PHYSICS

The \( p \)-orbital magnetic topological states on a square lattice

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ABSTRACT

Honeycomb or triangular lattices were extensively studied and thought to be proper platforms for realizing the quantum anomalous Hall effect (QAHE), where magnetism is usually caused by \( d \) orbitals of transition metals. Here we propose that a square lattice can host three magnetic topological states, including the fully spin-polarized nodal loop semimetal, QAHE and the topologically trivial ferromagnetic semiconductor, in terms of the symmetry and \( k \cdot p \) model analyses that are material independent. A phase diagram is presented. We further show that the above three magnetic topological states can indeed be implemented in the two-dimensional (2D) materials ScLiCl\(_5\), LiScZ\(_5\) (Z=Cl, Br) and ScLiBr\(_5\), respectively. The ferromagnetism in these 2D materials is microscopically revealed from \( p \) electrons of halogen atoms. This present study opens a door to explore the exotic topological states as well as quantum magnetism from \( p \)-orbital electrons by means of the material-independent approach.

Keywords: \( p \)-orbital magnetism, square lattice, topological states

INTRODUCTION

In two-dimensional (2D) systems, the coexistence of magnetism and non-trivial topological states can induce many novel physical phenomena. A typical example is the quantum anomalous Hall effect (QAHE), where the combination of ferromagnetism and topological insulator can generate dissipationless edge states at boundaries \([1–6]\). The quantized Hall conductivity is carried by the edge states, which are robust against disorders and impurities. Owing to the dissipationless chiral edge states, QAHE would have potential applications in ultralow-power consumption spintronic devices \([7]\). Thus, the search for materials with QAHE has attracted extensive interest \([3,8–12]\). Since the seminal work of Haldane \([1]\), the honeycomb lattice is thought to be a proper platform to realize the QAHE, e.g. several ferromagnetic transition metal trihalides with honeycomb lattice were proposed to be candidates for the implementation of QAHE \([13–19]\). Besides, the experimental observation of QAHE was realized in magnetic atom doped systems \([10,20–22]\), and recently in the few layers of magnetic semiconductor MnBi\(_2\)Te\(_4\) \([23–26]\) with triangular lattice. In these materials, \( d \) orbitals of the transition metal play important roles in realizing QAHE. The following two interesting questions then arise. Can the QAHE be realized in other lattices, such as the square lattice? Can the QAHE be obtained in materials with \( p \) orbitals? The studies of these questions not only give a further understanding of topological states and quantum magnetism, but also offer a new family of materials to search for possible room-temperature QAHE.

In this work, we address these appealing issues by revealing a square lattice with the space group P\( /4n\) (No. 85) that can accommodate three different \( p \)-orbital magnetic topological states, i.e. the fully spin-polarized nodal loop semimetal, QAHE and the ferromagnetic semiconductor. These three quantum states can be obtained by the symmetry and \( k \cdot p \) model analysis, which can be implemented in the 2D materials ScLiCl\(_5\), LiScZ\(_5\) (Z=Cl, Br) and ScLiBr\(_5\). It is shown that the ferromagnetism in these 2D materials is attributed to \( p \) orbitals. Our findings provide a new mechanism of magnetic topological states from \( p \)-orbital electrons on square lattices, and also present a novel family of 2D magnetic topological materials with high Chern number.
Figure 1. The square lattice with three magnetic topological states. (a) Schematic illustration of the double degeneracy at high-symmetry points (red dots). (b) Schematic depiction of hourglass dispersion along $\Gamma - X$ and $\Gamma - M$ high-symmetry lines. The labels indicate the eigenvalues of $\tilde{M}$. (c) Schematic phase diagram with respect to the parameters $\eta$ and $\beta/b$ in (1), where yellow, red and green regions represent topological semimetal, topologically trivial ferromagnetic semiconductor and QAHE states, respectively.

### THREE MAGNETIC TOPOLOGICAL STATES ON SQUARE LATTICES

Let us consider a square lattice with the space group of $P/4n$ (No. 85) for $p$ orbitals. There are three generators: four-fold rotation symmetry $C_4: (x, y, z) \rightarrow (y, -x, z)$, roto-inversion symmetry $\tilde{I}: (x, y, z) \rightarrow (\frac{1}{2} - x, \frac{1}{2} - y, -z)$ and glide mirror symmetry $\tilde{M}_z: (x, y, z) \rightarrow (\frac{1}{2} + x, \frac{1}{2} + y, -z)$. The symmetry-protected double degeneracy appears at high-symmetry points in the absence of spin-orbit coupling (SOC), as shown in Fig. 1(a). Without SOC, the spin and orbital parts of the electronic wave functions are decoupled, and hence all crystalline symmetries are preserved for each spin channel separately like spinless particles. For the whole Brillouin zone (BZ), the glide mirror $\tilde{M}_z$ is preserved. The high-symmetry points $\Gamma, X, Y$ and $M$ are invariant under the combined operation $T\tilde{M}_z$. We note that $(T\tilde{M}_z)^2 = T_{110}^2$, where $T^2 = 1$ for the spinless case, and $T_{110} = e^{-i\phi}$. The translation by one unit cell along the [110] direction.

At point $X$, we have $k_x = \pi$ and $k_y = 0$, while at point $Y$, we have $k_x = 0$ and $k_y = \pi$. Consequently, $(T\tilde{M}_z)^2 = -1$ for points $X$ and $Y$. This antunitary operator thus generates a Kramers-like double degeneracy at points $X$ and $Y$. One may note that point $M$ is invariant under both $C_4$ and $\tilde{I}$. The commutation relation between $C_4$ and $\tilde{I}$ is given by $C_4\tilde{I} = T_{0\pi}T^2\tilde{I}C_4$, where $T_{0\pi} = e^{i\phi}$. At point $M$, we have $k_x = \pi$ and $k_y = \pi$; hence, $T_{0\pi} = -1$. As a result, for any energy eigenstate $|u\rangle$ with $C_4$ eigenvalue $E_z$, it must have a degenerate partner $\tilde{I}|u\rangle$ with $C_4$ eigenvalue $-E_z$. This proves that the double degeneracy at $M$ is guaranteed by the symmetry. At point $\Gamma$, $p_x$ and $p_y$ orbitals should be degenerate, while $p_z$ orbitals are not.

Now we turn to discuss the possible band crossing at high-symmetry lines. Consider the $\Gamma - X$ and $\Gamma - M$ lines, which are invariant under $\tilde{M}_z$. The Bloch states along $\Gamma - X$ can be chosen as the eigenstates of $\tilde{M}_z$ with eigenvalues $E_z = \pm e^{-i\phi}$. The glide eigenvalues are $\pm 1$ at $X$ and $\pm 1$ at $\Gamma$. Because $\Gamma$ and $X$ are both time-reversal-invariant momenta, a Kramers pair has eigenvalues $(+i, -i)$ at $X$, and yet it has $(+1, +1)$ or $(-1, -1)$ at $\Gamma$. This suggests that the pairs must switch partners when going from $X$ to $\Gamma$, and the switching leads to the hourglass-type dispersion, as schematically shown in Fig. 1(b). A similar analysis applied for path $\Gamma - Y$ shows that the band crossing is also hourglass-type along $\Gamma - Y$. The Bloch states along $\Gamma - M$ have the eigenvalues of $\tilde{M}_z$ with eigenvalues $E_z = \pm e^{-i\phi}$. At the $\Gamma (0,0)$ point, the eigenvalues are $(+1, -1)$, while at the $M (\pi, \pi)$ point the eigenvalues are $(-1, +1)$. Focusing on the middle of the two bands, they have opposite eigenvalues, and their ordering is inverted between $\Gamma$ and $M$. As a result, there must be a cross along the $\Gamma - M$ path, and the crossing point is protected by $\tilde{M}_z$ [Fig. 1(b)]. Because $\Gamma$ and $M$ are both time-reversal-invariant momenta, a Kramers pair with eigenvalues $+1$ (or $-1$) is degenerate, leading to the crossing point of hourglass type. The crossing point may trace out a nodal loop centered at $\Gamma$.

To characterize the above discussed band crossing, we construct an effective $k \cdot p$ model for the low-energy band structure on a square lattice. We first consider the case without SOC. The four states at point $\Gamma$ correspond to two 2D irreducible representations $E_a$ and $E_c$. The model should respect the following symmetries: the four-fold rotation $C_{4z}$ and the mirror symmetry $M_z$. Expanding up to the $k$ quadratic order, we find that the effective Hamiltonian takes the form

$$H_0 = \begin{pmatrix} a(k) & -ic(k) & 0 & 0 \\ ic(k) & a(k) & 0 & 0 \\ 0 & 0 & b(k) & -id(k) \\ 0 & 0 & id(k) & b(k) \end{pmatrix},$$

where $a(k) = a_1 + a_2k_x^2, b(k) = b_1 + b_2k_x^2, c(k) = a_3 + a_4k_x^2, d(k) = b_3 + b_4k_x^2, k_x^2 = k_x^2 + k_y^2$ and the parameters $a_i$ and $b_i$ ($i = 1, 2, 3, 4$) are real. Considering a fully spin-polarized ferromagnet, the inclusion of SOC gives an additional contribution to the above Hamiltonian, which can be treated as a perturbation due to the relatively weak SOC strength. We
The structure of the ScLiCl$_5$ monolayer with a square lattice is depicted in Fig. 2(a). Each primitive cell contains two formula units of XYZ$_5$. To confirm the stability of the ScLiCl$_5$ monolayer, its phonon spectra have been calculated. There is no imaginary frequency mode in the whole Brillouin zone, indicating that this monolayer is dynamically stable. The structural stability of the ScLiCl$_5$ monolayer is also examined in terms of the formation energy. The obtained negative values of the formation energy (the energy difference between XYZ$_5$ and X, Y crystals, $\frac{5}{2} Z_2$ molecule) for XYZ$_5$ monolayers are indicative of an exothermic reaction. Moreover, the thermal stability of the ScLiCl$_5$ monolayer is tested using molecular dynamics simulation by considering a $3 \times 3 \times 1$ supercell of ScLiCl$_5$ with 126 atoms. After being heated at 300 K for 6 ps with a time step of 3 fs, no structural changes occur, indicating that this monolayer is also thermodynamically stable. More details can be found in the online supplementary material.
The optimized lattice constant of the ScLiCl$_5$ monolayer is $d_0 = 7.9372$ Å. By a spin-polarized calculation, we note that the total spin magnetic moment carried by ScLiCl$_5$ is about 1.53 $\mu_B$ per unit cell, which is mainly attributed to the two Cl atoms at Wyckoff position 2$c$ [colored red in Fig. 2(a)], whose spin and orbital moments are of about 0.50 and 0.16 $\mu_B$ per atom, respectively. The ferromagnetism mainly originates from the $p$ orbitals of Cl atoms, whereas the spin magnetic moment on the Sc atom is calculated to be zero. It is well interpreted that, for ScLiCl$_5$, because Li and Sc have one and three valence electrons, respectively, Cl atoms possess unpaired electrons and thus should carry a non-zero spin magnetic moment. To determine the magnetic ground state, we compared the total energies between ferromagnetic (FM), antiferromagnetic (AFM) and non-magnetic (NM) states. The FM state is found to be more stable than the AFM and NM states.

The partial density of states (PDOS) and electronic band structure in the absence of SOC are shown in Fig. 2(b) and (c), respectively. One observes that the material is a half-metal, with only one spin channel (spin down) being metallic and another spin channel (spin up) being insulating. From the projected density of states as displayed in Fig. 2(b), one may see that the states around the Fermi energy are fully polarized in the spin-down channel, while the spin-up channel has a large gap. In addition, the low-energy states are dominated by the $p_{x,y}$ orbitals of the Cl atoms at Wyckoff position 2$c$. From Fig. 2(c), we find two features of the band structure. One is the double degeneracy at high-symmetry points $\Gamma$, $X$ and $M$, and another is the linear band-crossing points appearing on the paths $\Gamma - X$ and $\Gamma - M$. These crossing points are not isolated, and form a nodal loop around the $\Gamma$ point as shown in Fig. 2(d).

In the presence of SOC, magnetic anisotropy should be considered. In order to determine the easy axis of magnetization, we pin down the magnetization direction for FM configurations. By comparing the energies of different magnetization directions, we find that the out-of-plane direction is energetically preferred over the in-plane directions and along that are isotropic. We have also estimated the Curie temperature $T_C$ for the FM state by using the Monte Carlo simulation based on an effective Hamiltonian

$$H_{pin} = \sum_{\langle i, j \rangle} J_1 S_i^z S_j^z + \sum_{\langle i, j \rangle} J_2 S_i^z S_j^z,$$

(2)

where the spin vectors are normalized, the superscripts $i$ and $j$ label the $2c$ Cl sites, $\langle i, j \rangle$ and $\langle (i, j) \rangle$ indicate nearest-neighboring and next-nearest-neighboring sites, respectively, and $J_1$ and $J_2$ are the corresponding FM exchange integrals. The values of $J_1$ and $J_2$ extracted from DFT integrals are $-4.572$ and $-0.161$ meV, respectively. The calculated Curie temperature for monolayer ScLiCl$_5$ is about 123 K.

The electronic band structure with SOC for the ScLiCl$_5$ monolayer is shown in Fig. 2(e). Note that the SOC only breaks the degeneracy at point $\Gamma$, but keeps the degeneracies at points $X(Y)$ and $M$, which are protected by symmetry. The nodal loop is also preserved with SOC in monolayer ScLiCl$_5$, as shown in Fig. 2(f). Thus, the ScLiCl$_5$ monolayer exhibits a fully spin-polarized nodal loop semimetal (we could call it the nodal loop half-semimetal). By fitting the two bands near the Fermi level, we can obtain the parameters for ScLiCl$_5$: $\beta/a_1 = -1.4$ and $(\gamma + \eta)/a_1 = 0$. Thus, ScLiCl$_5$ locates in the region of the topological semimetal marked by a star in Fig. 1(c).

### Ferromagnetic semiconductor in ScLiBr$_5$

The ScLiBr$_5$ monolayer shares the same structure as ScLiCl$_5$, except a larger lattice constant $a = 8.4175$ Å. The stability of monolayer ScLiBr$_5$ is also checked by its phonon spectra, molecular dynamics and formation energy, indicating it is feasible in experiment (see the online supplementary material). The spin-polarized calculation shows that the total spin magnetic moment of ScLiBr$_5$ is about $1.43 \mu_B$ per unit cell, and the two Br atoms at Wyckoff position 2$c$ possess spin and orbital magnetic moments of about 0.46 and 0.34 $\mu_B$ per atom, respectively, whereas spin and orbital magnetic moments on other atoms are negligible. By comparing the total energies between the FM, AFM and NM states, the FM state is found to be more stable than the antiferromagnetic and non-magnetic states. The ScLiBr$_5$ monolayer has a similar band structure in the presence of SOC, as shown in Fig. 3(a). In this case, the ScLiBr$_5$ monolayer possesses a ferromagnetic ground state with out-of-plane magnetization, and the Curie temperature was estimated to be 67 K by Monte Carlo simulation based on (2) with $J_1 = -1.990$ meV and $J_2 = -0.455$ meV. The band gap of about 60.2 meV is opened by SOC for monolayer ScLiBr$_5$, and it turns into a topologically trivial ferromagnetic semiconductor state with a zero Chern number. By fitting the two bands near the Fermi level, we can obtain the parameters for ScLiBr$_5$: $\beta/a_1 = -2.3$ and $(\gamma + \eta)/a_1 = -4.6$, which locates in the region of the ferromagnetic semiconductor marked by a dot in Fig. 1(c).
The electronic band structure and density of states for monolayer LiScCl$_5$ in the absence of SOC are shown in Fig. 4(a) and (b), respectively. It can be seen that monolayer LiScCl$_5$ holds a similar band structure as ScLiCl$_5$ in the presence of SOC except that the band crossing points are closer to the $\Gamma$ point. From the projected density of states as displayed in Fig. 4(b), we may observe that the states around the Fermi energy are fully polarized in the spin-down channel, while the spin-up channel has a large gap. The low-energy states are dominated by the $p$ orbitals of all Cl atoms.

In the presence of SOC, magnetic anisotropy should be considered. By comparing the energies of different magnetization directions, we uncover that the out-of-plane direction is energetically preferred over the in-plane directions, and the energy of out-of-plane magnetization is 1.10 meV lower than that of in-plane magnetization. The Curie temperature is estimated to be 28 K. After tuning on SOC, a gap of about 24.7 meV is opened, as shown in Fig. 4(c).

The topologically non-trivial band structure of the LiScCl$_5$ monolayer is characterized by a non-zero Chern number $C = 2$ with a quantized charge Hall plateau $2e^2/h$ and two gapless chiral edge states connecting the valence and conduction bands, as shown in Fig. 4(d) and (e), respectively. In addition to the QAHE with high Chern number, the magneto-optical Kerr effect, being a kind of non-contact (non-damaging) optical technique, is a powerful tool for measuring the magnetism in 2D materials [11,34]. It can be seen that a large Kerr angle $\theta_{\text{Kerr}}$ is obtained for the LiScCl$_5$ monolayer, particularly for photon energies $\omega$ near 1.7 eV, as shown in Fig. 4(f).

The maximal Kerr angle for the LiScCl$_5$ monolayer is an order of magnitude larger than that for the CrGeTe$_3$ monolayer [34], and about 3 times larger than that for bulk Fe [35]. By fitting the two bands near the Fermi level, we can obtain the parameters for LiScCl$_5$: $\beta/a_1 = -2.0$ and $(\gamma + \eta)/a_1 = -1.9$, which locates in the region of the QAHE marked by a triangle in Fig. 1(c). The high-Chern-number QAHE with a large band gap of about 113 meV can be also implemented in monolayer LiScBr$_5$ (see the online supplementary material).

QAHE in LiScX$_5$ (X=Cl, Br)

The LiScX$_5$ (X=Cl, Br) monolayer can be obtained by exchanging the positions of the Li and Sc atoms in the structures of ScLiCl$_5$ and ScLiBr$_5$ [Fig. 2(a)]. We take LiScCl$_5$ as a prototypical example because LiScBr$_5$ shares very similar features. The optimized lattice constant of the LiScCl$_5$ monolayer is $a_0 = 8.0048$ Å. The stability of monolayer LiScCl$_5$ is also checked by its phonon spectra, molecular dynamics and formation energy (see the online supplementary material), indicating it is feasible in experiment. By comparing the formation energy, we find that ScLiCl$_5$ ($-1.52$ eV) is a little more stable than LiScCl$_5$ ($-1.51$ eV). By a spin-polarized calculation, we find that the spin magnetic moment carried by LiScCl$_5$ is about 1.54 $\mu_B$ per cell shared by all Cl atoms, while the Cl atoms at Wyckoff position 2c have orbital moment (0.03 $\mu_B$) much larger than that (0.01 $\mu_B$) for the Cl atoms at Wyckoff position 8g. Our calculated results show that the FM state is more stable than the antiferromagnetic and non-magnetic states.

The electronic band structure and density of states for monolayer LiScCl$_5$ in the absence of SOC are shown in Fig. 4(a) and (b), respectively. It can be seen that monolayer LiScCl$_5$ holds a similar band structure as ScLiCl$_5$ in the presence of SOC except that the band crossing points are closer to the $\Gamma$ point. From the projected density of states as displayed in Fig. 4(b), we may observe that the states around the Fermi energy are fully polarized in the spin-down channel, while the spin-up channel has a large gap. The low-energy states are dominated by the $p$ orbitals of all Cl atoms.

In the presence of SOC, magnetic anisotropy should be considered. By comparing the energies of different magnetization directions, we uncover that the out-of-plane direction is energetically preferred over the in-plane directions, and the energy of out-of-plane magnetization is 1.10 meV lower than that of in-plane magnetization. The Curie temperature is estimated to be 28 K. After tuning on SOC, a gap of about 24.7 meV is opened, as shown in Fig. 4(c).

The topologically non-trivial band structure of the LiScCl$_5$ monolayer is characterized by a non-zero Chern number $C = 2$ with a quantized charge Hall plateau $2e^2/h$ and two gapless chiral edge states connecting the valence and conduction bands, as shown in Fig. 4(d) and (e), respectively. In addition to the QAHE with high Chern number, the magneto-optical Kerr effect, being a kind of non-contact (non-damaging) optical technique, is a powerful tool for measuring the magnetism in 2D materials [11,34]. It can be seen that a large Kerr angle $\theta_{\text{Kerr}}$ is obtained for the LiScCl$_5$ monolayer, particularly for photon energies $\omega$ near 1.7 eV, as shown in Fig. 4(f).

The maximal Kerr angle for the LiScCl$_5$ monolayer is an order of magnitude larger than that for the CrGeTe$_3$ monolayer [34], and about 3 times larger than that for bulk Fe [35]. By fitting the two bands near the Fermi level, we can obtain the parameters for LiScCl$_5$: $\beta/a_1 = -2.0$ and $(\gamma + \eta)/a_1 = -1.9$, which locates in the region of the QAHE marked by a triangle in Fig. 1(c). The high-Chern-number QAHE with a large band gap of about 113 meV can be also implemented in monolayer LiScBr$_5$ (see the online supplementary material).
Table 1. The differences in matrix elements squared between two directions of the magnetization \( \left| \langle \sigma^+ | L_z | \sigma^+ \rangle \right|^2 - \left| \langle \sigma^- | L_z | \sigma^- \rangle \right|^2 \) and \( \left| \langle \sigma^+ | L_z | \sigma^- \rangle \right|^2 - \left| \langle \sigma^- | L_z | \sigma^+ \rangle \right|^2 \) in (3), where \( \sigma \) and \( u \) are occupied and unoccupied orbitals, and + and − are majority and minority spin states, respectively.

| \( P_{u,+} \) | \( P_{u,+} \) | \( P_{u,+} \) | \( P_{u,-} \) | \( P_{u,-} \) | \( P_{u,-} \) |
|----------------|----------------|----------------|----------------|----------------|----------------|
| 1             | 1             | 0             | 0             | 0             | 0             |
| 0             | 0             | 0             | 0             | 0             | 0             |
| 1             | 1             | 0             | 0             | 0             | 0             |
| 0             | 0             | 0             | 0             | 0             | 0             |

Table 2. Orbital-resolved magnetic single-ion anisotropic energy \( E_{SIA} \) of Cl atoms in the fully spin-polarized nodal loop semimetal ScLiCl\(_5\), where the dominated \( E_{SIA} \) comes from the \( (p_x, p_y) \) or \( (p_x, p_z) \) matrix element of the Cl atoms at Wyckoff position 2c.

| Cl | \( (p_x, p_y) \) | \( (p_x, p_z) \) | \( (p_y, p_z) \) |
|----|----------------|----------------|----------------|
| 2c | 3.08           | 0.10           | -0.44          |
| 8g | -0.05          | 0.00           | -0.01          |

Table 3. The spin \( \langle S \rangle \) and orbital \( \langle O \rangle \) moments (in \( \mu_B \)) of \( Z \) (or Cr) atoms, MAE (in meV) per formula unit between the out-of-plane and in-plane FM configurations and the Curie temperature \( T_C \) (in kelvins) for \( XYZ_5 \) compounds as well as CrGeT\(_3\) [38] for comparison.

| Wyckoff position | \( \langle S \rangle \) | \( \langle O \rangle \) | MAE | \( T_C \) |
|------------------|----------------|----------------|-----|--------|
| ScLiCl\(_5\) 2c   | 0.497          | 0.159          | 3.512 | 123    |
| 8g              | 0.066          | 0.002          |      |        |
| ScLiBr\(_5\) 2c   | 0.460          | 0.337          | 7.919 | 67     |
| 8g              | 0.063          | 0.002          |      |        |
| LiScCl\(_3\) 2c   | 0.136          | 0.028          | 0.504 | 28     |
| 8g              | 0.162          | 0.006          |      |        |
| LiScBr\(_3\) 2c   | 0.166          | 0.069          | 5.418 | 20     |
| 8g              | 0.142          | 0.020          |      |        |
| CrGeTe\(_3\) 2c   | 3.614          | 0.004          | 1.850 | 19     |

MAGNETIC SINGLE-ION ANISOTROPY

We take ScLiCl\(_5\) as an example to discuss the microscopic mechanism of the out-of-plane magnetization and large magnetic anisotropy.

According to second-order perturbation theory, magnetic anisotropy from single-ion anisotropy (SIA) can be described as [36,37]

\[
E_{SIA} = \lambda \sum \frac{\left| \langle \psi_u | L_z | \psi_u \rangle \right|^2 - \left| \langle \psi_u | L_z | \psi_u \rangle \right|^2}{\epsilon_u - \epsilon_o},
\]

where \( \lambda \) is the SOC constant, \( L_{z/u} \) represent the angular momentum operators, and \( \epsilon_u \) and \( \epsilon_o \) are the unoccupied and occupied energies, respectively. A positive value of \( E_{SIA} \) indicates the out-of-plane magnetization, and the in-plane magnetization otherwise. Equation (3) means that the orbitals near the Fermi energy mainly contribute to the magnetic anisotropy energy (MAE). By calculating the differences in matrix elements squared between two directions of the magnetization for \( p \) orbitals according to (3), as shown in Table 1, we note that the contributions to MAE from the same spins and opposite spins between occupied \( \langle | \psi_u \rangle \) and unoccupied \( \langle | \psi_u \rangle \) states have opposite signs. Positive and negative matrix elements prefer the out-of-plane magnetization and in-plane magnetization, respectively. In our systems, the states near the Fermi energy are mainly contributed by the same spin (spin down) of \( p_{u,0} \) orbitals, which should prefer an out-of-plane magnetization.

To confirm the above observation, the orbital-resolved \( E_{SIA} \) was calculated for the ScLiCl\(_5\) monolayer as listed in Table 2. It is seen that the Li and Sc atoms as well as the Cl atoms at Wyckoff position 8g make no contribution to \( E_{SIA} \), while the main contribution comes from the Cl atoms at Wyckoff position 2c, as revealed in Table 2. The value of the \( (p_x, p_y) \) matrix element is dominated and positive, indicating an out-of-plane magnetization, which is consistent with the above analysis. For monolayers ScLiBr\(_5\) and LiScX\(_3\) (\( X=\text{Cl}, \text{Br} \)), the same analysis applies and we find that they all prefer the out-of-plane magnetization, which is consistent with our DFT results.

The atomic SOC is calculated as \( H_{SOC} = \lambda |S| \cdot L \), where \( \lambda \) related to the atomic number is the coefficient of SOC and \( S \) and \( L \) represent the spin and orbital angular momentum operators, respectively. Although \( \lambda \) is small for the Cl atom (\( \sim 331 \text{ cm}^{-1} \)) [39], its orbital moment is large (\( \sim 0.16 \mu_B \)) as shown in Table 3, while in many \( d \)-orbital magnetic materials though their \( \lambda \) is much larger than that of the Cl atom, their orbital magnetic moments are quenched, and thus in our compounds, the atomic SOC of \( p \) orbitals is large and opens a gap of tens of meV, which is comparable with and even larger than many \( d \)-orbital magnetic materials. For example, for monolayer CrGeTe\(_3\), its \( \lambda \) is \( 740 \text{ cm}^{-1} \) [39], while its orbital moment is quenched (\( \sim 0.004 \mu_B \)) [38], leading to a small SOC. Because of the large SOC in our compounds, the obtained single-ion anisotropy is large.

From Table 3, we note that the estimated Curie temperature for our compounds is higher than that of CrGeTe\(_3\), especially for ScLiCl\(_5\) and ScLiBr\(_5\). Moreover, for ScLiCl\(_5\), ScLiBr\(_5\) and LiScBr\(_5\), their magnetic anisotropies are much higher than CrGeTe\(_3\) because the former has larger orbital...
magnetic momenta. Thus, the $p$-orbital magnetism in our compounds is reliable and stable.

CONCLUSION

In this work, we propose the $p$-orbital topological magnetic states on a square lattice with space group $P/4n$ by means of the symmetries and $k \cdot p$ model analyses that are material independent. Three currently interested topological states, including topological semimetal, QAHE and the topologically trivial ferromagnetic semiconductor, can be obtained on the square lattice, depending on the interplay between different SOC parameters. A phase diagram is presented. As examples, we show that the above three different topological states can indeed be implemented in the 2D materials ScLiCl$_2$, ScLiBr$_3$ and LiScCl$_3$ (or LiScBr$_3$), respectively. Furthermore, the ferromagnetism of these 2D ferromagnets is unveiled from the $p$ orbitals of halogen elements, and the microscopic origin of ferromagnetism from $p$ electrons is elaborated. This present study opens a door to explore not only exotic topological states (e.g. nodal loop half-semimetal), but also the quantum magnetism from $p$-orbital electrons in terms of the model and material-independent analyses.

SUPPLEMENTARY DATA

Supplementary data are available at NSR online.

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AUTHOR CONTRIBUTIONS

J.Y.Y., B.G. and G.S. designed the research. J.Y.Y. did the calculation and wrote the paper. All authors discussed the results and revised the paper.

Conflict of interest statement. None declared.

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