First-Principles Design of a Half-Filled Flat Band of the Kagome Lattice in Two-Dimensional Metal-Organic Frameworks

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We design from first principles a new type of two-dimensional metal-organic frameworks (MOFs) using phenalenyl-based ligands to exhibit a half-filled flat band of the kagome lattice, which is one of the lattice family that shows Lieb-Mielke-Tasaki’s flat-band ferromagnetism. Among various MOFs, we find that trans-Au-THTAP(trihydroxytriaminophenalenyl) has such an ideal band structure, where the Fermi energy is adjusted right at the flat band due to unpaired electrons of radical phenalenyl. The spin-orbit coupling opens a band gap giving a non-zero Chern number to the nearly flat band, as confirmed by the presence of the edge states in first-principles calculations and by fitting to the tight-binding model. This is a novel and realistic example of a system in which a nearly flat band is both ferromagnetic and topologically non-trivial.

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Introduction. — The exploration and discovery of new strongly-correlated or topologically non-trivial materials drive much of modern condensed-matter physics, yet an experimental design of such materials is still challenging. One class of materials that could greatly extend possibilities of material designing is, in our view, metal-organic frameworks (MOFs). These are crystalline materials composed of metal ions and bridging organic molecules, which have been the subject of numerous investigations in inorganic and materials chemistry [1]. Owing to their typically trivial and localized electronic states, MOFs have not attracted much attentions from condensed-matter physicists. However, recent experimental success in fabricating atomically layered two-dimensional (2D) MOFs with kagome lattice structures, initiated by the Nishihara group [2, 3], is bridging the gap between condensed-matter physics and chemistry. The Dincă group also succeeded in creating 2D MOFs [3]. (Similar 2D MOFs have been fabricated by other groups [3, 10].) Some of these new 2D MOFs have been theoretically proposed to become organic Z2 topological insulators [11, 12] or half-metallic ferromagnets [13, 14].

The kagome lattice has a virtue of its electronic structure exhibiting a flat band at the highest (or lowest) energy. It has been proven that the tight-binding Hubbard model on the kagome lattice has a non-trivial ground state (far different from the atomic limit) showing itinerant ferromagnetism at arbitrary on-site Coulomb repulsion \( U > 0 \) when the flat band is half-filled [20]. While several classes of lattices are known to show the flat-band ferromagnetism, as proposed by Lieb, Mielke and Tasaki [21, 24], the kagome lattice has an advantage that it is a realistic structure from a synthetic point of view, and does not require a fine tuning for hopping parameters to accommodate a flat band [23]. When the spin-orbit coupling (SOC) is introduced, the energy gap opens between neighboring bands, and the flat band becomes topologically non-trivial with a non-zero Chern number [26]. It has been discussed that if the nearly flat band with a non-zero Chern number is fractionally filled and is well separated from other bands, the system could be a fractional Chern insulator [26, 28].

So far, it has been difficult to make the flat band partially filled for the 2D kagome lattice (i.e., the Fermi energy adjusted to the flat band). While some quasi-1D systems have been regarded as possible candidates for nearly-flat-band ferromagnetism [29–31], there is no experimental realization in 2D crystalline systems, as originally proposed by Lieb, Mielke, and Tasaki. Theoretically, Liu et al. [32] proposed that an In-based 2D organic nanosheet could possess a topologically non-trivial flat band near the Fermi energy, but this material still needs an additional hole doping to be ferromagnetic. Moreover, it may be difficult to experimentally realize such a 2D structure because the highly covalent bond between In and C would not lead to the formation of crystalline materials.

In this Letter, we propose new 2D MOFs with kagome structures from first principles, where we can show that the flat band of a kagome lattice is indeed expected to be half-filled with the appropriate choice of the organic ligand. The essential idea is to use an organic neutral radical called phenalenyl as a building block. In the absence of such an organic radical, the hole doping would be necessary as described in the previous research [32, 33]. Based on first-principles electronic structure calculations, we discuss that the proposed phenalenyl-based 2D MOF becomes ferromagnetic with the flat band having a non-
zero Chern number if SOC is taken into account.

New 2D Metal-Organic Frameworks. — We employ phenalenyl-based ligands [see Z = C\(^*\) in Fig. 1(a)], where \(\bullet\) represents an unpaired electron, which are appropriate for creating kagome structures with its triangular shape. These ligands are envisaged to be polymerized with a transition metal (M = Ni, Cu, Pt, Au), where X and Y(= O, S, NH) coordinate to M [see Fig. 1(b)]. When we choose M as spin-1/2 metal (Cu or Au in this case), there is one unpaired electron per orbital on each kagome site. An important advantage of a phenalenyl-based ligand is that we can automatically adjust the Fermi energy from the brown dashed line in Fig. 1(c) to the desired position to make the flat band of the kagome lattice half-filled by transferring two unpaired electrons per unit cell to M due to the neutral stable radical resonance structure of phenalenyl [see the arrow in Fig. 1(c)]. In the absence of this radical (such as when Z = N), the electron or hole doping would be necessary, as described previously\(^{14,32,33}\). Specifically, a chemical formula of trans-Au-THTAP (trihydroxytriaminophenalenyl)\(^{35}\) with M = Au, X = NH, Y = O is shown in Fig. 1(d). Its whole real-space structure and its unit cell (solid line) are illustrated in Fig. 1(e), with Au atoms occupying each vertex of the kagome lattice.

Electronic Structures From First Principles. — The above expectations to realize a half-filled flat band have to be confirmed with a first-principles electronic structure analysis. To this end, we used the first-principles electronic state calculation code called OPENMX\(^{36}\), based on density functional theory (DFT). With the repeated slab construction\(^{37}\), we first calculated possible phenalenyl-based MOFs with M = Cu, Au as spin-1/2 metal. In order to conserve a parity symmetry and break other symmetries to lift the degeneracy, we preferred X \(\neq Y\) and a trans-structure\(^{13,38}\). Then, we found that compounds with M = Cu tend to have a bended band, so we preferred M = Au. Finally, we found that all three remaining candidates (with M = Au and (X, Y) = (O, S), (S, NH), (NH, O)) have a nearly flat band exactly lying on the Fermi energy\(^{39}\). Among these, trans-Au-THTAP \([(X,Y) = (NH, O)]\) has the best band structure in the sense that its band structure is accurately fitted to that obtained from a tight-binding model on the kagome lattice around the Fermi energy.

trans-Au-THTAP. — We then concentrated on the band structure of trans-Au-THTAP and, after the geometric optimization, the system completely favors a planar structure with an optimized lattice constant of 16.91 Å. The band structure calculated without SOC is shown in Fig. 2(a). The black solid lines display the spin-up bands, while the blue dashed lines display the spin-down bands. The system shows a metallic nature and the nearly flat band near the Fermi energy \([E = 0\) in Fig. 2(a)] arising from the kagome lattice is approximately half-filled and indeed spin-polarized. This gives a ferromagnetic behavior with a total spin moment of 1.00 \(\mu_B\)/unit cell. We have to note that this spin moment exactly coincides with the expected value for the flat-band ferromagnetism\(^{20}\), and the reason why there is a flat band near the Fermi energy is not accidental as happened in Ref. 19. Remarkably, the analysis of the partial density of states (PDOS) for each element clearly shows that the kagome bands near the Fermi energy mostly come from C and N atoms [see Fig. 2(b)]. This real-space property is further confirmed by the analysis of the spin density\(^{40}\). Moreover, by carrying out a fully relativistic self-consistent calculation on this system, we find that SOC opens a gap of 7.8 meV between the nearly flat band and the lower dispersive band at \(\Gamma\) [compare Fig. 2(c) without SOC to (d) with SOC]. This system has turned out to

![FIG. 1](Image)
FIG. 2. (Color online) (a) Band structure of trans-Au-THTAP calculated without SOC. The black solid (blue dashed) lines show the spin-up (spin-down) bands. (b) PDOS of the spin-down bands for each element in trans-Au-THTAP calculated without SOC. (c) Zoom up of (a) around the Fermi energy. (d) Zoom up of the band structure calculated with SOC. (e) Comparison between the band structures calculated with SOC by DFT (solid lines) and by TB (circles) with the calculated Chern numbers (Ch). In each panel, the Fermi energy is taken to be zero.

be still metallic, due to a slight warping of the nearly flat band. The spin-up and spin-down bands are no longer separable when we calculate with SOC, but the $z$-component of the spin ($\sigma_z$) is approximately a conserved quantum number because the calculated magnetic order is always along the $z$-direction in the case of including SOC. This gives a total spin moment of 0.99 $\mu_B$/unit cell, a total orbital moment of 0.02 $\mu_B$/unit cell along the $z$-direction, and an exchange splitting of 159.5 meV.

Topological Properties From a Tight-Binding Model. — In order to show the topological non-triviality of the gap between the nearly flat band and the dispersive band of trans-Au-THTAP, we first considered a single-orbital tight-binding (TB) model on the kagome lattice, where each single orbital is assumed to be localized around Au. Actually, the wave functions forming the kagome bands are not completely localized on Au and spreading over the $\pi$-conjugated system, but we can still assume a single-orbital TB model as long as the lattice symmetry is preserved and the parameters of the TB model are somehow renormalized by the effect of spreading. We added a Zeeman term (exchange splitting) to the Hamiltonian considered in Ref. [11–13, 26, 41] to include the effect of ferromagnetism (a similar term was already considered in Ref. [13]). We considered a complex nearest-neighbor (NN) hopping and a real next-nearest neighbor (NNN) hopping in a Hamiltonian, $H = E_0 + H_0 + H_{SO} + H_Z$,

$$H_0 = -t_1 \sum_{\langle ij \rangle \sigma} c^\dagger_{i\sigma} c_{j\sigma} - t_2 \sum_{\langle\langle ij\rangle\rangle \sigma} c^\dagger_{i\sigma} c_{j\sigma},$$  (1)

$$H_{SO} = i\lambda_1 \sum_{\langle ij \rangle \alpha \beta} \nu_{ij} \sigma^z_{\alpha\beta} c^\dagger_{i\alpha} c_{j\beta},$$  (2)

$$H_Z = b \sum_i (-1 - \sigma^z_{\alpha\beta}) c^\dagger_{i\alpha} c_{i\beta}.$$  (3)

Here $E_0$ is the energy offset and $c^\dagger_{i\sigma}$ and $c_{i\sigma}$ are the creation and annihilation operators of the $\sigma$-spin electron on the $i$th site of the kagome lattice, respectively. $\langle ij \rangle$ and $\langle\langle ij\rangle\rangle$ denote the NN and NNN bonds, respectively, while $t_1$ and $t_2$ are the corresponding real-valued hopping parameters. $\lambda_1$ is the NN intrinsic spin-orbit coupling and $b$ is the Zeeman splitting, while $\sigma_{\alpha\beta}$ is the Pauli matrix for the spin component. $\nu_{ij}$ is 1 for the counterclockwise hopping and $-1$ for the clockwise hopping when viewed from above. For simplicity, we have omitted the NNN imaginary hopping parameter $\lambda_2$, which is expected to be much smaller than the others [12].

This TB Hamiltonian conserves the $z$-component of the spin, so we can divide the one-particle Hilbert space $\mathcal{H}$ into $\mathcal{H}_+ \oplus \mathcal{H}_-$ by the eigenvalue of $\sigma^z$. We only consider the space $\mathcal{H}_+$ because there are only spin-down bands
near the Fermi energy. In other words, we have projected out the spin-up states by taking the limit $b \to \infty$ first. We can then accurately fit the kagome bands in the DFT calculation with SOC (solid line) to that obtained from the TB model (circles) around the Fermi energy as shown in Fig. 2(e) with the parameters $E_0 = -87.4$ meV, $t_1 = 45.1$ meV, $t_2 = 1.0$ meV, $\lambda_1 = 1.2$ meV. Based on the TB model, we can calculate a topological Chern number ($C_h$) for each band. From the results displayed in Fig. 3(e), we can conclude that the nearly flat band is indeed topologically non-trivial with a Chern number of $-1$ within this TB framework.

**Edge States.** — The topological non-triviality of the system is also endorsed by a DFT calculation for the edge states. We again used OPENMX with the repeated ribbon construction. Because we are now interested in the topological properties of the designed bulk structure, we assumed the magnetic easy axis to be the same as the bulk, and imposed the spin orientation on the constraint to be along the $z$-direction, which is taken to suppress the boundary effect that destabilizes the bulk magnetic structure. The calculated band structure is shown in Fig. 3 where the red and blue dashed lines clearly show the chiral edge states for each boundary and cannot be gapped away. Since we took a different boundary condition for each edge (one with Au aligning along the boundary and the other without Au), they are asymmetric against $k_x = 0$. These edge modes are similar to the ones in Ref. 15 except for the spin-polarization due to its ferromagnetism. The wave functions for the dashed bands are exponentially localized along the boundaries, while there are also trivial flat bands in the Fig. 3 localized along the boundaries. If we exclude such trivial bands as states in the atomic limit, the emergence of the non-trivial states again confirms the topological nature of the nearly flat band.

**Conclusion.** — We have proposed several new 2D MOFs, and found that trans-Au-THTAP has a topologically non-trivial nearly flat band from the DFT calculations. This is a novel and realistic example of a system in which a nearly flat band at the Fermi energy is both ferromagnetic and topologically non-trivial. From a synthetic standpoint, we have to note that Au indeed prefers a square planar coordination environment in the +2 formal oxidation state, as discussed in Ref. 46. We have to note that there would be electron correlation effects for the flat-band ferromagnetic ground state that are not properly captured within the DFT framework, which is worthwhile to study in the future.

Although the proposed system, being metallic, does not have a quantized Hall current, a topologically non-trivial phase realized in the proposed material (called a Chern metallic phase in Ref. 47) is still worth investigating. At this moment we have only confirmed the existence of the edge states with a constraint on the spin structure, and it would be a future work to clarify whether there is a magnetic order along the boundaries and how it affects the topological edge states. It would be more interesting if one could enhance the band gap by further material exploration using the flexibility of MOFs to make the system a ferromagnetic insulator with a quantized Hall conductance (quantum anomalous Hall effect 48) or a fractional Chern insulator 28, 29 or with a large ratio of (band gap)/(band width), although we have to note that a no-go theorem has been proven mathematically for topologically non-trivial perfectly flat bands within local tight-binding models.

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MOF is defined as a coordination compound continuously extending in 1, 2 or 3 dimensions through coordination bonds with an open framework containing potential voids by IUPAC. See S. R. Batten, N. R. Champness, X.-M. Chen, J. García-Martínez, S. Kitagawa, L. Ohrström, M. O’Keeffe, M. P. Suh, and J. Reedijk, Pure Appl. Chem. 85, 1715 (2013).


[34] Here M = Ag is excluded since a homologous element Ag is known to have a tendency to form straight complexes.

[35] An IUPAC name for the ligand is 3,6,9-triaminophenol-1,4,7-triol.


[38] A cis-structure will break the structure’s parity symmetry.


[40] See Supplemental Material for the real-space spin density and Bloch functions.


In this Supplemental Material we would have Section A: calculation (simulation) details of the band structures and the edge states of the proposed materials within the framework of density functional theory (DFT). Section B: calculated band structures for all the proposed materials explaining the emphasis on trans-Au-THTAP (trihydroxytriaminophenalenyl) as a target metal-organic framework (MOF). Section C: discussion on the exchange-correlation-potential dependence of the calculated band structures of trans-Au-THTAP. Section D: bulk spin density and bulk Bloch functions of trans-Au-THTAP. Section E: wave functions of the edge states of trans-Au-THTAP and distinction of the topological edge states from the trivial ones.

Section A: PROFILES FOR THE DFT CALCULATIONS

In OPENMX [1], one-particle wave functions are expressed by the linear combination of pseudoatomic basis functions, where the norm-conserving pseudopotentials are used. In our simulation, the generalized gradient approximation represented by Perdew, Burke and Ernzerhof (GGA-PBE) [2] was employed for the exchange-correlation potential. A self-consistent loop was iterated until the energy was relaxed with the error of $10^{-7}$ Hartree. The geometric (structure) optimization of internal coordinates was also iterated until the force became smaller than $10^{-4}$ Hartree/Bohr, and the lattice constant was optimized with the error of 0.01 Å. The plot of the density of states (in Fig. 2(b) in the main text) used a Gaussian broadening of 0.01 eV.

In order to capture the two-dimensional (2D) nature of the proposed MOFs, we defined a repeated slab construction as follows. The distance between the neighboring MOFs is 10 Å to neglect the interlayer interaction. We used a $30 \times 30 \times 1$ $k$-points mesh in the first Brillouin zone and an energy cutoff of 500 Ry for the numerical integrations and the solution of the Poisson equation using the fast Fourier transformation algorithm. The semicore $5p$ electrons of Au were treated as valence electrons. As basis functions, $5p$, $5d$, $5f$, $6s$, $6p$, $6d$, $7s$, $7p$, and $8s$ orbitals for Au, $2s$, $2p$, $3s$, $3p$, and $3d$ orbitals for C, N, O, and $1s$, $2s$, $2p$, $3p$ for H were employed. Initial crystal structures for the geometry optimization were constructed from molecular structures [3] in the Cambridge Crystallographic Data Center [4].

In order to calculate the edge states of trans-Au-THTAP, we defined a repeated ribbon construction as follows. We considered a ribbon of the proposed MOF with the width of 292.8 Å (which corresponds to 20 unit cells) along the $y$-direction [i.e., perpendicular to both the $z$-direction and the $a$-direction. See Fig. 4(d)]. We used a $30 \times 1 \times 1$ $k$-points mesh. We did not execute the optimization of the atomic configuration and the lattice constant for simplicity here.
TABLE I. Lattice constants and total spin moments obtained from the geometric optimization and the DFT calculation with spin polarization for the proposed materials.

<table>
<thead>
<tr>
<th>Name</th>
<th>Ligand</th>
<th>M (X, Y)</th>
<th>Lattice constant (Å)</th>
<th>Total spin moment (µB/unit cell)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a)</td>
<td>trans-Cu-THTTP</td>
<td>trihydroxytrithiophenalenyl Cu (O, S)</td>
<td>17.29</td>
<td>1.00</td>
</tr>
<tr>
<td>(b)</td>
<td>trans-Cu-TATTP</td>
<td>triaminotrithiophenalenyl Cu (S, NH)</td>
<td>17.50</td>
<td>1.00</td>
</tr>
<tr>
<td>(c)</td>
<td>trans-Cu-THTAP</td>
<td>trihydroxytriaminophenalenyl Cu (NH, O)</td>
<td>16.62</td>
<td>4.80</td>
</tr>
<tr>
<td>(d)</td>
<td>trans-Au-THTTP</td>
<td>trihydroxytrithiophenalenyl Au (O, S)</td>
<td>17.70</td>
<td>1.00</td>
</tr>
<tr>
<td>(e)</td>
<td>trans-Au-TATTP</td>
<td>triaminotrithiophenalenyl Au (S, NH)</td>
<td>17.90</td>
<td>1.00</td>
</tr>
<tr>
<td>(f)</td>
<td>trans-Au-THTAP</td>
<td>trihydroxytriaminophenalenyl Au (NH, O)</td>
<td>16.91</td>
<td>1.00</td>
</tr>
</tbody>
</table>

FIG. S1. (a)-(f) Band structures for (a) trans-Cu-THTTP, (b) trans-Cu-TATTP, (c) trans-Cu-THTAP, (d) trans-Au-THTTP, (e) trans-Au-TATTP and (f) trans-Au-THTAP. In each panel, black solid (blue dashed) lines show the spin-up (spin-down) bands, and the Fermi energy is taken to be zero. (g) Unit cell of trans-Au-THTAP and the definition of the axes. (h) Band structure for trans-Au-THTAP calculated without spin polarization.

Section B: CALCULATED BAND STRUCTURES FOR ALL THE PROPOSED MATERIALS

With the repeated slab construction, we have calculated all the proposed materials without a spin-orbit coupling (SOC) and found that all of these favor a planar structure and ferromagnetism. The calculated lattice constants and total spin moments are listed in Table I and the calculated band structures are shown in Figs. S1(a)-(f). It can be easily seen that (f) trans-Au-THTAP has the best band structure in the sense that its band structure is accurately fitted to that obtained from a tight-binding model on the kagome lattice around the Fermi energy, as shown in Fig. 2(e) in the main text. In (d) trans-Au-THTTP and (e) trans-Au-TATTP, the lowest band of the kagome bands near the Fermi energy is warping around Γ due to the interference with the lower irrelevant bands, and these bands will not be described by a simple single-orbital kagome tight-binding model. Before introducing spin polarization, the flat band of the kagome lattice is completely half-filled in trans-Au-THTAP [see Fig. S1(h)]. This unpolarized state will be destabilized and a ferromagnetic order appears in the calculation with spin polarization [see Fig. S1(f)].
FIG. S2. Band structures for trans-Au-THTAP in (a) GGA, (b) GGA+SOC, (c) GGA+U and (d) GGA+SOC+U calculations. In calculations without SOC, black solid (blue dashed) lines show the spin-up (spin-down) bands. In each panel, the Fermi energy is taken to be zero.

Section C: COMPARISON BETWEEN GGA AND GGA+U FOR trans-Au-THTAP

With the repeated slab construction for trans-Au-THTAP, we have compared the band structures of trans-Au-THTAP between GGA(+SOC) and GGA(+SOC)+U, where +SOC means calculations with a spin-orbit coupling. In GGA(+SOC)+U calculations, we added a Hubbard term of $U = 2$ eV for Au 5$d$ orbitals (which is a value used often for 5$d$ transition metals, such as Ir [{5}]). As shown in Fig. S2, there is no qualitative difference between GGA and GGA+U for the kagome bands, but by including $U$ the exchange splitting grows from 159.5 meV to 162.5 meV, and the SOC gap between the flat band around the Fermi energy and the lower dispersive band changes from 7.8 meV to 6.5 meV.
Section D: BULK SPIN DENSITY AND BULK BLOCH FUNCTIONS OF \textit{trans-}Au-THTAP

With the repeated slab construction for \textit{trans-}Au-THTAP, we calculated the bulk spin density without SOC [see Fig. S3(b)]. In addition to the partial density of states (PDOS) analysis shown in Fig. 2(b) in the main text, the spin density clearly shows the real-space profile of the flat band because almost only the flat band is spin-polarized in the ground state of \textit{trans-}Au-THTAP. It can be clearly seen that the flat band comes mostly from C \textit{2p\textsubscript{z}} and N \texti{2p\textsubscript{z}} orbitals, with a smaller contribution from Au \texti{5d} orbitals. We also calculated the real parts of the spin-down Bloch functions (the periodic part of the one-particle wave function for each \textit{k}-point) for the flat band near the Fermi energy with SOC [see Figs. S3(d)-(f)]. There is a strong momentum (\textit{k}) dependence in the Bloch functions, which is a feature inherent in the nearly-flat-band ferromagnetism \cite{cite}, clearly shows that the flat band is not in the trivial atomic limit.
Section E: REAL-SPACE PROPERTIES OF THE EDGE STATES OF trans-Au-THTAP

As mentioned in the main text, there are both non-trivial edge states from the topological property and trivial edge states when we calculate the edge states with the repeated ribbon construction. The band structure shown in Fig. S4(a) is the same band structure as in the main text [see Fig. 3 in the main text], but we highlighted only the non-trivial edge state along the bottom edge (red dashed line) and the trivial edge state along the top edge (blue solid line). Figures S4(b)-(e) display a real part of a typical spin-down Bloch function for the non-trivial edge state at each \( k_x \)-point, while (f)-(i) for the trivial edge state. There is a significant momentum \( k_x \) dependence in the Bloch functions for the non-trivial edge state, whereas for the trivial edge state the real-space structure is almost the same.
for different $k_x$. Thus, we can conclude that the trivial edge state is actually in the atomic limit, so we can exclude such states when we consider the topological property of the proposed system.

[3] CCDC structure codes: XIGZAQ and MEYZUN.