Screening two-dimensional materials with topological flat bands

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The topological flat band (TFB) has been proposed theoretically in various lattice models, to exhibit a rich spectrum of intriguing physical behaviors. However, the experimental demonstration of flat band (FB) properties has been severely hindered by the lack of materials realization. Here, by screening materials from a first-principles materials database, we identify a group of two-dimensional materials with TFBs near the Fermi level, covering some simple line-graph and generalized line-graph FB lattice models. These include the kagome sublattice of O in TiO_2 yielding a spin-unpolarized TFB, and that of V in ferromagnetic V_3F_8 yielding a spin-polarized TFB. The monolayer Nb_3TeCl_7 and its counterparts from element substitution are found to be breathing-kagome-lattice crystals. The family of monolayer III_2VI_3 compounds exhibit a TFB representing the coloring-triangle lattice model. ReF_3, MnF_3, and $MnBr_3$ are all predicted to be diatomic-kagome-lattice crystals, with TFB transitions induced by atomic substitution. Finally, HgF_2, CdF_2, and ZnF_2 are discovered to host dual TFBs in the diamond-octagon lattice. Our findings pave the way to further experimental exploration of eluding FB materials and properties.

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Introduction. The destructive interference of wave functions in a crystal lattice gives rise to a type of electronic band without dispersion, dubbed as topological/singular flat bands (TFBs) [1–5]. Without spin-orbit coupling (SOC), the TFB can be identified by the presence of a band touching point with a dispersive band at a high-symmetric $\boldsymbol{k}$ point, where its singular Bloch wave functions in reciprocal space manifest the emergence of topological noncontractible edge states in real space [4,5]; it differs from an isolated trivial flat band (FB) with nonsingular Bloch wave functions, such as the dangling-bond states. With SOC, the degeneracy at the touching point is lifted, leading to the gapped 2D TFB with a nonzero (spin) Chern invariant [4]. The nontrivial topology and the inherently strong electron-electron interaction due to vanishing electron kinetic energy render the TFB a rich spectrum of physical phenomena, such as fractional quantum anomalous Hall effect [6–8], ferromagnetism [9,10], Wigner crystallization [11,12], superconductivity [13], excitonic insulator state [14,15], and excited quantum anomalous/spin Hall effect [16].

Various lattice models have been theoretically proposed to host the TFB. These models are generally based on line-graph construction [17–19], such as the kagome [6,19,20], Lieb [21,22], breathing-kagome [23–26], diatomic-kagome [16,27], coloring-triangle lattices [28], and the diamond-octagon lattice (i.e., line-graph lattice of Lieb lattice) [2,29]. Also, the square and honeycomb lattices, with multiple atomic orbitals on each lattice site, can host TFBs [3,11,30–32]. On the other hand, however, very few electronic FB materials [33–42] have been identified, either experimentally or computationally, and realizations of the known FB lattice models are rather limited in general, with only a few examples such as the breathing-kagome lattice in Fe_3Sn_2 [41], and the coloring-triangle lattice in Cu-dicyanobenzene monolayer [42] presented so far. This, apparently, has severely hindered the experimental realization of eluding FB properties. It is worth noting that the experimental discovery of FB-associated superconductivity in twisted bilayer graphene [43] has generated a lot of excitement recently, and a surprising ferromagnetic covalent-organic framework without transition-metal atoms [44] has been shown to be originated from a FB [10]. Therefore, computational design and identification of new FB materials, already exist or to be fabricated, is highly desirable to significantly advance the study of TFB physics, materials, and devices.

On the other hand, the recent establishment of materials databases has enabled a high-throughput screening approach to discovering new materials in batches. For example, all the three-dimensional (3D) nonmagnetic topological crystals are screened for compiling a complete catalog of topological materials [45–47]; various nontrivial magnetic crystals are identified from 2D and 3D materials databases [48,49]. Here, by screening the 2DMatPedia database [50] for 2D crystalline materials, we have identified 15 monolayer atomic crystals hosting TFBs near the Fermi level, which cover six different FB lattice models. Among them, the kagome sublattice of O in TiO_2 supports a spin-unpolarized TFB, and that of V in ferromagnetic V_3F_8 supports a spin-polarized TFB. Monolayer

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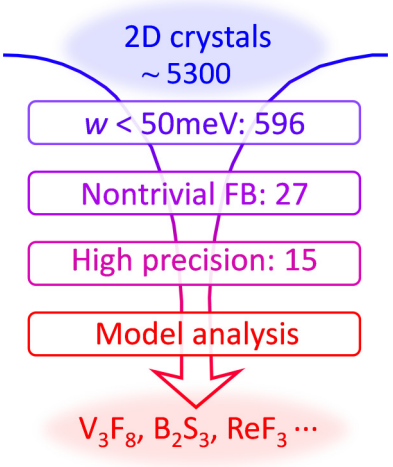

FIG. 1. Flowchart of computational screening for 2D crystals with ideal TFBs.

Nb_3TeCl_7 and its counterparts from element substitution, whose layered 3D structures were already synthesized over two decades ago, are found to be breathing-kagome crystals with possible high-order topology. The family of monolayer III_2VI_3 compounds exhibit a TFB representing the coloring-triangle lattice model. ReF_3, MnF_3, and $MnBr_3$ are all predicted to be diatomic-kagome crystals, with TFB transitions induced by atomic substitution. HgF_2, CdF_2, and ZnF_2 are discovered to host dual TFBs in the diamond-octagon lattice. Overall, some existing simple line-graph and generalized line-graph FB lattice models have been identified with several candidate materials.

Screening procedure. Our screening for 2D materials having TFBs starts with the database of 2DMatPedia, which contains electronic band structure without SOC for ~5300 monolayer atomic crystals [50], as shown in Fig. 1. The bandwidth w of 11 bands (chosen for FB searching in this work) around the Fermi level is calculated, from which 354 nonmagnetic and 242 magnetic materials are revealed to possess bands whose w is less than 50 meV. We note that the choice of this bandwidth threshold could be somewhat arbitrary, and we used a value that lies in the typical range of the SOC-induced energy gap in realistic solid materials. In general, using a larger (smaller) value one would find more (less) FB materials from screening but a higher (lower) percentage of trivial ones. Next, if the narrow band is identified with a degenerate point with another dispersive band, its host material is selected, amounting to 27 candidate TFB materials [see Tables S1 and S2 in the Supplemental Material (SM) [51]]. Finally, these 27 candidates were further double-checked by *ab initio* calculations with high precision and lattice model analysis (see the computational methods in the SM), which confirmed 15 TFB materials (nine nonmagnetic and six magnetic) representing respectively six different TFB lattice models, as listed in Table I. In the following, we choose the representative examples to show ideal TFBs discovered in (i) kagome lattice, (ii) related breathing-kagome, coloring-triangle, and diatomic-kagome lattices, (iii) diamond-octagon lattice, and (iv) d-orbital graphene lattice.

TABLE I. The discovered monolayer atomic crystals with ideal TFBs.

Model	Nonmagnetic	Magnetic	Space group
Kagome	TiO_2, $BaYSn_4O_7$	V_3F_8, $Li_2Fe_3F_8$	P-3m1
Breathing kagome	Nb_3TeCl_7, Ta_3SBr_7	Nb_3Cl_8	
Coloring triangle	B_2S_3 (III_2VI_3)		P-62m
Diatomic kagome	ReF_3	MnF_3, $MnBr_3$	P-3m1
Honeycomb		VF_3	
Diamond octagon	CdF_2, ZnF_2, HgF_2		P-4m2

Kagome crystals with the ideal TFB. The well-known kagome lattice consists of three sites in a unit cell, exhibiting two Dirac bands touched with a TFB [Fig. 2(a); see the tight-binding Hamiltonian in the SM]. As shown in Fig. 3(a), the monolayer TiO_2, a widely studied compound, has O atoms forming an upper and lower kagome layer bridged by O atoms in the middle. It turns out to host a typical kagome band structure from p orbitals of O atoms, with a well separated FB right below the Fermi level while the Dirac bands mix with other trivial bands. As expected, an isolated TFB arises upon a nontrivial gap opening in the presence of SOC [Fig. S1(a) in the SM]. There are also two additional sets of kagome bands far below the Fermi level arising from O 2s orbitals in the upper and lower kagome layer, respectively (Fig. S2), sharing a doubly degenerate TFB. Another nonmagnetic kagome crystal is found in monolayer $BaYSn_4O_7$, featuring a TFB around the Fermi level arising from Sn atoms sitting on a kagome lattice [Fig. S3(a)]. Previously, kagome bands have been mostly shown in metal-organic and covalent-organic frameworks [35–39]. Here we discover the unknown inorganic 2D materials hosting well-separated kagome bands.

Spin-polarized TFBs are discovered in magnetic kagome crystals with transition-metal elements. As shown in Fig. 3(b), three V atoms in monolayer V_3F_8 (shown to be stable in the literature [55]), with magnetic moment $M = 7\,\mu_B$, sit in a kagome sublattice. Naturally, spin-up kagome bands from d orbitals of vanadium atoms arise, with the TFB lying close to the Fermi level based on electron counting. With SOC, a topological gap opens to isolate a Chern TFB [Fig. S1(b)], affording an intriguing possibility of exploring fractional quantum anomalous Hall effect. Another ferromagnetic kagome material is found in monolayer $Li_2Fe_3F_8$, where the kagome sublattice consists of Fe atoms each with $M = 4\,\mu_B$ [Fig. S3(b)]. The above-mentioned kagome materials are all intrinsic, without the need of doping, and ideal, without band overlapping with other trivial bands, superior over previous computationally and experimentally identified electronic kagome metals [40,56–60]. For the kagome materials of TiO_2 and V_3F_8, their SOC-induced gap opening between the TFB and dispersive bands (Fig. S1) confirms the gapped TFB to possess a nonzero (spin) Chern invariant [4]. Similarly, the TFBs in other identified 2D materials are checked, where the gap is larger for materials with heavier atoms having stronger SOC.

Breathing-kagome crystals with potential second-order topology. When one triangle in a kagome lattice shrinks

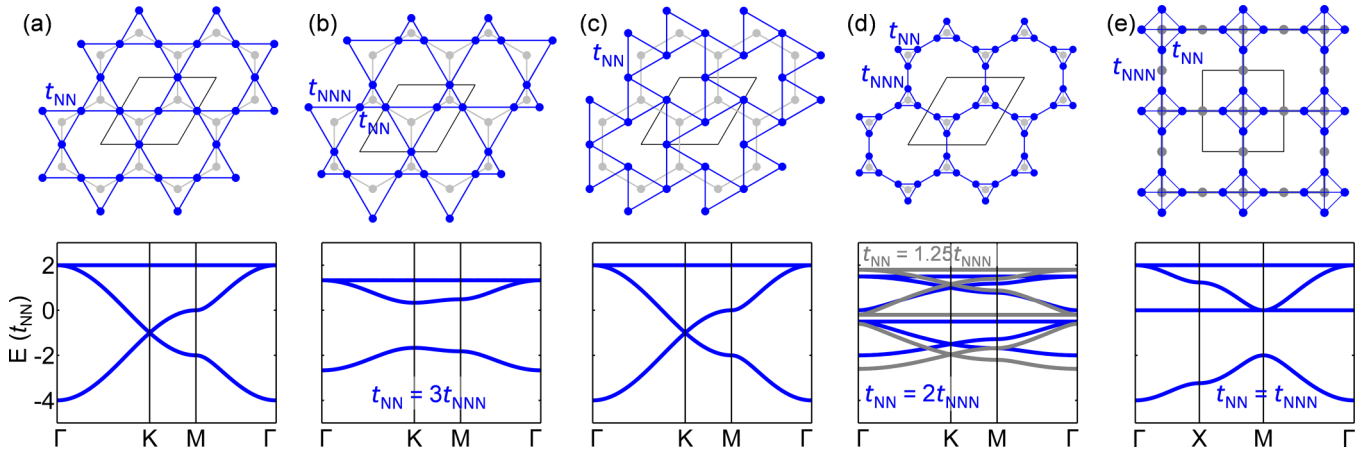

FIG. 2. Lattice structure and tight-binding bands of (a) kagome lattice, and its derivatives including (b) breathing-kagome, (c) coloring-triangle, (d) diatomic-kagome lattices, and (e) diamond-octagon lattice. t_{NN} and t_{NNN} represent the NN and NNN hopping integral, respectively. Gray and blue dots/lines in upper panels show the original lattices and their (generalized) line-graph lattices, respectively. For standard construction, (a) and (e), a line graph takes edge centers of the original graph as its vertices, while for generalized construction, a line graph takes off-edge-center positions, along the edge (b) or off the edge (c), as its vertices. Panel (d) can be viewed as two copies of the generalized line graph in (b), leading to dual FBs.

and the other one expands, a so-called breathing-kagome lattice model is constructed as a generalized line graph of hexagonal lattice [Fig. 2(b)]. This breathing mode does not affect the TFB, but opens a gap at Dirac point to induce the second-order topological corner states [23–25,61,62]. As shown in Fig. 4, monolayer Nb_3TeCl_7 is discovered to have Nb atoms locating at the breathing-kagome lattice sites, where Nb d orbitals constitute a clean set of kagome bands with a Dirac gap at the Fermi level. Such a breathing-kagome state is also discovered in 2D Ta_3SBr_7 with Ta atoms locating at the breathing-kagome sites [Fig. S4(a)]. Furthermore, spin-polarized breathing-kagome states with $M = 1\,\mu_B$ are discovered in monolayer Nb_3Cl_8 with three Nb atoms sitting at three breathing-kagome sites, respectively [Fig. S4(b)]. While magnetic interaction separates the spin-up and -down bands in energy space, all the features of breathing-kagome bands remain intact. Compared with Nb_3TeCl_7, the ferromagnetism originates from substituting a Te atom with a Cl atom; compared with V_3F_8, the breathing deformation in Nb_3Cl_8 originates from substituting V atoms by Nb atoms.

Coloring-triangle crystals with kagome bands. When one triangle in the kagome lattice rotates 30° clockwise and the other triangle 30° counterclockwise, a so-called coloring-triangle lattice, which is another generalized line graph of hexagonal lattice by an off-edge-center construction

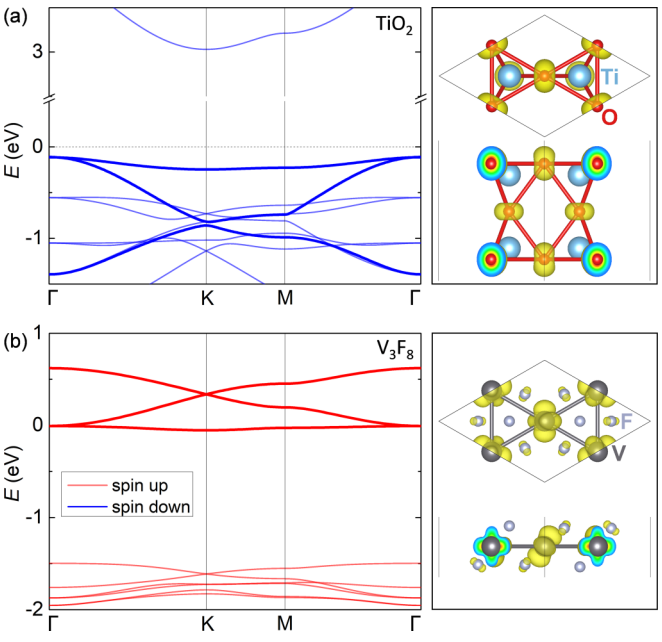

FIG. 3. Ideal kagome bands in monolayer kagome atomic crystals. (a) Oxygen atoms in monolayer TiO_2 form upper and lower kagome layers connected by middle sites, with titanium atoms locating at the center of oxygen tetrahedrons. The electronic structure exhibits obvious kagome bands (bold lines in left panel), and the charge of TFB near the Fermi level is contributed by p orbitals of oxygen atoms (right panel). (b) Vanadium atoms in monolayer V_3F_8 form the perfect kagome lattice, resulting in the typical kagome bands (bold lines in left panel) with a TFB exactly locating at Fermi level. The d orbitals of vanadium atoms contribute to the electron charge of the spin-up kagome bands (right panel).

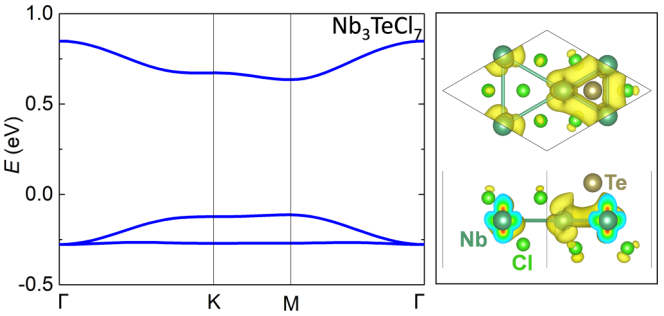

FIG. 4. Left: The electronic structure of monolayer Nb_3TeCl_7 with niobium atoms sitting on a breathing-kagome lattice. Right: Electron charge distribution contributing to the breathing-kagome bands with a gapped Dirac cone in the left.

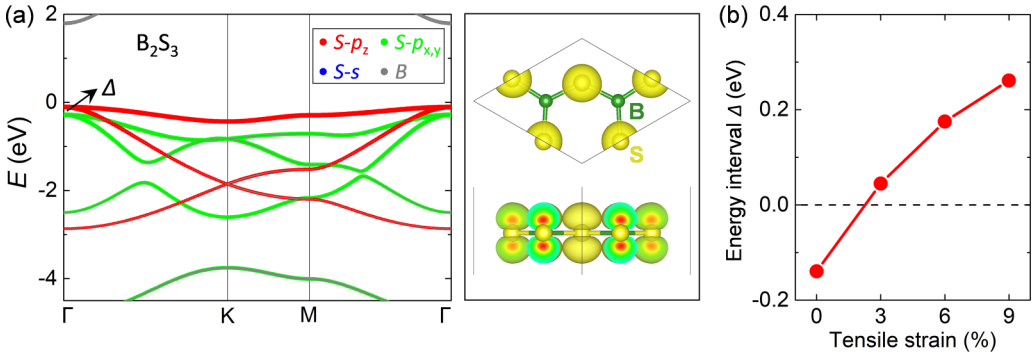

FIG. 5. (a) Electronic structure and orbital composition of monolayer B_2S_3 with +6% tensile strain (left panel). Δ marks the Γ-point energy interval between p_z and $p_{x,y}$ bands just below the Fermi level. The p_z orbitals of sulphur atoms, occupying the sites of a coloring-triangle lattice (right panel), contribute to the TFB of kagome bands near the Fermi level. (b) The variation of Δ with external tensile strain.

[Fig. 2(c)], forms to also host identical kagome bands [28]. The lattice can also be viewed as a triangle lattice with part of nearest-neighbor (NN) hopping being blocked, so that its realization in real crystalline materials is supposed to be quite difficult. Surprisingly, we have overcome this difficulty "accidentally." As shown in Fig. 5(a), within each unit cell of monolayer B_2S_3, S atoms constitute a triangle sublattice while B atoms are located at the center of two S triangles. Consequently, via the bridging of B atoms, the NN hopping between those S atoms in the two triangles with B are much stronger than those S atoms without bridging B atoms. So, effectively this provides a unique mechanism to selectively block part of NN hopping in a triangle S lattice, as required by the construction of a coloring-triangle lattice. This is clearly confirmed by the prefect kagome bands arising from S p_z orbitals in B_2S_3 [Fig. 5(a)]. The kagome bands are actually embedded in other bands from S $p_{x,y}$ orbitals in the equilibrium B_2S_3 (Fig. S5), but can be separated out by applying a small biaxial tensile strain [Fig. 5(b)], which simultaneously moves the TFB upward closer to the Fermi level. Deep-energy kagome bands from S 3*s* orbitals manifest also the coloring-triangle lattice in monolayer B_2S_3 (Fig. S5).

Similar to B_2S_3, our search reveals that most of the monolayer III_2VI_3 compounds, made from elements in the III and VI main group, exhibit the ideal kagome FBs satisfying the coloring-triangle model upon strain-induced band separation [Table S3, Fig. S6(a)]. An exception is monolayer Tl_2Te_3, for which the kagome states disappear due to the destruction of the desired coloring-triangle hopping, but on the other hand, its strong SOC induces a large topological gap of 0.32 eV [Fig. S6(b)], affording a candidate for high-temperature topological insulator. Also, the FB is absent in monolayer In_2Se_3, Tl_2S_3, and Tl_2Se_3 with large atoms, because of the structure-induced destruction of the desired coloring-triangle hopping (see details in Fig. S7).

TFBs evolution in diatomic-kagome crystals. When every kagome site is replaced by a pair of lattice sites, a diatomic-kagome lattice is formed, which is yet another generalized line graph of hexagonal graph constructed with two copies of breathing-kagome lattices [Fig. 2(d)], leading to intriguing evolution of TFBs and phase transitions [16,27]. For example, with a small next-NN (NNN) hopping integral, two sets of kagome bands coexist [blue lines in Fig. 2(d)]; as the NNN hopping becomes stronger, the bands evolve into a combination of Dirac bands and $p_{x,y}$-orbital hexagonal lattice bands [gray lines in Fig. 2(d)], labeled as (D; $p_{x,y}$) phase [11,30]. Interestingly, our screening process leads to the discovery of three diatomic-kagome crystals, exhibiting the TFB transitions as proposed in tight-binding models. As shown in Fig. 6(a), monolayer ReF_3 has Re atoms sitting on the diatomic-kagome sites, resulting in the (D; $p_{x,y}$) bands mainly contributed from Re-*d* orbitals. In contrast, the monolayer MnF_3 and $MnBr_3$ are ferromagnetic with $M = 4\,\mu_B$ on each Mn atom, possessing two sets of spin-up kagome and (D; $p_{x,y}$) bands, respectively [Figs. 6(b) and 6(c)]. From ReF_3 to MnF_3, the (D; $p_{x,y}$) bands transform into two sets of kagome bands, which in turn transform back into the (D; $p_{x,y}$) bands from MnF_3 to $MnBr_3$. This indicates that atomic substitution is an effective way to tune the hopping integrals in diatomic-kagome lattices, providing a promising strategy for topological and FB engineering [63].

Dual TFBs in diamond-octagon crystals. Lieb lattice contains a sublattice of checkerboard lattice which is the line graph of square lattice, and hence possesses FB. The line graph of Lieb lattice (dubbed as diamond-octagon lattice) also possesses FB [2,29]. With four lattice sites in a square unit cell [Fig. 2(e)], when the NN hopping integral is equal to the NNN hopping integral, two perfect TFBs can appear, which are degenerate with one parabolic band in between, with the band touching at Γ and M points, respectively. Due to the usual exponential decay of lattice hopping with distance in real materials, this peculiar model with equal hopping for both closer and farther sites appeared difficult to be realized. Interestingly, in monolayer HgF_2 (Fig. 7), two Hg atoms on the horizontal boundaries of the unit cell are located at different heights from the other two on vertical boundaries. This leads to an almost equal distance between the NN Hg-Hg sites in the same plane to the NNN Hg-Hg sites in the different planes, so as to satisfy the desired electron hopping condition prescribed in the above model [Fig. 2(e)], similar to the case of bilayer Ni(CH) for realizing a diatomic-kagome lattice [16]. Consequently, monolayer HgF_2 exhibits two TFBs inside four bands which are mainly contributed by *s* orbitals of mercury atoms. Also, this type of dual TFBs is found in monolayer CdF_2 and ZnF_2 (Fig. S8).

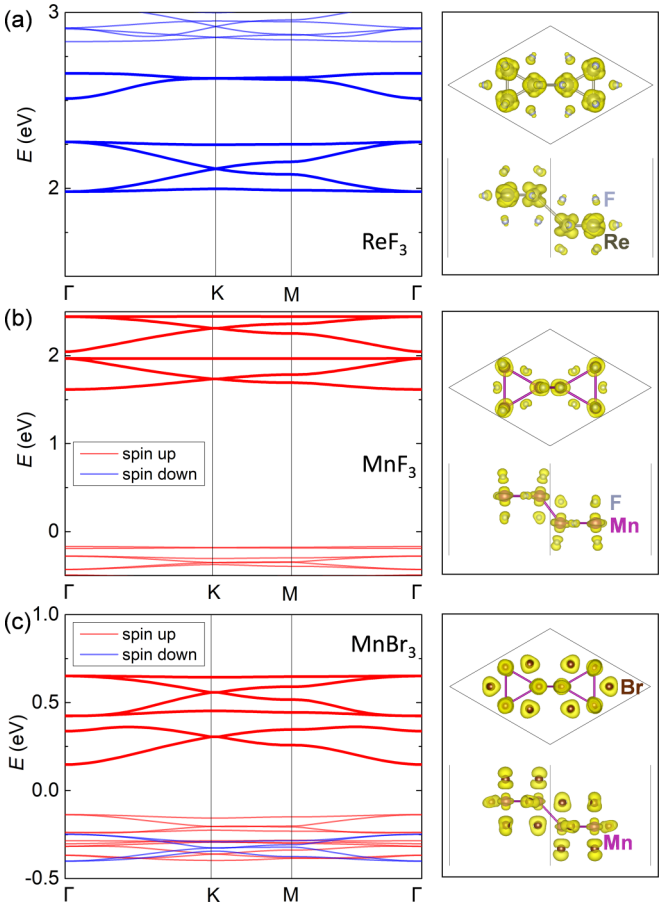

FIG. 6. Atomic and electronic structure for diatomic-kagome crystals of (a) ReF_3, (b) MnF_3, and (c) $MnBr_3$. Left: Band structure with the diatomic-kagome bands highlighted by thick lines. Right: The distribution of electron charge density for the diatomic-kagome bands highlighted in the left.

Orbital-enabled TFBs in honeycomb crystals. Beyond TFBs from line-graph construction, atomic/molecular orbitals in non-line-graph lattices can also be exploited to produce TFBs [3,11,30–32]. Organometallic framework [11,30,31] and bismuthene on semiconductor substrates [64,65] have been predicted to realize the honeycomb lattice with TFBs

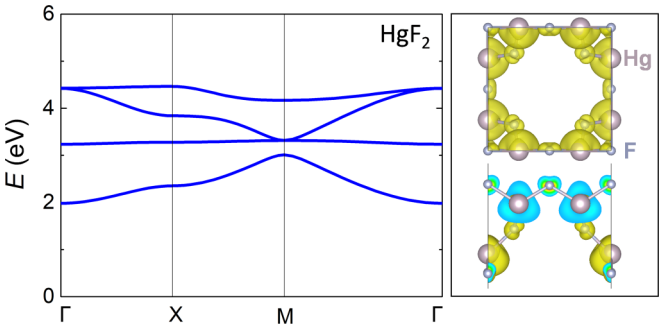

FIG. 7. Dual TFBs in monolayer HgF_2 with a diamond-octagon lattice. Left: Electron band structure. Right: Charge density plot showing the bands in the left are mainly contributed from *s* orbitals of mercury atoms.

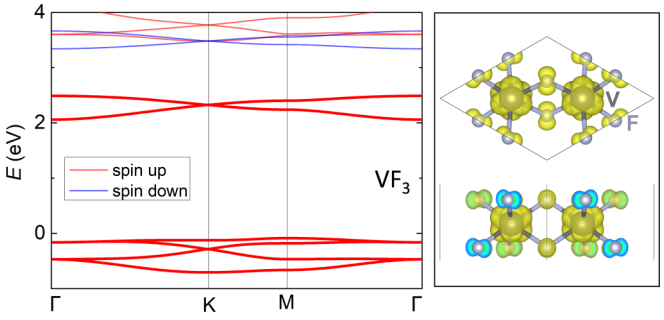

FIG. 8. Honeycomb TFB crystal of monolayer VF_3. Left: Band structure. Right: Electron charge density distribution contributing to the highlighted bands in the left.

from *p* orbitals. Here, as shown in Fig. 8, we discover monolayer VF_3 has V-*d*-orbital TFBs and Dirac bands, which conforms to the recently proposed TFBs from d_{xy} and $d_{x^2-y^2}$ orbitals [3] and Dirac bands from d_{z^2} in a honeycomb lattice, respectively. The TFB is just below the Fermi level and spin polarized, with $M = 2\,\mu_B$ on each V atom. In contrast, a previous sd^2-orbital honeycomb lattice with TFB has been shown in W overlayer on a halogenated Si(111) surface invoking an electronic kagome lattice from the coupled *s* and *d* orbitals [32].

In all the above cases, the screening condition for bandwidth $w < 50$ meV is used (Fig. 1). One might relax this condition to discover more FB materials. For example, we also found the monolayer II_1VII_2 compounds, made from elements in the II and VII main groups, have the checkerboard and diamond-octagon FBs with larger bandwidth, where the former arises from an intriguing *p*-orbital configuration that is equivalent to the diagonal *p*-orbital orientation in a square lattice [3], such as $MgCl_2$ shown in Fig. S9 of SM. Totally, two monolayer crystals of VF_3 and $MgCl_2$ are identified to be non-line-graph materials having the FB near the Fermi level, while others are line-graph FB materials. Beyond the analysis by using tight-binding lattice models, the Wannier construction for FBs of the non-line-graph VF_3 and line-graph V_3F_8 is carried out, which again confirms our finding that they respectively conform to the kagome and honeycomb FB model (see details in Fig. S10).

Potential experimental fabrication and measurement. We note that the 3D layered Nb_3TeCl_7 and Ta_3SBr_7 have been experimentally synthesized over twenty years ago [66,67], making experimental observation of these material-specific breathing-kagome states highly promising. The 2D film of breathing-kagome crystal Nb_3Cl_8 with controllable thickness has been already realized in experiment [68], which calls for immediate experimental confirmation of its predicted TFB. Also, some of the coloring-triangle crystals, such as Ga_2S_3, are potentially experimentally achievable [69–71]. For other 2D FB crystals predicted here, we expect that they can be exfoliated from layered bulk materials by the mechanical, electrical, or chemical method [72,73], or can be grown on suitable prescreened substrates by the molecular beam epitaxy, or chemical vapor deposition technology [74,75].

The TFBs predicted in this work, all around Fermi level, can be directly probed by angle resolved photoemission

spectroscopy [76], where the momentum-space band dispersion can be measured. Also, the FBs can be probed by scanning tunneling spectroscopy (STS), where a sharp peak of density of states could be shown as a signature for FBs [40], and the edge/corner states from electron topology can be directly imaged by the space-resolved STS. The measurement of quantum Hall conductance can demonstrate topological transport properties [77], which would potentially confirm the intriguing fractional quantum anomalous Hall effect from partially occupied TFBs [6–8].

Conclusions. By employing the data screening calculations on materials from a first-principles materials database, we have discovered 15 inorganic 2D crystals (Table I) with ideal TFBs representing six FB lattice models, which opens a door towards experimental observation of TFBs in real materials, and exploration/substantiation of their intriguing physics and applications. In particular, some of these realistic 2D crystals have already been made in experiments, which will hopefully draw immediate attention. Moreover, the screening approach developed here for searching 2D FB crystals can be extended to systematically uncovering 3D FB materials from the well-established databases [78–81].

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