

Stacking-insensitive ferroelectricity in layered ScPS_3 : Lone-pair-driven polarization and strain-stabilized ferroelectricity against the antiferroelectric ground state

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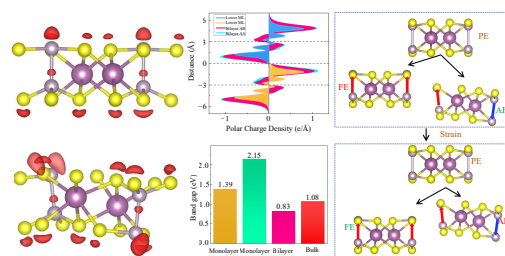
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ABSTRACT

Interlayer stacking dependence of polarization remains a critical bottleneck for 2D vdW ferroelectrics in nanoelectronics. Herein, first-principles calculations reveal stacking-insensitive ferroelectricity in layered ScPS₃, driven by out-of-plane perturbation of the P–P dimers. Unlike conventional ion-displacement ferroelectrics such as CuInP₂S₆, where polarization comes from metal-ion off-centering, the polarization of ScPS₃ arises from local ionic displacement within the P–P dimers together with the the P lone pairs. This polarization originates from the intralayer asymmetry of the monolayer and remains nearly unchanged for different stacking orders (AA, AB), circumventing the stringent fabrication tolerances of sliding ferroelectrics. Although the antiferroelectric (AFE) phase is identified as the thermodynamic ground state and the ferroelectric (FE) phase is metastable at zero strain, biaxial strain beyond $\pm 6\%$ stabilizes the FE phase and enables a reversible AFE-to-FE phase transition. Together with the low switching barriers and the stacking-independent polarization ($\sim 0.73\text{--}0.74\text{ }\mu\text{C}/\text{cm}^2$), these results establish ScPS₃ as a versatile platform for strain-engineered memories and flexible optoelectronics.

Keywords two-dimensional ferroelectrics, antiferroelectricity, lone-pair activity, first-principles calculations, ScPS₃, strain engineering



1 Introduction

Two-dimensional van der Waals (vdW) ferroelectrics are promising for ultra-scaled, low-power non-volatile memories thanks to their atomically thin profiles, dangling-bond-free surfaces, and compatibility with existing semiconductor technologies [1–3]. However, practical deployment faces two bottlenecks: polarization suppression near the monolayer limit and extreme sensitivity to

interlayer stacking [4, 5]. In most vdW systems, even a slight lateral shift between adjacent layers can reverse or quench the net polarization, imposing fabrication tolerances that impede scalable integration.

The MPX₃ family (M = metal, P = phosphorus, X = chalcogen) offers a wide range of properties, from semiconductors and magnets to multiferroics, owing to its layered structure and diverse elemental combinations [6, 7]. Recent advances in synthesis enable the controlled



growth of dozens of MPX_3 compounds by chemical vapor deposition (CVD) via competitive reaction mechanisms [8]. Under pressure or strain, they display an exceptionally wide range of phenomena, including magnetic and multiferroic properties [9, 10], spin-flop transitions [11], the anomalous Hall effect [12], strong anisotropy in carrier effective mass [13], and an insulator-to-superconductor transition [14].

Among these, CuInP_2S_6 (CIPS) is the most extensively studied ferroelectric member. Its ferroelectricity originates from the off-center displacement of Cu^+ ions: in the high-temperature paraelectric phase, Cu^+ sits at the inversion center; below the Curie temperature it shifts off-center, breaking inversion symmetry and generating spontaneous polarization [15–17]. This polarization is switchable under an external electric field and exhibits good stability [18, 19]. However, CIPS suffers from high switching barriers and a strong dependence of polarization on stacking order — a common drawback of ion-displacement layered ferroelectrics.

Notably, a large family of 2D ferroelectric materials was discovered in bimetallic phosphates via machine learning. Their ferroelectricity originates from spontaneous symmetry breaking induced by the opposite displacements of bimetal atoms [20]. In contrast, recent first-principles calculations attribute the ferroelectricity of ScPS_3 to out-of-plane P–P dimer perturbation and the associated charge redistribution, rather than to metal-ion migration [20, 21]. This mechanism, enabled by the structural flexibility of the dimers and the light Sc mass, may favor fast switching. On the other hand, experiments have observed the coexistence of ferroelectric (FE) and antiferroelectric (AFE) states in $\text{CuInP}_2\text{Se}_6$ [22]. In low-dimensional structures, the AFE phase is often lower in energy than the FE phase and thus thermodynamically more favorable [23]. Nevertheless, the AFE phase of ScPS_3 remains poorly studied, and its relative stability is unclear.

Interlayer stacking order is known to be a critical factor for the ferroelectric stability of 2D vdW materials, and inappropriate stacking can eliminate ferroelectricity entirely. For instance, in typical MPX_3 members and other 2D ferroelectrics, centrosymmetric stackings (e.g., AB) preserve the inversion symmetry that ferroelectricity must break, thereby disfavoring a net out-of-plane polarization and quenching the spontaneous polarization [4, 5]. In ScPS_3 , by contrast, the polarization is expected to arise from intralayer lone-pair activity rather than from the interlayer stacking itself, and should therefore be far less sensitive to the stacking order. Although this ferroelectricity is proposed to originate from P–P dimer perturbation, how interlayer stacking affects its FE stability and how stacking configurations suppress ferroelectricity remain unclear. This gap, together with the poorly characterized AFE phase, limits the understanding of ferroelectric modulation and hinders practical device applications of ScPS_3 .

In this work, we perform systematic first-principles calculations and reveal three key findings. First, the AFE phase is the thermodynamic ground state, yet the FE phase can be stabilized by biaxial strain, enabling a reversible $\text{AFE} \rightarrow \text{FE}$ transition with a barrier as low as 0.41 eV. Second, the spontaneous polarization arises mainly from intralayer lone-pair stereochemical activity combined with P–P dimer perturbation; remarkably, its magnitude is nearly identical for AA and AB stackings, showing a stacking insensitivity that circumvents the stringent fabrication requirements of conventional sliding ferroelectrics. Third, the switching barriers for $\text{FE} \leftrightarrow \text{AFE}$ (0.28 eV for $\text{FE} \rightarrow \text{AFE}$ and 0.41 eV for $\text{AFE} \rightarrow \text{FE}$) are much lower than those of perovskite ferroelectrics and comparable to the best sliding-type 2D systems; the polarization ($\sim 0.74 \mu\text{C}/\text{cm}^2$) lies well within the detectable range and can be tuned continuously by strain. In addition, the band gap and optical absorption depend systematically on layer thickness and strain, offering new routes for strain-engineered optoelectronics. Collectively, these characteristics establish ScPS_3 both as a model system for lone-pair-driven ferroelectricity in vdW materials and as a practical candidate for low-power memory and flexible optoelectronics with high tolerance to processing variations.

2 Computational methods

All calculations are performed within the framework of density functional theory (DFT) as implemented in the Vienna Ab initio Simulation Package (VASP) [24, 25]. Structural optimizations use the Perdew–Burke–Ernzerhof (PBE) exchange–correlation functional [26] within the generalized gradient approximation (GGA) [27]. The electron–ion interactions are described by the projector augmented wave (PAW) pseudopotentials. The band structures presented in Figs. 1(f) and (g) are obtained at the PBE level, whereas the electronic and optical properties reported in Section 3.3 are from HSE06 hybrid functional [28] with the exact exchange mixing parameter α set to the default value of 0.25. A 15 Å vacuum layer is inserted perpendicular to the ab plane to eliminate spurious interlayer interactions.

Atomic positions and lattice parameters are fully relaxed until the maximum force on each atom is less than $1 \times 10^{-3} \text{ eV}/\text{\AA}$ and the total energy difference between consecutive steps is smaller than $1 \times 10^{-8} \text{ eV}$. A plane-wave cutoff energy of 500 eV is adopted to balance computational accuracy and efficiency. The Brillouin zone is sampled using a $9 \times 9 \times 1$ Monkhorst–Pack k -point mesh [29]. The van der Waals interactions were accounted for by employing the DFT-D3 method with Becke–Johnson damping [30, 31]. The energy barriers for the transition between the ferroelectric (FE) and antiferroelectric (AFE) phases are determined using the climbing-image nudged elastic band (CI-NEB) method.



VASPKit was used to process and analyze the calculated data [32]. The spontaneous polarization was obtained from the Berry phase approach (modern theory of polarization) implemented in VASP. Unless otherwise specified, all energy values in this work are normalized to one monolayer unit cell (2 f.u.).

3 Results and discussion

3.1 Lone-pair polarization origin

The crystal structure of monolayer ScPS_3 is characterized by a two-dimensional van der Waals layered architecture (Fig. S1). As depicted in Fig. 1(a), the Sc atoms form a hexagonal lattice, with P–P dimers positioned at the center of the hexagonal rings. S atoms bridge the metal (Sc) and P atoms, and each Sc atom is coordinated by six S atoms in an octahedral geometry via strong ionic-covalent bonds. This configuration renders the P–P dimers highly flexible, facilitating lattice distortion. The ferroelectric phase exhibits a non-centrosymmetric structure with the P–P dimers displaced out-of-plane along the *c*-axis, whereas the antiferroelectric phase is centrosymmetric with no net polarization.

The mechanism driving the spontaneous polarization was investigated through electron localization function (ELF) analysis and projected density of states (PDOS). The ELF isosurfaces (red lobes) in Figs. 1(b) and (c) visualize the stereochemically active lone pairs of the P atoms, which are the primary origin of ferroelectricity in ScPS_3 . This instability arises from a synergistic effect: the size mismatch between the P–P dimer triangular sublattice and the Sc hexagonal sublattice induces geometric strain, and the lone pairs respond to the local chemical environment of the Sc atoms. This broken inversion symmetry is corroborated by the PDOS in Fig. 1(d), which shows distinct electronic states for the upper and lower P atoms.

The lone pairs of the P atoms drive a charge reconstruction within the $[\text{P}_2\text{S}_6]^{4-}$ clusters, which breaks the inversion symmetry and produces an asymmetric charge distribution with separated charge centers [Fig. 1(e)]. The inequivalence of the upper and lower P-site states in the PDOS [Fig. 1(d)] confirms the loss of centrosymmetry, validating that the asymmetric lone-pair activity and the resulting charge separation constitute the microscopic origin of the spontaneous polarization.

Monolayer ScPS_3 exhibits indirect band-gap semiconductor behavior in both ferroelectric and antiferroelectric phases. At PBE level, the FE phase (0.76 eV, VBM at Γ , CBM at M) and AFE phase (1.35 eV, VBM at Y, CBM at S) exhibit markedly different band gaps and band edge positions. Both phases share Sc d-derived CBM and S p-derived VBM, which remain unchanged due to the preserved planar Sc–S bonding, while the P–P dimer distortion only affects non-band-edge states.

The PBE gaps here are systematically lower than the HSE06 values presented in Section 3.3.

3.2 Stacking-insensitive sliding

Interlayer sliding is recognized as a defining characteristic of two-dimensional van der Waals ferroelectrics, serving as a primary mechanism for modulating their performance. To investigate this, we fixed the top layer of bilayer ScPS_3 and tracked the structural evolution and energy variation of the bottom layer along the $[1\bar{1}]$ and $[10]$ directions. These pathways reveal a systematic evolution of the interlayer stacking sequence (Fig. 2 and Table S2). Throughout, interlayer interactions are dominated by weak vdW forces, with no bond breaking or reconstruction.

Significant anisotropy in the energy barriers is revealed in Fig. 2. Sliding along the $[10]$ direction occurs readily, whereas a much higher barrier is encountered along the $[1\bar{1}]$ direction [Fig. 2(b)]. This difference stems from the distinct intermediate stacking states formed along each path. Along the $[10]$ trajectory, the AC and AD stackings show low atomic steric hindrance and high lattice matching, acting as metastable states with low barrier peaks. Conversely, the AA stacking configuration was traversed along the $[1\bar{1}]$ path. Although the AA stacking itself is the lowest-energy configuration (see below), the saddle points connecting AB with AA and AA with AB' along this path involve strong atomic repulsion and pronounced lattice distortion, which raise the barrier peaks. Consequently, although both pathways permit periodic reversible sliding, the stacking transition along the $[1\bar{1}]$ direction requires stronger external fields.

In the sliding-energy profile [Fig. 2(b)], the total energy of AA-stacked bilayer ScPS_3 is lower than that of the AB-stacked counterpart by ~ 50 meV per monolayer unit cell (25 meV/f.u.), so the AA stacking is thermodynamically more favorable. Since this difference is comparable to the thermal energy at room temperature, both stacking forms may coexist. The total energy of AA is lower than that of the initial AB stacking and of the AC, AD, and AB' intermediates. This stability comes from the perfect vertical alignment of interlayer atoms, which maximizes the out-of-plane orbital overlap between Sc, P, and S atoms across adjacent layers and thus strengthens interlayer bonding. The calculated sliding barriers confirm that bilayer ScPS_3 has a much lower barrier than conventional perovskite ferroelectrics, so interlayer sliding is readily achieved under weak external fields.

3.3 Layer-dependent optoelectronic properties

Figure 3 presents the structural stability, interfacial charge dynamics, and ferroelectric polarization of bilayer ScPS_3 for the AA and AB stackings. As shown in Section 3.2, the AA stacking is only marginally more

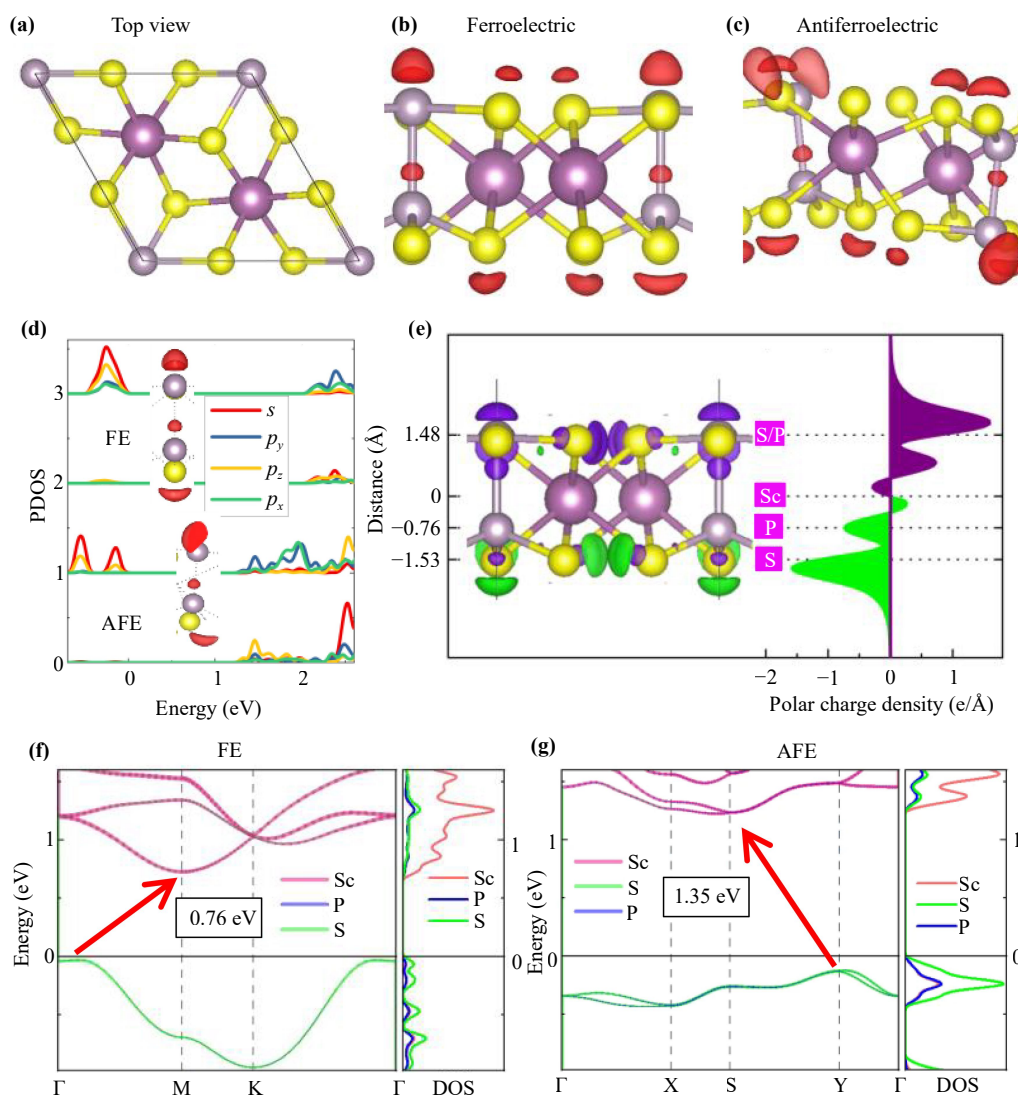


Fig. 1 Crystal structure and electronic properties of ScPS₃. (a) Top view of the monolayer phase. (b, c) Side views of the monolayer in the ferroelectric (FE) and antiferroelectric (AFE) phases, respectively. Electron localization function (ELF) isosurfaces (red) in both panels visualize the stereochemical activity of P lone pairs. (d) Projected density of states (PDOS) of the upper and lower P atoms in the monolayer FE phase. (e) Planar-averaged polar charge density along the *c*-axis for the monolayer FE and AFE phases. (f, g) Projected electronic band structures of the monolayer in the FE and AFE phases, respectively.

stable than AB, so both stacking modes may coexist. The charge density difference, which reflects the weak interlayer interactions, is shown in Figs. 3(a) and (b), with its planar average in Fig. 3(c) and the corresponding bilayer and bulk structures in Fig. S3: a subtle charge redistribution (localized near S and Sc atoms) occurs without covalent bonding, confirming van der Waals coupling.

Sliding is central to ferroelectric storage, flexible electronics, and energy conversion [33]. The spatial distribution of polarized charge around P atoms is illustrated in Figs. 3(d) and (e): it remains strikingly consistent across AA and AB stackings despite their distinct geometries. Quantitative integration in Fig. 3(f) confirms nearly

identical polarization magnitudes, with spontaneous polarization values of $\sim 0.73 \mu\text{C}/\text{cm}^2$ for the AA stacking and $\sim 0.74 \mu\text{C}/\text{cm}^2$ for the AB stacking. The negligible difference ($<2\%$) confirms the stacking insensitivity of the ferroelectricity.

The origin of polarization must be distinguished from the effect of sliding. The ferroelectricity in ScPS₃ is driven by the stereochemical activity of P lone pairs, which induces intralayer P–P dimer distortion and generates a spontaneous out-of-plane polarization. This mechanism differs fundamentally from sliding ferroelectricity, where polarization arises solely from symmetry breaking by interlayer lateral shifts. The observed stacking insensitivity refers specifically to the robustness of the

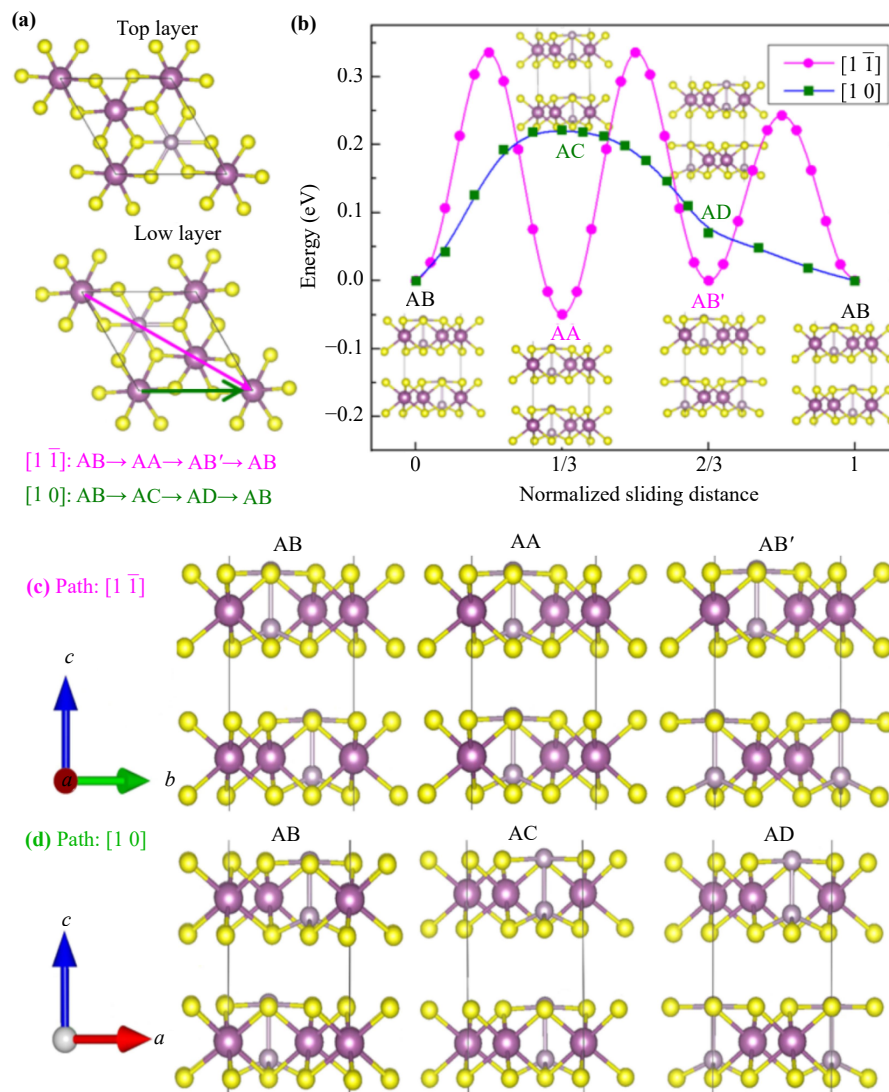


Fig. 2 Interlayer sliding phase transitions in two-dimensional bilayer ScPS₃. **(a)** Schematic diagram of the bilayer stacking configuration: the top atomic layer remains stationary while the low layer undergoes lateral sliding. The red arrow denotes the sliding path along the $[1 \bar{1}]$ crystallographic direction, and the green arrow represents the sliding trajectory along the $[1 0]$ crystallographic direction. **(b)** energy barrier via interlayer sliding. The energy is referenced to the AB-stacking configuration and normalized to one monolayer unit cell (2 f.u.). **(c, d)** Structure evolution via layer sliding along the $[1 \bar{1}]$ and $[1 0]$, respectively.

polarization magnitude against different stacking orders (AA vs. AB). By contrast, sliding modulates the total energy, as evidenced by the calculated barriers, and thus governs the mechanical stability and phase transition pathways. The weak interlayer vdW coupling preserves polarization stability while allowing structural flexibility, ensuring robust performance under strain. This relaxes the strict stacking control required in conventional layered ferroelectrics.

Electronic structure determines the optoelectronic response and device applications of ferroelectric materials. To correct the band-gap underestimation of pure DFT, the HSE06 hybrid functional is used for the electronic structure of ScPS₃; all band gaps and optical properties

in this section are therefore HSE06 values, systematically larger than the PBE values of Section 3.1. The band structures of ScPS₃ at different dimensionalities show that all ferroelectric phases are indirect band-gap semiconductors, with the valence band maximum and conduction band minimum at different high-symmetry points of the Brillouin zone.

Figure 3(g) shows the HSE06 band gap as a function of layer thickness and phase. The band gap of ScPS₃ follows the order: Monolayer AFE > Monolayer FE > Bulk > Bilayer (Fig. S4). The monolayer AFE phase has the widest band gap (2.15 eV), and the bilayer FE phase the narrowest (0.83 eV). The atomic displacement and charge rearrangement induced by ferroelectric polarization

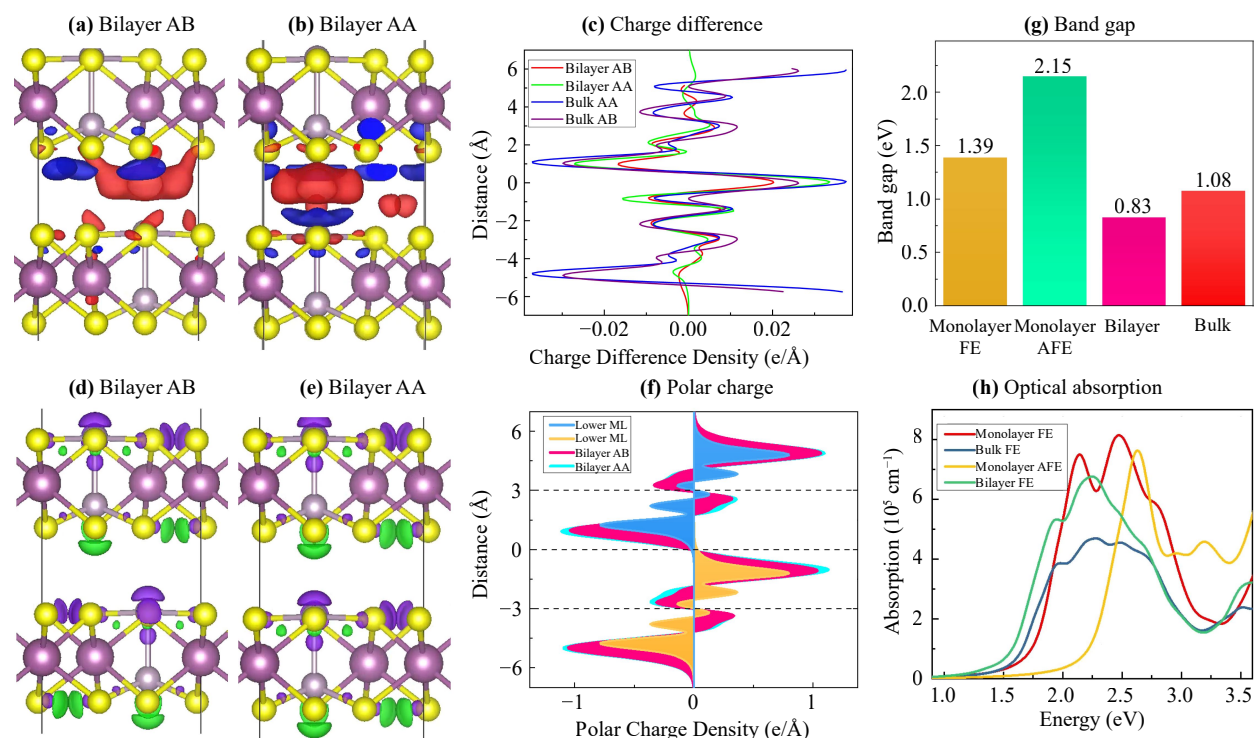


Fig. 3 Structural, electronic, and polarization properties of layered ScPS₃ across AA and AB stackings. (a, b) Charge density difference of ferroelectric bilayer AA and AB, respectively. (c) Planar-averaged differential charge density along the *c* direction. (d, e) Polar charge density of ferroelectric bilayer AA and AB, respectively. (f) Planar-averaged polar charge density with respect to that of monolayers. (g, h) Band gap variation and optical absorption spectrum of monolayer, bilayer, and bulk ScPS₃, respectively.

and interlayer interactions reduce the band gap via enhanced orbital hybridization, showing that dimensionality and phase state strongly regulate the electronic properties.

Energy research, spanning photovoltaics [34, 35], photocatalysis [36, 37], and electrochemical energy storage [38–40], is increasingly important for sustainable development. To evaluate the optoelectronic potential of ScPS₃, the optical absorption was calculated [Fig. 3(h)]. All configurations absorb strongly in the visible and ultraviolet, with absorption coefficients of the order of 10⁵ cm⁻¹, and the spectral shape correlates with the calculated band gap. Specifically, the bilayer FE phase, with a moderate band gap of 0.83 eV, shows broad absorption in the lower-energy (visible) range, with a maximum absorption coefficient of $\sim 7.0 \times 10^5$ cm⁻¹. This red-shifted edge, enabled by its narrow band gap, gives the bilayer FE phase broad-band light harvesting across the near-infrared and visible ranges, making it promising for broadband photodetection and photovoltaics. A rigorous assessment of any such application would also require the band-edge positions relative to the redox potentials, which are not accessible from the present calculations.

The monolayer FE phase has an indirect HSE06 band gap of 1.39 eV, with the VBM at Γ and the CBM at M

[Fig. S4(b)], and shows the strongest optical response of all phases, with a peak absorption exceeding 8.0×10^5 cm⁻¹. This strong response stems from a gap well matched to the visible spectrum, which yields a high joint density of states for band-edge transitions. The monolayer AFE phase, with the largest band gap of 2.15 eV, shows a sharp high-energy absorption peak, indicating a shift of spectral weight toward the higher-energy side of the visible region. This blue shift is consistent with its wide band gap, which suppresses low-energy visible absorption and makes it a candidate for blue and near-ultraviolet photodetection. The bulk FE phase (1.08 eV) shows an intermediate absorption profile of moderate intensity.

These results show that the band gap, absorption intensity, and spectral shape of ScPS₃ can be tuned continuously by layer thickness and ferroelectric phase. The correlation between band structure and optical response clarifies the structure–property relationship in ScPS₃ and offers a strategy for designing ScPS₃-based optoelectronic devices with tailored performance, from near-infrared/visible photodetection to blue and near-ultraviolet optoelectronics.

3.4 Strain-tunable AFE–FE transition

The energy curves along the ferroelectric switching path-

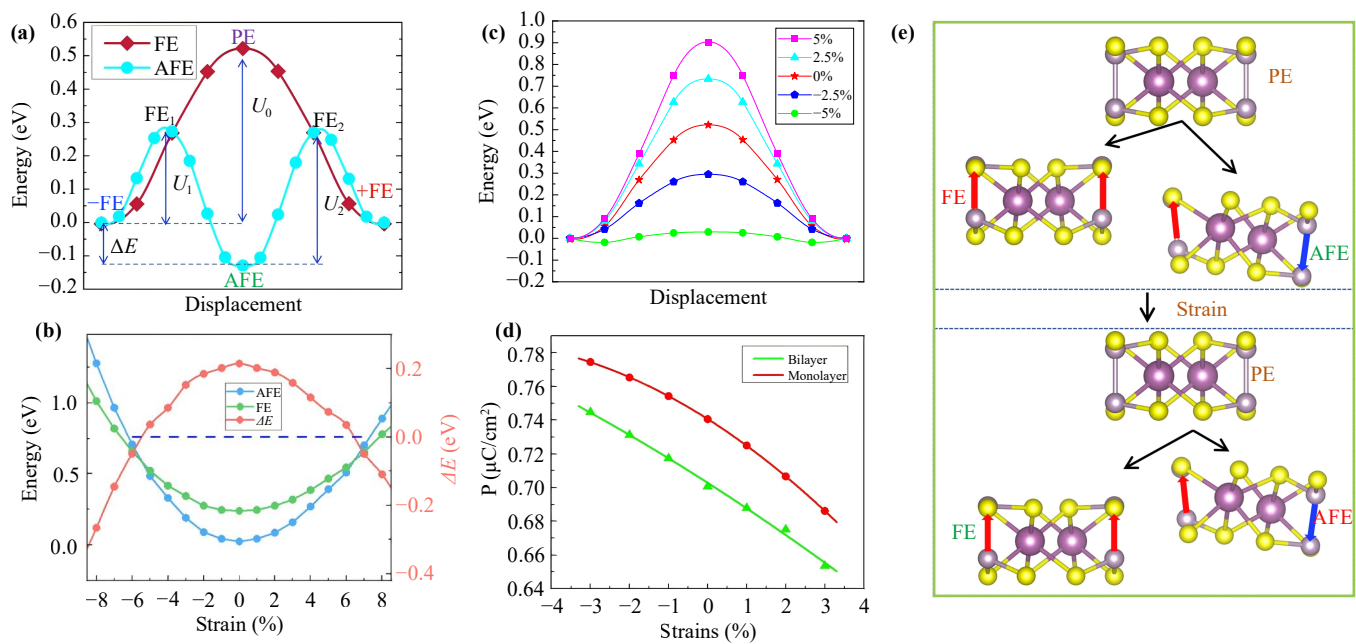


Fig. 4 (a) The ferroelectric switching pathways of monolayer ScPS₃. (b) Energy evolution of ferroelectric monolayer ScPS₃ under biaxial strain. (c) Ferroelectric switching barriers under different applied strains. (d) Polarization versus strain curves for monolayer and bilayer ScPS₃. (e) Atomic structural schematics showing strain-induced transformation between ferroelectric (FE) and antiferroelectric (AFE) phases as well as the paraelectric (PE) reference structure, in which red and blue line segments denote the polarization directions, respectively.

way [Fig. 4(a)] show a characteristic double-well profile and clear bistability. The antiferroelectric phase is confirmed as the thermodynamic ground state, while the ferroelectric phase is identified as metastable, with an energy difference of 0.13 eV between them (Table S1). The FE→PE (paraelectric) barrier is 0.52 eV and the FE→AFE relaxation barrier only 0.28 eV, whereas the reverse AFE→FE barrier is as high as 0.41 eV, indicating considerable kinetic stability of the AFE phase and the need for a strong external stimulus to reverse it.

Unlike conventional three-dimensional perovskite ferroelectrics such as BaTiO₃ and PbTiO₃ [41], ScPS₃ switches through local P–P dimer perturbation and weak interlayer vdW sliding, without long-range ion migration, with barriers much lower than those of perovskites (>1.2 eV). Compared with two-dimensional sliding ferroelectrics such as MoS₂, h-BN and representative CuInP₂S₆ [42–44], ScPS₃ achieves lower barriers through the synergistic effect of structural distortion and interlayer sliding.

Strain engineering is also an effective tool for modulating the electronic structure of 2D systems. For instance, shear strain switches spin splitting and activates piezomagnetic responses in altermagnetic materials, highlighting the versatility of strain-induced symmetry breaking [45]. The biaxial strain effect on phase stability is summarized in Fig. 4(b). AFE remains the energetically favored ground state within −6% to +6% strain, while the system switches to FE phase once strain exceeds

±6%, as elastic stress overcomes the 0.41 eV zero-strain AFE→FE barrier. The transition originates from the competition between elastic energy and dipole alignment energy, establishing strain engineering as an effective band-tuning strategy for 2D ferroelectrics [46].

The switching barrier increases monotonically from −5% (compressive) to +5% (tensile) strain [Fig. 4(c)]. Compressive strain strongly suppresses the barrier (<0.1 eV at −5%), favoring polarization switching, whereas tensile strain raises it markedly (~0.9 eV at +5%), stabilizing the polarized state. At the critical strain of ±6%, the energy difference between the AFE and FE phases approaches zero, providing the thermodynamic condition for the transition.

Figure 4(d) plots the polarization as a function of strain for monolayer and bilayer ScPS₃. For both systems, the polarization decreases monotonically. At the same strain, the polarization of the monolayer is comparable to or slightly higher than that of the bilayer, reflecting the influence of dimensionality. The monolayer polarization ranges from ~0.68 to 0.78 $\mu\text{C}/\text{cm}^2$, lower than that of typical displacement-type ferroelectrics such as CuInP₂S₆ (~3–5 $\mu\text{C}/\text{cm}^2$) [47], but considerably higher than that of weakly polar 2D systems such as α -In₂Se₃ (~0.1–0.3 $\mu\text{C}/\text{cm}^2$) and nonpolar h-BN [48]. Moreover, the polarization of bilayer ScPS₃ is independent of stacking configuration, with nearly identical polar charge distributions for AA and AB.

The strain-driven AFE→FE transition is directly

revealed by the atomic schematics in Fig. 4(e). Red and blue line segments denote the polarization directions of the FE and AFE phases, respectively. The paraelectric reference structure is shown for comparison [Fig. S2(c)]. At zero strain and within -6% to $+6\%$, the system adopts the AFE ground state. Adjacent P–P dimers alternate in antiparallel displacement along the c -axis, so their dipoles cancel macroscopically and match the lattice periodicity. When compressive strain exceeds -6% (i.e., $<-6\%$), the in-plane contraction breaks the antiparallel balance. All P–P dimers then shift in the same direction along the c -axis, yielding a net polarization and driving the transition to the FE phase. Meanwhile, asymmetric distortion of the S coordination polyhedra around Sc atoms further stabilizes the non-centrosymmetric structure. Similarly, when tensile strain exceeds $+6\%$ (i.e., $>+6\%$), the excessive in-plane stretching destabilizes the antiparallel arrangement. The dimers adopt parallel alignment to lower the energy, again driving the AFE→FE transition.

The atomic-scale picture is quantitatively corroborated by the strain dependence of the P–P bond length l and the layer thickness h summarized in Table S3. In the FE phase, l contracts slightly from 2.30 Å at -6% strain to 2.28 Å at $+6\%$, while h decreases markedly from 3.26 to 2.76 Å, reflecting a flattened layered framework and a more symmetric P–P dimer with parallel dipoles. In contrast, the AFE phase shows a much stronger bond-length response (2.31→2.25 Å) and a comparable reduction in layer thickness (3.27→2.79 Å). Such strain-induced changes in the P–P dimer geometry alter the orientation of the lone-pair electrons and thus the local dipole moment, in quantitative agreement with the monotonic strain dependence of the polarization in Fig. 4(d).

4 Conclusions

In summary, first-principles calculations reveal that ScPS₃ hosts robust ferroelectricity whose polarization originates in the monolayer from out-of-plane P–P dimer perturbation coupled with stereochemical lone-pair activity, rather than from interlayer sliding or metal-ion displacement; this polarization is also remarkably insensitive to the interlayer stacking order in the bilayer. The antiferroelectric phase is identified as the thermodynamic ground state, with low switching barriers (0.28 eV for FE→AFE and 0.41 eV for AFE→FE) that favor low-power operation. Biaxial strain can drive a reversible AFE–FE transition and tune both polarization and band gap continuously. The polarization magnitude remains nearly identical for AA and AB stackings, greatly relaxing fabrication constraints. These findings establish ScPS₃ as a promising platform for strain-engineered, low-power ferroelectric and optoelectronic devices, and provide a theoretical basis for exploring stacking-insensitive 2D ferroelectrics.

Declarations The authors declare that they have no competing interests and there are no conflicts.

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