Antiferromagnetic topological crystalline insulator and mixed Weyl semimetal in two-dimensional NpAs monolayer

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Abstract

Magnetic topological states have attracted significant attentions due to their intriguing quantum phenomena and potential applications in topological spintronic devices. Here, we propose a two-dimensional material NpAs monolayer as a candidate for multiple topological states accompanied with the changes of magnetic structures. Under the antiferromagnetic configuration, the long-awaited topological crystalline insulator (TCI) emerges with a nonzero mirror Chern number \( CM = 1 \) and a giant band gap of 630 meV, and remarkably a pair of gapless edge states can be tailored by rotating the magnetization directions while the TCI phase survives. Moreover, we establish the existence of quantum anomalous Hall effect and nontrivial nodal points under the ferromagnetic configuration, thereby giving rise to the mixed Weyl semimetal after adding the magnetization direction to topological classification. Our findings not only provide an ideal candidate for uncovering exotic topological characters with magnetism but also put forward potential applications in topological spintronics.

1. Introduction

Magnetic topological states are currently attracting widespread interest in condensed matter physics with a variety of exotic topological phenomena constantly proposed and intensively explored \([1, 2]\), and in which, the anomalous Hall effect \([3–7]\), spin–orbit torques \([8]\), magneto-optical effect \([9, 10]\), and Dzyaloshinskii–Moriya interaction \([11, 12]\) can exceed by far that of conventional compounds, suggesting a huge potential for spintronic devices. Antiferromagnetic (AFM) topological insulator (TI) \([13]\), a conceptual milestone, has been demonstrated in three-dimensional (3D) \( \text{MnBi}_2\text{Te}_4 \) \( n \) \((n = 1, 2, 3) \) family of materials with axion topology and Möbius fermion ensured by a combined symmetry \( T \) \( T \) \( T \), where \( T \) is the time-reversal symmetry and \( T_{1/2} \) represents the half-primitive-lattice translation \([14–21]\). Only recently, AFM TI has started to reach out to two dimension \([22–24]\), and, remarkably, the AFM quantum spin Hall effect can coexist with the superconductivity in FeSe monolayer \([25]\), but the materials realization of two-dimensional (2D) AFM TI is still a challenging task. Therefore, it is essential to search for new candidates and novel formation mechanisms to protect the nontrivial topology. Crystalline mirror symmetry, which greatly enriches the classification of topological phases \([26–30]\), as commonly believed can give rise to the AFM topologically insulating phase, i.e. AFM topological crystalline insulator (TCI) \([28, 31]\). Indeed, a 3D AFM TCI has been theoretically proposed in \( \text{EuIn}_2\text{As}_2 \) \([32]\). However, a 2D AFM TCI has not yet been revealed, and it would be of great significance to obtain it in intrinsic antiferromagnets.

On the other hand, magnetism provides an efficient path for tailoring the band topology, meaning that topological phase transitions can be gained via manipulating the spin degree of freedom. For example, together with a magnetic transition from AFM to ferromagnetic (FM) configuration, the AFM TI \( \text{MnBi}_2\text{Te}_4 \)
changes into a Weyl semimetal [14, 15], and the quantum spin Hall effect (QAH) [14, 15], and the quantum spin Hall effect transitions into the quantum anomalous Hall effect (QAHE) in functionalized Sn [33]. Furthermore, topological phase transition is also often accompanied with manipulating the directions of spin polarization, especially for the breaking of crystalline mirror symmetry, where the related nodal-line semimetals change into Weyl semimetals [4, 34], and TCIs change into second-order TIs [32, 35, 36]. Remarkably, while adding the directions of spin polarization to the topological analysis, a natural classification of mixed topological semimetals is obtained [8, 37]. However, limited by the contrasted interplay of magnetism and topology, the magnetism-mediated phase transitions have been rarely reported up to date.

In the present work, using first-principles calculations, we show that the band topology can be effectively controlled by the magnetic configurations and directions in NpAs monolayer with the realization of TCIs, QAHE, and mixed Weyl semimetal. Correlated with the mirror symmetry $M_z$, a 2D TCI phase, characterized with the nonzero mirror Chern number and gapless edge states, emerges in AFM NpAs monolayer when the magnetic moment points out-of-plane. The TCI phase can still be obtained while rotating the magnetic direction into the in-plane, however, unlike the former case, the edge states can be either gapless or gapped depending on the interplay between magnetization direction and mirror symmetry $M_x \text{ or } M_y$. For the FM ordering with out-of-plane direction, the NpAs monolayer exhibits QAHE, characterized with a quantized Chern number $C = -1$ and chiral edge state. Remarkably, a topological phase transition from $C = -1$ to $C = 1$ accompanied with a closing of the band gap occurs as the magnetization direction is varied, thereby promoting the NpAs monolayer as a promising material candidate for understanding the complex interplay between magnetism and topology.

2. Computational details

Our calculations based on density functional theory were carried out for structural relaxations and electronic structure calculations as implemented in the Vienna $ab$ initio simulation package [38]. The generalized gradient approximation with Perdew–Burke–Ernzerhof exchange-correlation functional was adopted to describe the electronic interaction [39]. A vacuum thickness of 20 Å was introduced to avoid the interaction between the nearest slabs. The kinetic cutoff energy was set to 500 eV. All structures were relaxed until the convergence threshold of the maximum forces on each atom was less than 0.01 eV Å$^{-1}$, and the criterion of total energy for convergence was set as $10^{-6}$ eV. A Monkhorst–Pack $k$-point mesh of $11 \times 11 \times 1$ was utilized. The GGA + $U$ method is used for f orbitals to calculate the modified Coulomb repulsive interaction of Np atom with $U = 4$ eV [4], and the phonon calculations were carried out based on a supercell approach using the PHONOPY code [40]. The topological edge states were calculated using effective tight-binding Hamiltonians from maximally localized Wannier functions (MLWFs) as implemented in the WANNIER90 [41, 42] and WANNIER tools packages [43].

3. Results and discussion

Bulk NpAs has long been studied both theoretically and experimentally due to the exotic electronic and nonlinear magnetic properties [44, 45], that crystallizes in the face-centered-cubic NaCl-type structure with space group $Fm\overline{3}m$. The Np atoms are at 4c Wyckoff position and As at 4b Wyckoff position, and remarkably they are coplanar. Here, we focus on the (001)-oriented monolayer of NpAs with coplanar Np and As atoms positioned in the mirror plane $z = 0$. As depicted in figure 1(a), NpAs monolayer forms a 2D square lattice with one Np and one As in the unit cell, and the corresponding space group is $P4/mmm$. The optimized lattice constant is $a = 4.11$ Å, which is basically consistent with the experimental bulk structure [44]. Moreover, as verified by the explicit calculations of phonon spectrum, displayed in figure 1(b), there is no imaginary frequency over the whole Brillouin zone, suggesting that NpAs monolayer is dynamically stable and difficult to destroy once formed.

The total energy investigations show that ground state of NpAs monolayer is spin-polarized with an AFM ordering along out-of-plane direction, which is energetically preferred by 41 meV. The magnetic moment on each Np is about 4 $\mu_B$ with a 5f$^4$ configuration. Figure 1(c) presents the total density of states (DOS) and partial DOS of each Np atom. Clearly, the partial DOS of two Np atoms are spin-polarized, but their magnetic moments are in opposite direction, hence the total DOS is spin unpolarized resulting in a zero net magnetic moment. In the absence of SOC, as illustrated in figure 2(a), the conduction and valence bands overlap each other around the $\Gamma$ point, revealing a band inversion with the Np-$d_{xy}$ orbitals being lower by 94 meV than the As-$p_x$, and a gapless nodal line is obtained. Switching on SOC leads to a gap opening as shown in figure 2(b). The bands are always degenerate in this case due to the presence of
combined time-reversal and spatial inversion symmetries, and, remarkably, the system is insulating with an inverted gap as large as 630 meV.

It is well known that band inversion is an important mechanism for the realization of a topologically nontrivial insulator. The magnetic space group of the NpAs monolayer is $P_{4/mnm}'$, in which the AFM ordering with out-of-plane alignment is perpendicular to the mirror plane $z = 0$, i.e. the system possesses
$M_z$ symmetry that is symmetric under the reflection $z \rightarrow -z$. The existence of $M_z$ suggests a high possibility for the formation of a 2D AFM TCI in NpAs monolayer. To show this explicitly, we construct a tight-binding Hamiltonian based on the MLWFs of NpAs monolayer, and meanwhile a mirror operator $M$ distinguishes the occupied Bloch states with different mirror eigenvalues $-i$ and $+i$ into two manifolds. For each manifold, the Wilson-loop calculations are carried out with the Wilson-loop matrix given by

$$W_{(k_x+2\pi,k_y)}-(k_x,k_y) = \lim_{\eta \rightarrow -i \infty} \int_{-F_{N-1}F_{N-1} \cdots F_{N-1}} \langle \eta \rangle = \langle \eta \rangle (2\pi i/N) \langle \eta \rangle |u_m(k_x,k_y)| u_n(k_x,k_y) \rangle$$

where $|u_m(k_x,k_y)|$ is the lattice-periodic part of Bloch state at $(k_x,k_y)$, $m, n$ are band indices, and $N$ is the number of $k_z$ points used in the calculations. Figures 2(c) and (d) indicate that Chern number of the all occupied bands for each manifold is, respectively, $C_i = 1$ and $C_{-i} = -1$, resulting in a total Chern number $C = C_i + C_{-i} = 0$ and a mirror Chern number $C_M = (C_i - C_{-i})/2 = 1$. More precisely, figure 2(e) displays the Berry curvature $\Omega(k)$ of a given subset of occupied states with mirror eigenvalue $+i$, calculated according to [46, 47]

$$\Omega(k) = \sum_{m \neq n} -2 \text{Im} \left\langle \psi_{mk} | \psi_{nk} \right\rangle \left( \psi_{mk} | \psi_{nk} \right\rangle^2, (1)$$

where $\psi_{mk}$ and $\epsilon_{mk}$ are the Bloch wavefunctions and corresponding eigenvalues of band $m/n$, respectively, and $\psi_{kx}, \psi_{ky}$ are the velocity operators. With the dominant contribution to $\Omega(k)$ coming from the region around $\Gamma$, opposite signs for opposite mirror eigenvalues $\pm i$ $C_{\pm i}$ indeed acquire integer values of $\pm 1$, obtained by a $k$-space integral $C_{\pm i} = \frac{1}{2\pi} \int_{k_{BZ}} \Omega(k) d^2k$, which is in good agreement with our Wilson-loop calculations and uncover the 2D AFM TCI nature of the NpAs monolayer.

We then investigate the nontrivial edge states of NpAs monolayer, which is the hallmark of a 2D TCI, and the calculated value of $C_M = 1$ indicates that there is one pair of gapless edge states in the bulk inverted gap. To show this, we performed calculations of the edge states using MLWFs and the edge Green’s function. Figure 2(f) presents the results of the semi-infinite NpAs monolayer terminated with As atoms. Clearly, a pair of gapless edge states emerge around the $\Gamma$ point that bridge the bulk conduction and valence bands. Further analysis shows that the same behavior appears for all directions of edges due to the surviving of mirror symmetry $M_z$, confirming explicitly that the NpAs monolayer with out-of-plane magnetization is a 2D AFM TCI.

Generally, mirror symmetry breaking opens a gap in the edge/surface states of TCIs [26, 28]. For the AFM NpAs monolayer, one way to destroy the $M_z$ symmetry is switching the directions of spin polarization, such as from out-of-plane to the in-plane direction. Figures 3(a) and (b) present the orbitally resolved bulk band structures and the edge states, respectively. Although the gap is reduced, it maintains the insulating property with a band gap of 220 meV that is still large enough for the room-temperature applications. Remarkably, similar to the above configuration with out-of-plane direction, the band order is inverted at the $\Gamma$ point, indicating that the system is topologically nontrivial. This is due to the fact that mirror symmetry $M_z$ ($M_{\perp}$) is hosted in the NpAs monolayer with in-plane AFM alignment. Moreover, despite the broken $M_z$, the gapless edge states can still be obtained around the $\Gamma$ point, as shown in figure 3(b), demonstrating further the 2D AFM TCI phase in NpAs monolayer with respect to the $M_{\perp}$. Unlike the $M_z$ symmetry, edge states remain gapless only along the direction perpendicular to the magnetic directions, while the others are gapped, i.e. the gapless edge states can be tailored via the magnetic directions while the nontrivial topology survives. Interestingly, this is maybe used as the magnetically engineered topological spintronic sensors and memory.

When the magnetic moments on Np, with an amplitude of $4 \mu_B$, are coupled parallel to each other, as illustrated in figure 1(d), both the total and partial DOS are spin-polarized. The corresponding band structures of FM NpAs monolayer without SOC are displayed in figures 4(a) and (b). Clearly, the spin-up bands are gapless with a band touching at the $\Gamma$ point, while a gap appears in the spin-down bands with an
amplitude of 262 meV, suggesting a pronounced half-metallic character near the Fermi energy. Taking SOC into consideration, figure 4(c) shows that the nodal point of spin-up bands opens a gap of 83 meV, indicating preliminary nontrivial topological property of the FM NpAs monolayer. Different from the band inversions in AFM configuration that emerge in the degenerated two spin channels, here, it occurs only in the spin-up bands. Therefore, the spin-up channel will carry a nonzero Chern number, while a zero one for the spin-down channel, for example $C^\uparrow = \pm 1$ and $C^\downarrow = 0$, leading to the exotic QAHE with an integer total Chern number $C = \pm 1$. We thus check the nontrivial property by anomalous Hall conductivity given by

$$\sigma_{xy}^{A} = C e^2/h,$$

where $C$ is the total Chern number defined as $C = \frac{1}{2\pi} \int_{BZ} \Omega(k) d^2k$ and $\Omega(k)$ is the Berry curvature over all the occupied states [46, 47]. The bottom panel of figure 4(g) presents the $\sigma_{xy}^{A}$ as a function of the Fermi level and the reciprocal-space distribution of Berry curvature $\Omega(k)$ around the $\Gamma$ point, is indeed acquired with an integer value of $-1$, demonstrating the QAHE in NpAs monolayer. The QAHE is further confirmed by the calculated edge states, as shown in figure 4(h), where one chiral edge state connects the conduction and valence bands.

As displayed in figure 4(d), after including SOC, the band structures of NpAs monolayer with in-plane ferromagnetism reveals band crossings along the $\Gamma - X$ direction near the Fermi level. There are two semimetallic points in the 2D Brillouin zone as clearly shown by the 3D illustration of band dispersions in figure 4(f). To uncover the topological nature of these crossing points, we evaluate the Berry phase along a closed loop in momentum space around one of the points, and find that both of them are topologically nontrivial with a Berry phase of $\pi$. Moreover, the nontrivial points are connected with emergent edge states as illustrated by the edge dispersion of a semi-infinite 1D ribbon in figure 4(i). They remain intact as long as the magnetization direction is fixed, but a gap opens up by magnetization switching from in-plane to out-of-plane directions, whereby the gap opening mediates a topological phase transition corresponding to a change of the Chern number from $C = -1$ to $C = 1$. The top panel of figure 4(g) plots the $\sigma_{xy}^{A}$ with the Chern number $C = 1$ accompanied by the changing of the chiral edge state in figure 4(h), establishing the emergence of mixed Weyl semimetal for FM NpAs monolayer within the 3D mixed phase space $(k_x, k_y, \theta)$, where $(k_x, k_y)$ is the 2D crystal momentum $k$ and polar angle $\theta$ represents the magnetization direction from out-of-plane to in-plane [37]. Additionally, similar to the [100] directions, the mixed Weyl points emerge as well while the in-plane magnetization along the [010] direction due to the underlying $M_{x/y}$ symmetry in figure S1 (https://stacks.iop.org/NJP/23/123018/mmedia).

4. Conclusion

In conclusion, we have demonstrated the emergence of AFM TCI and mixed Weyl semimetal in 2D NpAs monolayer. The AFM TCI phase is protected by the crystalline mirror symmetry with the magnetic moments along both the out-of-plane and in-plane direction associating with the $M_z$ and $M_x$ ($M_y$) respectively. On the other hand, FM NpAs monolayer holds insulating and semimetallic topological states,
respectively, when the direction of the magnetization is varied from out-of-plane to in-plane directions. This mediates a topological phase transition between QAHE with opposite chirality, which corresponds to a change of the Chern numbers. Our results not only broaden the research field of 2D magnetic topological states, but also further improve the understanding of the combination of topology and magnetism.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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