Two-dimensional CoSe structures: Intrinsic magnetism, nonsymmorphic magnetic nodal line, and antiferromagnetic metal state

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The interplay between magnetism, band topology, and electronic correlation in low dimensions has been a fascinating subject of research. Here, we propose two-dimensional (2D) material systems which demonstrate such an interesting interplay. Based on first-principles calculations and structural search algorithms, we identify three lowest energy 2D CoSe structures, termed as the α-, β-, and γ-CoSe. We show that α- and γ-CoSe are ferromagnetic metals. They possess rich topological band features, including the nonsymmorphic magnetic nodal line, the magnetic Weyl point, and the magnetic Weyl loop. Remarkably, all these features are robust against spin-orbit coupling. Meanwhile, β-CoSe is a rare example of a 2D antiferromagnetic metal, which is related to a Fermi surface nesting feature for its three conduction band valleys. The possible phase transitions and the experimental aspects have been discussed.

I. INTRODUCTION

Two-dimensional (2D) materials has been attracting tremendous research interest in recent years [1–4]. With a thickness of only one or a few atomic layers, 2D materials naturally enjoy the advantages of large surface-to-volume ratio, excellent mechanical flexibility, and easy tunability via applied fields or chemical functionalization. Besides, the intricate interplay between quantum effects and reduced dimensionality may give rise to a range of surprising physics, especially for 2D materials with transition metal elements. For example, it has been reported that the superconductivity in layered FeSe can be remarkably enhanced when approaching the 2D limit. Its transition temperature can be increased from 8 K for the bulk to 65–109 K for a single layer [5–10]. The recent discovery of intrinsic magnetism in 2D layers of CrI3 [11, 12], Cr2Ge2Te5 [13], and Fe3GeTe2 [14] is another example. The magnetic ordering from the transition metal d orbitals is surprisingly robust, with Curie temperatures above the liquid helium or even liquid nitrogen temperature. These fascinating findings have triggered a surge of efforts to explore novel 2D transition metal compounds.

Recently, Zhou et al. [15] successfully synthesized a new transition metal compound, the tetragonal CoSe, by using a topochemical deintercalation approach. The bulk material has a layered structure, and is isostructural to the famous FeSe superconductor. Although superconductivity has not been detected, measurements have shown that bulk CoSe is a ferromagnetic metal with in-plane magnetic moments [16–18]. Because of its layered structure, it is possible to thin down the material towards the 2D regime. Indeed, very recently, Ma et al. [19] have fabricated ultrathin CoSe nanoplates with a thickness down to 2.3 nm (about 4-5 layers), using a chemical vapor deposition method, and revealed its interesting thickness-dependent transport property. Despite these exciting progress, there are important questions awaiting to be answered. First, the monolayer CoSe has not been demonstrated yet, it is therefore natural to ask whether the tetragonal phase can be stabilized in the monolayer limit? Second, besides the tetragonal phase, is there any other competing metastable 2D structural phases? Finally, what are the special physical properties of these 2D CoSe phases?

Motivated by these questions and the experimental advances mentioned above, in this work, we theoretically explore the possible 2D CoSe structures in the monolayer limit and investigate their electronic properties. By carrying out comprehensive structural search combined with first-principles calculations, we identify the single layer of the tetragonal CoSe phase (termed as α-CoSe) as the global minimum in the 2D monolayer limit. Meanwhile, we also predict another two metastable 2D CoSe structural phases (termed as β and γ phases). These structures are illustrated in Fig. 1. We show that these monolayer structures have good dynamical and thermal stabilities. Interestingly, the different crystal structures result in different magnetic ground states: α- and γ-CoSe.
FIG. 1. Top and side views of the atomic structures of (a,b) α-CoSe, (c,d) β-CoSe, and (e,f) γ-CoSe.

respectively show out-of-plane and in-plane ferromagnetism (FM), whereas β-CoSe shows out-of-plane antiferromagnetism (AFM). From Monte-Carlo simulations, we find that their magnetic orderings are fairly robust, with estimated transition temperatures above the liquid nitrogen temperature, ranging from \( \sim 140 \text{ K} \) to \( \sim 200 \text{ K} \). More importantly, we discover in these 2D materials several unusual features. In α-CoSe, we find the existence of nodal lines around the Fermi level located at the boundary of the 2D Brillouin zone (BZ). Remarkably, these nodal lines are protected by a nonsymmorphic symmetry, and are robust against spin-orbit coupling (SOC) as well as magnetic ordering. Such a kind of nonsymmorphic magnetic nodal line has not been reported before. In β-CoSe, we find its AFM coexists with a metallic ground state, which is rather unusual. By analyzing its band structure, we connect this feature to the possible spin density wave formation associated with a Fermi surface nesting character. γ-CoSe is a ferromagnetic metal. It features a pair of 2D magnetic Weyl points below the Fermi level, protected by a twofold rotational symmetry. And when the magnetization is rotated to the \( z \) direction, the two Weyl points will evolve into a single Weyl loop, which is 100% spin polarized. Again, both the magnetic Weyl points and the magnetic Weyl loop found here are robust against SOC. Our work unveils multiple 2D structural phases for a new transition metal compound. Their rich physical properties may lead to promising spintronic and electronic applications.

II. METHOD

The 2D crystal structural search was performed by using the evolutionary algorithm implemented in the USPEX code [20–22] combined with the density functional theory (DFT) calculations using the Vienna \textit{ab-initio} simulation package (VASP) [23, 24]. In the search, the number of atoms in a unit cell was limited to 12 (i.e., six formula units), and the thickness of the 2D layer was limited to \( 4 \text{ Å} \). The single layer tetragonal CoSe structure (α-CoSe) was used as a seed in the first generation. New structures were generated by carefully designed variation operators, such as heredity and soft mutation, and were fully relaxed. The relaxed energy was used for selecting structures as parents for the next generation of structures. The whole search evolved 50 generations with 40 structures in each generation.

In our DFT calculation, the projector augmented wave method [25] was used to describe the electron-ion interactions. The generalized gradient approximation with the Perdew-Burke-Ernzerhof (PBE) realization [26] was adopted for the exchange correlation functional. To account for the important correlation effect associated with the Co-3d orbitals, we included the Hubbard \( U \) correction via the PBE+\( U \) method [27]. The \( U \) value was taken to be 4 eV according to Ref. A vacuum layer of 15 Å was added to avoid artificial interactions between periodic images. The plane wave energy cutoff was set to be 600 eV, and the BZ was sampled with Γ-centered \( k \) mesh with size of \( 15 \times 15 \times 1 \). The structures were fully optimized with the energy and force convergence criteria of \( 10^{-7} \text{ eV} \) and \( 10^{-4} \text{ eV/Å} \), respectively. Our band structure results have also been verified by using the hybrid functional (HSE06) approach [28].

The phonon spectra of the materials were calculated using the PHONOPY code through the DFPT approach [29] on a \( 4 \times 4 \times 1 \) supercell. The thermal stability was checked by performing the \textit{ab-initio} molecular dynamics (AIMD) simulations on a \( 5 \times 5 \times 1 \) supercell. In the simulation, the NVT canonical sampling was performed by integrating the equations of motion at 2 fs time intervals, and the temperature was controlled via a Nosé-Hoover thermostat. At each time step, the total energy was evaluated to an accuracy of \( 10^{-4} \text{ eV/cell} \) with a plane-wave energy cutoff of 300 eV.

III. 2D COBALT SELENIDE STRUCTURES

We have performed comprehensive global minimum structural search for 2D CoSe structures using the evolutionary algorithm. The search has been run for 50 generations. Each generated structure has been fully relaxed using VASP, and the structures have been sorted by energy. Figure 2 gives an overview of our structural search.

We find that the lowest-energy structure [α-CoSe, see Fig. 1(a,b)] is isostructural to the single layer of the bulk tetragonal CoSe synthesized in the previous exper-
FIG. 2. Overview of the structural search. The structures are ordered by their energies. The three low energy structures are highlighted.

FIG. 3. Calculated phonon spectra for (a) α-CoSe, (b) β-CoSe, and (c) γ-CoSe.

The three 2D CoSe structures are illustrated in Fig. 1. Both α- and γ-CoSe have a square type lattice, whereas β-CoSe has a hexagonal lattice. α-CoSe is isostructural to the single layer FeSe. Each Co atom is surrounded by four Se atoms, forming a tetrahedral coordination. In comparison, for γ-CoSe, the top view of the lattice [Fig. 1(e)] resembles a Lieb lattice. While the Co atoms all lie in the same atomic plane, each Se site has two atoms lying on top of each other, off the Co plane. As for β-CoSe, its top view shows a bipartite honeycomb lattice. However, each site in this honeycomb is actually occupied by a Co-Se pair lying on top of each other [see Fig. 1(c,d)]. We note that for α- and γ-CoSe, the Co-Co distance (2.307 Å for α, 2.358 Å for γ) is less than that in the Co metal (2.506 Å), whereas for β-CoSe, this distance (2.607 Å) is slightly larger. The detailed structural parameters are presented in Table I.

To assess the stability of these 2D structures, we have calculated their phonon spectra. As shown in Fig. 3, the phonon spectra exhibit no soft mode, indicating that the three structures are dynamically stable. The thermal stability is investigated by the AIMD simulations. We have performed the simulation for each of the three CoSe structures at 300 K. The result confirms that all the three lattice structures are well maintained against the thermal fluctuations at room temperature (see the Supplemental Material [30]).
TABLE I. Structural and magnetic properties of the three 2D CoSe structures. Here, the magnetic moment is the one on each Co site. The moment on the Se site is negligible.

| Structure  | Layer group | Space group | Lattice constant | Magnetism | Magnetic moment | Easy axis |
|------------|-------------|-------------|-----------------|-----------|-----------------|-----------|
| α-CoSe     | Square      | P4/nmm      | 3.74 Å          | FM        | 2.3 µB          | ⟨001⟩     |
| β-CoSe     | Hexagonal   | P̅3m1       | 3.74 Å          | AFM       | 2.2 µB          | ⟨001⟩     |
| γ-CoSe     | Square      | P4/mmm      | 3.51 Å          | FM        | 1.9 µB          | ⟨100⟩     |

IV. MAGNETIC PROPERTY

Co is a 3d transition metal element. Materials containing Co often exhibit magnetic orderings in the ground state. Hence, we need to first pin down the ground-state magnetic configuration for each of the three CoSe 2D structures.

With first-principles calculations, we have compared the energies of the different types of magnetic configurations, including the nonmagnetic (NM), the FM, and several possible AFM configurations. In the process, we have included the SOC to determine the magnetic anisotropy. The results are illustrated in Fig. 4 and presented in Table I. One observes that both α- and γ-CoSe have FM ground state; whereas β-CoSe is AFM. The magnetic moments for α- and β-CoSe prefer the out-of-plane (z) direction; whereas for γ-CoSe, the preferred direction is in-plane along the x direction. For all three magnetic structures, the magnetic moments are mainly distributed on the Co ions, with a magnitude ~ 2 µB.

We have estimated the magnetic transition temperatures for these states by performing the Monte-Carlo simulations based on a classical spin model [31]:

\[
H = - \sum_{\langle i,j \rangle} J_{ij} \mathbf{S}^i \cdot \mathbf{S}^j - K \sum_i (S_{i\alpha}^2).
\]  

(1)

Here, the spin vectors are normalized, i and j label the Co sites, the first term represents the exchange coupling between the nearest neighbors i and j, the second term represents the magnetic anisotropy, and α refers to the easy axis direction. The values of the parameters \( J_{ij} \) and K are determined from the DFT calculation (see [30] for details). From the Monte-Carlo simulations, we obtain that the Curie temperatures for α- and γ-CoSe are 143 K and 195 K, respectively; while the Neel temperature for β-CoSe is about 186 K. These values are all above the liquid nitrogen temperature, indicating that the magnetic orderings in these 2D CoSe structures are fairly robust.

V. ELECTRONIC PROPERTY

After fixing the magnetic ground states, in the following, we shall turn to the electronic band structure properties of these 2D CoSe structures.

![FIG. 5. (a) Calculated band structure and (b) PDOS for α-CoSe in the absence of SOC. In (a), the two spin channels are marked by the red and the blue colors, respectively. (c) Brillouin zone of α-CoSe with high symmetry points labeled. The red lines illustrate the nonsymmorphic magnetic nodal lines [indicated by the red arrow in (a)], and the blue circle illustrates the magnetic Weyl loop [indicated by the blue arrows in (a)].](image)

A. α-CoSe: Nonsymmorphic magnetic nodal lines

Let us first consider α-CoSe. As we have discussed above, its ground state is FM with magnetization along the z direction. Figure 5(a) shows the calculated band structure for this ground state in the absence of SOC, and Fig. 5(b) shows the corresponding spin and orbital projected density of states (DOS). One observes that the α-CoSe is a FM metal, and the low-energy bands around the Fermi level are dominated by the Co-3d orbitals. Both spin channels exist on the Fermi level. The spin polarization at \( E_F \) is about 60%.

Interestingly, one observes that the low energy bands
in the spin minority channel form a nodal line along the BZ boundary. The blue arrows indicate the magnetic nonsymmorphic magnetic nodal lines found in \(-\text{CoSe}\) here are magnetic, robust regardless of SOC, and are protected by a nonsymmorphic crystal symmetry. Such a kind of nodal lines has not been discovered before.

Remarkably, these nodal lines remain robust even when SOC is turned on. Figure 6 shows the DFT result with SOC included. Compared with Fig. 5(a), the change is small. The main difference is that some original band crossings are lifted (e.g., those on the \(\Gamma-X\) path). However, one clearly observes that the nodal lines on the BZ boundary are preserved. With SOC, although both \(T\) and \(S_{2x}\) are individually broken, \(TS_{2x}\) is preserved. Importantly, the relation in Eq. (2) remains to be valid in the presence of SOC. To see this, note that by including SOC, \(T^2\) becomes \(-1\) instead of \(+1\); meanwhile, \(S_{2x}\) also need to rotate spin by \(\pi\), such that \((S_{2x})^2\) receives an extra \(-1\) factor due to the \(2\pi\) spin rotation. Thus, the two \(-1\) factors cancel out, making the relation (2) intact. This demonstrates that the nodal lines in \(-\text{CoSe}\) are protected band features both without and with SOC.

Nodal lines have been proposed in a few 2D material examples before [34–41]. However, for most cases, the nodal lines are only stable in the absence of SOC. The exceptions are only reported very recently. For example, a so-called hourglass Weyl loop protected by a glide mirror symmetry was proposed [40, 41], but it was realized for a nonmagnetic system. Wang et al. [41] identified a magnetic nodal loop in monolayer MnN, which is protected by a symmorphic mirror symmetry. In comparison, the nodal lines found in \(-\text{CoSe}\) here are magnetic, robust regardless of SOC, and are protected by a nonsymmorphic crystal symmetry. Such a kind of nodal lines has not been discovered before.

In addition, we note that there is another Weyl loop below the Fermi level (by \(\sim 0.25\) eV) centered around the \(\Gamma\) point, as indicated by the blue arrow in Fig. 5(a). This loop is protected by the glide mirror symmetry, and the valence band maximum (VBM) at \(\Gamma\). Thus, the ground state of \(-\text{CoSe}\) is an AFM metal.

As we have analyzed in Sec. IV, \(-\text{CoSe}\) is AFM with the magnetic configuration shown in Fig. 4(b). Figure 7(a) shows its calculated band structure (SOC included). One notes that the system is a metal. The band gap closes indirectly due to the overlap between the conduction band minimum (CBM) at \(M\) and the valence band maximum (VBM) at \(\Gamma\). Thus, the ground state of \(-\text{CoSe}\) is an AFM metal.

This is a very interesting observation, because the AFM ordering is usually accompanied by an insulating state. AFM metals are quite rare, not to mention that our current example is in 2D. As pointed out in Ref. [42],

**B. \(\beta\)-CoSe: AFM metal state**

As we have analyzed in Sec. IV, \(\beta\)-CoSe is AFM with the magnetic configuration shown in Fig. 4(b). Figure 7(a) shows its calculated band structure (SOC included). One notes that the system is a metal. The band gap closes indirectly due to the overlap between the conduction band minimum (CBM) at \(M\) and the valence band maximum (VBM) at \(\Gamma\). Thus, the ground state of \(\beta\)-CoSe is an AFM metal.

This is a very interesting observation, because the AFM ordering is usually accompanied by an insulating state. AFM metals are quite rare, not to mention that our current example is in 2D. As pointed out in Ref. [42],
FIG. 7. (a) Band structure of $\beta$-CoSe with SOC included. (b) Fermi surface of $\beta$-CoSe. $n_1$ and $n_2$ are the two nesting vectors. (c) Structure of $\beta$-CoSe, with the lattice vectors $a_1$ and $a_2$ labeled.

FIG. 8. Phase diagram of $\beta$-CoSe with respect to $U$ and the lattice strain.

AFM metals are usually associated with spin density waves connected with certain Fermi surface nesting features. Indeed, the wavelength for the AFM ordering [as shown in Fig. 7(c)] is determined by the lattice vectors $a_1$ and $a_2$, with a magnitude of $a = |a_1| = |a_2|$. In Fig. 7(b), we plot the Fermi surface of $\beta$-CoSe. It has a hole pocket at $\Gamma$ and three electron pockets at the three $M$ points. One observes the nesting feature among the three electron pockets. The nesting vectors are given by $n_1$ and $n_2$. One then immediately notices that $n_i|a_i|$ ($i = 1, 2$), and $n = 2\pi/a$, where $a$ is the magnitude of the nesting vector. This suggests that the AFM ordering in $\beta$-CoSe is closely connected with its Fermi surface nesting feature. Here, it is likely that the spin density wave is not strong enough to gap the whole Fermi surface (one important factor is the existence of itinerant carriers from the VBM), so that the system remains metallic. The case is similar to that for the AFM state of metallic Cr [42].

We note that in Fig. 7(a), the lowest conduction band and the highest valence band only have a small overlap in energy. This suggests that the state could be quite sensitive to perturbations. Here, we consider two system parameters. One is the electron correlation parameter $U$ (here for the Co-3$d$ orbitals), and the other is the lattice strain. The calculated phase diagram is shown in Fig. 8. Three phases appear in the diagram: the AFM metal, the nonmagnetic metal, and the AFM insulator. With increasing $U$, the system transforms from nonmagnetic metal to AFM metal and finally to AFM insulator, reflecting the increasing importance of electron correlation effects. Increasing the lattice strain shows the similar trend, because the strain suppresses the electron kinetic energy, hence effectively enhancing the electron correlation effects. These behaviors are consistent with the typical picture based on the Hubbard model [42].

C. $\gamma$-CoSe: Weyl point versus Weyl loop

$\gamma$-CoSe has a FM ground state with in-plane magnetization (along $x$). Figure 9 shows its band structure in the absence of SOC. One observes that $\gamma$-CoSe is a FM metal. From the projected DOS, the low-energy bands are dominated by the Co-3$d$ orbitals. Meanwhile, the Se-4$p$ orbitals also give a sizable contribution. The spin polarization at the Fermi level is about 42%.

One notices that in Fig. 9(a), slightly below the Fermi level, the spin-minority bands form linear band crossings around the $M$ point. Scanning the band structure around this region shows the existence of a Weyl loop centered at $M$. As a Weyl loop, it is twofold degenerate. The loop is formed by an electron-like band and a hole-like band, hence it belongs to type-I, according to its dispersion [43].

Like most previous 2D examples, the loop is destroyed when SOC is considered. However, as shown in Fig. 10(a), although most points on the loop are gapped out, there remains a pair of Weyl points on $M$-$Y$. Note that distinct from Weyl points in 3D systems which are topologically stable, Weyl points in 2D must require extra symmetry protections. We find that the key symmetry here is the $C_{2x}$ symmetry, i.e., the twofold rotation along the magnetization direction. The two Weyl points are protected, as the two crossing bands have opposite $C_{2x}$ eigenvalues along $M$-$Y$. Stable Weyl points in 2D under
FIG. 9. (a) Band structure and (b) PDOS of $\gamma$-CoSe in the absence of SOC. In (a), the two spin channels are marked by the red and the blue colors, respectively. The red arrows in (a) indicate the magnetic Weyl loop centered around $M$.

FIG. 10. Band structures of $\gamma$-CoSe (with SOC included) with magnetic moments in the (a) $x$ and (b) $z$ direction, respectively. For better comparison, we repeat the path $X-M$ in (b). The arrow in (b) indicates the Weyl point. The arrows in (a) indicate the points on a Weyl loop.

FIG. 11. (a) Illustration of the location of the two magnetic Weyl points (red dots) in the ground state of $\gamma$-CoSe with moments along the $x$ direction. (b) The two Weyl points transform into a Weyl loop (red loop) when the moments are oriented along the $z$ direction.

SOC were only recently found in very few realistic examples, such as in monolayer PtCl$_3$ [44] and in monolayer GaTeI [40]. Our work offers another example, with the protecting symmetry different from the previous cases.

More interestingly, if we rotate the magnetization to the $z$ direction, the Weyl loop around $M$ can be recovered [see Fig. 10(b)]. This is because in such a case, the system has a horizontal mirror $M_z$ preserved. The two crossing bands have opposite $M_z$ eigenvalues, hence the loop is protected even under SOC. This is a very interesting magneto band structure effect, namely, by controlling the magnetization direction, one can tune the transformation between a pair of magnetic Weyl points and a magnetic Weyl loop, as illustrated in Fig. 11.

VI. DISCUSSION AND CONCLUSION

We have a few remarks before closing. First, in this work, we have discussed in detail three 2D CoSe structures, corresponding to the three with the lowest energies in our structural search. There exist more possible metastable structures with higher energies. (One of them, the $\delta$-CoSe, is discussed in [30]) Although the low energy ones have a better chance to be achieved, it is possible that, depending on the growth condition, the higher energy ones may also have an opportunity to be realized in experiment.

Second, as well developed methods for growing 2D materials, the chemical vapor deposition (CVD) and molecular beam epitaxy (MBE) methods may be good choices for realizing the discussed 2D CoSe structures. Notably, the bulk phase for $\alpha$-CoSe has been synthesized by a topochemical deintercalation approach [15]. Hence, $\alpha$-CoSe monolayer may also be obtained by the exfoliation method, which is commonly applied for making other 2D materials.

Finally, we comment on a few experimental aspects for detecting our predicted effects. The band topology features including the nodal lines in $\alpha$-CoSe, the Weyl points and the Weyl loop in $\gamma$-CoSe can be directly imaged by the angle resolved photoemission spectroscopy (ARPES).
technique. This technique has been successfully applied for many cases before, such as imaging the nodal loops in monolayer Cu$_2$Si [35] and CuSe [36]. One advantage of 2D materials is that its Fermi level can be tuned, e.g., by gating, in a wide range, so even the band features slightly above the Fermi level can be imaged. For $\beta$-CoSe, we have shown that strain can drive a phase transition from an AFM metal to an AFM insulator. For 2D materials, strain can be readily applied, e.g., by using a beam-bending apparatus [45] or by using an atomic-force microscope tip [46]. For $\gamma$-CoSe, to control its magnetization direction, one can use an applied magnetic field or couple it to a magnetic substrate.

In conclusion, we have performed a comprehensive search for CoSe 2D structures. We have identified three lowest energy candidates and revealed their rich physical properties. We show that they possess different magnetic ground states. $\alpha$- and $\gamma$-CoSe are FM with different easy axis, whereas $\beta$-CoSe is AFM. $\alpha$-CoSe has nonsymmetric magnetic nodal lines around the Fermi level, which are stable both without and with SOC. Such a kind of nodal lines has not been reported before. $\beta$-CoSe is a rare example of a 2D AFM metal. We show that this peculiar state is associated with the Fermi surface nesting features in its band structure, and by tuning the interaction or strain, one can achieve multiple phase transitions. $\gamma$-CoSe hosts a pair of magnetic Weyl points in its ground state, and a transformation from Weyl points to a Weyl loop can be induced by rotating the magnetization direction. Importantly, all these topological band features are robust against SOC. Our work uncovers a fascinating material platform for studying the interplay between magnetism, correlation effects, and band topology in 2D. These predicted materials could also have promising applications in electronics and spintronics.

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