

Co-emergence of Dirac and multi-Weyl Topological Excitations in Pnictide Antiperovskites

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The relatively unexplored family of pnictide-based antiperovskites has been shown to harbor prospects for topological phases. Using the example of Ca_3BiP , we demonstrate a cascade of phases as the initial cubic symmetry is broken successively. Initially a small gap Z_2 topological insulator, spin-orbit coupling leads to band re-ordering resulting in a topological semimetal phase. Compressive uniaxial (001) strain leads back to a small gap Z_2 topological insulator, with the expected gapless boundary modes. Tensile (001) strain leaves the system with a pair of Dirac points along $(0,0,\pm k_z)$ pinning the Fermi level, producing an unusual double meniscus of connected Fermi arcs on the (100) and (010) surfaces. Finally, breaking time-reversal symmetry by an applied Zeeman field produces a pair of multi-Weyl nodes (massive or massless depending on direction) simultaneous with a sister pair of Dirac modes along each $\pm k_z$ axis, combining to pin the Fermi level in the close vicinity of these varied single-particle excitations.

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Topological bulk phases have a valence-conduction band inversion that must become disentangled at an interface to vacuum (*i.e.* at a surface), giving rise to topologically protected gapless, conducting surface states.[1] In three dimensions (3D), a Z_2 topological insulator (TI) is characterized by a $Z_2 = 0$ or 1 topological invariant. Semimetals can also be classified by topological invariants, with the four-fold degenerate (including spin) Dirac point being the canonical example. Symmetry lowering has a strong impact. For a non-centrosymmetric (broken inversion symmetry (IS)) or magnetic (broken time reversal symmetry [TRS]) material, the effective low energy Hamiltonian for linear band crossing $\hbar v_F(\vec{\tau} \cdot \vec{k})$, where $\vec{\tau}$ are the Pauli matrices, has eigenvalues $E = \pm \hbar v_F |\vec{k}|$, with the sign providing the helicity of the Weyl particles. There is an additional type of degeneracy, that of a conventional zero-gap semiconductor with quadratically touching bands that has no net topological character.

Intermediate between these degeneracies lie multi-Weyl points[2], from which dispersion proceeds as *either linear (massless) or quadratic (massive)* depending on direction. Multi-Weyl points are 3D generalizations of the semi-Dirac point in 2D that displays linear or quadratic dispersion depending on direction.[3, 4]. Such points reflect a universe in which the form of the kinetic energy operator depends on direction. These points are protected by symmetry, however the semi-Dirac point in a broken TRS system can be driven by spin-orbit coupling to a Chern insulator phase.[5]

To date several structural classes of topological materials have been discovered or predicted, beginning with the topological insulator $\text{Bi}_{1-x}\text{Sb}_x$ alloys,[6] Bi_2Se_3 and related families.[7–11] Other materials, *viz.* HgTe [12], α - Sn (gray tin) and Halide perovskites [13], are not intrinsically topological but become so by tuning with strain to induce a topological phase. Likewise, families of topo-

logical semimetals have been predicted and pursued.[14–18] Surface probes such as angle resolved photoemission spectroscopy (ARPES) and scanning tunneling microscopy (STM) are used to identify the corresponding topological surface states. In all of these systems, it is *sp* bands that provide the topological character.

The antiperovskite (APv) structure class of alkaline earth and pnictides compounds has been proposed to harbor potential topological insulating phases.[19, 20] Our density functional theory based survey of the entire class of $3 \times 5 \times 5$ possible alkaline earth-pnictide APv compounds,[20] *viz.* $Ae_3Pn_A Pn_B$, where $Ae = \text{Ca, Sr, Ba}$ and $Pn_A, Pn_B = \text{N, P, As, Sb, Bi}$, confirmed that some of these compounds in the assumed cubic structure host, or are very close to, non-trivial topological phases. In this paper we choose a representative member of this new class and demonstrate that a sequence of symmetry lowering changes can propel a given compound through a cascade of phases that includes all of the above mentioned phases and into a new class in which both Dirac and multi-Weyl excitations lie in the close vicinity of the Fermi level, thus determining the transport and low-energy bulk excitations and providing connected Fermi arcs in the boundary spectrum.

This cubic APv structure (space group $Pm\bar{3}m$) has vertex-sharing Ae_6Pn_B octahedra, with Pn_A^{3+} ions in the interstitial A sites surrounded symmetrically by eight octahedra. Several nitrides in this class were surveyed by Niewa.[21] Our notation of elements Ae , Pn_A , and Pn_B follows that of an antiperovskite with general chemical formula X_3AB , where the A cation is 12-fold cubo-octahedral coordinated and B is 6-fold coordinated by an octahedron of X cations. The central role of the anion was discussed previously; unlike in the oxide perovskites, the A site rather than the B site plays a central role in the electronic structure as it provides the uppermost valence bands that border the gap or invert with the conduction

bands.

To obtain the electronic structure of the APv compounds $Ae_3Pn_A Pn_B$, the all-electron DFT method full-potential local orbital (FPLO) method,[22] was used with the generalized gradient approximation (GGA) exchange correlation of Perdew, Burke, and Ernzerhof (1996).[23] A dense $20 \times 20 \times 20$ k -mesh was used for self-consistency because of the delicate band overlap near Γ . Spin-orbit coupling (SOC) was included by using the fully relativistic four component Kohn-Sham-Dirac equation implemented in FPLO, without resorting to the intermediate scalar relativistic approximation.

Analysis of the band topology was done by calculating the Z_2 topological invariants following the Fu-Kane prescription, based on parity eigenvalues at the time-reversal invariant momenta (TRIM),[24] which are available from the FPLO code. About a dozen compounds have non-zero Z_2 invariants in the assumed cubic structure.[20] The surface spectrum was obtained by first obtaining the maximally-localized Wannier functions and corresponding hopping amplitudes.[25, 26] A semi-infinite system is then constructed and the surface spectrum is calculated using the iterative surface Green's function method [27, 28] with a tight binding hopping cut-off of 4 unit cells.

Our computational survey[20] established that of two given pnictides, the stable structure always has the largest of the pair occupying the A site. This anion provides the valence band maximum states; being the heaviest of the two pnictide atoms it provides the strongest SOC splitting, thus tending to promote inversion of valence and conduction bands and hence topological character. The lower conduction bands have Ae d orbital character. The cubic aPV compounds are mostly narrow gap semiconductors, with both conduction band minimum and valence band maximum occurring at Γ , giving SOC a central role in band inversion.

When band inversion occurs, Pn p orbitals with odd parity at Γ are shifted above the lowest a band which has even parity, due to p eigenvalue splitting by SOC. Fig. 1 shows a rapid, almost abrupt, shift in the s special character of Bi along $X - \Gamma - Z$. The three-fold cubic degeneracy at the Γ point has to be connected by one conduction and two valence bands and results in a TSM state. By symmetry breaking, these TSM can be transformed into TI and DSM.

Uniaxial compression. As a specific example we consider Ca_3BiP , one of the narrow gap APv semiconductors, as an example of a TI with Z_2 indices (1;000) before SOC is included. It has a calculated direct band gap of 0.16 eV at the Γ point and the p orbitals of the A site anion, Bi, lie at the bottom of the gap. SOC inverts the band ordering with an inversion energy of 0.28 eV, producing a topological band-touching semimetal due to degenerate $p_{3/2}$ states pinning the Fermi level. A compressive uniaxial strain in the range 1 – 7% (the range we have studied) breaks the crystal symmetry while maintaining

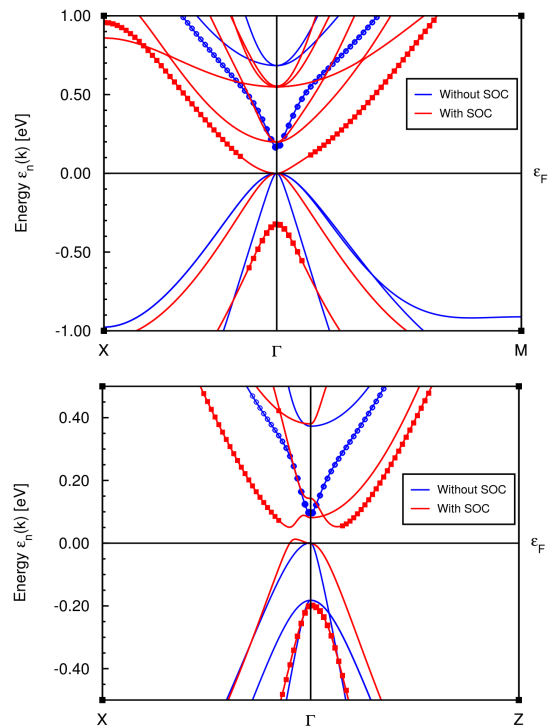


FIG. 1: Top panel: without SOC (blue) cubic Ca_3BiP is a topological insulator with an anomalously small electron mass. SOC (red) re-orders the bands leaving a zero-gap semimetal. The large symbols denote Bi $6s$ character, demonstrating the sp band re-ordering with SOC. Bottom panel: 5% tetragonal compressive strain in Ca_3BiP results in a TI phase. The valence-conduction inversion energy is 0.28 eV.

the inverted band ordering, and opens up a gap of 40 meV at the maximum strain, resulting in a TI phase. A representative band structure is shown in Fig. 1.

Topologically protected gapless surface states serve as direct confirmation of the topological bulk bands, and also serve directly as a bridge to experiment. The conducting character of these surface states is known to be robust against disorder, except when introduced by magnetic impurities (breaking TRS).[29] As shown in Fig. 2, both (001) and (100) surfaces have surface states crossing the bulk gap of 30 meV. The (001) surface shows bands straightforwardly connecting the valence and conduction states, while the topological surface bands on the (100) surface incorporate a single Dirac point. These topological surface states are similar to those observed in Bi_2Te_3 , Sb_2Te_3 and Bi_2Se_3 .

Uniaxial Expansion. Expansion of the lattice parameter c of Ca_3BiP by 1 – 5% (the range we studied) also retains the band inverted ordering, but the different sign of eigenvalue splitting by SOC produces a different result. The surface spectrum displays a (two-fold) linear band crossing along $\Gamma - Z$ and gapped elsewhere as shown in Fig. 3. The band crossing point (4-fold degenerated including spins) is a signature of a quadruple Dirac point,

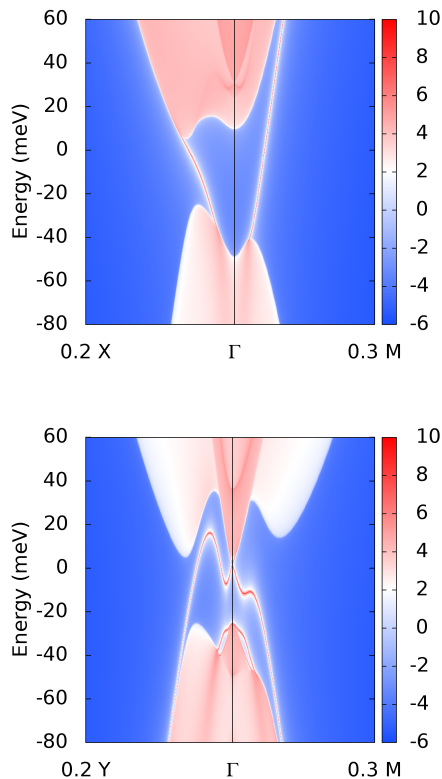


FIG. 2: Energy and momentum dependent spectral density of a semi-infinite crystal of (001)-compressed (5%) Ca_3BiP terminated on (001) [top panel] and (100) [bottom panel] surfaces. Warmer colors represent higher intensity.

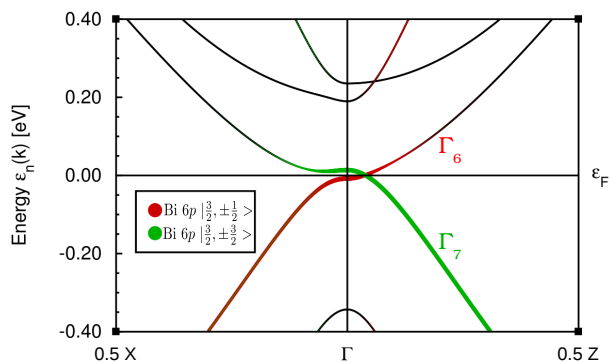


FIG. 3: Band structure of Ca_3BiP with tensile strain of 1% along (001) shows a Dirac point at $\vec{k} = (0, 0, k_o)$, where $k_o = 0.022 \text{ \AA}^{-1}$. Around k_o , the bands have $\text{Bi } 6p \left| \frac{3}{2}, \pm \frac{1}{2} \right\rangle$ and $\left| \frac{3}{2}, \pm \frac{3}{2} \right\rangle$ characteristics.

and unavoidable as the bands have different irreducible representations, i.e. Γ_6 and Γ_7 , as characterized and protected by the C_{4v} rotational point group symmetry.

The (100) surface shows topological surface bands

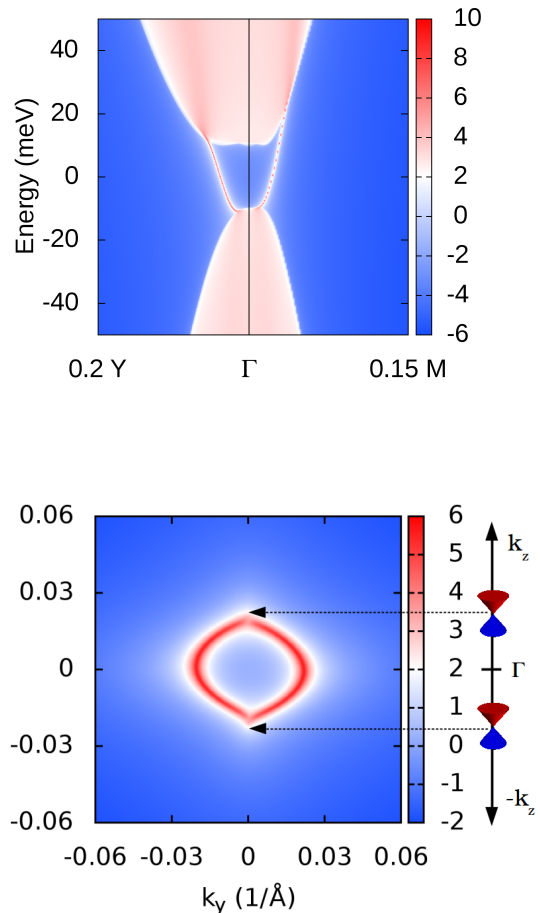


FIG. 4: Top panel: energy and momentum dependent spectral density of a semi-infinite crystal of (001)-expanded Ca_3BiP terminated on the (100) surface. Bottom panel: the (100) surface shows two half-circle Fermi arcs connecting projections of Dirac points along $\pm k_z$.

crossing the projected bulk band gap of about 20 meV (Fig. 4). The Fermi surface exhibits two topologically non-trivial Fermi arcs touching at the Dirac nodes $\vec{k} = (0, 0, \pm k_o)$ protected by both IS and TRS. This single pair of Fermi arcs connecting two Dirac points, like the Fermi arc connecting Weyl points in a WSM, is possible to exist, as a Dirac point can be treated as two Weyl points with opposite chirality, which in this case do not annihilate each other since the degeneracy is protected by the crystal symmetry. One notable property of the Fermi arc is that, ideally, electron density on the Fermi arc of one surface can sink through the bulk to the Fermi arc of the opposite surface, while eigenvalues remain continuous.

Low energy description. An effective 4×4 Hamiltonian generalized from those used for Na_3Bi [30] and

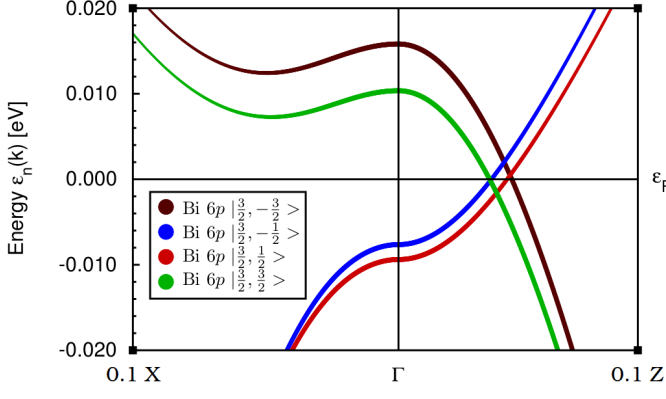


FIG. 5: Weyl nodes at $k_o^* = (0, 0, k_o^*)$ under the external magnetic field of $B = 24\text{T}$. The bands are mostly split along k_z and minimally in energy.

Cd_3As_2 [31] can be adopted to describe the Dirac band crossing at k_o in Fig. 3, with Bi $6p$ $|\frac{3}{2}, \pm\frac{1}{2}\rangle$ and $|\frac{3}{2}, \pm\frac{3}{2}\rangle$ states as the basis, in the order of $J_z = \frac{1}{2}, -\frac{3}{2}, -\frac{1}{2}, \frac{3}{2}$, after SOC. The Hamiltonian for the C_4 symmetry of Ca_3BiP around Γ reads, after (001) strain,

$$H_\Gamma(k) = \varepsilon_0(k) + \begin{pmatrix} M(k) & Ak_+ & 0 & 0 \\ Ak_- & -M(k) & 0 & 0 \\ 0 & 0 & M(k) & -Ak_- \\ 0 & 0 & -Ak_+ & -M(k) \end{pmatrix} \quad (1)$$

where the bands are $M(k) = M_0 - M_1k_z^2 - M_2(k_x^2 + k_y^2)$ with negative constants to reproduce band inversion, $k_\pm = k_x \pm ik_y$ and A provides the scale for coupling to uniaxial strain along \hat{z} . $k_o = \sqrt{M_0/M_1}$ gives the band crossing point along \hat{z} , and the dispersion around k_o yields $E_{k_o} = \pm\sqrt{[2k_o\delta k_z - M_2(\delta k_x^2 + \delta k_y^2)]^2 + A^2(\delta k_x^2 + \delta k_y^2)}$.

Time reversal symmetry breaking. The Hamiltonian in Eq. 1 remains block-diagonal, containing two multi-Weyl Hamiltonian systems of opposite chirality. Symmetry can be broken further by violating IS or TR symmetry. Breaking IS is not an easy route, since the four-fold symmetry of Eq. 1 is to be preserved, otherwise it will open a gap and generate massive fermions. A simpler means is to apply an external Zeeman magnetic field B coupling to the SOC basis sets, which preserves the crystal symmetry but breaks TRS,[32]

$$H_{\text{Zeeman}}(k) = H_\Gamma(k) + \begin{pmatrix} \Delta p_1 & 0 \\ 0 & \Delta p_2 \end{pmatrix} \otimes \sigma_z \quad (2)$$

where $\Delta p_{1,2} = \frac{1}{2}g_{1,2}\mu_0 B$ for Bi $6p$ $|\frac{3}{2}, \pm\frac{1}{2}\rangle$ and $|\frac{3}{2}, \pm\frac{3}{2}\rangle$ states respectively. Eq. 2 can be block-diagonalized into 2×2 matrices,

$$H_\pm(k) = \left[M(k) \pm \frac{(\Delta p_1 - \Delta p_2)}{2} \right] \sigma_z \pm \left[\frac{(\Delta p_1 + \Delta p_2)}{2} \right] I \pm A(k_x \sigma_x + k_y \sigma_y) \quad (3)$$

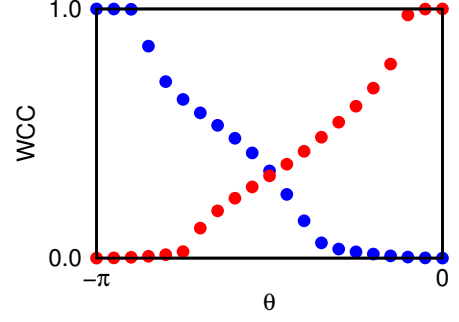


FIG. 6: The Wannier charge center (WCC) is computed for each loop at a constant azimuthal angle θ around a sphere surrounding Weyl points at $k_z = 0.020\text{\AA}^{-1}$ (blue) and 0.024\AA^{-1} (red). The change in the charge polarization for each loop from south ($\theta = -\pi$) to north ($\theta = 0$) for blue and red curves indicates -1 and $+1$ chiralities, respectively.

The Weyl nodes are displaced to $k_o^* = (0, 0, k_o^*)$, where $k_o^* = \sqrt{\frac{M_0}{M_1} \pm \frac{\Delta p_1 - \Delta p_2}{2M_1}}$. Expansion around k_o^* gives $E_{k_o^*} = \pm\sqrt{[2k_o^*\delta k_z - M_2(\delta k_x^2 + \delta k_y^2)]^2 + A^2(\delta k_x^2 + \delta k_y^2)} \pm (\Delta p_1 + \Delta p_2)/2$, where the Weyl Hamiltonians are decoupled along \hat{z} into k_o^* and shifted in energy by the average of the magnetic energies.

Fig. 5 illustrates the final spectrum for $B=24\text{ T}$ ($\mu_B B \approx 1\text{ meV}$), for which the degeneracies are located at $k_{z,j} = \pm 0.020\text{\AA}^{-1}, \pm 0.022\text{\AA}^{-1}, \pm 0.023\text{\AA}^{-1}, \pm 0.024\text{\AA}^{-1}$. The former and latter lie at E_F and occur for bands of opposite sign in J_z , while the middle two are for equal sign in J_z occur above and below E_F . We observe that the nodes lying at E_F are *multi-Weyl points*: linear dispersion along k_z as shown in Fig. 5, and quadratic in the k_x, k_y plane. A quantitative description in a tight binding scheme derived from maximally localized Wannier functions[25] is provided in the Supplemental Material. Our calculation on Na_3Bi also shows similar behavior.

The flow of the hybrid Wannier charge center (WCC) provides direct confirmation of the topological character. By constructing a closed 2D cut of the BZ containing a Weyl node, the evolution of the band sum of the hybrid WCCs around the closed cut can be calculated to determine the Weyl chirality.[33] Fig. 6 shows the evolution of the charge polarization of the two Weyl nodes at E_F , which indicates their corresponding chiralities $C_w = (-1, +1)$.

In summary, we have shown that members of this class of APv materials can be driven into various topological phases by symmetry breaking. SOC, strain, and magnetic field can drive a cascade through a variety of topological phases. SOC drives or deepens band inversion by splitting the $P_n p_{3/2}$ eigenvalues upward beyond the s conduction band at Γ , leaving a topological band-touching semimetal. Uniaxial strain breaks the degeneracy

acy and, if compressive, produces a topological insulator, while if tensile, gives a Dirac semimetal. At that point, application of a magnetic field breaks TRS and splits each Dirac node into a pair of Weyl nodes with opposite chirality, yielding a multi-Weyl points whose bounding projections are joined by Fermi arcs.

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