

Manipulation of the anomalous Hall effect in magnetic topological insulator heterostructure $\text{MnBi}_2\text{Te}_4/\text{Bi}_2\text{Te}_3$ by Si substrate surface engineering

K. Ishihara,¹ S. Ichinokura,^{1, a)} S. V. Eremeev,² T. T. Sasaki,³ R. Takada,¹ H. Nishimichi,¹ R. Akiyama,¹ E. V. Chulkov,^{4,5,6} and T. Hirahara¹

¹Department of Physics, Institute of Science Tokyo, Tokyo 152-8551, Japan

²Institute of Strength Physics and Materials Science, Tomsk, 634055, Russia

³Research Center for Magnetic and Spintronic Materials, National Institute for Materials Science, Tsukuba 305-0047, Japan

⁴Donostia International Physics Center (DIPC), Paseo de Manuel Lardizabal, 4, 20018 San Sebastián/Donostia, Basque Country, Spain

⁵Tomsk State University, Tomsk, 634050, Russia

⁶Saint Petersburg State University, Saint Petersburg, 198504, Russia

(*Electronic mail: hirahara@phys.sci.isct.ac.jp)

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We developed an *in situ* Hall measurement setup and measured the anomalous Hall effect (AHE) in magnetic topological insulator heterostructures $\text{MnBi}_2\text{Te}_4/\text{Bi}_2\text{Te}_3$ grown on different Si(111) substrate surfaces. For the sample grown on the Si(111)- 7×7 surface, the AHE signal appears at 15 K and becomes larger by further cooling, showing that the Curie temperature T_c is 15 K. In contrast, although the T_c is the same, the AHE signal shows a local maximum at 10 K for the sample grown on the $\beta\text{-Bi}/\text{Si}(111)\text{-}\sqrt{3} \times \sqrt{3}$ surface. A plausible explanation for this peculiar behavior is the enhanced skew scattering caused by the Bi layer, or the presence of the states localized at the interfacial Bi layer, which will affect the Berry curvature of the system. Our results demonstrate the possibility to artificially control the property of a two-dimensional magnet by modification of the substrate surface with a single monatomic layer.

The Hall effect, which is a voltage generated transverse to an electrical current due to the application of a magnetic field or magnetization, is one of the key parameters to discuss quantum properties of materials^{1,2}. Among various types of Hall effect, the anomalous Hall effect (AHE) has been regarded as an important physical quantity not only to characterize magnetic systems³ but also to discuss the Berry curvature in non-magnets⁴.

While experiments to detect the Hall effect have been performed extensively, there are only a few reports that have addressed Hall effect measurements in ultrahigh vacuum (UHV). Even then, the Hall coefficient for non-magnetic systems has only been discussed⁵⁻⁸, and no AHE measurements have been performed up to now. Due to the rise of various two-dimensional magnets which can be derived from van der Waals layered crystals^{9,10} or intrinsic magnetic topological insulators^{11,12}, the need for a Hall effect measurement system in UHV environment has increased since some systems are not stable in air and capping the surface to prevent oxidation may change the sample magnetization. In this respect, *in situ* characterization is desired to discuss the intrinsic magnetic properties. However, there are hardly any facilities that are capable of performing such experiments. Furthermore, it is desired to simultaneously measure the electronic structure together with macroscopic magnetic properties since the difference in the sample quality as well as the degree of oxidation may also cause confusion in the data interpretation.

Therefore in the present study, we modified our all-in-one multimodal UHV system (Fig. S1) enabling sample fabrication and *in situ* angle-resolved photoemission spectroscopy (ARPES) as well as transport measurements^{13,14} and measured the AHE of an intrinsic magnetic topological insulator heterostructure $\text{MnBi}_2\text{Te}_4/\text{Bi}_2\text{Te}_3$ (MBT/BT) grown on Si(111) substrates¹⁵ with different surface structures. The Curie temperature was determined as 15 K for all the samples, but we found that the AHE shows a peculiar behavior when the heterostructure was grown on the $\beta\text{-Bi}/\text{Si}(111)\text{-}\sqrt{3} \times \sqrt{3}$ surface. This likely originates from the skew scattering from the Bi monolayer, or the change in the Berry curvature of the system since the states stemming from this interfacial Bi layer are present near the Fermi level (E_F) and show a crossing with the Dirac cone of MBT/BT. Our results show that a single monatomic layer decoration at the substrate surface can significantly alter the magnetotransport property of a two-dimensional magnet, which is a potentially important point in developing atomic-layer spintronic devices.

The heterostructure samples were prepared by molecular beam epitaxy equipped with a reflection-high-energy electron diffraction (RHEED) system. First, a clean Si(111)- 7×7 surface was prepared on an *n*-type substrate (1.5-5 Ωcm resistivity at room temperature) by a cycle of resistive heat treatments. The $\beta\text{-Bi}/\text{Si}(111)\text{-}\sqrt{3} \times \sqrt{3}$ (hereafter $\beta\sqrt{3}\text{-Bi}$) surface was formed by 1 ML ($7.83 \times 10^{14} \text{ cm}^{-2}$) of Bi deposition on the 7×7 surface at 620 K monitored by RHEED. Then Bi was deposited either directly on the 7×7 surface or after the formation of the $\beta\sqrt{3}\text{-Bi}$ surface at $\sim 250^\circ\text{C}$ in a Te-rich condition to grow quintuple layer (QL) structured Bi_2Te_3 films and further annealed at $\sim 250^\circ\text{C}$ for 5 minutes. This results in a QL by QL growth and the thickness is monitored with

^{a)}Present Address: Center for Basic Research on Materials, National Institute for Materials Science, Tsukuba 305-0003, Japan

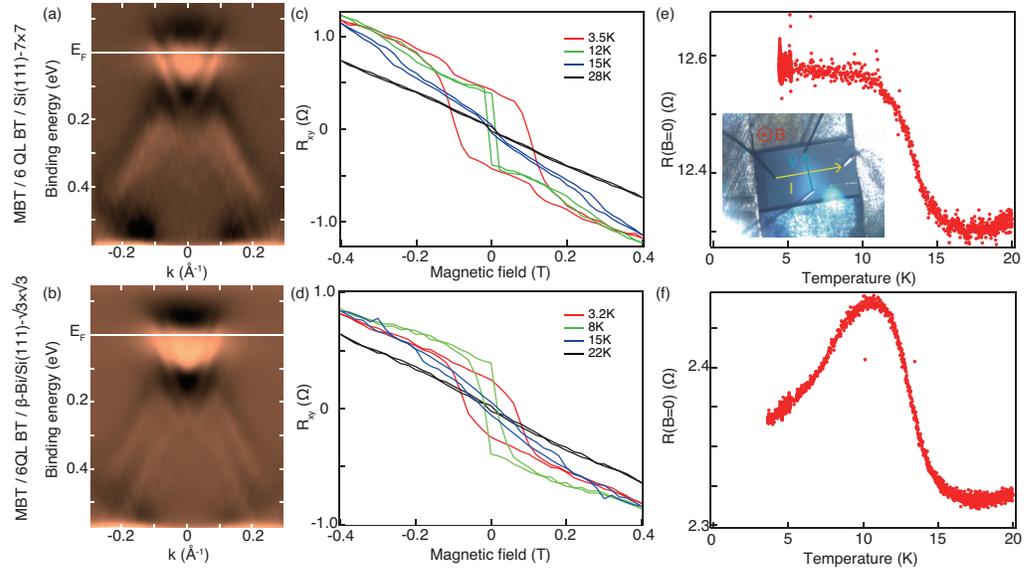


FIG. 1. (a, b) Band dispersion (second derivative) of MBT / 6 QL BT / Si(111)- 7×7 (a), and that of MBT / 6 QL BT / β -Bi/Si(111)- $\sqrt{3} \times \sqrt{3}$ (b), respectively, along the Γ -M direction measured at room temperature. (c, d) The asymmetric Hall resistance R_{xy} as a function of the applied magnetic field measured at various temperatures for MBT / 6 QL BT / Si(111)- 7×7 (c) and MBT / 6 QL BT / β -Bi/Si(111)- $\sqrt{3} \times \sqrt{3}$ (d), respectively. (e, f) Temperature dependence of the measured resistance R at zero field after magnetizing the sample for MBT / 6 QL BT / Si(111)- 7×7 (e) and MBT / 6 QL BT / β -Bi/Si(111)- $\sqrt{3} \times \sqrt{3}$ (f), respectively. The inset in (e) shows the measurement configuration.

RHEED oscillations. Finally, Mn was deposited on Bi₂Te₃ film in a Te-rich condition at ~ 260 °C. The 1×1 periodicity with the same lattice constant is maintained during this process for the samples we have fabricated^{15,16}.

The ARPES measurements were performed after the sample preparation with a commercial hemispherical photoelectron spectrometer equipped with angle and energy multidetectors (ScientaOmicron R4000) with He I α radiation (21.2 eV) at room temperature. Some additional ARPES measurements were performed in a different UHV chamber (ScientaOmicron DA20).

Then, the transport measurements were performed in our custom-made system^{13,14} which has four probes that can move independently and by touching the surface of the sample, four-point probe (4PP) resistance measurements can be performed in UHV (Figs. S2, S3). To measure the Hall effect, the probes were aligned in a cross geometry as shown in the inset of Fig. 1(e). The samples were cooled down to ~ 3 K in the present study and a magnetic field as large as 0.4 T was applied perpendicular to the sample surface. Although it is not possible to deduce physical parameters quantitatively since we have not fabricated a Hall bar structure, it is still possible to make a qualitative discussion as we will show in the following.

The samples were capped with ~ 10 nm of Te before taking

them out of the UHV chamber for scanning transmission electron microscopy (STEM) measurements. Electron transparent specimens for STEM observations were prepared by the standard lift-out technique using an FEI Helios G4-UX dual-beam system. Probe aberration corrected STEM, FEI TitanTMG2 80-200 microscope, was used. Chemical compositions were measured by energy-dispersive X-ray spectroscopy (EDS).

For structural optimization and electronic band calculations we used the Vienna Ab Initio Simulation Package^{17,18} with generalized gradient approximation (GGA-PBE)¹⁹ to the exchange-correlation potential. The interaction between the ion cores and valence electrons was described by the projector augmented-wave method^{20,21}. Relativistic effects were taken into account, including the spin-orbit interaction. In order to describe the vdW interactions we made use of the DFT-D3 functional with Becke-Johnson dumping scheme²². The geometry optimization was performed until the residual forces on atoms became smaller than 1 meV/Å. To correctly describe the highly correlated Mn-*d* electrons of MBT we include the correlation effects within the GGA+*U* method²³ within the Dudarev scheme²⁴. The $U_{\text{eff}} = U - J$ value for the Mn 3*d*-states was chosen to be equal to 5.34 eV^{11,25}. For analysis of interactions between atoms at the interfaces we use the projected crystal orbital Hamilton population (pCOHP) method^{26,27} implemented within Local-orbital basis suite to-

wards electronic structure reconstruction (LOBSTER) code²⁸.

Figures 1(a) and (b) show the band dispersion (second derivative) of MBT / 6 QL BT / 7×7 (a), and that of MBT / 6 QL BT / $\beta\sqrt{3}$ -Bi (b), respectively, along the $\bar{\Gamma}$ -M direction²⁹. One can find the linearly dispersion Dirac cone near the Fermi level (E_F) together with the bulk bands, showing that the films are n -doped. For the higher binding energy side, there are two bands dispersing away from the $\bar{\Gamma}$ point. These features are basically the same as what is shown in Ref. 15 and demonstrates that two samples apparently do not exhibit significant difference in the surface electronic structure.

However, some difference was observed in the transport data for the two samples. Figures 1(c) and (d) show the Hall resistance R_{xy} as a function of the applied magnetic field measured at various temperatures for MBT / 6 QL BT / 7×7 (c) and MBT / 6 QL BT / $\beta\sqrt{3}$ -Bi (d), respectively. These data were asymmetrized from the measured raw data R as explained in Fig. S5 to deduce the Hall resistance. It can be noticed that above 20 K, both samples exhibit the normal Hall effect, showing a linear dependence. The negative Hall coefficient shows that the carriers are electrons, consistent with the band dispersion measured with ARPES (Figs. 1(a) and (b)). Upon lowering the temperature, a hysteresis behavior can be observed, demonstrating the detection of the AHE. In Fig. 1(c), the value of R_{xy} at zero field does not change so much for the data at 12 K and 3.5 K, but the value of the magnetic field that R_{xy} crosses zero Ω increases. This shows that the residual magnetization does not increase but the coercive field increases below 12 K. This is a typical behavior of ferromagnets and one can say that the measured AHE is reflecting the magnetization of the MBT/BT heterostructure. Since the contribution of the longitudinal resistance R_{xx} can be expected to be very small in the measured R as shown in Fig. S5, we measured the temperature dependence of $R(B=0)$ after the field sweep to deduce the Curie temperature T_c . As shown in Fig. 1(e), one can find a sharp rise around 15 K by lowering the temperature and this shows that T_c is ~ 15 K. Although we could not detect any signal of ferromagnetism for this sample with *ex situ* XMCD measurements down to 6 K¹⁵, the present measurements unambiguously suggest that the MBT/BT heterostructure grown on Si(111)- 7×7 is ferromagnetic below 15 K, which is similar to the report in Ref. 12. The origin of this discrepancy cannot be clearly elucidated at this time.

A qualitatively different behavior was observed for the MBT/BT heterostructure grown on $\beta\sqrt{3}$ -Bi. As shown in Fig. 1(d), the clear detection of the hysteresis loop shows that this system is also ferromagnetic. However, although the coercive field is larger for the data at 3.2 K compared to that at 8 K, R_{xy} at zero field is smaller at 3.2 K. This characteristic can be seen more vividly in the temperature dependence of $R(B=0)$ as shown in Fig. 1(f). The signal shows a sharp rise at 15 K, but starts to decrease again below 10 K. This feature is clearly different from the behavior shown in Fig. 1(e) and cannot be explained by a simple effect of typical ferromagnetism.

To elucidate the origin of this peculiar behavior found in AHE, we have performed STEM measurements. Figure 2(a) shows the STEM image for the MBT / 6 QL BT / 7×7 . Between the Te capping layer and the Si substrate, the struc-

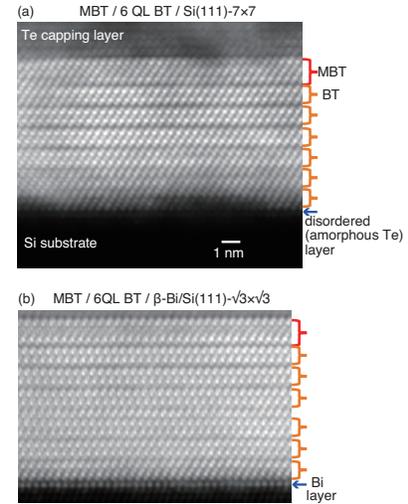


FIG. 2. TEM image of MBT / 6 QL BT / Si(111)- 7×7 (a), and that of MBT / 6 QL BT / β -Bi/Si(111)- $\sqrt{3} \times \sqrt{3}$ (b), respectively. The MBT and BT layers are explicitly indicated and the difference in the interface between the Si substrate and the film is emphasized.

ture can be mostly identified as the designed one. It should be emphasized that at the heterostructure/substrate interface, the image is somehow disturbed. This is due to the fact that disordered Te will build up as a wetting or a buffer layer to saturate dangling bonds of the 7×7 surface^{30,31} before the BT growth. In contrast, the heterostructure/substrate interface for the MBT / 6 QL BT / $\beta\sqrt{3}$ -Bi shown in Fig. 2(b) has a sharp appearance with a well-defined monolayer. This is likely due to the Bi-trimer layer which originally formed the $\beta\sqrt{3}$ -Bi surface reconstruction³², although we have no evidence whether the in-plane arrangement has changed or not upon the deposition of BT. But the EDS mapping shows that this is a Bi layer as shown in Fig. S6. Since there is hardly any difference in the distribution of unintentionally displaced Mn atoms that can influence the magnetic property as measured by EDS (Fig. S7)³³⁻³⁷, we conclude that the only clear difference in the atomic structure between the two samples is found at the BT/Si interface.

It can be anticipated that since the top MBT surface layers which is probed by ARPES is not different for the two systems, the band dispersion shown in Figs. 1(a) and (b) demonstrate no difference. However, because our films are n -doped and the bulk carriers are also involved in transport, it can be speculated that the difference in the interface structure can influence the AHE signal. It is known that the band dispersion of BT films grown on Si(111)- 7×7 films can be reproduced by DFT calculations for free-standing BT slabs³⁸. However, it seems that we need to take into account the Bi layer at the BT/Si interface to describe the electronic structure of BT /

$\beta\sqrt{3}$ -Bi. To this end, we have constructed a model shown in Fig. 3(a), in which 5 QL of BT was placed on top of $\beta\sqrt{3}$ -Bi. The in-plane lattice constants of BT(111) and Si(111)- $\sqrt{3} \times \sqrt{3}$ differ approximately by factor 1.5 (4.38 vs. 6.69 Å) and hence the better matching between them is realized with superlattices ratio of 3/2, *i.e.* BT(111)-3 \times 3 should be placed on a β -Bi/Si(111) with $2\sqrt{3} \times 2\sqrt{3}$ periodicity with 1.9% compression. To simulate the Si(111) substrate we used a slab of two bi-layers (BL) thickness and Hydrogen atoms were used to passivate the Si dangling bonds at the bottom (unreconstructed) surface of the slab. The atomic positions of the bottom BL were fixed at their bulk positions while atoms of the top Si BL as well as β -Bi layer and in the interface QL of BT were relaxed. The calculated band dispersion is depicted in Fig. 3(b) and the different markers show the states localized at different positions in the slab structure.

The bonding energy between Bi-trimer atoms with the underlying Si atoms and that with the nearest Te atoms of the BT were calculated and it turns out that the interaction of BT with the Bi-trimer layer is derived to be about 9 times weaker compared to that between the Bi-trimers and the Si(111) substrate. Due to this weak interaction at the BT/ $\beta\sqrt{3}$ -Bi interface, the variation in the interface potential is relatively small and limited to the first (closest to the interface) QL, as shown in Fig. 3(c). This makes the seemingly complicated band dispersion of Fig. 3(b) easy to understand. The states marked by red circles in Fig. 3(b) have their origin as the Rashba-split surface states near the \bar{K} and \bar{M} points (Fig. S8(a)) of the $\beta\sqrt{3}$ -Bi surface³², but the energy position has shifted closer to E_F due to the change in the potential at the interface. It should also be noted that due to the imposed $2\sqrt{3} \times 2\sqrt{3}$ periodicity, the Rashba states of $\beta\sqrt{3}$ -Bi are folded to the $\bar{\Gamma}$ point. Similarly, the energy level of the Dirac cone at the top surface (grey circles in Fig. 3(b)) and that at the interface (blue circles) is shifted due to the potential difference at the interface and at the vacuum side of BT.

As discussed above, we are not sure if the above structure model is actually reflecting the situation in the experiment. However, the presence of the bands near the Fermi level localized at the interface Bi layer in addition to the BT Dirac cone seems to be universal irrespective of the presence of the MBT septuple layer at the top (Fig. S8(b)) or whether the BT film thickness is only 1 QL nor the change in stacking between the BT and the β -Bi/Si(111)- $2\sqrt{3} \times 2\sqrt{3}$ surface (Figs. S8(c)-(e)). Therefore, we have experimentally tried to confirm the presence of this interface state for the 1 QL BT/ $\beta\sqrt{3}$ -Bi system. As shown in Fig. S9, there are indeed some features near the Fermi level other than the Dirac cone, but since the intensity is quite weak, we cannot make a direct comparison to the calculated band dispersion. Nevertheless, this shows that interface states localized at the β -Bi interface layer can affect the film property for the MBT / BT grown on the $\beta\sqrt{3}$ -Bi surface.

The most simple explanation concerning the unique behavior of the AHE shown in Fig. 1(d) and (f) is that the carriers undergo skew scattering at the interfacial layer and this can give rise to extrinsic AHE due to the strong SOC of Bi. However, since it is well known that the extrinsic mechanism is

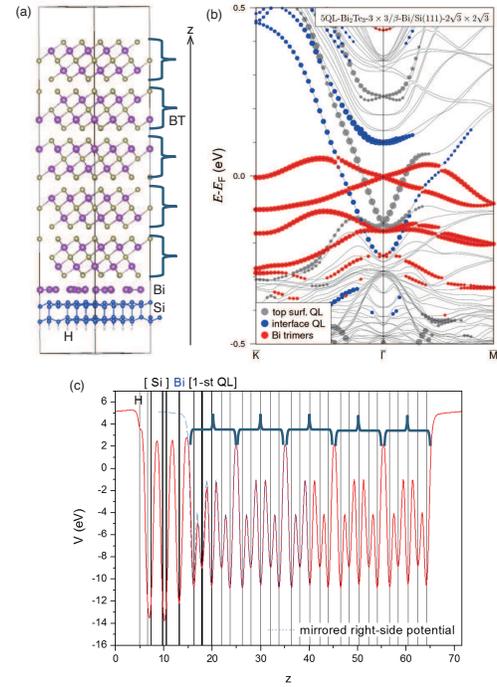


FIG. 3. (a) Atomic structure model (the presented structure was visualized with VESTA³⁹) and (b) calculated band dispersion near E_F of the 5 QL BT / β -Bi/Si slab. The different markers show the states localized at different positions within the model shown in (a). (c) In-plane (x, y) averaged electrostatic potentials $V(z)$ within the slab. Since the interaction at the Bi/BT interface is weak, there is only a slight change compared to the vacuum/BT interface.

only dominant for low-mobility (side jump) or high-mobility (skew scattering) systems and most materials can be described by intrinsic effects due to the Berry curvature^{2,40}, we speculate that the AHE of MBT / 6 QL BT / $\beta\sqrt{3}$ -Bi may also be affected by the band structure. One can obviously find that the states originating from the β -Bi layer and the interfacial Dirac cone cross each other (Fig. 3(b)). Such crossing may be a source of Berry curvature, although the actual calculation is beyond the scope of the present work⁴¹. Nevertheless, we emphasize that the present results demonstrate a convenient way to manipulate the Berry curvature artificially using a single layer at the heterostructure/substrate interface, which can potentially be used to develop spintronic devices using atomically thin magnets.

In summary, by means of the developed *in situ* Hall measurement setup, we have shown that the AHE in magnetic topological insulator heterostructures MnBi₂Te₄/Bi₂Te₃/Si(111) can be manipulated by controlling the BT/Si interface. The formation of the β -Bi/Si(111)-

$\sqrt{3} \times \sqrt{3}$ surface reconstruction before the heterostructure growth results in appearance a local maximum in the AHE signal at 10 K. This behavior can stem from the skew scattering at the interface, or the effect of the states localized at the Bi interfacial layer that resides at the same energy as the Dirac state of the bottom side of the MBT/BT film, which can affect the Berry curvature of the system.

See supplementary material for details on the instrumental apparatus, linearity of the measured IV curves, raw ARPES data of Figs. 1(a) and (b), data processing in the Hall measurements, EDS mapping of the samples, additional results of DFT calculations, and ARPES data of 1QL BT / $\beta\sqrt{3}$ -Bi.

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AUTHOR DECLARATIONS

CONFLICT OF INTEREST

The authors have no conflicts of interest to disclose.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

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