Unique surface-state connection between Weyl and nodal ring fermions in ferromagnetic material Cs_2MoCl_6

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For topological materials with a coexistence of Weyl nodes and nodal rings, their unique surface-state configuration and connection still need to be studied and discussed. In this Letter, we predict a ferromagnetic (FM) material, Cs_2MoCl_6 , with a coexistence of Weyl and nodal ring fermions in its spinful FM electronic band structure, which is unusual since FM materials are very rare in nature and nodal ring band crossings will usually open a gap when spin-orbit coupling is taken into consideration. We find that the surface states of Cs_2MoCl_6 show different properties along different directions, i.e., the surface states are in a drumhead shape showing the nodal ring property on the (001) surface and in a helicoid shape showing the Weyl property on the (010) surface. Interestingly, both the drumhead surface states and the helicoid surface states will cross the projected points of the Weyl and nodal ring along different directions. In particular, helicoid surface states on the (010) surface will meet the nodal ring tangentially, with their shapes changing abruptly as a function of the energy. We implement both first-principles calculation and an analytical model to understand the unique surface-state connection for systems with a coexistence of Weyl nodes and nodal rings (or nodal lines). This result is universal and irrespective of the presence or absence of time-reversal symmetry ($\mathcal{T}$).

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Introduction. In the recent years, topological magnetic materials [1–17] have drawn much attention due to their exotic properties, e.g., topological axion insulator states [18–20], an intrinsic quantum anomalous Hall effect without external magnetic fields [6,7,9,11–13], and the display of magnetism, topology, spin dynamics, and quantum transport. However, experimentally synthesized and verified magnetic topological materials are very limited, especially for magnetic topological semimetals [8,10–16]. Furthermore, compared to the bulk topology, the properties of topological surface states such as the configuration and connection are not so well understood in some systems. In this Letter, we propose a “high-quality” ferromagnetic (FM) material Cs_2MoCl_6 [21] with space group $Fm\bar{3}m$ that has both Weyl and nodal ring fermions in its spinful electronic band structure. We find a unique surface-state connection between the Weyl and nodal ring fermions along different surface directions by both density functional theory (DFT) and model analysis due to an intriguing interplay between drumhead surface states (DSSs) from the nodal rings and helicoid surface states (HSSs), i.e., Fermi-arc surface states, from the Weyl nodes. We show that the DSSs always go across the Weyl nodes, when the projection of the Weyl nodes is inside that of the nodal ring. We also

show that how the HSSs connect the projection of the nodal ring and the curvature of HSSs depends sensitively on the energy. Such a unique surface-state connection also appears in several previous studies, yet lacks discussion [6,22–25]. Our results show that the connection pattern of the surface states is surface dependent for systems with both Weyl and nodal ring fermions, and it is universal for systems with or without considering spin-orbit coupling (SOC) and time-reversal ($\mathcal{T}$) symmetry.

Crystal and magnetic structure for Cs_2MoCl_6 . Cs_2MoCl_6 belongs to space group No. 225, and the lattice constant we use in this Letter is $a = 10.57 \text{ \AA}$ after a relaxation on the atomic positions, which is slightly different from the experimental data $10.21 \text{ \AA}$ at room temperature [21]. For most of the compounds having a similar crystal structure as Cs_2MoCl_6 (e.g., replacing Cs, Mo, and Cl by other alkali metal elements, transition metal elements, and halogen elements, respectively), their magnetic properties have yet to be explored by experiments except for Cs_2CoF_6 and Rb_2CoF_6 , which have a FM phase at low temperature [26], and Cs_2MoCl_6 is also among the ones lacking exploration.

In order to explore the magnetic structure of the ground state in Cs_2MoCl_6 , we calculate the total energy of the systems with three different magnetic structures for No. 225 by DFT, which are shown in Figs. 1(c)–1(e). The total energy for the FM phase in Figs. 1(c) and 1(d) is about 20 meV lower than the antiferromagnetic phase in Fig. 1(e) per unit cell, and the FM phase with magnetic order on the (001) surface [Fig. 1(c)] will have a smaller total energy of about 1 meV per unit cell than that along the [111] direction [Fig. 1(d)]. Thus, we will choose the one with a magnetic moment

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FIG. 1. Crystal structure, Brillouin zone (BZ), and splitting of d orbitals of Cs_2MoCl_6 . (a) and (b) are the crystal structure and (surface) Brillouin zone of Cs_2MoCl_6 . (c)–(e) are three different types of magnetic structures of Cs_2MoCl_6 with a ferromagnetic phase along the [001] direction, [111] direction, and antiferromagnetic phase, respectively. (f) shows the splitting of d orbitals under different fields.

along the highest-symmetry direction, i.e., the [001] direction, to explore the topological properties in Cs_2MoCl_6 . In the following, we will introduce the topology of the electronic band structure of Cs_2MoCl_6 both in the nonmagnetic (NM) phase and in the FM phase separately.

Band structure of Cs_2MoCl_6 . In the spinless NM electronic band structure, d orbitals from Mo will split into e_g and t_{2g} orbitals due to the octahedral crystal field environment, and the splitting is very huge due to the large p - d hybridization of Mo and Cl, as shown in Figs. 1(f) and 2(a). Thus, we only need to study the t_{2g} orbitals located near the Fermi energy, and they exhibit nontrivial topologies, i.e., nodal chains formed by the two lowest t_{2g} orbitals crossing the Fermi level. Due to the existence of mirror symmetries,

FIG. 2. Spinless electronic band structure and the nodal chain distribution of Cs_2MoCl_6 in the NM phase. (a) is the spinless electronic band structure of Cs_2MoCl_6 in the NM phase, where the t_{2g} orbits are magnified in (b). (c) shows the nodal chain band crossing formed by the lowest two bands of t_{2g} orbits.

FIG. 3. Spinful electronic band structure and topological calculations on Cs_2MoCl_6 in the FM phase. (a) is the spinful electronic band structure of Cs_2MoCl_6 in the FM phase with a magnetic moment along the [001] direction. (b) shows the positions of a pair of Weyl points (red and green hexagons) and nodal rings in the bulk, (001), and (010) surface BZ. (c) Schematic figure of the projected positions of the nodal ring and Weyl points on the (001) and (010) surfaces. (d) shows the surface states, together with the projection of the Weyl points (red and green hexagons) and the nodal ring (red circles) on the (001) surface. (e) shows the real and imaginary parts for λ_+ obtained by the $k \cdot p$ model with parameters of $\tilde{D} = 1$ and $\sqrt{m} = 5$, where $\text{Im}(\lambda_-) = -\text{Im}(\lambda_+)$. (f) is the absolute value of the real part $|\text{Re}(\lambda)|$ and the imaginary part $\text{Im}(\lambda)$ obtained by the DFT calculation, where the k points are labeled in (d). (g) and (h) are the DSSs with different energies labeled in (d), where E_1 corresponds to the energy of nodal rings.

all the nodal rings will be pinned on the M_x , M_y , and M_z planes, and meet with each other at the high-symmetry point W , forming a nodal chain semimetal, as shown in Figs. 2(b) and 2(c).

Figure 3(a) show the spinful electronic band structure of Cs_2MoCl_6 with the FM phase by the DFT+ U method [27], and the Hubbard U term for Mo is 0.33 eV obtained by the linear response method [28]. After considering the FM structure with a magnetic moment on the (001) surface and SOC, M_z and C_{4z} will still be preserved, while $M_{x,y}$ and $C_{4x,4y}$ will be broken. Thus, the nodal chain on the $M_{x,y}$ plane will open a gap but still leave a single nodal ring protected by the M_z symmetry. However, the nodal ring will move slightly away from the W point due to the symmetry breaking, and cross the high-symmetry line W_1 - W_2 as shown in Fig. 3(b). Furthermore, SOC will make a change on the band structure along the Γ - Z direction, and generate a pair of Weyl points protected by C_{4z} symmetry, as shown by the red and green hexagons in Fig. 3(b). Therefore, the t_{2g} bands will be

significantly affected after considering the FM order and SOC, which make the nodal chains into a pair of Weyl points and a single nodal ring, as shown in Fig. 3(b).

k · p model analysis. Since two bands forming the Weyl points and a nodal ring are well separated from other bands, an effective 2×2 Hamiltonian based on the basis set $\{d_{xz}, d_{yz}\}$ can be constructed to describe the topological and other physical properties of the system. As discussed above, SOC and FM order will break the spin-rotation symmetry, $M_{x,y}$ and $C_{4x,4y}$ of the system, reducing the symmetry of the system to C_{4h} . Furthermore, the eigenvalues of the $\hat{C}_{4z}$ operator for those two bands along the Γ -Z direction are $e^{-\frac{\pi i}{4}}$ and $e^{-\frac{3\pi i}{4}}$, so the $k \cdot p$ Hamiltonian describing the system around Z with C_{4h} double group symmetry can be written to the lowest order in $\mathbf{k}$ as

$$H(k_x, k_y, k_z) = \begin{pmatrix} m - |\mathbf{k}|^2 & \tilde{D}k_z k_+ \\ \tilde{D}k_z k_- & -m + |\mathbf{k}|^2 \end{pmatrix}, \quad (1)$$

where $k_{\pm} = k_x \pm ik_y$ and $\mathbf{k}$ is a wave vector measured from the Z point. $\tilde{D}$ and m are positive constants. We derived this Hamiltonian from the irreducible representations of the two bands around Z after proper rescaling of the wave vector (see the Supplemental Material for details [29]). The eigenvalues of H are $E = \pm \sqrt{(m - |\mathbf{k}|^2)^2 + \tilde{D}^2 k_z^2 (k_x^2 + k_y^2)}$, which form a pair of Weyl points and a single nodal ring located at $(k_x = 0, k_y = 0, k_z = \pm \sqrt{m})$ and $(k_x^2 + k_y^2 = m, k_z = 0)$ with $E = 0$, respectively, as shown in Fig. 3(c).

As you may notice, the projections of topological band crossings on different surfaces will have different configurations. Namely, the two Weyl points will be projected inside of the nodal ring on the (001) surface, and to different sides of the projection of the nodal ring on the (010) surface [Fig. 3(c)]. This difference will lead to different types of surface-state connections, which will be explored by both DFT calculation and model analysis in the following. Topological surface states obtained by DFT are calculated by the Green's function method [30,31] with the tight-binding Hamiltonian obtained from maximally localized Wannier functions [32,33].

Surface states of FM Cs₂MoCl₆ on the (001) surface. Figure 3(d) shows the surface states on the (001) surface, where two Weyl points will be projected onto the center of the nodal ring, i.e., $\bar{\Gamma}$, as shown in Fig. 3(b). It is worth mentioning that the topological surface states as well as the bulk bands are fully spin polarized in Cs₂MoCl₆. In order to get a better understanding of the surface-state connection and configuration, we also calculate the Fermi arcs with different energies, as shown in Figs. 3(g) and 3(h). The calculations show that the surface states on the (001) surface are in a drumhead shape, which is a characteristic of the nodal ring semimetal, and they will cross the projection of the Weyl points for systems with a coexistence of Weyl points and a nodal ring. Thus, we can control the density of the surface states or the Fermi velocity of the topological surface states on the (001) surface by modulating the energy of Weyl points and nodal rings. In the following, we will also use the analytical model for a further discussion on the universal connection pattern between a pair of Weyl points and a nodal ring.

According to the $k \cdot p$ model in Eq. (1), the nodal ring will be projected onto a circle $k_x^2 + k_y^2 = m$ and two Weyl points will be projected onto the $k_x = k_y = 0$ point on the (001) surface. In order to obtain the eigenvalues and eigenvectors of the surface states, we will deal with a semi-infinite system where the bulk and the vacuum lie in $z \leq 0$ and in $z > 0$, respectively. In this case, k_z is no longer a good quantum number, so we need to replace k_z by $-i\partial_z$ [34,35] and obtain the Hamiltonian of the semi-infinite system,

$$H_z(k_x, k_y) = \begin{pmatrix} m - R^2 + \frac{\partial^2}{\partial z^2} & -i\tilde{D}Re^{i\theta} \frac{\partial}{\partial z} \\ -i\tilde{D}Re^{-i\theta} \frac{\partial}{\partial z} & -m + R^2 - \frac{\partial^2}{\partial z^2} \end{pmatrix}, \quad (2)$$

where $k_{\pm} = Re^{\pm i\theta}$ with $R (>0)$ and θ being real. By introducing a trial wave function proportional to $e^{\lambda z}$ and a parameter λ satisfying $\text{Re}(\lambda) > 0$ to represent a surface state, we obtain the corresponding eigenvectors

$$\psi_{\pm}(z) = \begin{pmatrix} i\tilde{D}Re^{i\theta} \lambda_{\pm} \\ m - R^2 + \lambda_{\pm}^2 - E \end{pmatrix} e^{\lambda_{\pm} z}, \quad (3)$$

where $\lambda_{\pm}^2 = \frac{1}{2} \tilde{D}^2 R^2 - (m - R^2) \pm \sqrt{E^2 - (m - R^2) \tilde{D}^2 R^2 + \frac{1}{4} \tilde{D}^4 R^4}$. If we impose a boundary condition $\psi(z=0) = 0$ for an eigenvector of the form $\psi = C_+ \psi_+ + C_- \psi_-$ ($C_{\pm}$: constant), we obtain dispersionless DSSs with $E = 0$, and $\lambda_{\pm}$ becomes $\lambda_{\pm} = \frac{\tilde{D}R}{2} \pm \sqrt{(\frac{\tilde{D}R}{2})^2 + R^2 - m}$ (see the Supplemental Material for details [29]). In order to meet the condition of $\text{Re}(\lambda) > 0$, R should satisfy $|R| \leq \sqrt{m}$. Therefore, the DSSs cross the projection of the Weyl points lying at $E = 0$, which match with our DFT calculations very well.

Next, we will study the penetration length for the surface states on the (001) surface, which have a unique connection between Weyl points and the nodal ring, by both the analytical model and DFT calculation separately. The penetration length l for a wave function proportional to $e^{\lambda z}$ ($\lambda = \lambda_{\pm}$) is given by $l_{\pm} = \frac{1}{\text{Re}(\lambda_{\pm})}$. Here, we consider l_- as the penetration depth because $l_- \geq l_+$. When the surface state approaches the Weyl points or the nodal ring, the penetration length l_- is expected to be divergent due to the bulk nature of these bands. Figure 3(e) shows the values of $\lambda_{\pm}$ as a function of R , which is between the projection of the Weyl points ($R = 0$) and the nodal ring ($R = \sqrt{m}$). We notice that $R = \tilde{R} = \sqrt{\frac{4m}{\tilde{D}^2 + 4}}$ is a special point in that $\lambda_+ = \lambda_-$ is satisfied and the behavior of $l_{\pm}$ changes drastically. When $R > \tilde{R}$, $\lambda_{\pm}$ are real and the surface state decays into the bulk without oscillation. On the other hand, when $R < \tilde{R}$, $\lambda_{\pm}$ are complex and the surface states show an oscillating decay into the bulk. Further calculations obtained by a 47-layer slab with the tight-binding Hamiltonian obtained from maximally localized Wannier functions also show the same trend with the model calculation for both the real and imaginary parts of λ , which are shown in Fig. 3(f).

Surface states of FM Cs₂MoCl₆ on the (010) surface. On the (010) surface, two Weyl points will be projected on the $\bar{\Gamma}$ - $\bar{Z}$ high-symmetry line, while the nodal ring will be projected to a line near $\bar{X}$ and $\bar{Z}$, as shown in Figs. 3(b) and 3(c). Figures 4(b)–4(d) show the Fermi arcs with different energies labeled in Fig. 4(a), where E_2 corresponds to the energy of the

FIG. 4. Surface-state calculation on the (010) surface for Cs_2MoCl_6 . (a) is the surface state on the (010) surface. (b)–(d) are the Fermi arcs with different energies labeled in (a), where E_2 corresponds to the energy of the nodal ring projected at $\bar{Z}$. (e)–(g) are Fermi-arc calculations by the $k \cdot p$ model with $E > 0$, $E = 0$, and $E < 0$. Parameters are $\tilde{D} = 1$ and $\sqrt{m} = 5$. The red and green hexagons represent the Weyl nodes with chiralities $+1$ (monopole) and -1 (antimonopole), respectively.

nodal ring projected at $\bar{Z}$. When the energy for the isoenergetic contour decreases from E_1 to E_3 , the Fermi arcs from the Weyl points will cross through the projected points of the nodal ring, and rotate around two Weyl points. Thus, surface states on the (010) surface are in a helicoid shape and show the feature of Weyl points, which is different with the surface states on the (001) surface showing the feature of a nodal ring.

These Fermi-arc surface states agree with expectations from bulk quantities. At the Γ point ($k_z = 0$), the Γ_6^+ ($C_4 = e^{-\frac{\pi i}{4}}$) irreducible representation has a lower energy than Γ_7^+ ($C_4 = e^{-\frac{3\pi i}{4}}$), and at the Z point ($k_z = \pi$), the Z_7^+ ($C_4 = e^{-\frac{3\pi i}{4}}$) irreducible representation has a lower energy than Z_6^+ ($C_4 = e^{-\frac{\pi i}{4}}$). From this band inversion between different C_4 eigenstates, we can derive the difference of Chern numbers $[\text{Ch}(k_z)]$ between the $k_z = 0$ and $k_z = \pi$ subspaces. From Refs. [36,37], we get $\frac{i\text{Ch}(k_z=\pi-\delta)}{i\text{Ch}(k_z=\delta)} = \frac{e^{-\frac{3\pi i}{4}}}{e^{-\frac{\pi i}{4}}} = -i$. Namely, $\text{Ch}(k_z = \pi - \delta) - \text{Ch}(k_z = \delta) \equiv -1 \pmod{4}$. Here, δ is a small constant, which is introduced to make the spectrum gapped by avoiding the nodal ring. This difference of Chern numbers is caused by the Weyl nodes at $\mathbf{k} = (0, 0, \pm k_0)$ ($k_0 > 0$), with a monopole charge ∓ 1 , respectively. In the present case, $\text{Ch}(k_z) = 0$ for $0 < |k_z| < k_0$ and $\text{Ch}(k_z) = -1$ for $k_0 < |k_z| < \pi$, leading to topological chiral surface states with a negative velocity v_x on the (010) surface. It perfectly agrees with Figs. 4(b)–4(d).

Further model calculations on the (010) surface also verify this point, as shown in Figs. 4(e)–4(g) (see the Supplemental Material for details [29]). The projection of the Weyl

points and the nodal ring are located at $(k_x, k_z) = (0, \pm\sqrt{m})$ and $(k_z = 0, -\sqrt{m} \leq k_x \leq \sqrt{m})$, respectively. When $E = 0$ [Fig. 4(f)], the Fermi arcs cross the projected nodal ring and connect the two Weyl points straightly; when $E > 0$ and $E < 0$ [Figs. 4(f) and 4(g)], the Fermi arcs rotate around two Weyl points and cross through the nodal rings. Remarkably, when $E \neq 0$, the Fermi arc meets the nodal ring tangentially, and the direction of the Fermi arc near the nodal ring changes abruptly across $E = 0$, as shown in Figs. 4(e)–4(g). It is due to an interplay between the helicoid nature of the surface state and the tangential property between the surface Fermi arc and a bulk Fermi surface. This abrupt change of the Fermi arc also appears in Figs. 4(b)–4(d) from the DFT. Both the DFT calculation and the model calculation on the Fermi arcs show a perfect match with each other, which proves the correctness of our theory on the unique surface-state connection.

We notice that such a unique surface-state connection behavior can be found in other materials with different symmetries, such as in the electronic band structure of FM HgCr_2Se_4 (with SOC and without $\mathcal{T}$) [6], the electronic band structure of FM double perovskites ($\text{Ba}_2\text{NaOsO}_6$, $\text{Sr}_2\text{SrOsO}_6$, $\text{Ba}_2\text{ZnReO}_6$, $\text{Ba}_2\text{MgReO}_6$, and $\text{Ba}_2\text{CdReO}_6$, with SOC and without $\mathcal{T}$) [22,23], and the phonon band structure of oxide perovskites (BaTiO_3 , PbTiO_3 , without SOC and with $\mathcal{T}$) [25]. However, none of those works discuss the connection of the unique surface states, and they mainly focus on the topology of the bulk bands. We demonstrate that both the drumhead surface states and the helicoid surface states will cross the projected points of the Weyl and nodal ring along different directions. Such a surface-state connection for systems with a coexistence of Weyl points and nodal rings (and nodal lines) is universal, and it is also applicable to systems with or without considering SOC and $\mathcal{T}$.

Conclusion. We propose a FM topological half metal Cs_2MoCl_6 with spin-polarized bulk bands and surface states near the Fermi level. Due to the coexistence of Weyl points and a nodal ring, the surface states of Cs_2MoCl_6 show different properties along different directions. On the (001) surface, the surface states are in the drumhead shape, and they always go across the Weyl nodes. On the (010) surface, the Fermi-arc surface states are in a helicoid shape, and they meet the nodal ring tangentially, with their shapes changing abruptly as a function of the energy. An analytical model calculation also matches with the DFT calculation on both surface-state connections and configurations, which helps to verify our theory on the topological surface states. Such a unique feature of the surface-state connection is universal for systems with a coexistence of Weyl points and nodal rings, irrespective of the presence or absence of time-reversal symmetry and spin-orbit coupling.

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