

Effect of symmetry breaking on Dirac points in topological semimetal EuAgBi

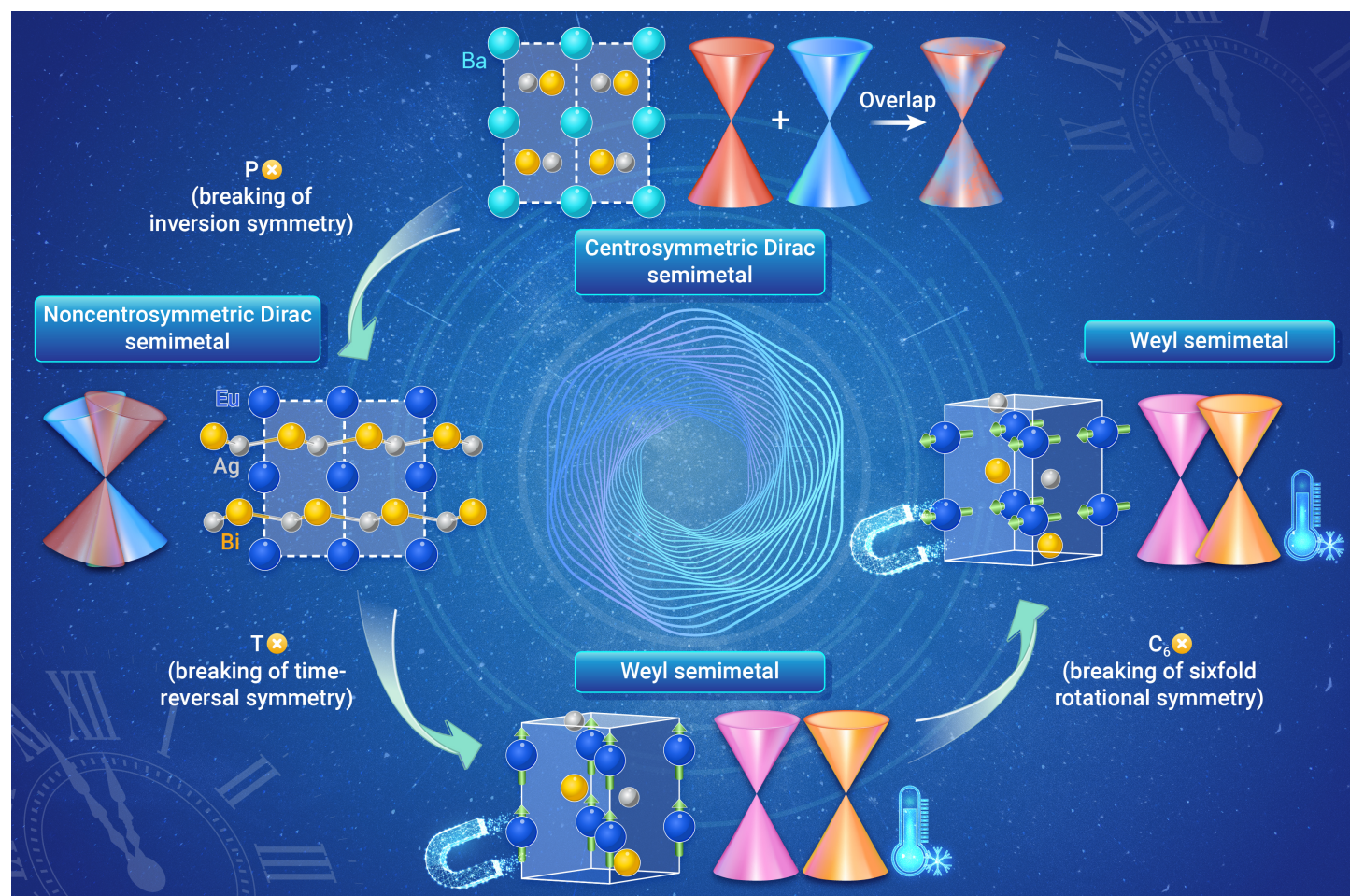
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GRAPHICAL ABSTRACT



PUBLIC SUMMARY

- Symmetry-breaking-driven topological phase transitions were observed in EuAgBi and BaAgBi.
- Transformations among centrosymmetric and noncentrosymmetric Dirac points and Weyl points were observed.
- Noncentrosymmetric Dirac points were confirmed by angle-resolved photoemission spectroscopy.

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Combining symmetry breaking with topological theory constitutes a promising approach for advancing the understanding and control of topological phase transitions, which calls for experimental evidence from suitable material systems. We investigated Dirac points (DP) in noncentrosymmetric EuAgBi through the breaking of inversion symmetry (P), time-reversal symmetry (T), and sixfold rotational symmetry (C_6), using centrosymmetric BaAgBi as the reference. The analysis of various magnetic structures in EuAgBi achieved by controlling temperature and magnetic field shows that breaking P does not affect the existence of DP; breaking both P and T transforms DP into Weyl points (WP); and despite the reduced symmetry when P, T, and C_6 are all broken, WP remains formally unaffected in the field-induced ferromagnetic phase. Additionally, we have realized the possible coexistence of WP and DP along distinct directions. Furthermore, the noncentrosymmetric (P-broken) DP, previously predicted theoretically but lacking experimental evidence, were observed in paramagnetic EuAgBi using angle-resolved photoemission spectroscopy. Our results provide realistic examples of how distinct topological states can emerge under different symmetry-breaking scenarios.

INTRODUCTION

Topological band states are described by using global topological invariants derived from the distribution of eigen wavefunctions over the whole Brillouin zone (BZ). They are robust against changes induced by local perturba-

tions and discussions of topological phase transitions typically do not center on changes in symmetry.^{1,2} However, with the discovery of numerous topological states, research has shown that symmetry plays a crucial role in the formation of these states.^{3,4} The seemingly contradictory nature of these two aspects suggests that combining the perspective of symmetry breaking with topological theory holds promise for constructing a unified framework to tune the topological phase transition. Regarding the topological states, such as topological insulators, topological semimetals, and topological superconductors, previous research has mainly focused on their classification based on symmetries.⁵⁻⁸ However, realistic materials exhibiting the effects of symmetry breaking on specific topological states remain rare, highlighting the need for further experimental studies to provide crucial references and validation.

In the study of Dirac points (DP), inversion symmetry (P) and time-reversal symmetry (T) are crucial for both two-dimensional (2D) DP and three-dimensional (3D) DP.⁹⁻¹⁵ Additional lattice symmetry (ALS) contributes to the stability of 3D DP.^{5,16-21} The transformation of DP through the breaking of P, T, and ALS (Figure 1A) is highly expected but not realized. This challenge stems from several factors. First, Dirac semimetals without P are quite rare. Only materials such as SrHgPb, KSnSe₂, and Cd₃As₂ with C_{4v}^4 symmetry have been proposed, but none have been experimentally verified.^{19,22-26} Second, the predicted materials are all non-magnetic, preventing further exploration of T-broken effects. Finally, it remains experimentally challenging to manipulate symmetry within a single material, such as achieving precise control of doping.^{27,28} Therefore, the lack of appropriate materials allowing for controlled

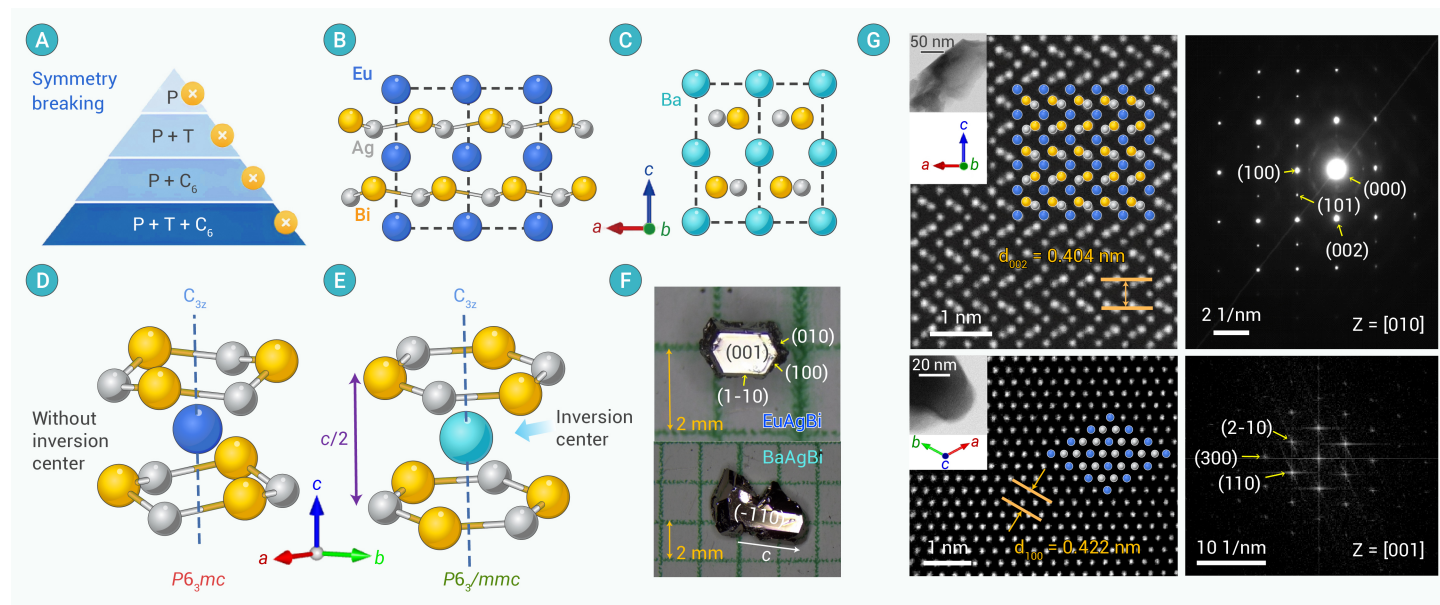


Figure 1. Crystal structures of EuAgBi and BaAgBi (A) Schematic of symmetry breaking. Crystal structures of EuAgBi in (B) and (D); BaAgBi in (C) and (E). The blue dashed lines represent the threefold rotation axis C_{3z} . (F) Optical photographs of high-quality EuAgBi (top) and BaAgBi (bottom) single crystals marked with crystal planes (The grid size is 2 mm). (G) HAADF-STEM images, corresponding selected area electron diffraction pattern and fast Fourier transform pattern of EuAgBi single crystal along [010] and [001].

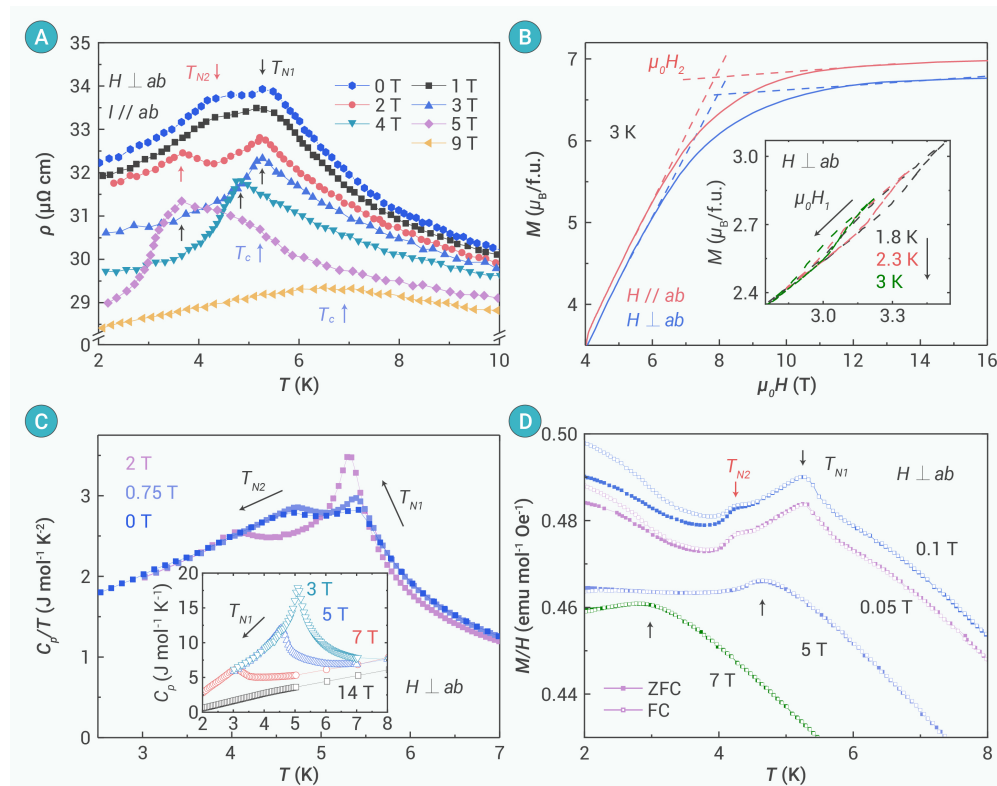


Figure 2. Electrical, magnetic, and specific heat measurements of EuAgBi (A) Temperature-dependent resistivity at low temperature under magnetic field. (B) Field-dependent isothermal magnetization. The inset shows a magnified view around 3 T. (C) Temperature-dependent specific heat capacity $C_p(T)/T$ and $C_p(T)$ in the inset. (D) Temperature-dependent magnetic susceptibility.

allows for preparation of centrosymmetric EuAgBi (space group $P6_3/mmc$, No. 194) powder as included in the database (PDF 04-019-4231).^{30,31} By using the high-temperature flux method, we obtained high-quality EuAgBi single crystal (CCDC number: 2419060; Figure 1F) having noncentrosymmetric crystal structure. It crystallizes in the $P6_3mc$ space group (No. 186) with Ag and Bi atoms buckling along the c axis (Figures 1B & D), resulting in a lack of an inversion center (without P), as directly confirmed by high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) (Figure 1G). The known BaAgBi has Ag and Bi atoms lying within the same plane with an inversion center at the Ba atom's position (Figures 1C & E). EuAgBi and BaAgBi single crystals own similar elemental compositions and hexagonal honeycomb structures, with distinction in P, suggesting that these two

symmetry breaking hinders this in-depth study into DP.

In this letter, we study the changes in DP induced by breaking P, T, and sixfold rotational symmetry (C_6), based on two structurally similar materials, EuAgBi and BaAgBi. BaAgBi preserves P, hosting centrosymmetric DP. The Ag- s and Bi- p orbitals dominate near the Fermi level in both compounds, providing a practical platform to study the effects of symmetry. As known, when the point group is neither C_{4v} nor C_{6v} , breaking P causes DP to become Weyl points (WP).²⁴ This occurs due to the absence of C_4 and C_6 , which serve as ALS contributing to the stability of the P-broken DP.²⁴ In the case of noncentrosymmetric EuAgBi with the C_{6v} point group, DP persist despite breaking P. Subsequently, both P and T are broken in ferromagnetic (FM) [001] EuAgBi (spins ferromagnetically align along [001]), resulting in the dissolution of DP into WP, indicating that C_6 alone cannot protect DP. Moving forward, P, T, and C_6 are all broken in antiferromagnetic (AFM) [110] (Néel vector along [110]) and FM [100] EuAgBi, where WP persist (FM [100]) despite the further reduction in symmetry caused by breaking C_6 . Additionally, WP are found in the $k_z = 0$ plane of paramagnetic (PM) EuAgBi, where P and C_6 are broken, achieving the possible coexistence of DP and WP. P-broken DP are remarkably demonstrated by angle-resolved photoemission spectroscopy (ARPES) measurements, using BaAgBi with P as a reference.

MATERIALS AND METHODS

The EuAgBi and BaAgBi single crystals were grown using the high-temperature solution method with Bi as the flux. For EuAgBi, europium ingots (99.9%, Alfa Aesar), silver shots (99.999%, Alfa Aesar), and bismuth granules (99.995%, Alfa Aesar) were mixed in a molar ratio of 1:1:9. For BaAgBi, barium rods (99.9%, Alfa Aesar), silver shots (99.999%, Alfa Aesar), and bismuth granules (99.995%, Alfa Aesar) were used in a molar ratio of 2:1.6:9. The crystal growth conditions for both materials were identical. The mixtures were placed in a fritted alumina crucible set, sealed in fused silica ampoules under vacuum, and heated to 1283 K for 12 hours.²⁹ The ampoules were then slowly cooled to 723 K at a rate of 3 K/h. At this temperature, the crystals were separated from the remaining liquid by centrifugation.

RESULTS AND DISCUSSION

Considerable effort has been devoted to grow EuAgBi single crystal by varying methods. It is proved that the synthesis method used before only

materials provide platforms for studying P breaking effects (more details in Sections 1 of SI).

The resistivity $\rho(T)$, isothermal magnetization $M(H)$, specific heat capacity $C_p(T)$, and magnetic susceptibility $\chi(T)$ of EuAgBi under applied magnetic field are shown in Figure 2. There are two magnetic transition peaks ($T_{N1} \sim 5.3$ K; $T_{N2} \sim 4.5$ K; 0 T) that shift to lower temperature with increasing magnetic field, a typical behavior of AFM transition.^{33,32} Under magnetic field of 5 T, a kink (T_c) appears at temperature higher than T_{N1} , shifting to higher temperature as the magnetic field increases from 5 T to 9 T (Figure 2A). This corresponds to a magnetic transition between field-induced FM phase and high-temperature PM phase.³³ Additionally, a small magnetic hysteresis appears around 3 T ($\mu_0 H_1$), moving to lower field with increasing temperature (the inset of Figure 2B), corresponding to the suppression of T_{N2} around 3 T in $\rho(T)$ and $C_p(T)$ (Figure 2A and the inset of Figure 2C). These mark the boundary between AFM I and AFM II. The magnetization parallel to the ab plane saturates more easily, with a smaller saturation field ($\mu_0 H_2$) (Figure 2B), indicating that the easy magnetization axis lies within the ab plane. We established a magnetic phase diagram as shown in Figure 3, combining all the measurement results (more details in Section 2 of SI).

Since compounds with structures similar to EuAgBi have been reported to have A-type AFM structure, we performed calculations on the total energy, band structure, and topological properties for several possible magnetic structures.³³⁻³⁵ As shown in Table I, the total energy of AFM phase is lower than that of FM phase more than 1 meV, which indicates that the ground state is AFM. It is also noted that the magnetic anisotropic energy is very small and the total energy with the magnetic moments lying in the ab plane is slightly lower than that being upright, suggesting that the easy magnetization axis is within the ab plane. All of these are consistent with the experimental results. Among all magnetic structures, AFM [110] is the lowest energy state and the likely ground state. AFM [100] and FM [110] are difficult to converge, reflecting their unfavorable formation. FM [001] and FM [100] can be achieved by applying magnetic field along the [001] and [100] to saturated value, respectively (more details in Sections 2 of SI).³⁶

To investigate the DP though symmetry breaking, the case of P breaking is first analyzed. When the point group is neither C_{4v} nor C_{6v} , the breaking of P directly leads to the disappearance of DP as reported in our previous work.²⁴ Here, the discussion focuses on EuAgBi, which satisfies the C_{6v} and breaks P

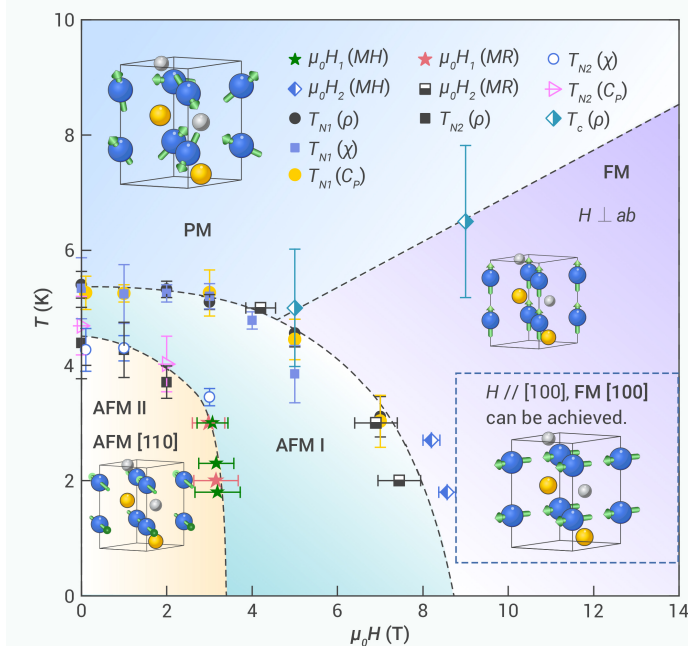


Figure 3. Magnetic phase diagram of EuAgBi with possible magnetic structures Characteristic temperatures and magnetic fields obtained from different measurements are marked with different symbols.

compared to BaAgBi. For PM EuAgBi, only bands along the Γ -A are doubly degenerate due to C_6 (Figures 4A & D), while all bands in BaAgBi are doubly degenerate due to P and T (Figures 4B & E). PM EuAgBi and BaAgBi both have DP along the Γ -A, as shown by the blue circles in Figures 4A-B. The 2D irreducible representations DT7 and DT8 form the doubly degenerate bands that intersect to create the fourfold degenerate DP in Figure 4G, showing that P breaking does not affect the DP's existence. Furthermore, the ALS that contributes to the stability of the P-broken DP can be specifically identified as C_6 here.²⁴ The bands of PM EuAgBi are non-degenerate without C_6 . There might be WP in generic k point in noncentrosymmetric PM phase. Indeed, it is found that along a non-high-symmetry path at $k_z = 0$ between the Γ -K and Γ -M, WP exist. Considering the C_6 symmetry, there are totally twelve WP in the $k_z = 0$ plane. The positions of all these WP (red spheres for positive chirality and black spheres for negative chirality) and DP (purple spheres) in the BZ for PM EuAgBi are shown in Figure S10(a) of SI. The surface states corresponding to the WP are shown in Figure S10(b) of SI.

Next, different magnetic structures of EuAgBi are analyzed, as which can further break T and C_6 .^{4,37,38} For EuAgBi, Bi- p and Ag- s orbitals dominate the bands near the Fermi level, while Eu f -electrons lie well below. These Eu f -electrons are responsible for the magnetism and alter the symmetry of the system. In field-induced FM [001], the presence of magnetism breaks P and T, but spins along the c axis retain C_6 . The two doubly degenerate bands still split into four non-degenerate bands (corresponding 1D irreducible representations are DT7, DT8, DT9, and DT12, respectively), causing the DP to disperse into four WP (Figure 4G). In FM [100] and AFM [110], the spins lie within the ab plane, thus breaking P, T, and C_6 entirely (Figure 4F). Strictly speaking, FM [100] has only 1D irreducible representations, and the crossing points are four WP. Since the energies of two bands that represent as B3 and B4 under magnetic space group are very close with energy difference of 2.03×10^{-4} eV, they can be approximated as doubly degenerated bands (represented as B3 + B4), leading to the four WP being considered as two nearly triple points (NTP) (Figure 4G).^{39,40} Although AFM [110] forms a gap along the Γ -A, the bands only have 2D irreducible representations, indicating they remain doubly degenerate (more details on Sections 4 of SI).⁴¹

As far as we know, there is currently no experimental evidence verifying the theoretically predicted noncentrosymmetric DP (P-broken DP). We carefully conducted ARPES measurements on EuAgBi and BaAgBi. The cleavage planes are determined to be the (100) plane for both materials by analyzing the distribution of Fermi surface (Figures 5A-B). Since the measurement was

Table 1. Total energy of various magnetic structures of EuAgBi. DP and WP refer to the Dirac points and Weyl points, respectively.

Phase	Topology	Total Energy (eV)
AFM [110]	gap	-38.48795
AFM [001]	DP	-38.48768
FM [100]	nearly triple points	-38.48634
FM [001]	WP	-38.48611
PM	DP	
AFM [100]	Non-convergence	
FM [110]	Non-convergence	

taken along the sixfold rotational axis, the distribution exhibits a periodicity of Γ -A- Γ (Figures 5C-F). The red dotted lines, obtained by periodically replicating the Γ -A band structure of PM EuAgBi and BaAgBi from Figure 4G, represent the theoretical calculations, which show excellent agreement with the measured band dispersion. This demonstrates the reliability of our calculation. The DP are marked by red arrows and prove the existence of noncentrosymmetric DP in EuAgBi and centrosymmetric DP in BaAgBi in Figures 5C-F. As shown in Figure S10(a) of SI, the noncentrosymmetric DP marked by purple spheres are located at $(0, 0, \pm 0.63\pi/c)$, making them closer to the A point than to Γ . This proximity leads to DP from neighboring periods clustering in pairs in Figures 5C & E. Additionally, in PM EuAgBi, the DT7 band at the A point is positioned below the Fermi level (Figure 4G), leading to the formation of clear degeneracy points at A in the experimentally observed Γ -A- Γ band structure (Figures 5C & E). For the first-principles calculations not using HSE06 hybrid functional, the band structures near the Γ point are truncated (red dotted line) as highlighted by the yellow dashed boxes for both EuAgBi and BaAgBi, while ARPES measurements show bands extending further into higher energy (Figure S7). Our previous work found that whether to use the HSE06 hybrid functional affects the presence of noncentrosymmetric DP when symmetry requirements are satisfied. For example, Bi_2PdO_4 opens a gap with HSE06.²⁴ Our results confirm experimentally that HSE06 hybrid functional improves the accuracy of DFT band calculations, though prior predictions of intrinsic noncentrosymmetric DP did not use this approach.^{22,23,25,26} The bands perpendicular to the $\bar{\Gamma}$ - $\bar{Z}$ across the DP (cut 2) are mapped in Figures 5G-H, seeing the location of the DP from the side. In PM EuAgBi, P breaking results in the formation of four singly degenerate bands (splitting the DP bands) (Figure 5G), unlike the two doubly degenerate bands in BaAgBi (Figure 5H). Though the four bands are too close to be resolved by ARPES, P-breaking-induced splitting along K- Γ is observable: in Figures 5I-J, short (long) yellow arrows mark the singly degenerate LD4 (LD3) band from the splitting of the doubly degenerate DT8 of Γ -A. A magnified view of bands along K- Γ -A is shown in Figures S8-S9.

EuAgAs and EuCuAs with similar crystal structure to EuAgBi have both been reported to host nontrivial magnetic structures, with neutron diffraction experiments confirming the case for EuCuAs.^{15,34} Compared to Cu and As, Ag and Bi in EuAgBi have larger atomic numbers, leading to stronger spin-orbit coupling. Additionally, EuAgAs and EuCuAs have centrosymmetric structures that cannot generate Dzyaloshinskii-Moriya interactions.^{42,43} These two factors make EuAgBi more favorable for the emergence of nontrivial magnetic structures, though this requires verification through further experiments. Nontrivial magnetic structures such as skyrmions and biskyrmions hold great promise for racetrack memory applications, as they can be driven by extremely low density current.^{44,45} Furthermore, charge pumping between WP due to chiral anomaly can enhance the control of skyrmions in racetrack memory.⁴⁶ The P-broken structure provides a platform for controlling topological magnetic structures via topological band structures. Last but not least, considering the DP is far from Fermi level and K atoms can form KZnBi having the same structural type as BaAgBi, introducing hole through K doping is a feasible experimental approach to shift the DP closer to the Fermi level.^{47,48}

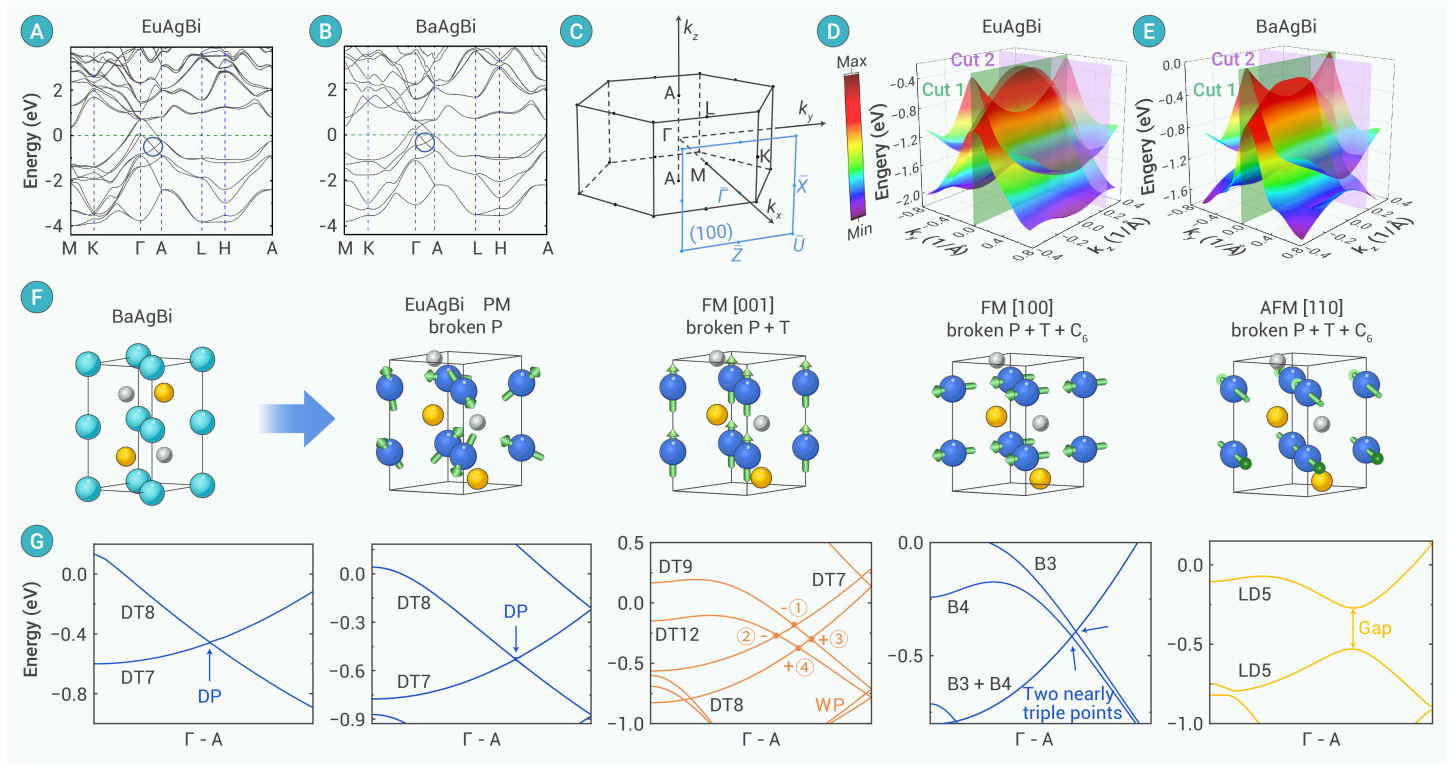


Figure 4. Calculated band structures and topological states Band structures for PM EuAgBi in (A) and BaAgBi in (B). The DP are marked with blue circles. (C) Bulk and (100) surface projected BZ. 3D band structures of k_y - k_z plane are shown in (D) and (E), where cut 1 (along the k_z) and cut 2 (through the DP along the k_y) are marked with green plane and purple plane, respectively. (F) Schematic of crystal structure and magnetic structures. BaAgBi is taken as the starting point for symmetry breaking. (G) Effect of different magnetic structures on the bands along Γ -A. In the band structure of FM [001], four WP are labeled 1-4, with their corresponding chiralities (± 1) indicated next to each point.

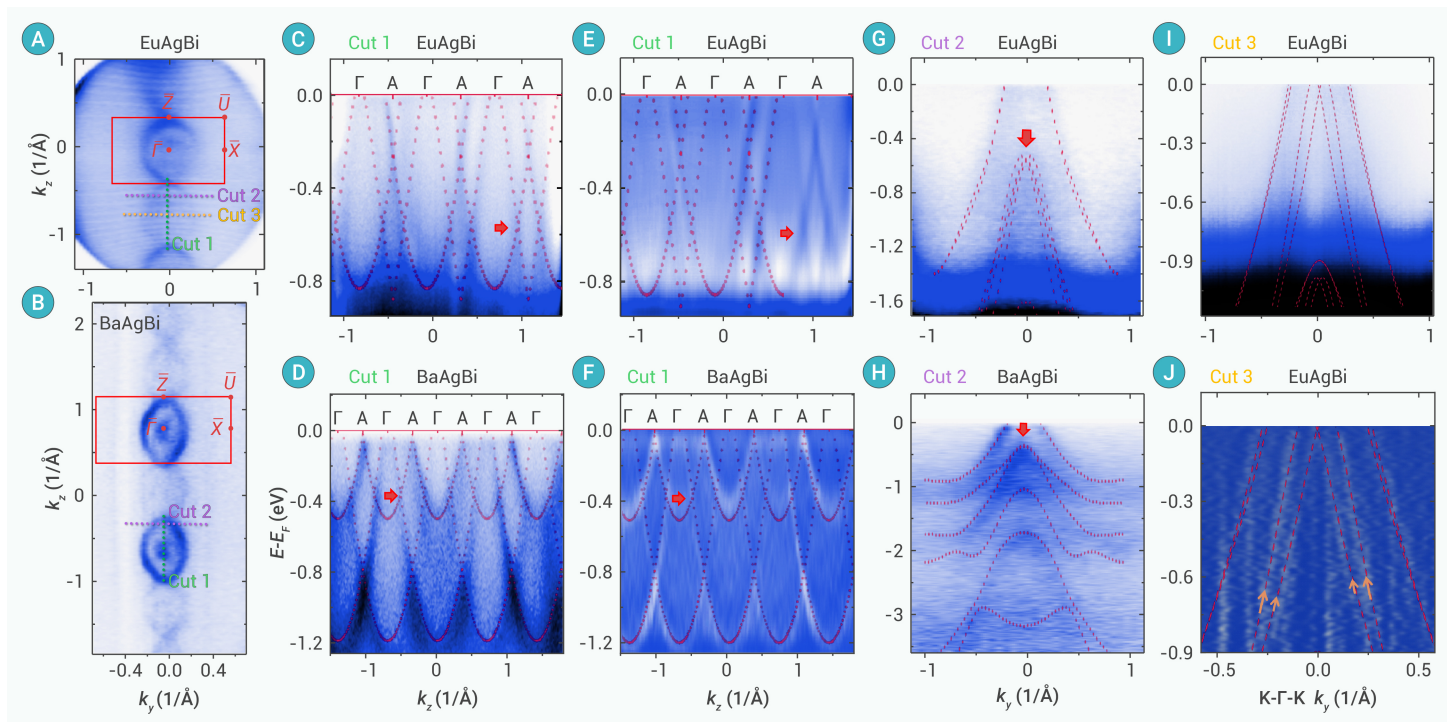


Figure 5. Existence of noncentrosymmetric DP ARPES intensity maps of the Fermi surface on the (100) plane of (A) EuAgBi and (B) BaAgBi, respectively. The red boxes represent the BZ boundary. ARPES intensity maps along Γ -A (cut 1) in (C) and (D) and second derivative analysis in (E) and (F); cut 2 in (G) and (H) are presented, corresponding to EuAgBi and BaAgBi, respectively. The DP are marked by the red arrows. (I) ARPES intensity map along the K- Γ -K path of EuAgBi (cut 3). (J) Curvature intensity plot of momentum distribution curves of (I). Short (long) yellow arrows indicate the LD4 (LD3) band.

CONCLUSIONS

The DP of EuAgBi was systematically studied by breaking P, T, and C_6

symmetry, taking BaAgBi as the starting point. P-broken DP is shown in PM EuAgBi using ARPES, compared to BaAgBi with P, providing the previously

lacking experimental evidence. This indicates that P is not necessary for the existence of DP. Subsequently, both P and T are broken in FM [001] EuAgBi; moreover, P, T, and C_6 are all simultaneously broken in FM [100] and AFM [110] EuAgBi. The simultaneous breaking of P and T causes the DP to split into four WP, and further breaking of C_6 does not necessarily eliminate these WP. Additionally, possible coexistence of DP and WP is demonstrated in PM EuAgBi. These results indicate that EuAgBi and BaAgBi are good platforms for studying the symmetry breaking of DP and potential applications in next-generation spintronics.

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AUTHOR CONTRIBUTIONS

X. H. W., Z. Y. S., W. L., Y. P., and B. J. contributed equally to this work. G. W., H. M. W., and T. Q. coordinated and supervised the project. X. H. W. conceived the idea, carried out the crystal growth and the electrical and magnetic property measurements, and wrote the raw manuscript. Z. Y. S. carried out the First-principles calculations and symmetry analysis; W. L. and B. J. collected the ARPES data; Y. P. collected the STEM data; C. L. assisted with the collection of the X-ray photoemission spectroscopy data. All authors

participated in the discussion and contributed to the texts in this manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.

DATA AND CODE AVAILABILITY

Data are available from the corresponding author upon reasonable request.

SUPPLEMENTAL INFORMATION

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