Quantum anomalous Hall effect in two-dimensional magnetic insulator heterojunctions

Jinbo Pan1,6, Jiabin Yu2,3,6, Yan-Fang Zhang1,4,5, Shixuan Du4, Anderson Janotti5, Chao-Xing Liu1✉ and Qimin Yan1✉

Recent years have witnessed tremendous success in the discovery of topological states of matter. Particularly, sophisticated theoretical methods in time-reversal-invariant topological phases have been developed, leading to the comprehensive search of crystal database and the prediction of thousands of topological materials. In contrast, the discovery of magnetic topological phases that break time reversal is still limited to several exemplary materials because the coexistence of magnetism and topological electronic band structure is rare in a single compound. To overcome this challenge, we propose an alternative approach to realize the quantum anomalous Hall (QAH) effect, a typical example of magnetic topological phase, via engineering two-dimensional (2D) magnetic van der Waals heterojunctions. Instead of a single magnetic topological material, we search for the combinations of two 2D (typically trivial) magnetic insulator compounds with specific band alignment so that they can together form a type-III broken-gap heterojunction with topologically non-trivial band structure. By combining the data-driven materials search, first-principles calculations, and the symmetry-based analytical models, we identify eight type-III broken-gap heterojunctions consisting of 2D ferromagnetic insulators in the MXY compound family as a set of candidates for the QAH effect. In particular, we directly calculate the topological invariant (Chern number) and chiral edge states in the MnNF/MnNCl heterojunction with ferromagnetic stacking. This work illustrates how data-driven material science can be combined with symmetry-based physical principles to guide the search for heterojunction-based quantum materials hosting the QAH effect and other exotic quantum states in general.

npj Computational Materials (2020) 6:152; https://doi.org/10.1038/s41524-020-00419-y

INTRODUCTION

The quantum anomalous Hall (QAH) effect1–3, a zero magnetic field manifestation of the integer quantum Hall effect, originates from the exchange interaction between electron spin and magnetism and exhibits quantized Hall resistance and zero longitudinal resistance. Similar to the quantum Hall effect, the QAH effect harbors dissipationless chiral edge states, thus providing an energy-efficient platform for state-of-the-art applications in spintronics4,5 and quantum computing6,7. Guided by theoretical predictions8, the QAH effect was experimentally demonstrated in the magnetically (Cr or V) doped (Bi, Sb)2Te3. However, magnetic doping reduces the spin-orbit coupling (SOC) strength and thus may drive the system into a trivial phase. It also degrades the sample quality by bringing a large amount of disorder scatterings into the system. Consequently, the critical temperature of the QAH state through the magnetic doping approach is usually below 2 K, an order-of-magnitude lower than the Curie temperature of these compounds. Therefore, it is highly desirable to search for new platforms consisting of stoichiometric materials with intrinsic magnetism to realize high temperature QAH state.

Recent rapid development of two-dimensional (2D) layered magnetic insulators has provided an exciting opportunity for the realization of QAH effect in un-doped material systems. The QAH effect has been proposed to appear in the heterostructures of topological insulator (TI) and magnetic insulator through the magnetic proximity effect across the interface18–21. Another recent advance is the successful synthesis of a class of intrinsic magnetic TI materials, including MnBi2Te4, MnBi2Se4, and MnSb2Te418–20, in which A-type antiferromagnetism and topologically nontrivial band structure coexist, leading to the successful observation of the QAH state20–22. The QAH effect has also been observed in twisted bilayer graphene23,24.

RESULTS

Discovery workflow for QAH heterojunctions

Despite several recent works25–26, the search for more QAH materials is still challenging because it requires the coexistence of ferromagnetism and topological band structure, which is rare in a single material. In this study, we propose an alternative strategy to realize the QAH effect in 2D magnetic van der Waals (VdW) heterojunctions by combining data-driven discovery of 2D magnetic compounds with a theoretical analysis of topological properties. All the previous works18–20,25,26 on the QAH heterojunctions require at least one intrinsic topological material. In contrast, every QAH VdW heterojunction predicted by our approach can be constructed with two trivial 2D ferromagnetic (FM) insulators. There are three types of heterojunctions, defined by the band alignment between the two constituting materials, as shown in Supplementary Fig. 1. The key idea is to make sure the two FM 2D materials form the so-called type-III broken-gap junction, in which the conduction-band minimum (CBM) of one 2D material is lower in energy than the valence-band maximum (VBM) of the other compound at certain high-symmetry momentum, as shown in Fig. 1a. In other words, an inverted (topological) band structure is formed by the heterojunction, rather than one individual material. As this strategy only requires the (valence or conduction) band offset to be larger than the band gap of one of
the two 2D materials, it greatly relaxes the stringent conditions for the QAH effect and allows us to develop a systematic approach to identify the candidate compounds for the QAH heterojunctions in 2D magnetic VdW material database.

This approach has been successfully utilized to search for the quantum spin Hall insulators, such as InAs/GaSb quantum wells, and other semiconductor heterojunctions, but it has not been applied to the search of QAH systems, mainly due to the lack of 2D magnetic materials database. Therefore, a data-driven computational and theoretical search of 2D magnetic materials is called for. Figure 1b summarizes the workflow for our data-driven computational approach, which is carefully designed to incorporate high-throughput calculations at multiple computational levels and symmetry-based physical principles that are critical to quickly identify the most promising candidates hosting the QAH effect. In this initial work, we focus on a family of 2D monolayer MXY compounds (M = metal atoms, X = N, S, Se, Te, Y = F, Cl, Br, I) and predict 46 potential 2D magnetic semiconductors in this family. A database including the information of CBM, VBM, band offset, the orbital nature and the symmetry properties of the conduction and valence bands, and the magnetic anisotropy energies and exchange coupling for each candidate compound is created. Based on this 2D magnetic material database, we identify eight broken-gap VdW heterojunctions made from different combinations of these compounds to possess inverted band structure.

We next perform a generic symmetry analysis of topological properties for the VdW heterojunctions. This analysis is based on the relationship between topological invariant, the Chern number (CN) that characterizes the QAH effect, and the two-fold rotation (C₂) eigenvalues. We demonstrate that the CN for the heterojunction must be changed by an odd number when the magnetizations in two magnetic VdW compounds are switched from FM to anti-FM (AFM) stacking order. This suggests that the QAH phase must exist for at least one magnetic stacking order.

Finally, we choose MnNF/MnNCl heterojunctions as an example and construct a tight-binding model that captures the main features of the band structure from the realistic material simulations. Based on this tight-binding model, we explicitly compute the CN and the chiral edge states of this heterojunction. Our results show that the QAH phase occurs for the FM stacking order of the magnetization between two materials of the heterojunction and is consistent with our symmetry analysis by computing the C₂ eigenvalues from first-principles calculations.

Prediction of 2D magnetic insulators in MXY compounds

In the past decade, a tremendous amount of computed and experimental material data has been shared in the materials research community through multiple material databases for both bulk and 2D inorganic materials. This offered a vast inorganic materials space that is explorable by data-driven approaches and enabled the data-driven discovery of quantum materials hosting exotic topological phases. Here, our theoretical predictions of 2D heterojunctions that host the QAH effect is motivated by the data-driven discovery of a family of 2D monolayer MXY compounds (M = metal atoms, X = N, S, Se, Te, Y = F, Cl, Br, I) that are predicted to host both FM and nonmetallic ground states. We initiated our material discovery efforts from a material set of around 560 monolayer MXY compounds that are created through elemental substitution based on the existing MXY materials in public 2D materials databases. The data-driven materials discovery process is illustrated in Fig. 1b and more computational details can be found in Supplementary Fig. 2. With FM state as the initial magnetic configuration, through an energy minimization and electron density optimization process, we identified 46 potential magnetic semiconductors (see Supplementary Table 1). 2D FM insulators MnNX and CrCX (X = Cl, Br, I; C = S, Se, Te) have been predicted in previous work. Our comprehensive study puts this 2D compound family as a great platform for the discovery and design of quantum materials based on 2D magnetic heterojunctions.

Constructions of ten different FM MXY compounds, we propose eight VdW heterojunctions (as listed in Table 1), in which the CBMs of one 2D layer (called conduction layer) is below the VBM of the other 2D layer (called valence layer). Thus, these heterojunctions exhibit the desirable band inversion, as well as small enough (<6.5%) lattice mismatch, as shown in Table 1. The Curie temperatures (Tc) of the eight 10 monolayer FM MXY compounds are evaluated by performing Monte-Carlo (MC) simulations based on a 2D Heisenberg Hamiltonian. The interaction types, the values of exchange coupling J and magnetic anisotropy energy A are summarized in Fig. 1a and Table 2. Computational details related to magnetic anisotropy energies and exchange coupling interactions are included in the Supplementary Methods and Supplementary Fig. 3. Characterized by high Curie temperature as well as small band gaps, this set of magnetic semiconductor compounds provides a great platform for the design of VdW heterojunctions to realize the QAH effect. The CBM of all the conduction layers and the VBM of all the valence layers are both located at Γ point, facilitating the analysis of broken-gap junctions.

Symmetry analysis for the VdW heterojunctions

We next analyze the topological properties of the proposed eight VdW heterojunctions based on the symmetry aspect. The magnetic moments are in the z direction (normal to the 2D planes) for all these magnetic MXY compounds, which reduce
Using quantum numbers (see Fig. 2a, b), and then we can split equalizes the chemical potentials of the two layers. Such VdW heterojunctions but include the charge transfer that neglect the SOC and the interlayer coupling between two layers in principle calculations (see details in the Supplementary Fig. 8), the switching from the FM stacking to the AFM stacking, as the orbital nature is unchanged while the spin direction is reversed. It results in \( n_{\text{AFM}}^{\text{high}} = 4N_{\text{high}} - n_{\text{FM}}^{\text{low}} \), where the pre-factor 4 comes from the four \( C_2 \)-invariant momenta and \( N_{\text{high}} \) is the number of occupied bands fully below \( E_F \) in the conduction layer. Since only one band from the conduction layer is partially filled and it only contributes to \( n_{-c} \) at \( \Gamma \) for the heterojunctions of interest, \( n_{-c} \) is either 0 or 1, leading to \( n_{\text{AFM}}^{\text{low}} = 1 - n_{-c} \cdot n_{\text{FM}}^{\text{low}} \). In sum, we have \( n_{\text{AFM}} = n_{\text{FM}} + 1 + 4N_{\text{high}} - 2n_{\text{FM}}^{\text{low}} \), resulting in

\[
\pi_{\text{AFM}}^{\text{mod2}} = n_{\text{FM}} + 1 + \text{mod}2.
\]

Now we discuss the SOC and the interlayer coupling. The energy scales of these two effects are typically much smaller than the energy scale of the band inversion (i.e., the energy gap at \( \Gamma \) in this case), which is determined by the hopping and the magnetic exchange coupling. Indeed, as shown by the results from first-principle calculations (see details in the Supplementary Fig. 8), the SOC and interlayer coupling do not cancel the existing band inversion or introduce new band inversion in the heterostructures of interest, leaving Eq. (2) unchanged. However, the two effects are still important for our analysis since they open the gap at generic momenta (not \( C_2 \)-invariant momenta) and enable us to combine Eq. (2) with Eq. (1). (See schematic examples in Fig. 2c, d). As a result, we conclude that the parity of the CN is changed when switching the stacking order of the 2D heterojunction (from FM to AFM or the inverse), and thus at least one of the stacking orders can give non-zero CN. The remaining question is which stacking order guarantees a nonzero CN for the heterojunctions of interest. Let us assume that the CNs of the two individual materials have the same parity, e.g., both materials have zero CNs, so that the total \( n_{-c} \) of the two materials must be even before forming the heterojunction. Then, the key lies on the \( C_2 \) eigenvalues of the two states involved in the band inversion at \( \Gamma \), i.e., CBM of the conduction layer and VBM of the valence layer in this case. Specifically, if the two states have opposite \( C_2 \) eigenvalues for a certain stacking order, \( n_{-c} \) of the

\[
\text{CN mod} 2 = n_{-c} \mod 2
\]

Table 1. Potential van der Waals heterojunctions based on ferromagnetic MXY compounds with electronic band inversions and lattice mismatches <6.5%.

| Material A | CBM (eV) | Band character (CBM) | Material B | VBM (eV) | Band character (VBM) | Lattice mismatch along a (%) | Lattice mismatch along b (%) | Nontrivial stacking order |
|------------|----------|---------------------|------------|----------|---------------------|-------------------------------|-------------------------------|--------------------------|
| FeNF       | −7.9     | Fe: \( d_{x^2-y^2} \) | MnNF       | −7.6     | N: \( p_z \)       | 2.3                          | 1.1                          | FM                       |
| VSI        | −5.9     | V: \( d_{x^2} \)    | MoSI       | −5.6     | Mo: \( d_{yz} \)   | 1.7                          | 3.2                          | FM                       |
| VSI        | −5.9     | V: \( d_{y^2} \)    | WSe        | −4.8     | W: \( d_{z^2} \)   | 3.9                          | 6.1                          | FM                       |
| MnNF       | −7.1     | Mn: \( d_{x^2-y^2} \)| MnCl       | −6.7     | N: \( p_z \)       | 6.5                          | 0.6                          | FM                       |
| TiTe       | −4.9     | Ti: \( d_{x^2} \)   | WTe        | −4.9     | W: \( d_{z^2} \)   | 4.0                          | 3.0                          | FM                       |
| VSe        | −5.1     | V: \( d_z^2 / d_{x^2-y^2} \)| WSe        | −4.8     | W: \( d_{z^2} \)   | 6.5                          | 0.1                          | FM                       |
| CrSeCl     | −5.6     | Cr: \( d_{x^2-y^2} \)| VSe        | −5.6     | Se: \( p_x / p_y \) | 4.1                          | 0.5                          | FM                       |

*Nontrivial stacking order* indicates the stacking order that guarantees a nonzero CN under the assumption that each individual material is topologically trivial. Valence-band maximum (VBM) and conduction-band minimum (CBM) are given with respect to vacuum level.

Table 2. Magnetic interaction parameters and Curie temperatures of MXY compounds.

| \( J_{12} \) | \( J_{13} \) | \( J_{34} \) | \( A_{100} \) | \( A_{010} \) | \( A_{001} \) | \( T_c \) (K) |
|-------------|-------------|-------------|-------------|-------------|-------------|-------------|
| TiTe        | 9.33        | −13.19      | −1.22       | 0.001 *     | 0.001 *     | 0           | 30          |
| FeNF        | 0.09        | −19.85      | −8.90       | −0.02 *     | 0.38 *      | 0           | 459         |
| VSI         | −93.41      | −73.03      | −43.59      | 1.85        | 1.27        | 0           | 1100        |
| VSe         | −7.03       | −34.49      | −24.83      | 2.89        | 1.57        | 0           | 913         |
| MnNF        | −16.39      | −8.73       | −2.46       | 0.06        | 0.07        | 0           | 671         |
| MnCl        | −16.13      | −5.67       | −2.91       | 0.06        | 0.11        | 0           | 592         |
| CrSeCl      | −3.79       | −6.13       | 1.69        | −0.06       | 0.002       | 0           | 270         |
| MoSi        | 1.07        | −3.08       | 1.07        | −0.08       | −0.45       | 0           | 66          |
| WSe         | 4.03        | −7.68       | −6.38       | 1.32        | −0.41       | 0           | 318         |
| WTe         | 2.12        | −11.86      | −6.26       | −0.10       | −0.91       | 0           | 517         |

Exchange coupling parameter \( J \) (in meV), magnetic anisotropy energy \( A \) (in meV), and estimated Curie temperature \( T_c \) of MXY compounds based on hybrid function (HSE06) calculations.

*Magnetic anisotropy energies for TiTe and FeNF were evaluated using the PBE + U method, as the HSE06 calculations did not converge well for these two compounds.*
heterojunction must differ by 1 from the total $n$ of the two materials before forming the heterojunction (see Fig. 2a). Thus, such stacking order must yield a QAH heterojunction according to Eq. (1). With this principle in mind, we list the $C_2$ eigenvalues of the VBM and CBM for all FM materials with all magnetic moments fixed in the positive $z$ direction (see Supplementary Table 2). From this list, we can directly read out the $C_2$ eigenvalues of the two states involved in the band inversion at the $\Gamma$ point for any heterojunction with FM stacking, and if they are opposite (same), the heterojunction has non-zero CN for the FM (AFM) stacking.

The QAH effect in MnNF/MnNCl heterojunction
The discussion above based on symmetry eigenvalues is valid for all the eight VdW heterojunctions identified if the magnetic moments are aligned along the $z$ direction, while the topological properties of heterojunctions with in-plane magnetization is discussed in the Supplementary Discussion 1 and Supplementary Fig. 4. From this list, we can directly read out the $C_2$ eigenvalues of the two states involved in the band inversion at the $\Gamma$ point for any heterojunction with FM stacking, and if they are opposite (same), the heterojunction has non-zero CN for the FM (AFM) stacking.

small band gaps. Both compounds are predicted to host FM ground state, being dynamically and thermally stable (Supplementary Figs. 5–7). The Curie temperatures of these two compounds are evaluated by both HSE06 and PBE + U functionals. We observe that the hybrid function predicts lower transition temperatures than PBE + U for MnNF and MnNCl, owing to the fact that the inclusion of exact exchange in HSE06 functional generates more localized states on Mn atoms and weaker exchange interactions among them. Even in this case, the predicted Curie temperatures for MnNF and MnNCl are 671 K and 592 K, respectively (Fig. 3a), well above room temperature. Both partial density of states and orbital-projected band structures of MnNF and MnNCl (Fig. 3b) indicate that the conduction-band edges of these compounds are mainly $d$ states, while the valence-band edge is derived from $N \, p_y$ states.

Based on the calculated band alignment between the two compounds (as shown in Fig. 3b, c), we expect that a type-III broken-gap heterojunction can be formed with band inversion. As shown in Fig. 3d, we create MnNF/MnNCl VdW heterojunctions with both FM and AFM stacking orders. Atomic structures are relaxed using the hybrid functional (HSE06). The interlayer VdW interaction is evaluated by using the DFT-D3 correction method of Grimme et al. In the case of FM stacking, without SOC, the crossing between two bands in the same spin channel happens around $\Gamma$ point, leading to the band inversion of 14.0 meV (Supplementary Fig. 8b), and this crossing is protected by mirror
of the heterojunction is initialized, the magnetic stacking order magnetic couplings indicate that, once a FM (or AFM) stacking order. This combination of strong intralayer and weak interlayer magnetic interaction is very weak. The total energy of the MnNF/MnNCl heterojunction with FM stacking order is only 0.1 meV per primitive cell lower than that with AFM stacking order. The interlayer magnetic interaction is thus providing effective ways to manipulate both electronic and magnetic properties of VdW heterojunctions. As a result, one can control the electronic band inversion at external electric field along z direction (see Supplementary Fig. 9), thus providing effective ways to manipulate both electronic structures and topological properties of VdW heterojunctions. As compared to the strong intralayer magnetic interaction, the interlayer magnetic interaction is very weak. The total energy of the MnNF/MnNCl heterojunction with FM stacking order is only 0.1 meV per primitive cell lower than that with AFM stacking order. This combination of strong intralayer and weak interlayer magnetic couplings indicate that, once a FM (or AFM) stacking of the heterojunction is initialized, the magnetic stacking order will be rather stable, because the flip of one spin would be restrained by the strong intralayer magnetic interaction.

In MnNF and MnNCl, within the p2nm plane group, Mn cations are confined in a quasi-octahedral bonding environment associated with a $t_{2g}$-$e_g$ d state splitting. This crystal field splitting (1~2 eV) provides an essential electronic structure scaffold for small-gap FM semiconductors. Orbital analysis confirms that the CBM of MnNF or MnNCl is composed of Mn $e_g$ states, whereas the VBM is derived from the interaction between occupied N $p$ states and Mn $t_{2g}$ states. This unique electronic structure enforces the VBM and CBM of MX$_Y$ compounds different $C_2$ rotational eigenvalues (see Supplementary Tables 1 and 2), which is essential for the emergence of QAH effect in their heterojunctions. From our first-principles calculations, the CBM and VBM at the Γ point (zone center) of MnNF and MnNCl have opposite $C_2$ eigenvalues in the FM configuration, indicating that the heterojunction with FM stacking must have non-zero CN according to the above symmetry analysis. To better understand the result of first-principles calculation, we construct effective models around Γ point near the Fermi energy. Near $E_F$, the conduction and valence bands of both materials are composed of $p_y$ and $d_{x^2−y^2}$ orbitals, respectively (as shown in Fig. 3b, c; $x$, $y$, $z$ directions are defined in Fig. 3d). For the FM stacking, the basis can be chosen as $\langle d_{x^2−y^2}, | (|p_y, k\rangle + |p_y, k\rangle) \rangle$, resulting in the effective model:

$$
\begin{align}
E_F + B_{vx} k_x^2 + B_{vy} k_y^2 + \gamma d_{x^2−y^2} k_x k_y + \lambda_1 k_y^2,
\end{align}
$$

where $\lambda_1$ is given by SOC, all other parameters are dominated by the hopping. More details are provided in the Supplementary Methods 2. According to the band structure from first-principles calculations, we may choose $E_F − E_{C1} > 0$, $B_{vx} − B_{vy} < 0$, and $\gamma d_{x^2−y^2} < 0$. Then, the direct calculation of the CN yields $CN^{FM} = \text{sgn}(\lambda_1)$, which is non-zero. On the other hand, the effective model for AFM stacking has the form:

$$
\begin{align}
\begin{cases}
E_F + B_{vx} k_x^2 + B_{vy} k_y^2 − \lambda_0 k_x^2, \\
\lambda_0 E_F + B_{vx} k_x^2 + B_{vy} k_y^2
\end{cases}
\end{align}
$$

with bases $\langle d_{x^2−y^2}, | (|p_y, k\rangle + |p_y, k\rangle) \rangle$ and $\lambda_0$ induced by SOC. The $k$-independent off-diagonal term leads to CN equals zero. As the occupied bands fully below $E_F$ have the same CNs for different stacking orders, the total CNs should differ by one, confirming the above conclusion.

We then construct a tight-binding model for further clarification of the topological properties of the heterostructure. The basis of the model is given by the $d_{x^2−y^2}$, $d_{xy}$ and $d_{xz}$ orbitals of the Mn atoms and the $p_y$ orbital of the N atoms. The FM of each layer is included by adding the localized magnetic moments on the Mn atoms and the on-site SOC is also included. With reasonable parameter values, the bulk band structures of the FM and AFM stacked heterostructures are shown in Fig. 4a, b, respectively, which qualitatively match those obtained by the first-principle calculations (see the Supplementary Methods 2 and Supplementary Fig. 10 for more details). Direct calculations for the FM and AFM stacked heterostructure yield CN = 1 and CN = 0, respectively, coinciding with the chiral edge mode for FM stacking in

Fig. 3 Computational data for a representative material system: MnNF, MnNCl, and their VdW heterojunctions. a Estimation of Curie temperatures for MnNF and MnNCl by Monte-Carlo simulations based on a Heisenberg Hamiltonian model that is fitted to both HSE06 and PBE+U results. b, c Orbital-projected band structures of MnNF and MnNCl; d The relaxed structure of MnNF/MnNCl heterojunction. Band structures of MnNF/MnNCl heterojunction (with SOC) with FM stacking order e without and f with a shift of the MnNF layer relative to the MnNCl layer by 0.5 Å along the y direction are shown.
Fig. 4c and the fully gapped edge for AFM stacking in Fig. 4d. The tight-binding model calculation verifies different topological properties of the FM and AFM stacked heterostructures.

DISCUSSION

In summary, we proposed a new approach to realize the QAH effect in 2D magnetic VdW heterojunctions and conclusively demonstrate the existence of the QAH effect in MnNF/MnNCl heterojunction with FM stacking by combining the first-principles calculations and the symmetry analysis. We also proposed other VdW heterojunctions that can host the QAH effect, as listed in Table 1, and the FM stacking always guarantees non-zero CN for them if each individual material is topologically trivial. It is important to note that Mn, Cr, Mo, and W atoms in these compounds that form VdW heterojunctions are all in a d^3 electronic configuration with t^2g states occupied and e^g states empty. Given the high Curie temperatures (above room temperature) of many compounds in the MXY family (see Table 2), these VdW heterojunctions provide an ideal platform to explore the possibility of room-temperature QAH state. The electronic structures and topological properties of VdW heterojunctions can be effectively controlled by strain and electric field (see Supplementary Fig. 9). Moreover, the design principles proposed in this work is not limited to this family of materials and can be extended/applied to a large variety of 2D magnetic systems in multiple plane groups protected by various symmetry operations, which will greatly expand the family of heterojunction-based QAH systems in the future. Therefore, our work will stimulate experimental synthesis and measurement efforts in this class of heterojunction systems and accelerate the discovery and design of new QAH materials, as well as quantum systems for other magnetic topological phases.

Note added
During the final stage of this work, a related work appeared on arXiv, which performed a systematic search for magnetic topological materials based on magnetic topological quantum chemistry approach52.

METHODS

First-principles calculations based on density functional theory

In our calculations, geometric relaxations and total energy calculations were performed using density functional theory within projector-augmented wave (PAW) potentials53,54 as implemented in the VASP code55. A vacuum slab of 20 Å and a plane-wave basis set with an energy cutoff of 520 eV were used. A 10 × 8 × 1 Γ-point centered k-point was applied to sample the Brillouin zone. For the initial steps of discovery process, GGA + U was employed to optimize the geometric structures56. The U-values are 3.25, 3.7, 3.9, 5.3, 3.32, 6.2, 4.38, and 6.2 eV for V, Cr, Mn, Fe, Co, Ni, Mo, and W, respectively, which are outlined in the work by Wang et al.57. The structures were fully relaxed until energy and force were converged to 10^-6 eV and 0.01 eV/Å, respectively. The screened hybrid functional of Heyd, Scuseria, and Ernzerhof (HSE06)58,59 was used to compute the band structures of MXY monolayers. For the 10 promising FM MXY scaffolding compounds, HSE06 calculations were performed to optimize the geometric structures, and compute the magnetic interactions. To verify the dynamical stability of monolayer MnNF and MnNCl, phonon dispersion analysis was performed by using the density functional perturbation theory method as implemented in the Phonopy code60, interfaced with VASP. In phonon and formation energy calculations, GGA + U along with the PAW potentials were employed, with a convergence criterion of 10^-8 eV for energy. For the ab initio molecular dynamics (MD) simulations61,62, a canonical (NVT) ensemble was used. Time step for MD simulations is 1 fs.

DATA AVAILABILITY

The authors declare that the main data supporting the findings of this study are available within the paper and its Supplementary files. Other relevant data are available from the corresponding author upon reasonable request.

Received: 27 May 2020; Accepted: 4 September 2020; Published online: 13 October 2020

REFERENCES

1. Liu, C.-X., Zhang, S.-C. & Qi, X.-L. The quantum anomalous Hall effect: theory and experiment. Annu. Rev. Condens. Matter Phys. 7, 301–321 (2016).
2. Wang, J., Lian, B. & Zhang, S.-C. Quantum anomalous Hall effect in magnetic topological insulators. Phys. Scr. T164, 014003 (2015).
3. Chang, C.-Z. & Li, M. Quantum anomalous Hall effect in time-reversal-symmetry breaking topological insulators. J. Phys. Condens. Matter 28, 123002 (2016).
4. Wu, J., Liu, J. & Liu, X.-J. Topological spin texture in a quantum anomalous Hall insulator. Phys. Rev. Lett. 113, 136403 (2014).
5. Zhang, R.-X., Hsu, H.-C. & Liu, C.-Y. Electrically tunable spin polarization of chiral edge modes in a quantum anomalous Hall insulator. Phys. Rev. B 93, 235315 (2016).
6. Chen, C.-Z., Xie, Y.-M., Liu, J., Lee, P. A. & Law, K. T. Quasi-one-dimensional quantum anomalous Hall systems as new platforms for scalable topological quantum computation. Phys. Rev. B 97, 104504 (2018).
7. Zeng, Y., Lee, C., Chaudhary, G. & MacDonald, A. H. Quantum anomalous Hall Majorana platform. Phys. Rev. B 97, 081102 (2018).
8. Yu, R. et al. Quantized anomalous Hall effect in magnetic topological insulators. Science 329, 61–64 (2010).
9. Chang, C.-Z. et al. High-precision realization of robust quantum anomalous Hall state in a hard ferromagnetic topological insulator. Nat. Mater. 14, 473–477 (2015).
10. Chang, C.-Z. et al. Experimental observation of the quantum anomalous Hall effect in a magnetic topological insulator. Science 340, 167–170 (2013).
11. Checkelsky, J. et al. Trajectory of the anomalous Hall effect towards the quantized state in a ferromagnetic topological insulator. Nat. Phys. 10, 731 (2014).
12. Bestwick, A. J. et al. Precise quantization of the anomalous Hall effect near zero magnetic field. Phys. Rev. Lett. 114, 187201 (2015).
13. Li, M. et al. Proximity-driven enhanced magnetic order at ferromagnetic-insulator-magnetic-topological-insulator interface. Phys. Rev. Lett. 115, 087201 (2015).
14. Tang, C. et al. Above 400-K robust perpendicular ferromagnetic phase in a topological insulator. Sci. Adv. 3, e1700307 (2017).
15. Katsnelson, M. F. et al. A high-temperature ferromagnetic topological insulating phase by proximity coupling. Nature 533, 513–516 (2016).
16. Fu, H., Liu, C.-X. & Yan, B. Exchange bias and quantum anomalous Hall effect in the MnBi2Te4–Cr1–x system. Phys. Rev. B 98, 024204 (2018).
17. Liu, C. et al. Intrinsic quantum anomalous Hall phase induced by proximity in germanene/Gr/GeTe3/ van der Waals heterostructure. Sci. Adv. 6, eaaz0948 (2020).
18. Zou, R., et al. Intrinsic quantum anomalous Hall phase induced by proximity in germanene/Gr/GeTe3/van der Waals heterostructure. Phys. Rev. B. 101, 161108 (2020).
19. Lu, L. J. et al. Intrinsic magnetic topological insulators in van der Waals layered MnBi2Te4–family materials. Adv. Sci. 5, eaaw6685 (2019).
20. Otrokov, M. M. et al. Prediction and observation of an antferromagnetic topological insulator. Nature 576, 416–422 (2019).
21. Liu, C. et al. Quantum phase transition from axion insulator to Chern insulator in MnBi2Te4. Preprint at https://ui.adsabs.harvard.edu/abs/2019arXiv19040715L (2019).
22. Deng, Y. et al. Quantum anomalous Hall effect in intrinsic magnetic topological insulator MnBi2Te4. Science 367, 895–900 (2020).
23. Ge, J. et al. High-chem-number and high-temperature quantum Hall effect without Landau levels. Nat. Sci. Rev. 7, 1280 (2020).
24. Serlin, M. et al. Intrinsic quantized anomalous Hall effect in a moire heterostructure. Science 367, 900–903 (2020).
25. Shaper, A. L. et al. Emergent magnetism near three-quarters filling in twisted bilayer graphene. Science 365, 605 (2019).
26. Choudhary, K., Garrity, K. F., Jiang, J., Pletcher, R. & Tavares, F. Computational search for magnetic and non-magnetic 2D topological materials using unified spin-orbit spillage screening. NPJ Comput. Mater. 6, 1–8 (2020).
27. Olsen, T. et al. Discovering two-dimensional topological insulators from high-throughput computations. Phys. Rev. Mater. 3, 024005 (2019).
28. Liu, C., Hughes, T. L., Qi, X.-L., Wang, K. & Zhang, S.-C. Quantum spin Hall effect in inverted Type-II semiconductors. Phys. Rev. Lett. 100, 236601 (2008).
29. Zhang, H., Xu, Y., Wang, J., Chang, K. & Zhang, S.-C. Quantum spin Hall and quantum anomalous Hall states realized in junction quantum wells. Phys. Rev. Lett. 112, 216803 (2014).
30. Turner, A. M., Zhang, Y., Meng, R. S. K. & Vishwanath, A. Quantized response and topology of magnetic insulators with inversion symmetry. Phys. Rev. B 85, 165120 (2012).
31. Hughes, T. L., Prodan, E. & Bernevig, B. A. Inversion-symmetric topological insulators. Phys. Rev. B 83, 245132 (2011).
32. Bradlyn, B. et al. Topological quantum chemistry. Nature 547, 298–305 (2017).
33. Po, H.-C., Vishwanath, A. & Watanabe, H. Symmetry-driven indicators of band topology in the 230 space groups. Nat. Commun. 8, 50 (2017).
34. Crutchfield, J., de Boer, J., van Wezel, J., Kane, C. L. & Slager, R.-J. Topological classification of crystalline insulators through band structure combinatorics. Phys. Rev. X 7, 041069 (2017).
35. Bergerhoff, G., Hundt, R., Sievers, R. & Brown, I. The inorganic crystal structure data base. J. Chem. Inf. Comput. Sci. 23, 66–69 (1983).
Research (Grant number N00014-18-1-2793), as well as Kaufman New Initiative research grant of the Pittsburgh Foundation. A.J. acknowledges support from U.S. DOE SC-00014388. S.X.D. thanks the International Partnership Program of Chinese Academy of Sciences, Grant number 112111KYSB20160061. It benefitted from the supercomputing resources of the National Energy Research Scientific Computing Center (NERSC), a U.S. Department of Energy Office of Science User Facility operated under Contract number DE-AC02-05CH11231.

AUTHOR CONTRIBUTIONS
Q.Y. and C.X.L. conceived and coordinated the research project. J.B.P. and J.Y. contributed equally to this work. J.B.P., Y.F.Z., and Q.Y. implemented the discovery workflow and performed the DFT calculations. J.Y. and C.X.L. performed the symmetry analysis and constructed the effective and tight-binding models. All authors participated in discussing the data and editing the manuscript.

COMPETING INTERESTS
The authors declare no competing interests.

ADDITIONAL INFORMATION
Supplementary information is available for this paper at https://doi.org/10.1038/s41524-020-00419-y.

Correspondence and requests for materials should be addressed to C.-X.L. or Q.Y.

Reprints and permission information is available at http://www.nature.com/reprints

Publisher’s note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. The images or other third party material in this article are included in the article’s Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article’s Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/.

© The Author(s) 2020