Realization of semimagnetic and magnetic topological insulators via topological surface states floating

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The observation temperatures of quantum anomalous Hall effect in semimagnetic and magnetic topological insulator (MTI) heterostructures are always much lower than Curie temperature. Here, we theoretically demonstrate that floating of topological surface states (TSSs) into magnetic insulators is not only able to produce ideal semi-MTI heterostructures, but also provides insight into the origin of extremely low observation temperature of the quantum anomalous Hall effect in different MTIs. We show that the emergence of diverging and floating of TSSs can be observed in MnBi2Te4/Bi2Se3 family heterostructures. Within the TSSs floating systems, MnBi2Te4/Sb2Te3 and NiBi2Te4/Sb2Te3 exhibit characteristics of ideal semi-MTIs, featuring a tunable Fermi level, and MnBi2Te4/Bi2Te3/NiBi2Te4 (NiBi2Te4/Sb2Te3/VBi2Te4) is found to be MTI with weak (strong) long-range ferromagnetic coupling and large (small) surface gap. Our findings reveal that the observation temperature of quantum anomalous Hall effect is governed by the smaller of the two surface gaps, rather than being directly linked to the Curie temperature of MTIs.

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I. INTRODUCTION

Topological insulators (TIs) exhibit robust topologically protected surface states along with insulating bulk states [1–3]. When the ferromagnetism is introduced, the time-reversal symmetry breaking leads to the emergence of a new topological state characterized by massive Dirac fermions on the nontrivial topological surface [4]. Exploring such a kind of magnetic topological insulator (MTI) systems is one of the most researched frontiers in condensed-matter physics [5]. In a perpendicularly magnetized MTI film, the gapped surface bands at top and bottom surfaces contribute to a total Hall conductance of $e^2/h$, giving rise to quantum anomalous Hall effect (QAHE) [6–8]. The pioneering theoretical proposal for QAHE was Haldane’s honeycomb-lattice model [9]. Extensive efforts have been devoted to exploring realistic materials systems for realizing QAHE [10–16]. Doping magnetic elements into TIs leads to the first successful observation of QAHE in experiment [17–21]. However, all the experimentally observed QAHEs in doped TIs to date have been limited to extremely low temperatures. Several studies indicated that inhomogeneous ferromagnetism plays a crucial role in determining the low temperature [22–26]. Furthermore, QAHE has also been observed in other systems, including Te-based heterostructures involving Cr-doped ZnTe magnetic semiconductors [27], intrinsic magnetic TIs such as MnBi2Te4 [28], twisted bilayer or trilayer graphene [29,30], and transition-metal dichalcogenide [31]. Despite the presence of homogeneous magnetism in some MTIs, the observation temperature in these systems remains pretty low, farther below the corresponding Curie temperature (T_C) of the MTIs.

Aside from MTIs, the semi-MTIs have been theoretically proposed, wherein magnetization in a three-dimensional TI induces a gap at the Dirac surface band perpendicular to the magnetization vector, resulting in a quantum Hall system with half-quantized Hall conductance ($e^2/2h$) [32,33]. However, experimentally observing the half-quantized Hall conductance is challenging due to the simultaneous appearance of paired Dirac cones. Several approaches have been proposed to measure the half-quantized Hall conductivity in different systems, such as Co/BiSbTeSe2 [34], Cr2Ge2Te6/BiSbTeSe2 [35], and MnBi8Te13 [36]. Recently, Mogi et al. successfully observed the half-quantized Faraday and Kerr rotations, as well as half-quantized Hall conductance in zero magnetic field transport experiments conducted on semi-MTI films comprised of (Bi,Sb)2Se3 and Cr doped (Bi,Sb)2Se3 [37]. However, similar to QAHE, the quantization occurs at temperatures below 2 K, primarily due to the random-doping induced magnetic inhomogeneity. Furthermore, it is shown that the precision of the half quantized $\sigma_y$ increases when the Fermi level approaches the Dirac point of the other surface state. Therefore, it is highly desirable for high-temperature and high-precision semi-MTIs possessing a single large surface gap and a tunable Fermi level.
Increasing the observation temperature of quantized Hall conductance, whether in MTIs or semi-MTIs, is a practical challenge. In this work, we design Bi$_2$Se$_3$/MnBi$_2$Te$_4$ family heterostructures by utilizing the topological proximity effect, which transfers TSSs from topological insulators to conventional materials [38–41]. It not only facilitates the creation of ideal semi-MTI heterostructures, but also provides valuable insights into the low observation temperature of various QAHE systems. Specifically, from first-principles calculations, we find that MnBi$_2$Te$_4$/Sb$_2$Te$_3$ and NiBi$_2$Te$_4$/Sb$_2$Te$_3$ exhibit characteristics of high-temperature semimagnetic TIs. Moreover, we identify MnSb$_2$Te$_4$/Bi$_2$Te$_3$/NiBi$_2$Te$_4$ (NiBi$_2$Te$_4$/Sb$_2$Te$_3$/VBi$_2$Te$_4$) as MTIs with weak (strong) long-range ferromagnetic coupling and large (small) surface gaps. Further analysis demonstrates that the QAHE observation temperature is determined by the smaller of the two opened surface gaps and is independent of T$_C$ of MTIs.

II. METHODS

Our first-principles calculations were performed by using the projected augmented-wave method [42] as implemented in the Vienna ab initio simulation package (VASP) [43]. The generalized gradient approximation (GGA) of Perdew-Burke-Ernzerhof (PBE) type was used to treat the exchange-correlation interaction [44]. The lattice constants of Bi$_2$Se$_3$, Bi$_2$Te$_3$, and Sb$_2$Te$_3$ were adopted from experiments, which were 4.14 Å, 4.26 Å, and 4.38 Å, respectively. A vacuum buffer space of 20 Å was used to prevent the coupling between adjacent slabs. The kinetic energy cutoff was set to be 350 eV. During structural relaxation, all atoms were allowed to relax until the Hellmann-Feynman force on each atom is smaller than 0.01 eV/Å. The Brillouin zone integration was carried out by using $9 \times 9 \times 1$ Monkhorst-Pack grids for different heterostructure systems. Unless mentioned otherwise, spin-orbit coupling and the GGA + U method was used to treat the Coulomb interaction of 3d transition metals, i.e., Mn, Ni, and V with U = 4.0 eV, 4.0 eV, and 3.0 eV, respectively [45]. In addition, we use the DFT-D3 method to describe the van der Waals interaction in our numerical calculations [46]. The most stable stacking of different heterostructures was found according to a previous study from a similar system MnBi$_2$Te$_4$/Bi$_2$Te$_3$ [47–51].

III. RESULTS AND DISCUSSION

A. Systems and their electronic structures

For MI/TI heterostructures as displayed in Fig. 1, TSSs respectively exhibit different behaviors, i.e., diving into inner quintuple layer (QL) of TI, being pinned at the interface, or floating into MI, depending on the coupling strength (strong, weak, or moderate) between MI and TI. In the case of diving MI/TI heterostructures, the realization of QAHE is challenging since the magnetic proximity effect has a relatively short length scale (a few angstroms) and TSSs are farther away from the MI/TI interface. To achieve a moderate interaction between magnetism and TSSs, the pinning or floating TSSs are better choices. Therefore, we focus on different van der Waals (vdW) layered semi-MTIs and MTIs constructed by single layer ferromagnetic insulators [MnBi$_2$Te$_4$, MnSb$_2$Te$_4$, NiBi$_2$Te$_4$, or VBi$_2$Te$_4$] and 5QLs TIs [Bi$_2$Se$_3$, Bi$_2$Te$_3$, and Sb$_2$Te$_3$] (see Fig. 1) with similar interfacial elements to investigate the topological proximity effect phenomenon.

Figures 2(a) and S1 [52] display the atom-resolved band structures of MnBi$_2$Te$_4$/Bi$_2$Se$_3$, MnSb$_2$Te$_4$/Bi$_2$Se$_3$, NiBi$_2$Te$_4$/Bi$_2$Se$_3$, and VBi$_2$Te$_4$/Bi$_2$Se$_3$ heterostructures, respectively. In MnBi$_2$Te$_4$/Bi$_2$Se$_3$, the degeneracy of two TSSs from Bi$_2$Se$_3$ is clearly lifted due to strong interaction between MnBi$_2$Te$_4$ and the upper surface of Bi$_2$Se$_3$, while preserving two Dirac points. To find out the spatial location of TSSs, the atom-specific character of each band is plotted, denoted by dots in Fig. 2(a). It is observed that the Dirac-cone-like TSSs are located at the interface of the fourth and fifth QLs (upper surface) and the first QL (lower surface) of Bi$_2$Se$_3$. It is noted that there is no electron weight from MnBi$_2$Te$_4$ contributing to Dirac points, implying a negligible interaction between magnetism and TSSs. Figure 2(b) reveals that the upper-surface Dirac point primarily resides at the interface of the fourth and fifth QLs, with no spatial superposition occurring between Mn atoms of MnBi$_2$Te$_4$ and the upper surface. Similar findings are observed in MnSb$_2$Te$_4$/Bi$_2$Se$_3$, NiBi$_2$Te$_4$/Bi$_2$Se$_3$, and VBi$_2$Te$_4$/Bi$_2$Se$_3$ heterostructures. Thus, in Bi$_2$Se$_3$-based heterostructures, no topological proximity effect manifests since the upper-surface Dirac points are shifted to deeper QLs of Bi$_2$Se$_3$, resulting in a negligible interaction between magnetism and TSSs.

As displayed in Figs. 2(c)–2(h), S2 [52], and S3 [52], one can see that, compared to Bi$_2$Se$_3$ based heterostructures, the upper-surface TSSs of Bi$_2$Te$_3$ and Sb$_2$Te$_3$ float into FMIIs. This floating behavior of TSSs leads to a strong interaction between magnetism and TSSs, resulting in opening of surface band gaps ranging from 72.9 to 148.1 meV. The atom-specific character of each band reveals that these surface band gaps are solely attributed to MIIs and the fifth QL (upper surface) of TIs. On the other hand, the Dirac points of the other surface state of TIs remain perfectly preserved, particularly...
FIG. 2. Atom-resolved band structures of (a) MnBi$_2$Te$_4$/Bi$_2$Se$_3$, (c) MnBi$_2$Te$_4$/Bi$_2$Te$_3$, (e) MnBi$_2$Te$_4$/Sb$_2$Te$_3$, and (g) NiBi$_2$Te$_4$/Sb$_2$Te$_3$ heterostructures. Size and color of dots denote different spectral weights and contributions from different atoms, respectively. Gray denotes the opened surface band gap due to interaction between ferromagnetic insulator and TSSs. The real-space charge density distribution of the upper-surface TSSs at the $\Gamma$ point around the Dirac points in (b) MnBi$_2$Te$_4$/Bi$_2$Se$_3$, (d) MnBi$_2$Te$_4$/Bi$_2$Te$_3$, (f) MnBi$_2$Te$_4$/Sb$_2$Te$_3$, and (h) NiBi$_2$Te$_4$/Sb$_2$Te$_3$ heterostructures. The gray and bars indicate the spacial locations of the different QLs and 1 SL ferromagnetic insulator. Purple line indicates the position of Mn or Ni in ferromagnetic insulator.

for the MnBi$_2$Te$_4$/Sb$_2$Te$_3$ and NiBi$_2$Te$_4$/Sb$_2$Te$_3$ heterostructures. The real-space DOS of the upper-surface TSSs at Dirac points [see Figs. 2(d), 2(f), and 2(h)] demonstrates that the upper-surface Dirac points float into MIs via the topological proximity effect, resulting in spatial superposition between MnBi$_2$Te$_4$, NiBi$_2$Te$_4$, and the upper surfaces. Interestingly, there exists a quantitatively correlation between the surface band gap and the spatial superposition of Mn or Ni with TSSs. For instance, the maximum peak of the floating TSS aligns precisely with the Ni position [see Fig. 2(h)], leading to a maximum surface band gap around 148.1 meV.

B. Semimagnetic topological insulators

Although semi-MTI has been achieved in magnetic-doped TI/TI heterostructures [37], it is still demanding for high-temperature and high-precision semi-MTIs with a single large surface gap and a tunable Fermi level close to the Dirac point of the other surface state. Among our TSSs floating systems, MnBi$_2$Te$_4$/Sb$_2$Te$_3$ and NiBi$_2$Te$_4$/Sb$_2$Te$_3$ stand out as the most promising candidates (as shown in Fig. S4 [52], the calculated Hall conductivities of the MBT/ST and NBT/ST are all equal to $e^2/2h$). In order to observe high-precision half-quantized Hall conductivity, it is essential to have a sufficiently large surface band gap opening. For MnBi$_2$Te$_4$/Sb$_2$Te$_3$ and NiBi$_2$Te$_4$/Sb$_2$Te$_3$ systems, their surface band gaps are 99.6 and 148.1 meV, respectively. These not only make them feasible to observe high-temperature half-quantized Hall conductivity, but also provide ample energy scales to shift the Fermi level. Furthermore, achieving high-precision half-quantized Hall conductivity requires a tunable Fermi level, which is achievable in these two systems by tuning the number of QLs of Sb$_2$Te$_3$. In Figs. S5(a) [52] and S6 [52], we present the opened surface band gap range of 60.0–99.6 meV and the energy difference between the Dirac point and Fermi level of 29–45 meV for MnBi$_2$Te$_4$/Sb$_2$Te$_3$ heterostructures. To further validate MnBi$_2$Te$_4$/Sb$_2$Te$_3$ as a semi-MTI, we provide atom-resolved band structures and real-space DOS of the upper-surface TSS in Figs. S5(b) [52] and S5(c) [52]. In comparison with MnBi$_2$Te$_4$/5-QLs Sb$_2$Te$_3$, the opened surface band gap primarily originates from the interaction between MnBi$_2$Te$_4$ and the sixth QL, independent of the fifth QL, which reinforces the notion that the opened band gap indeed originates from the interaction between TSSs and magnetism. Regarding the Dirac point, it is situated at the first QL of Sb$_2$Te$_3$. To demonstrate evidence of the topology of the opened surface band gap, we observe the band inversion between Bi and Te $p_z$ orbitals in MnBi$_2$Te$_4$/6-QLs Sb$_2$Te$_3$ heterostructures. Conversely, for other Bi$_2$Te$_3$ or Sb$_2$Te$_3$ based heterostructures, as depicted in Figs. S2 [52] and S3 [52], they do not qualify as ideal semi-MTIs due to varying degrees of interaction between the other surface state and MI, which would drive the quantized Hall conductivity to deviate from 1/2 in experimental observation.

C. Topological proximity effect

Now, let us move to explore the distinct behaviors exhibited by TSSs in Bi$_2$Se$_3$ (without topological proximity effect) and Bi$_2$Te$_3$ or Sb$_2$Te$_3$ (with topological proximity effect) based
heterostructures. We examine the work function, difference of work functions, and lattice mismatch of each heterostructure, which are presented in Table S1 [52]. By comparing with Bi$_2$Te$_3$ based heterostructures, a smaller difference of work functions and lattice mismatch are achieved in Bi$_2$Te$_3$ or Sb$_2$Te$_3$ based heterostructures. Similar findings have been reported in conventional insulator/TI heterostructures, where smaller lattice mismatch and work function difference lead to more pronounced topological proximity effect [38]. Otrokov et al. found that the MI/TI heterostructure with the same or an affine crystal structure will benefit for strong interaction between magnetism and TSS [26]. It is noteworthy that a small lattice mismatch is also present in all-Te based heterostructures, where Bi$_2$Te$_3$ and Sb$_2$Te$_3$ based heterostructures, we further shown in Fig. S4 [52], the Hall conductivities are identified as long-range FM systems, making them similar findings have been with Bi$_2$Se$_3$ based heterostructures, a smaller difference of more pronounced topological proximity effect [38]. Otrokov et al. smaller lattice mismatch and work function difference lead to reported in conventional insulator/TI heterostructures. Consequently, MnSb$_2$Te$_4$ and MnBi$_2$Te$_4$ via topological proximity effect, resulting in spatial superposition between MnSb$_2$Te$_4$, NiBi$_2$Te$_4$, and the first/fifth QLs. Furthermore, a quantitative correlation between the opened surface band gap and the superposition of Mn or Ni in the MI is observed. Similar conclusions can be drawn for the VBi$_2$Te$_4$/Sb$_2$Te$_3$/NiBi$_2$Te$_4$ heterostructures. Consequently, MnSb$_2$Te$_4$/Bi$_2$Te$_3$/NiBi$_2$Te$_4$ and VBi$_2$Te$_4$/Sb$_2$Te$_3$/NiBi$_2$Te$_4$ can be classified as MTIs.

However, a significant distinction arises between MnSb$_2$Te$_4$/Bi$_2$Te$_3$/NiBi$_2$Te$_4$ and VBi$_2$Te$_4$/Sb$_2$Te$_3$/NiBi$_2$Te$_4$, i.e., the energy difference between corresponding FM and AFM states are −7.79 and −168.96 meV, respectively. To uncover the physical origin, Fig. S8 [52] displays the real-space DOS of the upper and lower surface TSSs in three different heterostructures. The energy difference between FM and AFM states is proportional to the superposition strength between upper and lower surface states within third and fourth QLs. Notably, the most pronounced superposition between the upper and lower surface states occurs within the third and fourth QLs of VBi$_2$Te$_4$/Sb$_2$Te$_3$/NiBi$_2$Te$_4$, resulting in the strongest long-range ferromagnetic coupling. Kou et al. previously reported a typical penetration depth of 2–3 QLs for TSSs [53]. Based on this finding, for a 5 QLs Sb$_2$Te$_3$, both top and bottom layers exhibit surface states with a penetration depth of 2–3 QLs. Consequently, the long-range ferromagnetic coupling can be realized through the penetration of TSSs and, particularly, their strong superposition or resonance within these MTI systems.

**E. Origin of extremely low observation temperature of QAHE**

Regarding MnSb$_2$Te$_4$/Bi$_2$Te$_3$/NiBi$_2$Te$_4$, the minimum lower-surface band gap (43.8 meV) is theoretically large enough to allow for the manifestation of QAHE at room temperature. However, the $T_C$ of MnSb$_2$Te$_4$/Bi$_2$Te$_3$/NiBi$_2$Te$_4$ falls well below room temperature. As a result, the observation temperature of the QAHE will be determined by $T_C$. On the other hand, the analysis produces different results for VBi$_2$Te$_4$/Sb$_2$Te$_3$/NiBi$_2$Te$_4$. The minimum lower-surface band gap (13.5 meV) is sufficient to realize QAHE above 150 K (considering that the thermal motion energy at room temperature is approximately 26 meV). However, $T_C$ of VBi$_2$Te$_4$/Sb$_2$Te$_3$/NiBi$_2$Te$_4$ exceeds 150 K. Therefore, the observation temperature of QAHE in VBi$_2$Te$_4$/Sb$_2$Te$_3$/NiBi$_2$Te$_4$ will be determined by the minimum surface band gap (13.5 meV), implying a theoretically predicted observation temperature of approximately 150 K. Consequently, for the high-$T_C$ VBi$_2$Te$_4$/Sb$_2$Te$_3$/NiBi$_2$Te$_4$, the observation temperature of QAHE will be constrained by the smaller surface band gap associated with the upper-surface states of the MTI.
It is known that the observation temperature of QAHE in various MTIs is consistently lower than their respective $T_C$. Similarly, this situation arises in our predicted VBi$_2$Te$_4$/Sb$_2$Te$_3$/NiBi$_2$Te$_4$ system. In the case of magnetically doped TIs, previous studies have demonstrated that random distribution of local magnetic moments interacting with surface Dirac electrons leads to spatial fluctuations in the magnitude of the opened surface gap [22–25]. The observation temperature of QAHE in magnetically doped TIs is limited by the minimum surface gap associated with the smallest local magnetic moment. The extremely low observation temperature of QAHE is also attributed to the magnetic inhomogeneity in all-Te based heterostructures [27]. In line with these findings, the same scenario occurs in the VBi$_2$Te$_4$/Sb$_2$Te$_3$/NiBi$_2$Te$_4$ system characterized by homogeneous magnetism. Therefore, as depicted in Fig. 3(e), it can be inferred that the observation temperature of QAHE will always be determined by the minimum surface band gap resulting from the interaction between the minimum local magnetic moment and surface Dirac electrons when $T_C$ exceeds the surface band gap.

Based on the aforementioned discoveries, we propose a predictive mechanism, illustrated in Fig. 3(f), that relates the opened surface band gap to the occurrence of QAHE in sandwiched Bi$_2$Te$_3$ and Sb$_2$Te$_3$ based heterostructures. However, it is important to note that the existence of long-range ferromagnetism or an external magnetic field is a prerequisite for this predictive mechanism. For MnSb$_2$Te$_4$/Bi$_2$Te$_3$/NiBi$_2$Te$_4$, our results demonstrate the presence of large surface band gap MTIs, whereas for other considerations, they are confirmed to be small surface-band-gap MTIs (see Fig. S9 [52]). Additionally, the band structures of VBT/ST/BT/MBT and MBT/ST/BT/MBT as predictive systems have also been as test calculations (see Fig. S10 [52]), which agree with the predictive results provided in Fig. 3(f). By employing this predictive mechanism, it is possible to obtain high or low-temperature QAHE depending on the restricted surface band gap in various MTI heterostructures. Furthermore, this predictive mechanism can potentially be extended to other MTIs utilizing magnetic proximity effect and topological proximity effect by estimating the opened surface band gap of semi-MTIs.

**IV. CONCLUSIONS**

In conclusion, by manipulating floating of TSSs, we predict the occurrence of high-temperature half-integer quantized Hall conductance in MnBi$_2$Te$_4$/Sb$_2$Te$_3$ and NiBi$_2$Te$_4$/Sb$_2$Te$_3$. We confirm that MnSb$_2$Te$_4$/Bi$_2$Te$_3$/NiBi$_2$Te$_4$ and NiBi$_2$Te$_4$/Sb$_2$Te$_3$/VBi$_2$Te$_4$ can host high-temperature QAHE. Our findings reveal that the observation temperature of QAHE is determined by the smaller of the two opened surface gaps and is not directly correlated to the $T_C$ of the MTIs.
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[52] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.109.155427 for atom-resolved band structures of Bi2Se3, Bi2Te3, and Sb2Te3 heterostructures, Hall conductivity of semi-MTIs, variation of opened surface band gap and band structures of MBT/ST and NBT/ST semi-MTIs, work function and, work function difference and lattice mismatch of TI based heterostructures, schematic diagram depictions of the heterostructure between a MI and TI, energy difference between the FM and AFM ground states of the whole system in all sandwiched ST and BT based heterostructures, real-space DOSs of the upper and lower surface TSSs in three typical heterostructures, and atom-resolved band structures of magnetic topological insulators.