Realization of quantized anomalous Hall effect by inserting CrI\textsubscript{3} layer in Bi\textsubscript{2}Se\textsubscript{3} film

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Abstract

It is challenging to realize the quantum anomalous Hall effect (QAHE) at high operating temperatures using the two-dimensional (2D) Dirac surface states of three-dimensional (3D) topological insulators (TIs). Given the small non-trivial gap induced by adsorbing ferromagnetic (FM) CrI\textsubscript{3} monolayer (ML) on the surface of Bi\textsubscript{2}Se\textsubscript{3} films, we here propose another TI and FM semiconductor interfaced system to enhance the gap by inserting CrI\textsubscript{3} ML between the first top (bottom) quintuple layers (QL) and sub-top (sub-bottom) QL of Bi\textsubscript{2}Se\textsubscript{3} films symmetrically. The 2D non-trivial phase emerges in the Bi\textsubscript{2}Se\textsubscript{3} films with five or more QLs and the gap is enlarged to 30 meV in 1QL-Bi\textsubscript{2}Se\textsubscript{3}/CrI\textsubscript{3}/4QL-Bi\textsubscript{2}Se\textsubscript{3}/CrI\textsubscript{3}/1QL-Bi\textsubscript{2}Se\textsubscript{3}, which can be understood by the enhanced magnetic proximity effect. The topological non-triviality is confirmed by the nonzero Chern number and the existence of chiral edge state. Our finding will provide useful guidance to optimize the Bi\textsubscript{2}Se\textsubscript{3}–CrI\textsubscript{3} interface system for realizing QAHE at relatively high operating temperatures.

1. Introduction

Bi\textsubscript{2}Se\textsubscript{3} is well-known three-dimensional (3D) topological insulators (TIs). It was reported the two-dimensional (2D) helical surface states of Bi\textsubscript{2}Se\textsubscript{3} film will be gaped when the film thickness is reduced to less than six quintuple layers (QLs) due to the interactions between the top and bottom surface [1, 2]. Usually, the gap is considered as topologically trivial. The 2D gapless surface states may be opened up by magnetic impurities [3, 4]. The opened gap induced by magnetic dopants holds possibility of realizing quantum anomalous Hall effect (QAHE) [5], and has been successfully confirmed in Cr- or V-doped (Bi, Sb)\textsubscript{2}Te\textsubscript{3} thin films experimentally [6–8]. However, the operating temperatures for QAHE is extremely low (30 mK) [6]. Therefore, the long-range 2D ferromagnetic (FM) order is expected to magnetize the 2D surface state of Bi\textsubscript{2}Se\textsubscript{3} film, which is one necessary pre-condition of realizing QAHE.

There are theoretical works proposed the 3D TIs/magnetic insulator heterostructures possess a gaped surface states due to the magnetic proximity effect breaking time-reversal symmetry [9, 10]. However, how to realize the QAHE at high operating temperatures is still very challenging. Normally it is difficult to find 2D magnetic crystals [11–15] because magnetic order is forbidden by the 2D isotropic Heisenberg model at non-zero temperatures [16]. Despite this, magnetic anisotropy could lead to the occurrence of 2D Ising ferromagnetism. Monolayer (ML) chromium triiodide (CrI\textsubscript{3}) was observed as Ising ferromagnet with...
out-of-plane spin orientation by using magneto-optical Kerr effect microscopy [17, 18]. Recent calculation demonstrated the trivial gaps induced by the inter-surface interactions are converted into topologically non-trivial ones by adsorbing CrI$_3$ ML on the both surface of Bi$_2$Se$_3$ films [19]. Starting from five QLs of Bi$_2$Se$_3$, the QAHE is expected to be realized in the heterostructure of Bi$_2$Se$_3$ film sandwiched by CrI$_3$ ML (CrI$_3$/Bi$_2$Se$_3$-film/CrI$_3$). However, the non-trivial gap is small (<5.9 meV) and hence restrict the operating temperatures of QAHE, despite the nontrivial gap was proposed to be increased to ~10 meV by numerically decreasing the distance between the Bi$_2$Se$_3$ film and CrI$_3$ ML.

In this work, we try to find feasible route to enhance the non-trivial gaps in the heterostructures formed by the Bi$_2$Se$_3$ films and CrI$_3$ ML. Our calculated non-trivial gap of CrI$_3$/5(6)QL-Bi$_2$Se$_3$/CrI$_3$ is about ~2 meV, which is consistent with previous report [19] and hence confirms the reliability of our first-principles calculations. A larger band gap of nearly ~30 meV can be generated by inserting CrI$_3$ ML between the first top (bottom) and sub-top (sub-bottom) QL of six QL Bi$_2$Se$_3$ film (1QL-Bi$_2$Se$_3$/CrI$_3$/4QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$). Further analysis indicates the decreased distance between the CrI$_3$ ML and the inner 4QL-Bi$_2$Se$_3$ film, induced by the effective pressure of outermost Bi$_2$Se$_3$ QL, enhances the magnetic proximity effect and improves the non-trivial gap. The topological non-triviality is confirmed by the non-zero Chern number and the existence of chiral edge state. Our work may further promote the experimental observation and practical applications of QAHE based on the Bi$_2$Se$_3$–CrI$_3$ interfaced system.

2. Computational details

The electronic band structures and topological properties of Bi$_2$Se$_3$–CrI$_3$ interface system were calculated in the framework of the PBE-type generalized gradient approximation using VASP package, which is same as our previous works [20–22]. The plane-wave cutoff was set to 400 eV and the spin–orbit coupling (SOC) was included in the self-consistent electronic structure calculations on a 7 × 7 × 1 Monkhorst–Pack $k$-point mesh. The experimentally determined lattice constant of Bi$_2$Se$_3$ film is 4.14 Å [23], while that of CrI$_3$ is 6.87 Å [18]. Therefore, we constructed the Bi$_2$Se$_3$–CrI$_3$ interface system by using the unit cell of $\sqrt{3} \times \sqrt{3}R30^\circ$ Bi$_2$Se$_3$ film. We found the energy of 1QL-Bi$_2$Se$_3$/CrI$_3$/4QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$ with lattice constant of 6.87 Å is lower than that with the lattice constant (7.17 Å) of $\sqrt{3} \times \sqrt{3}R30^\circ$ Bi$_2$Se$_3$ film [23] so that we employed the lattice constant of 6.87 Å in our calculations. The vacuum layer is over 15 Å along $z$ direction to avoid the interaction between neighboring films due to the periodic boundary condition used in VASP package. For structural relaxation, DFT-D2 van der Waals (vdW) correction proposed by Grimme [24] was employed and all the atoms were allowed to relax until the atomic forces are smaller than 0.01 eV Å$^{-1}$. Since the CrI$_3$ ML is an Ising ferromagnet with out-of-plane spin orientation, we set the initial spin polarized direction of Cr atoms along $z$ axis. We tried LDA + $U$ method with different $U$, but the opened band gap increases slightly within 2 meV. Therefore, we neglected the $U$ correction in our calculations. We used the Wannier90 package [25] to fit the electronic band structures of the Bi$_2$Se$_3$–CrI$_3$ interfaced system, which will provide a model Hamiltonian and allow us to discuss its topological property in detail. Pyprocar package [26] was employed to check the spin texture of the states around the gap opened on the 2D Dirac surface states.

3. Results and discussion

To test the influence of FM semiconductor CrI$_3$ ML on the Dirac surface states of $\sqrt{3} \times \sqrt{3}R30^\circ$ Bi$_2$Se$_3$ film with 6QLs, we firstly constructed two kinds of Bi$_2$Se$_3$–CrI$_3$ interfaced systems: (1) CrI$_3$ ML adsorbed on one surface of Bi$_2$Se$_3$ film (CrI$_3$/6QL-Bi$_2$Se$_3$); (2) CrI$_3$ ML adsorbed on both surfaces of Bi$_2$Se$_3$ film (CrI$_3$/6QL-Bi$_2$Se$_3$/CrI$_3$). The first interface system composites a supercell with 98 atoms, including 36 Bi, 54 Se, 6 I and 2 Cr atoms, as shown in figure S1(a) in the supplementary materials (SM) (http://stacks.iop.org/NJP/22/073005/mmedia). Several interfaced configurations are considered: the Cr atom is directly above the top Bi in the first-top QL of Bi$_2$Se$_3$ film with the side view and top view being presented in figures S1(b) and (e) in SM, respectively; the Cr atom is above the top Se atom and middle Se atom in first-top QL of Bi$_2$Se$_3$ film, as shown by figures S1 (c) and (d) for side view, respectively, which are with same top view in figure S1(f). The energy is favorite for CrI$_3$/6QL-Bi$_2$Se$_3$ with the Cr atom sitting on the top of the first nearest Se [figure S1(c)]. This is consistent with previous reports [19]. The interface vertical distance between I and Se atoms is ~2.913 Å. The bond length between I and Cr atoms is 2.705 Å. The calculated magnetic moment is 3.000 $\mu_B$ on each Cr atom.

The corresponding band structures for CrI$_3$/6QL-Bi$_2$Se$_3$ with vdW correction is shown in figure S2(a). An obvious character is that one of the two spin degenerated Dirac states originally resided on the two
surfaces of Bi$_2$Se$_3$ film is lifted and the band gap is opened in one spin component. This is natural since only one surface of Bi$_2$Se$_3$ film is adsorbed with CrI$_3$ ML and the opened gap results from the magnetic proximity effect. This can be further understood based on the model Hamiltonian calculation [19]:

$$H(k_x, k_y) = \begin{bmatrix} \Delta_t & ik_x + k_y & 0 & 0 \\ -ik_x + k_y & -\Delta_t & 0 & 0 \\ 0 & 0 & \Delta_b & -ik_y - k_x \\ 0 & 0 & ik_y - k_x & -\Delta_b \end{bmatrix}.$$ 

Here $\Delta_t$ and $\Delta_b$ is the exchange field on the top and bottom surface states. The calculated band structures are shown in figure S3. It is clearly that one surface state cannot be gaped for figure S3(b). The absence of global gap makes the topological property of CrI$_3$/6QL-Bi$_2$Se$_3$ interfaced system being not well-defined.

We illustrate the band of CrI$_3$ ML with magenta color to compare its contributions with that of Bi$_2$Se$_3$ in dark yellow around Fermi level [figure S2(a)], thus further examining the influence of the CrI$_3$ on the band. The valence bands near the Fermi level mainly come from the $p_z$ orbitals of the Bi and Se atoms, while the energy of the $p_x$ and $p_y$ orbitals of I atoms at the $\Gamma$ point is 0.3 eV lower than the Fermi level, which indicate the hybridization between CrI$_3$ and Bi$_2$Se$_3$ is far from Fermi level. Therefore, the Dirac state at $\Gamma$ point is influenced slightly by the FM CrI$_3$ ML but important and essential for the emergence of non-trivial phase transition as we discussed below. The electronic band structure [figure S2(b)] of CrI$_3$/6QL-Bi$_2$Se$_3$/CrI$_3$ shows band inversion near the Dirac point. This is confirmed by the plot of the spin polarized dark yellow around Fermi level [figure S2(a)], thus further examining the influence of the CrI$_3$ on the band.

At present, the QAHE is realized at a very low temperature, which is still far from the practical applications. At extremely low temperature, surface states in the films are suppressed except for edge states. However, when the temperature is not zero, the carriers in the surface state will be thermally excited, and even the electrons in the edge state may be excited into the surface state. When the contribution of the surface state to the conductivity is close to or beyond that of edge states, the energy consumption of the whole system cannot be ignored, and the signal of the QAHE will also be destroyed. In order to solve this problem, the key is to increase the non-trivial gap of 2D electronic systems. However, the non-trivial gap in 1ML-CrI$_3$/5(6)QL-Bi$_2$Se$_3$/1ML-CrI$_3$ system is too small, which implies that could not be easily observed by combining the Hall transport measurements and surface quantum control. We hope to tune a big non-trivial gap in the interfaced systems so that surface states and their peculiar properties may be accessed.

To enhance the non-trivial gap in Bi$_2$Se$_3$–CrI$_3$ interfaced systems, we here propose the third interfaced system with the FM CrI$_3$ ML being inserted between the top (bottom) QL and sub-top (sub-bottom) QL of Bi$_2$Se$_3$ films. The stable interfaced configuration was estimated by comparing the total energy of the simplified heterostructure with the CrI$_3$ ML sandwiched by Bi$_2$Se$_3$ QL (1QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$), as shown in figure S4. Six configurations were considered respectively. Our calculations indicate the figures S4(a) and (b) are the two most energy favorable configurations. Then we insert the CrI$_3$ ML between the top (bottom) QL and sub-top (sub-bottom) QL of 6 QLs Bi$_2$Se$_3$ films (1QL-Bi$_2$Se$_3$/CrI$_3$/4QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$) with the interface configurations of figures S4(a) and (b). Total energy calculations indicate the configuration of figure S4(a) is more favorable than that of figure S4(b) by 0.564 eV per cell. Consequently, we will focus on the topological property of 1QL-Bi$_2$Se$_3$/CrI$_3$/4QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$ with the interfaced configuration shown in figure 1(a) in the following discussions. The calculated magnetic moment is 2.978 $\mu_B$ on each Cr atom. The interfacial Se atoms obtain a growing magnetic moment of 0.008 $\mu_B$ due to the increase of interfacial interaction. The optimized interlayer distance $d_t$ ($d_d$) between the CrI$_3$ ML and the top (below) QL of Bi$_2$Se$_3$ film is $\sim$2.89 Å ($\sim$2.80 Å), which is smaller than that ($\sim$3.10 Å) of CrI$_3$/Bi$_2$Se$_3$-film/CrI$_3$ [19] and will enhance the magnetic proximity effect. The electronic band structure of 1QL-Bi$_2$Se$_3$/CrI$_3$/4QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$ with considering SOC is shown in figure 2(a) with enlarged bands inside. We can clearly see the spin degeneracies of the Dirac surface states are lifted, and a gap of $\sim$30 meV is opened up near the Fermi level at the $\Gamma$ point [inset of figure 2(a)]. To further confirm this, we employ the in-plane lattice constant (7.17 Å) of $\sqrt{5} \times \sqrt{3}B$ 30° Bi$_2$Se$_3$ film [23], and the calculated gap is $\sim$30 meV, which is consistent our present results calculated by using the in-plane lattice constant of 6.87 Å. The valence bands below the Fermi level mainly come from the $p_z$ orbitals of the Bi and Se atoms. This magnetic proximity induced gap is the key to the QAHE [6]. Note that Dirac point near the Fermi level could not be gaped (figure S5) if the orientation of electron spin-polarization of Cr atoms is set to be in-plane. We also calculated the electronic band structures of 1QL-Bi$_2$Se$_3$/CrI$_3$/3QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$ (figure 1(b)) and 1QL-Bi$_2$Se$_3$/CrI$_3$/2QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$ seen from figures 2(b) and (c)
Figure 1. Interfacial atomic structure of Bi$_2$Se$_3$ film inserted with CrI$_3$ ML. (a) Side view of 1QL-Bi$_2$Se$_3$/CrI$_3$/4QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$. (b) Side view of the Cr atom in ML CrI$_3$ directly above (below) the nearest Bi (Se) in below (top) QL of Bi$_2$Se$_3$ film, and $d_1$ ($d_2$) indicating the interface distance between the CrI$_3$ and top (below) QL of Bi$_2$Se$_3$. Magenta, green, blue and red balls represent Bi, Se, Cr and I atoms, respectively.

Figure 2. Band structures (a) for 1QL-Bi$_2$Se$_3$/CrI$_3$/4QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$ interface system; (b) for 1QL-Bi$_2$Se$_3$/CrI$_3$/3QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$ interface system; (c) for 1QL-Bi$_2$Se$_3$/CrI$_3$/2QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$ interface system. (d)–(f) Spin projections of corresponding bands of (a)–(c) within $k_x$–$k_y$ plane around $\Gamma$ (0, 0) point with energy $-0.03$ eV for spin up in blue color and 0.03 eV for spin down in red color, respectively. The gap opened at the surface states is about $\sim 2$ meV [inset of figure 2(b)] and $\sim 1$ meV [inset of figure 2(c)], respectively.

Since the band inversion is critical for the emergence of non-trivial state and realization of QAHE, we analyze the spin-texture of the states near the opened gap. Spin projections of the two valence (conduction) bands within $k_x$–$k_y$ plane around $\Gamma$ (0, 0) point are shown in figures 2(d)–(f) at $-0.03$ eV (0.03 eV) energy, respectively. Note that the up (down) spin of two valence and conduction bands in the inset of figures 2(a)–(c) are in blue (red) color. The minimum of two conduction bands are mainly contributed by down-spin component with red color, while the maximum of two valence bands stems from the up-spin with blue color, presented in figure 2(d). Such feature of band inversion is same as that of CrI$_3$/6QL-Bi$_2$Se$_3$/CrI$_3$ [19], and the gap of $\sim 30$ meV should be topologically non-trivial. Figure 2(c) also shows there is a net out-of-plane spin polarization around gapped Dirac point. Through analyzing the spin-texture of the states near the opened gap, we can identify the gap of 1QL-Bi$_2$Se$_3$/CrI$_3$/3QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$ is topological non-trivial [figures 2(b) and (e)]. In figure 2(f), two conduction bands with red color are in down spin, while two valence bands with red and blue color are in down and up spin respectively. This shows that the system is in a transition state from a normal insulator to Chern insulator, so that the gap of 1QL-Bi$_2$Se$_3$/CrI$_3$/2QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$ is trivial. This demonstrate the non-trivial
Figure 3. (a) Berry curvature within the SOC gap in reciprocal-space; (b) Hall conductance as a function of the Fermi level within magnetic proximity effect induced gap; (c) 1D band structure of the nanoribbon of 1QL-Bi$_2$Se$_3$/CrI$_3$/4QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$ interface system.

Phase will emerge start from 5QL-Bi$_2$Se$_3$ film inserted by CrI$_3$ ML, same to CrI$_3$ adsorbed Bi$_2$Se$_3$ film.

To demonstrate the topological non-triviality of the gap opened on the 2D surface state of 1QL-Bi$_2$Se$_3$/CrI$_3$/4QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$ interface system, we calculated the Berry curvature and Chern number based on the Hamiltonian obtained by fitting the electronic band structures of 1QL-Bi$_2$Se$_3$/CrI$_3$/4QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$ using WANNIER90 package. The Berry curvature $\Omega(k)$ is defined by the following equation [27–30]

$$\Omega(k) = \sum_n f_n \Omega_n(k)$$

$$\Omega_n(k) = -\sum_{n' \neq n} 2 \text{Im} \left( \frac{\langle \psi_{nk} | \nu_x | \psi_{nk} \rangle \langle \psi_{nk} | \nu_y | \psi_{nk} \rangle}{\varepsilon_{nk} - \varepsilon_{nk'}} \right)$$

(2)

here $f_n$ is the Fermi distribution, $\psi_{nk}$ is the eigenstate of eigenvalue $\varepsilon_{nk}$ of band $n$, $\nu_x/\nu_y$ is the velocity operator. The distribution of total Berry curvature for the whole valence bands in the BZ is shown in figure 3(a). We can clearly see the Berry curvature mainly locating at the $\Gamma$ point, where the gap of $\sim30$ meV is opened. By calculating the Berry curvature at different energy and integrating it over the first Brillouin zone (BZ), the Chern number $C$ is defined by the following equation

$$C = \frac{1}{2\pi} \int_{BZ} d^2k \Omega(k).$$

(3)

We get the anomalous Hall conductance around the Fermi level [figure 3(b)].

$$\sigma_{xy} = \frac{e^2}{h} C.$$  

(4)

One can identify a quantized Hall plateau at a value of $e^2/h$ within the energy window of SOC gaps, indicating the Chern number of 1. Due to the bulk-boundary correspondence, 1D topologically protected edge states will exist in the band structure of the 1QL-Bi$_2$Se$_3$/CrI$_3$/4QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$ ribbon [figure 3(c)]. Obviously, the edge states connect the valence and conduction bulk bands. The existence of edge states is a manifestation of the non-trivial phase, which is the key for realization of QAHE.

Subsequently, we further explain how spin exchange splitting introduced by FM CrI$_3$ ML can lead to the formation of quantum anomalous Hall state from the 2D Dirac surface states of Bi$_2$Se$_3$ film. In the crystal field of 1QL-Bi$_2$Se$_3$/CrI$_3$/4QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$ without magnetic coupling and SOC, $d_{z^2}$ and $d_{xy}$ orbital of Cr in valence and conduction band are the closest to Fermi level seen from figure S6(a). Chemical bonding pushed $p_z$ states of I, Bi and Se are far away from Fermi level. Considered magnetic coupling, the inserted CrI$_3$ ML will introduce an effective spin exchange field, which will destroy the time inversion symmetry in the original system. Due to the magnetic polarization, $d_{z^2}$ and $d_{xy}$ orbital of Cr split into two branches away from Fermi level [figures S6(b) and (c)]. The $p_z$ orbital of Bi and Se in conduction band (valence band) split in a way that down (up) spin branch moves toward the Fermi level. With the magnetic exchange interactions, the $p_z$ band with down spin ($|p_z, \downarrow\rangle$) in valence band and $p_z$ band with up spin ($|p_z, \uparrow\rangle$) in conduction band are further closest to the Fermi level. The combination of magnetic exchange interaction and SOC leads to the band inversion of a pair of spin subbands [31] due to the exchange splitting can overcome the coupling-induced gap [figure S6(d)]. Figures 2(a) and (b) confirms the band
Figure 4. (a) Band gap versus the increase of interfacial distance $d_1$ between the CrI$_3$ and outermost 1QL-Bi$_2$Se$_3$ of 1QL-Bi$_2$Se$_3$/CrI$_3$/4QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$ interfaced system. (b) Band gap versus the increase of interfacial distance $d_2$ between the CrI$_3$ and inner 4QL-Bi$_2$Se$_3$ of 1QL-Bi$_2$Se$_3$/CrI$_3$/4QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$ interface system. Here the $d_0^1$ ($d_0^2$) is the optimized interlayer distance between the CrI$_3$ ML and the outermost (inner) 1QL-Bi$_2$Se$_3$.

inversion of a pair of spin sub-bands around Fermi level. This eventually leads to the emergence of non-trivial phase.

Another factor that determining the realization of QAHE is the temperature, where the out-of-plane FM order of CrI$_3$ ML should be formed first [32]. The ordering temperature $T_c$ in this geometry could be estimated based on the spin wave theory (SWT) with the approximate expression of

$$k_B T_c = \frac{\pi}{2} \log(1 + 2\pi J_1 \Delta_0)$$

where $\Delta_0$ can be roughly taken as magnetic anisotropy energy $\Delta E$ and $J_1$ is intralayer first neighbor exchange coupling parameter. The magnetic anisotropy energy $\Delta E$ is 0.55 meV/Cr in our calculation. $J_1$ is set to the value of 6.91 meV [33]. The evaluated $T_c$ using the calculated $\Delta E$ and $J_1$ is 66.11 K, which is agreement well with the corresponding experimental values (61 K) for bulk samples [18]. Since the non-trivial gap of $\sim 30$ meV corresponds to a temperature higher than 66.11 K, the operating temperature of QAHE is restricted by the $T_c$ of CrI$_3$ ML. It should note that the ordering temperature can be further improved by employing an external magnetic field.

Finally, we move to discuss the reasons for the enlarged non-trivial gap in 1QL-Bi$_2$Se$_3$/CrI$_3$/4QL-Bi$_2$Se$_3$/CrI$_3$/1QL-Bi$_2$Se$_3$ interface system with respect to that of CrI$_3$/6QL-Bi$_2$Se$_3$/CrI$_3$. We note the distance $d_1^0$ ($d_2^0$) between the CrI$_3$ ML and the outer 1QL-Bi$_2$Se$_3$ (inner 4QL-Bi$_2$Se$_3$) is optimized to be 2.89 Å (2.80 Å), which is smaller than that (2.90 Å) of CrI$_3$/Bi$_2$Se$_3$-film/CrI$_3$ [19]. Consequently, we analyze the variation of band topology in response to the increase of the distance. The magnitude of the gap changes slightly when increasing $d_1$ and the non-trivial topology remains robust against such variation [figure 4(a)], as confirmed by the existing band inversion [figures S7(a) and (b)]. Differently, with the increase of $d_2$, the band gap decreases first (up to 0.6 Å) and then increases monotonically (figure 4(b)). This means the gap is closed and then opened up during this process, indicating the occurrence of phase transition. Band inversion demonstrated the gap is topological non-trivial for $d_2 < d_2^0 + 0.5$ Å, while trivial phase will be evoked when $d_2$ is larger than $d_2^0 + 0.5$ Å [figures S7(c) and (d)]. This means the distance $d_2$ between the CrI$_3$ ML and the inner 4QL-Bi$_2$Se$_3$ is critical for the realization of QAHE.

We also make another test calculation based on CrI$_3$/4QL-Bi$_2$Se$_3$/CrI$_3$ interface system, which is topological trivial reported by previous work [19]. However, decreasing the interfacial distance between CrI$_3$ and Bi$_2$Se$_3$ film by 0.2 Å will lead to the emergence of band inversion and hence evoke the non-trivial phase (figure S8). For our proposed Bi$_2$Se$_3$–CrI$_3$ interfaced systems, the outermost Bi$_2$Se$_3$ QL provide an effective intrinsic pressure along the out-of-plane direction and hence decrease the distance between the CrI$_3$ ML and the inner Bi$_2$Se$_3$ film. The decreased distance will increase the magnetic proximity effect between the FM CrI$_3$ semiconductor and the Dirac surface state of Bi$_2$Se$_3$ film, which enhance the non-trivial gap of the inserted heterostructures.

4. Conclusions

In summary, we systematically investigated the possible realization of QAHE based on the topological insulator-ferromagnetic (Bi$_2$Se$_3$–CrI$_3$) interfaced system at relatively high operating temperatures by first-principles theory. Our calculations demonstrate the topological non-triviality of the interfaced system.
based on surface band inversion, nonzero Chern number and chiral edge state. Our result confirms that the inserted CrI3 magnetic interface resided in the range of 1 nm underneath top surface of Bi2Se3 films affects strongly the surface Dirac state and induces a larger bandgap of system. The induced QAHE hold high possibility to be detected in experiments through measurements of the Hall conductivity. Our work may promote the experimental observation and practical applications of QAHE in Bi2Se3–CrI3 interface system.

Conflicts of interest
The authors declare no competing financial interests.

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