasing disorder in Bi\(_2\)Te\(_3\), suggesting lesser reactivity between the diffused magnetic species and Bi\(_2\)Te\(_3\). Further, a significant enhancement in Gilbert damping, an out-of-plane (OP) canting of magnetization, and enhanced DL-SOT were observed in samples with higher \( c \)-axis-oriented textured TI. However, with significantly reduced crystallinity, surprisingly the granular Bi\(_2\)Te\(_3\) samples had a comparable enhanced spin-Hall conductivity as samples with the higher \( c \)-axis-oriented Bi\(_2\)Te\(_3\). This can be attributed to the quantum confinement effect in smaller crystallite grains in granular TI.\(^{26,27}\) Low-temperature \( m(H) \) and \( m(T) \) measurements revealed an AFM ordered phase in the predominantly Ni-diffused Bi\(_2\)Te\(_3\) interface from the formation of the topological AFM compound NiBi\(_2\)Te\(_4\).\(^{25}\) Interestingly, the strength of the exchange interaction of the interfacial AFM phase, as monitored by the exchange bias, was found to weaken significantly with the increase in disorder of the Bi\(_2\)Te\(_3\) layer. These results indicate a strong topological property of TIs with crystalline \( c \)-axis-oriented texture, which weakens considerably with increasing crystalline disorder. These experimental results show the possibility of tailoring topological properties of TIs by control of the crystalline structural disorder.

## RESULTS AND DISCUSSION

### Crystalline Structure—Properties of Sputter-Deposited Bi\(_2\)Te\(_3\)

Samples of \( \sim 30 \) nm Bi\(_2\)Te\(_3\) with varying crystalline disorder, (1) granular (GBT), (2) randomly oriented polycrystalline disordered (DBT), and (3) effectively \( c \)-axis-oriented crystalline (CBT), were grown using RF magnetron sputtering on amorphous thermally oxidized Si substrates (see FMR Measurements for material growth method and Supporting Information Section S1 for grain size characterization). The choice of amorphous thermally oxidized SiO\(_2\) substrates was to decouple Si lattice effects for disordered TI samples. Crystalline structural properties of the Bi\(_2\)Te\(_3\) samples were verified using X-ray diffraction (XRD) and high-resolution transmission electron microscope (HRTEM) imaging measurements, as shown in Figure 1a,b,d. The GBT samples did not show any significant diffraction peaks in the XRD measurement, suggesting a high amorphous content. Further, HRTEM images of the GBT, DBT, and CBT samples shown in Figure 1a verify the granular, randomly oriented crystallite domains and an improved \( c \)-axis-oriented vdW layered structure, respectively. The thickness and surface roughness of the samples were characterized using X-ray reflectometry (XRR) measurements, as shown in Figure 1e. From the fitting of XRR data, thickness of \( \sim 30 \) nm was obtained for all three samples. The fits to the XRR data also revealed surface roughness of 0.7, 1.7, and 1.0

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**Figure 1.** (a) Schematic model of three quintuple Bi\(_2\)Te\(_3\) unit cells. Cross-sectional HRTEM images showing structural disorder in (b) GBT, (c) DBT, and (d) CBT samples. (e) XRD data for the GBT, DBT, and CBT samples. Inset: normalized plots of symmetric XRD data. (f) XRR plots and theoretical fitting for GBT, DBT, and CBT samples used for characterization of thickness and surface roughness. The data for CBT samples are similar to the ones in ref 25.
nm for the GBT, DBT, and CBT samples, respectively, which are typical surface roughness values for sputtered-deposited thin films, confirming growth of high-quality TI films. The XRR measurements also reveal densities of 9.06 g/cm$^3$, 9.47 g/cm$^3$, and 9.31 g/cm$^3$ for the GBT, DBT, and CBT samples, respectively. These values of density of Bi$_2$Te$_3$ are comparable and do not follow any noticeable trend.

**Morphology of Interfacial Layer formed by Ni Diffusion into Bi$_2$Te$_3$.** Heterostructure samples of GBT/Py, DBT/Py, and CBT/Py were grown where the thickness of the layers was maintained at 30 and 20 nm for Bi$_2$Te$_3$ and Py, respectively (see FMR Measurements for the material growth method). HRTEM imaging and energy dispersive X-ray spectroscopy (EDS) measurements were performed to characterize the morphology and stoichiometric composition along the cross section of the samples, as shown in Figure 2a–f (see Supporting Information Section S2). The HRTEM images in Figure 2a–c clearly show the highly amorphous nature of GBT, randomly oriented vdW layered crystalline domains in DBT, and highly oriented vdW layers in the CBT layers. A closer examination of the interfaces of the heterostructures also reveal a rougher interface in the disordered GBT/Py and DBT/Py samples compared to the CBT/Py sample. The EDS cross-sectional profiles of atomic % of elements in Figure 2d–f show a significant diffusion of Ni (and smaller amounts of Fe) across the TI/FM interface into the Bi$_2$Te$_3$ layers, forming an interfacial layer denoted as Ni–Bi$_2$Te$_3$. In general, the Ni and Fe show a large gradient over a Bi$_2$Te$_3$ distance of 5–14 nm, where the Ni averages 30–40%, while the Fe diffusion is much smaller in the GBT/Py and CBT/Py samples. The more highly disordered GBT/Py and DBT/Py samples also have ~3% of Ni diffused throughout the thickness of the Bi$_2$Te$_3$ layer. However, the predominantly Ni–Bi$_2$Te$_3$ layer in the highly ordered CBT/Py sample acts as a barrier preventing further diffusion of Ni into the Bi$_2$Te$_3$ bulk. The formation of the Ni–Bi$_2$Te$_3$ layer is likewise accompanied by a thin Fe-enriched region in the intermediate Py layer (marked Py$^*$.). It must also be noted that the moderately disordered DBT/Py sample which has randomly oriented polycrystalline domains has developed a much higher Ni and Fe diffusion of ~47% and ~9% at the interface, respectively. The Fe diffusion, however, is only ~3–4% at the interface in the GBT/Py and CBT/Py samples.

The diffusion of Ni into the c-axis-oriented Bi$_2$Te$_3$ was previously shown to result from solid-state reactions leading to the formation of Ni–Te bonds and formation of the topological AFM compound, NiBi$_2$Te$_4$. Similar to that study, the room temperature $m(H)$ measurements can be used here as an indicator of the solid-state reaction of Ni with Bi$_2$Te$_3$ that is promoted by the delocalized TSS electrons. As shown in Figure 2h, all the Bi$_2$Te$_3$/Py samples show a clear decrease in the saturation magnetic moment for increasing disorder. This loss of saturation moment is compared to a control sample of Py by $\Delta m$. The $\Delta m$% values were found to be 13%, 37%, and 41% for the GBT, DBT, and CBT samples, respectively. This reduction in moments results from a change in valence state of the reacting magnetic species. Interestingly, in the samples with more highly disordered Bi$_2$Te$_3$, even though the diffusion of Ni is larger as observed in the EDS depth profiles in Figure 2d–f compared to the CBT/Py sample, the $\Delta m$% is lower. This clear enhancement in the loss of moments with crystalline order, and hence reactivity of Ni with Bi$_2$Te$_3$, is due to strengthening of TSS with increasing crystallinity of Bi$_2$Te$_3$. A lower number of the diffused magnetic species react and form compounds with Bi$_2$Te$_3$ due to weaker topological properties of highly disordered TI samples.

**Disorder Effects on Room-Temperature Magnetic Properties of Bi$_2$Te$_3$/Py.** To investigate the effects of disorder, $m(H)$ hysteresis loop and ferromagnetic resonance (FMR) experiments were performed on the three types of Bi$_2$Te$_3$ samples: highly disordered GBT/Py, moderately disordered DBT/Py, and c-axis-oriented CBT/Py. First, $m(H)$ hysteresis loop measurements were performed on the samples, with the magnetic field oriented in-plane (IP) and out-of-plane (OP) relative to the film plane, as shown in Figure 3a–c. For increasing c-axis-oriented growth of the Bi$_2$Te$_3$ layer, the saturation fields, $H_s$ measured in the IP and OP configurations show an increasing and decreasing trend,

![Figure 2](https://example.com/figure2.png)

**Figure 2.** Cross-sectional HRTEM images of (a) highly amorphous GBT/Py, (b) disordered DBT/Py with randomly oriented vdW domains, and (c) highly c-axis-oriented CBT/Py (similar to ref 25). The yellow dashed lines mark the approximate boundary between the Bi$_2$Te$_3$ and Py layers without considering the interface layers. Atomic % (At %) characterized using EDS for (d) GBT/Py, (e) DBT/Py, and (f) CBT/Py samples. The interface layers are highlighted in green and blue colors for the Ni–Bi$_2$Te$_3$ and Py$^*$, respectively. (g) Schematic of the Bi$_2$Te$_3$/Py samples with the layers marked. (h) $m(H)$ measurements for IP orientation at room temperature showing loss of moments in the Bi$_2$Te$_3$/Py samples due to interfacial Ni and Fe diffusion and reaction with Bi$_2$Te$_3$. Inset: comparison of % loss of moments ($\Delta m$%) in GBT/Py, DBT/Py, and CBT/Py samples compared to the control Py sample. The data presented for the CBT sample are similar to the ones in ref 25.
respectively. In addition, the ratio of remanence to saturation magnetization ($M_r/M_s$) in the IP $m(H)$ loop decreases for increasing crystalline order of Bi$_2$Te$_3$. These trends indicate an increase in effective OP magnetic easy-axis with the increased $c$-axis-oriented texture of Bi$_2$Te$_3$ in the Bi$_2$Te$_3$/Py heterostructure samples. This enhanced OP magnetic anisotropy is a characteristic of interaction of the magnetic moments with large SOC in the interfaces. Change in magnetic anisotropy has been previously predicted and observed in other TI/FM-based materials systems (also see Supporting Information Section S6). As shown in Figure 3b, the OP measured $m(H)$ loops for the Bi$_2$Te$_3$/Py samples also exhibit a smaller hysteresis loop in the low-field regions. These smaller components of the $m(H)$ loop are more prominent in the DBT/Py and CBT/Py samples, which otherwise exhibit a lower OP easy-axis of magnetic moments compared to the highly $c$-axis-oriented CBT/Py sample. The DBT/Py sample also had an unusually large coercive field ($H_c$) resulting from the randomly oriented vdW layered crystalline domains, as observed in both the IP and OP $m(H)$ loop measurements in Figure 3a,b. These effects in the samples with highly disordered polycrystalline TIs are possibly present due to the disordered magnetic texture that emerge in their interfaces. Further, interfacial magnetic exchange interactions exist between the FM and Py layer and the diffused magnetic species in the randomly oriented vdW crystallite domains of the DBT sample. The randomly oriented vdW domains of the DBT sample may also have significant localized variations in

Figure 3. Normalized $m(H)$ loops measured (a) in-plane (normalized plots of Figure 2h) and (b) out-of-plane for the GBT/Py, DBT/Py, and CBT/Py samples. Inset: Expanded low-field regions showing enhanced $H_c$ for the Bi$_2$Te$_3$/Py samples compared to the Py control sample. (c) Comparison of the saturation fields (IP and OP) and the $M_r/M_s$ ratio clearly highlighting an increase in OP anisotropy with crystalline $c$-axis-orientation of Bi$_2$Te$_3$. (d) FMR line width versus frequency for extraction of $\alpha$. (e) FMR resonance frequency versus field for extracting $H_{\perp}$ and $4\pi M_{\text{eff}}$. (f) Visual comparison of $\alpha$ and $H_{\perp}$ extracted from d and e, respectively. All measurements here were performed at 300 K.
TSSs. A combination of these effects possibly lead to an anomalously large coercive field in the DBT/Py samples even at 300 K.

Further information was obtained using ferromagnetic resonance (FMR) measurements to understand the changes in magnetization dynamics with changes in Tl disorder. The FMR line width (ΔH) and resonance field (H_0) were extracted from the FMR signal at different constant frequencies (f_res) (Supporting Information Section S4). The Gilbert damping parameter (α) was extracted by fitting a straight line to the FMR line width versus frequency plot using the equation, ΔH = ΔH_0 + γf_res. Here, ΔH_0 is the inhomogeneous line width and γ is the gyromagnetic ratio. As shown in Figure 3d, the values of α extracted for the Py, GBT/Py, DBT/Py, and CBT/Py samples were 0.0053, 0.0076, 0.0089, and 0.0123, respectively. This shows a progressive increase in α with increase in crystalline grain size of Bi$_2$Te$_3$ and a significant enhancement when the Bi$_2$Te$_3$ layer is highly c-axis-oriented (summarized in Figure 3f). This effect was also observed in other Bi$_2$Te$_3$/FM heterostructure materials (Supporting Information Section S6), which signals a large enhancement in SOC and the presence of robust TSS in c-axis-oriented Bi$_2$Te$_3$. In addition, the effective magnetization, 4πM_{eff} and the perpendicular magnetic anisotropy (PMA) field, H_L, were extracted by fitting the modified Kittel equation to the f_res versus H_res plots shown in Figure 3e,f, f_res = γ(ΔH + H_L)(H_res + H_L + 4πM_{eff})^{−1}, where 4πM_{eff} = 4πM_c − H_L and H_L is the uniaxial anisotropy field. The 4πM_c values were found to be 15.2, 14.7, 14.3, and 14.1 kOe, respectively, for Py, GBT/Py, DBT/Py, and CBT/Py samples. The decrease in 4πM_c for increasing crystalline order demonstrates the reduction in saturation magnetization due to interfacial diffusion of Ni and Fe from Py into Bi$_2$Te$_3$. In addition, the H_L values increased from 4.35, 5.07, 5.54, and 5.79 kOe, for the Py, GBT/Py, DBT/Py, and CBT/Py samples, respectively. This enhancement in H_L supports the m(H) results that show an increase in effective OP anisotropy with increasing c-axis-oriented texture of Bi$_2$Te$_3$. The magnetic properties measured using m(H) loops and FMR are summarized in Table 1.

Table 1. Summary of Room-Temperature Magnetic Properties of the GBT/Py, DBT/Py, and CBT/Py Heterostructure Samples

<table>
<thead>
<tr>
<th>Sample</th>
<th>H_{OOP} (kOe)</th>
<th>H_{IP} (kOe)</th>
<th>M_c/M_t (%)</th>
<th>4πM_c (kOe)</th>
<th>H_L-FMR (kOe)</th>
<th>α</th>
</tr>
</thead>
<tbody>
<tr>
<td>Py</td>
<td>11.9</td>
<td>0.95</td>
<td>96</td>
<td>1.52</td>
<td>4.35</td>
<td>0.0053</td>
</tr>
<tr>
<td>GBT/Py</td>
<td>11.2</td>
<td>1.22</td>
<td>96</td>
<td>1.47</td>
<td>5.07</td>
<td>0.0075</td>
</tr>
<tr>
<td>DBT/Py</td>
<td>9.7</td>
<td>2.01</td>
<td>90</td>
<td>1.43</td>
<td>5.54</td>
<td>0.0089</td>
</tr>
<tr>
<td>CBT/Py</td>
<td>8.1</td>
<td>3.05</td>
<td>84</td>
<td>1.41</td>
<td>5.78</td>
<td>0.0123</td>
</tr>
</tbody>
</table>

The enhancement in α for increasing c-axis texture of Bi$_2$Te$_3$ can be attributed to a large spin-pumping effect modeled by the spin-mixing conductance, g_{11}^c = \frac{4\pi M_c\Delta a}{k_B T_P} where T_PM is the thickness of the FM layer, Δa is the enhancement in Gilbert damping, and H is the reduced Planck’s constant. The resulting g_{11}^c values increased with increasing crystalline c-axis-orientation of Bi$_2$Te$_3$ and were 2.10 × 10^{-18}, 2.38 × 10^{-18}, and 4.08 × 10^{-18} m$^2$/A for the GBT/Py, DBT/Py, and CBT/Py samples, respectively. The g_{11}^c values were calculated assuming that the Gilbert damping enhancement in the Bi$_2$Te$_3$/Py samples is entirely due to spin-pumping. The loss of magnetization from interfacial diffusion of Ni from Py and spin-memory loss due to interfacial proximity-induced magnetization may also play a role in the enhancement of α in Bi$_2$Te$_3$/Py samples. However, these contributions toward enhancement in α could not be isolated because of complexity of interfaces in these heterostructure materials systems. However, the large enhancement in α with highly c-axis-oriented TIs is also observed in other TI/FM materials systems, including those which do not show interfacial diffusion (see Supporting Information Section S6). This suggests that for crystalline c-axis-oriented TIs, the TI/FM heterostructures experience a significant enhancement in spin-pumping predominantly from the presence of robust TSS. These results provide strong evidence of enhancement in SOC strength and topological properties in highly c-axis-oriented TI samples compared to disordered TIs.

Spin–Orbit Torque Properties of CBT/Py, DBT/Py, and GBT/Py Samples. The spin–orbit torque (SOT) characteristics were extracted from the symmetric and antisymmetric components of the fitted Lorentzian, as shown in Figure 4b–d, using the equations

\[ V_s = \frac{J_{eff}}{4} \frac{d\Delta H}{d\theta_0} \Delta\Theta \quad \text{and} \quad V_A = \frac{J_{eff}}{4} \frac{d\Delta\gamma}{d\theta_0} \left( \frac{1 + 2\Delta\Theta}{\Delta\Theta} \right) \Delta\Theta \]

Where, \( J_{eff} \) is the RF current injected, \( \theta_0 \) is the in-plane angle of the external DC field relative to the injected RF current, \( \Delta\Theta \) is the derivative of the anisotropic magnetoresistance (AMR) relative to \( \theta_0 \) (Supporting Information Section S5), \( \Delta\Theta \) is the line width in the frequency domain, \( \tau_{DL} \) is the damping-like DL-SOT, \( \gamma \) is the gyromagnetic ratio, \( \mu_0 \) is the permeability of vacuum, and \( \Delta H \) is the line width of the FMR signal. \( \tau_{DL} \) corresponds to the symmetric component of the Lorentzian, while \( \tau_{DL} \) corresponds to the antisymmetric components of the Lorentzian function. The DL-SOT was the largest in the CBT/Py sample with a value of 0.16 Oe, compared to 0.11 and 0.10 Oe in GBT/Py and DBT/Py, respectively, as shown in Figure 4e. The FL-torques must have a finite contribution from Oersted fields because of the large thickness (~20 nm) of the FM layers. The resistivity values of the Bi$_2$Te$_3$ samples capped with AlO$_x$ were 3.56 × 10^{-3} Ωm, 5.83 × 10^{-3} Ωm, and 2.45 × 10^{-3} Ωm, respectively, for the GBT, DBT, and CBT samples, respectively. The reduced resistivity in CBT can lead to a larger charge current and hence larger DL-SOT in the CBT/Py sample. However, the DL-SOT value in the CBT/Py sample is much larger than expected compared to the GBT/Py and DBT/Py samples and cannot be explained solely on the basis of Bi$_2$Te$_3$ resistivities. Further, the resistivity of the Bi$_2$Te$_3$ layers in the GBT/Py, DBT/Py, and CBT/Py samples must have been altered by the interfacial diffusion of Ni,Fe and formation of compounds. A combination of all these factors along with presence of robust TSSs possibly leads to an enhanced DL-SOT in the CBT/Py sample which is much larger than the GBT/Py and DBT/Py samples.

The spin-Hall conductivity, \( \sigma_{SHP} \), which measures the spin current, \( J_s \), generated from electric field, and \( E \) across the STFMR device is given by \( \sigma_{SHP} = \frac{J_s}{E} = \frac{4\pi M_c\Delta a}{k_B T_P} \). The average

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Figure 4. (a) Schematic for ST-FMR experimental setup with LIA and phase-locked RF current source. The brown and dark blue arrows signify up-spin and down-spin states, respectively. ST-FMR data and Lorentzian fitting for (b) GBT/Py, (c) DBT/Py, and (d) CBT/Py samples, measured at 4 GHz frequency. (e) DL-SOT (blue) and Oersted plus FL-SOT for the GBT/Py, CBT/Py, and DBT/Py samples extracted from b−d. (f) Values of $\sigma_{SH}$ for the GBT/Py, CBT/Py, and DBT/Py samples. The boxes in (e) and (f) represent quartile plots for 5 devices in each sample measured.

Figure 5. (a) Schematic of the AFM-FM heterostructure materials system and exchange bias in $m(H)$ loops. (b) $m(H)$ loops measured at 6 K under ZFC condition of the GBT/Py, DBT/Py, and CBT/Py samples showing spontaneous exchange bias. The size of the arrows qualitatively indicates the magnitude of shifts in the exchange bias. (c) $m(T)$ measurements of the GBT/Py, DBT/Py, and CBT/Py samples and their derivatives for characterization of $T_N$. (d) Exchange bias and $T_N$ values of the GBT/Py, DBT/Py, and CBT/Py samples extracted from b and c. The $m(H)$ and $m(T)$ data presented for the CBT sample are similar to the ones in ref 25.
σSH values shown in Figure 4f were calculated to be 8.1 × 10^5 b A/m and 8.1 × 10^5 b A/m for GBT/Py, DBT/Py, and CBT/Py samples, respectively. The GBT/Py and CBT/Py samples have a much larger σSH compared to the disordered polycrystalline DBT/Py sample. This points toward a reduction in charge-spin current conversion efficiency with degradation in crystalline ordering possibly due to scattering of spin current in the randomly oriented crystalline TI domains. The GBT/Py sample, however, has a larger σSH comparable to the DBT/Py sample and comparable to the CBT/Py sample. This happens possibly because of the quantum confinement effect in the smaller grain size of the GBT sample, resulting in larger charge-spin current conversion efficiency. The symmetric Lorentzian in ST-FMR also includes a contribution from spin-pumping due to the inverse spin-Hall effect (ISHE), which results in the α enhancement due to spin-pumping, as shown in the Figure 3d. However, because of the complexity of the interface in the Bi2Te3/Py samples, ISHE contribution to the symmetric component of FMR spectra could not be accurately isolated from the DL-SOT. Further, the direct determination of spin-charge current interconversion efficiency requires determination of the spin-Hall angle, θSH parameter.23–25 Due to the complexity of the interfacial structure, the effective resistivity or the thickness of the spin-Hall material cannot be determined which are required for calculation of θSH. Hence, the spin-charge conversion efficiency for the Bi2Te3/Py samples could not be determined and compared accurately.

**Effect of Crystalline Disorder in Interfacial Topological AFM Phase.** Creation of an interfacial AFM-ordered layer was reported in the interface of highly c-axis-oriented Bi2Te3/Py heterostructures.25 A fraction of the diffused magnetic species was found to undergo solid-state chemical reactions with Bi2Te3 promoted by the TSS electrons. With the increasingly crystalline c-axis-oriented texture of Bi2Te3, strengthening the topological property of Bi2Te3 led to an enhancement in reaction between the diffused species and Bi2Te3. This was revealed by the increase in magnetic moments in higher c-axis-oriented Bi2Te3/Py heterostructures. The increase in magnetic c-axis-orientation of Bi2Te3 also resulted in a notable enhancement in coercive field (Hc) and FMR measurements. Interestingly, polycrystalline disordered Bi2Te3 sample had a reduced spin-Hall conductivity possibly because of scattering of spins from polycrystalline grain boundaries, whereas the samples with granular and highly c-axis-oriented Bi2Te3 had comparable charge-Hall conductivities, which possibly resulted from quantum confinement effect in smaller crystalline grains and strong TSS, respectively. Furthermore, magnetization measurements at low temperatures revealed a spontaneous exchange bias in all the Bi2Te3/Py heterostructures including the samples with highly disordered Bi2Te3. This further corroborated the evidence for interfacial solid-state reactions and demonstrated a surprising resilience of the topological property of disordered Bi2Te3. However, the exchange interaction strength of the interfacial AFM phase was found to weaken significantly with the increase in structural disorder of Bi2Te3. This was verified by degradation in HEB and Tc with increase in disorder of the Bi2Te3. The weakened AFM exchange interaction and reduced magnetic moment loss suggested weakening of the topological property with an increase in the crystalline disorder of TI. These results will open the path for further exploration of the crystalline disorder in TIs and TI/FM interfaces and integration of TIs in practical spintronic devices.

**CONCLUSION**

Interfacial and magnetic properties of sputter-deposited TI/FM Bi2Te3/Py heterostructures were studied for three variations of the crystalline structural disorder of the Bi2Te3. An interface layer was found to form because of diffusion of Ni and smaller amounts of Fe into Bi2Te3. In conjunction with the experimental results reported in ref 25 a fraction of the diffused magnetic species was found to undergo solid-state chemical reactions with Bi2Te3 promoted by the TSS electrons. With the increasing crystallinity, c-axis-oriented texture of Bi2Te3, strengthening the topological property of Bi2Te3 led to an enhancement in reaction between the diffused species and Bi2Te3. This was revealed by the larger loss of magnetic moments in higher c-axis-oriented Bi2Te3/Py heterostructures. The increase in magnetic c-axis-orientation of Bi2Te3 also resulted in a notable enhancement in OP magnetic anisotropy, Gilbert damping, and DL-SOT as observed from the m(t) loop and FMR measurements. Interestingly, polycrystalline disordered Bi2Te3 sample had a reduced spin-Hall conductivity possibly because of scattering of spins from polycrystalline grain boundaries, whereas the samples with granular and highly c-axis-oriented Bi2Te3 had comparable charge-Hall conductivities, which possibly resulted from quantum confinement effect in smaller crystalline grains and strong TSS, respectively. Furthermore, magnetization measurements at low temperatures revealed a spontaneous exchange bias in all the Bi2Te3/Py heterostructures including the samples with highly disordered Bi2Te3. This further corroborated the evidence for interfacial solid-state reactions and demonstrated a surprising resilience of the topological property of disordered Bi2Te3. However, the exchange interaction strength of the interfacial AFM phase was found to weaken significantly with the increase in structural disorder of Bi2Te3. This was verified by degradation in HEB and Tc with increase in disorder of the Bi2Te3. The weakened AFM exchange interaction and reduced magnetic moment loss suggested weakening of the topological property with an increase in the crystalline disorder of TI. These results will open the path for further exploration of the crystalline disorder in TIs and TI/FM interfaces and integration of TIs in practical spintronic devices.
**EXPERIMENTAL SECTION**

**Material Growth.** Bi$_2$Te$_3$ thin films of thickness $\sim$30 nm of varying crystalline disorder were grown by cosputtering a composite Bi$_2$Te$_3$ target with a Te target, using RF magnetron sputtering at 90 and 20 W, respectively, with 4 mTorr Ar pressure on thermally oxidized Si substrates. The base pressure of the sputtering chamber was $\sim8 \times 10^{-6}$ Torr. The GBT, DBT, and CBT samples were grown with substrate maintained at 20, 160, and 250 °C, respectively. The DBT and CBT samples were further annealed at the growth temperatures inside the PVD process chamber in 45 mTorr pressure in Ar environment for 25 min. The CBT samples were grown using the same method as ref 25. The samples were capped with 2 nm Al at room temperature before breaking vacuum which oxidizes to Al$_2$O$_3$ on exposure to atmosphere. For the magnetic and ST-FMR experiments, 20 nm Py and 3 nm TiO$_2$ capping were deposited at room temperature after deposition of Bi$_2$Te$_3$. For the X-ray, TEM, and Hysteresis loop measurements, the same samples with precise dimensions of $5 \times 6$ mm$^2$ were used. Samples for STFMR measurements were deposited on 3 in. wafers.

**XRD Characterization.** X-ray diffraction was collected using a background-free, highly collimated beam of Cu–Kα1 radiation (wavelength $\lambda = 1.54056$ Å). The X-rays were captured by a 2D charged-coupled device (CCD). The Bragg reflections were indexed according to the Bi$_2$Te$_3$ bulk hexagonal unit cell, as indicated by ($h$, $k$, $l$) where $h$, $k$, and $l$ are the Miller indices.

**TEM and XEDS Characterization.** Samples for TEM investigations were prepared by focused ion beam milling (FIB) using a Ga+ ion source. Prior to TEM observation, an additional cleaning procedure was performed by Ar-ion milling to reduce a surface amorphous layer and residual Ga from the FIB process. The TEM observations were performed using a Talos 200-FX (ThermoFisher Scientific Inc.) TEM operated at an acceleration voltage of 200 kV. EDS measurements were performed using a ChemiSTEM (ThermoFisher Scientific), and processing of the spectra was performed using Esprit 1.9 (Bruker Inc.) software.

**FMR Measurements.** FMR measurements of $\alpha$, $4\pi M_d$, and $H_z$ were performed using a spin-torque FMR (ST-FMR) experimental setup. The analysis of the experiment is explained in Supporting Information Section S4. RF current is provided by a HP8350 RF source. A SR830 lock-in amplifier (LIA) provides reference low-frequency AC for modulation phase-locked with the RF current. The LIA was used for detection of ST-FMR signal. The bias DC field is provided by an Fe-core electromagnet on a rotating stage with precise angular control. The reported ST-FMR experiment was performed at a 45° angle of the microstrip relative to the DC bias field. Control of the experiment and data acquisition were done using NI LabVIEW. The S11, S12, and impedance values were used to calculate the RF current and the E-field using vector network analyzer (VNA). The total power lost in the electrical components, such as wires and connectors, was measured to be $\sim$60%; hence, 40% of 8 dbm power was used for RF current calculations. FMR characteristics in the Bi$_2$Te$_3$/Py heterostructure samples were also extracted from the ST-FMR experiment spectra as shown in Figure S2. The devices were patterned using an ion-milling process for the DBT/Py and CBT/Py samples and using a lift-off process for the Py and GBT/Py samples.

The FMR characteristics were extracted by fitting Lorentzian functions to the spectra, as shown in Figure S2a–d, using the equation:

$$V_{\text{min}} = V_F V_{\text{sym}} + V_{\text{fwm}}$$

which clearly shows broadening of the FMR line width progressively from Py, GBT/Py, and DBT/Py to CBT/Py samples. Here, $V_{\text{min}}$ is the DC voltage output recorded in the LIA, $F_{\text{sym}} = \frac{\Delta F}{(\mu H - \mu H_{\text{ms}})^2 + \Delta F^2}$ and $F_{\text{asym}} = \frac{\Delta F}{(\mu H - \mu H_{\text{ms}})^2 + \Delta F^2}$ are the symmetric and antisymmetric components of the Lorentzian function, $\Delta H$ is the line width of the FMR signal, and $H_{\text{ms}}$ is the FMR field. The ST-FMR measurements were performed at a 45° angle relative to the external applied magnetic field. The values of $\Delta H$, $H_{\text{ms}}$, $V_F$, and $V_{\text{fwm}}$ were extracted by fitting the ST-FMR signal using the equation above for the analysis of $\alpha$, $4\pi M_d$, $H_z$, and $\theta_{\text{SH}}$ reported in the main text.

**Hysteresis Loop Measurements.** Magnetization $m(H)$ and $m(T)$ measurements were obtained using a Quantum Design MPMS XL-7 superconducting quantum interference device (SQUID) magnetometer. Hysteresis loop $m(H)$ measurements were carried out at various temperatures between 6 and 300 K. The ZFC $m(T)$ measurements were obtained while increasing the temperature in an applied field of 50 Oe. Room temperature $m(H)$ measurements were taken using a vibrating sample magnetometer (VSM).

**ASSOCIATED CONTENT**

**Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsaelm.2c00523.

Additional experimental data with figures (PDF)

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**Author Contributions**

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Notes
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**ABBREVIATIONS**

AFM, Antiferromagnet; CBT, c-axis-oriented Bi$_2$Te$_3$; DBT, Disordered Bi$_2$Te$_3$; DL-SOT, Damping-like spin orbit torque; EDS, Energy-dispersive X-ray spectroscopy; FM, Ferromagnet; FMR, Ferromagnetic resonance; GBT, Granular Bi$_2$Te$_3$; HRTEM, High resolution transmission electron microscopy; IP, In-plane; MBE, Molecular beam epitaxy; OP, Out-of-plane; QAH, Quantum anomalous hall; RF, Radiofrequency; SOC, Spin orbit coupling; TI, Topological insulator; TSS, Topological surface states; XRD, X-ray diffraction; XRR, X-ray reflectometry; ZFC, Zero field cooled.

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