

Coexistence of Ferroelectricity and Metallicity in Weakly Coupled $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ Crystal

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Ferroelectric metals, traditionally considered mutually exclusive, face enduring challenges owing to screening effects of itinerant electrons on ferroelectric order. However, in certain van der Waals (vdW) heterostructures, two-dimensional materials with distinct structural and physical properties can be alternately arranged in layers, retaining their individual characteristics and thereby enabling the coexistence of ferroelectricity and metallicity. Here, we report the coexistence of robust in-plane ferroelectricity and exceptional metallicity in a natural vdW superlattice $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ crystal, exhibiting high-density carriers ($>10^{21} \text{ cm}^{-3}$), superconductivity ($T_c \sim 3.25 \text{ K}$), and Curie temperature up to 383 K. The weak-coupling electronic and phononic natures stabilize in-plane ferroelectricity against screening by high-density carriers, confirming the decoupled electron mechanism. Moreover, we demonstrate the first ferroelectric memristor, where the metallic channel conductance is manipulated through polarization dynamics. Our Letter establishes a universal platform for designing multi-order parameters quantum-confined materials, and opens new avenues for energy-efficient functional electronics.

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Introduction—The design of functional materials with exotic and innovative properties often requires the integration of multiple physical effects, particularly when these behaviors are traditionally deemed mutually exclusive. The incompatibility of ferroelectricity and metallicity arises from the screening effects of high-density itinerant electrons in metals. However, Anderson and Blount predicted the existence of “ferroelectric metals” from a symmetry perspective, where ferroelectriclike phase transition occurs within metallic systems [1]. Recent studies of polar metals have established new paradigms [2] and hold profound implications for the discovery of exotic quantum phenomena [3–7], such as

ferroelectric superconductivity [8–12] and nontrivial topology [13–15], as well as for the development of low-energy-consumption multifunctional electronic devices [16,17]. Significant progress was made half a century later with intrinsic polar metal LiOsO_3 at 140 K [18], but it struggles to achieve switchable electric polarization [19]. Subsequent artificial strategies, such as doping perovskite ferroelectrics [20,21] and constructing perovskite superlattices [22,23], have explored this approach, often enhancing metallicity at the expense of weakening or even suppressing ferroelectric stability suppressing ferroelectric stability [24]. Recently, two-dimensional (2D) semimetallic WTe_2 [25,26] and MoTe_2 [8] with low-density carriers, which relies on the confinement of electrons in in-plane and sliding ferroelectricity in out-of-plane [27], have opened new avenues for the exploration of coupled ferroelectricity and metallicity. However, integrating or reconciling excellent metallic states and robust switchable ferroelectric order into a single system at room temperature remains an ongoing challenge.

The *decoupled electron mechanism* (DEM) was proposed in which the design principle for polar metals lies in

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decoupling polar order from metallicity [28]. This decoupling requires precise control over chemical composition and crystal structure to separate the origins of polarity and metallicity while ensuring weak interactions between their sublattices [29]. Generally, achieving such decoupling in three-dimensional lattice materials is relatively difficult due to strong electron-lattice interactions. However, vdW superlattice crystals made from 2D materials benefit from weak vdW forces between sublattices, retaining intrinsic properties of different components and even exhibiting exotic properties beyond their parent materials [30–33]. Consequently, when vdW heterosuperlattices composed of ferroelectric and metallic layers are employed, the intrinsic coexistence of ferroelectricity and metallicity is expected to be attainable by decoupling polar sublattices from metallic sublattices and following the DEM principle.

Herein, we report the discovery of the coexisting metallicity and ferroelectricity in a natural vdW superlattice $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ crystal by multi-means investigation. Because of weak-coupled electronic and phononic properties, our system exhibits not only exceptional metallicity of high carrier density and superconductivity, but robust in-plane ferroelectricity with high Curie temperature, surpassing most known polar metals. As a verification, a first ultralow-resistance, low-operating-voltage, low-power ferroelectric memristor was realized in this metallic crystal at room temperature. This class of vdW superlattice establish a bridge for designing incompatible-orders quantum materials, and has potential for nonlinear optics and nonvolatile electronics.

Results—Coexistence of metallicity and ferroelectricity: A natural bulk vdW superlattice $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ crystal was synthesized via one-step chemical vapor transportation (CVT) (see Materials and Methods in Supplemental Material [34]). The distinct work functions of sublattices drive interlayer charge transfer forming vdW couplings, enabling self-assembly infinite-sublayer ordered growth. X-ray diffraction (XRD) patterns confirm the as-grown sample is highly oriented single-crystal [Fig. 1(a)]. The interlayer spacing was calculated to be 12.32 Å for $(\text{SnSe})_{1.16}(\text{NbSe}_2)$, compared with 6.27 Å in bulk 2H-NbSe₂. The cross-plane structures were investigated via high angle annular dark field scanning transmission electron microscope (STEM-HAADF). Figure 1(b) and Supplemental Material Fig. S1 [34] clearly show periodical vdW superlattice alternating stacked by SnSe and NbSe₂, with an interlayer spacing of 12.4 Å between NbSe₂ sublayers. Atom-resolved images of NbSe₂ [Fig. 1(c)] and SnSe sublattice [Fig. 1(d)] were obtained on a terrace morphology by scanning tunneling microscopy (STM) (see Supplemental Material Fig. S2 for details [34]). In NbSe₂ sublattice, the lattice constant of hexagonal Se atoms was measured to be 3.43 Å, consistent with that of monolayer 1H-NbSe₂ [43]. The SnSe sublattice shows a quasisquare lattice of Sn-Sn atoms, with lattice parameters of 4.34 Å

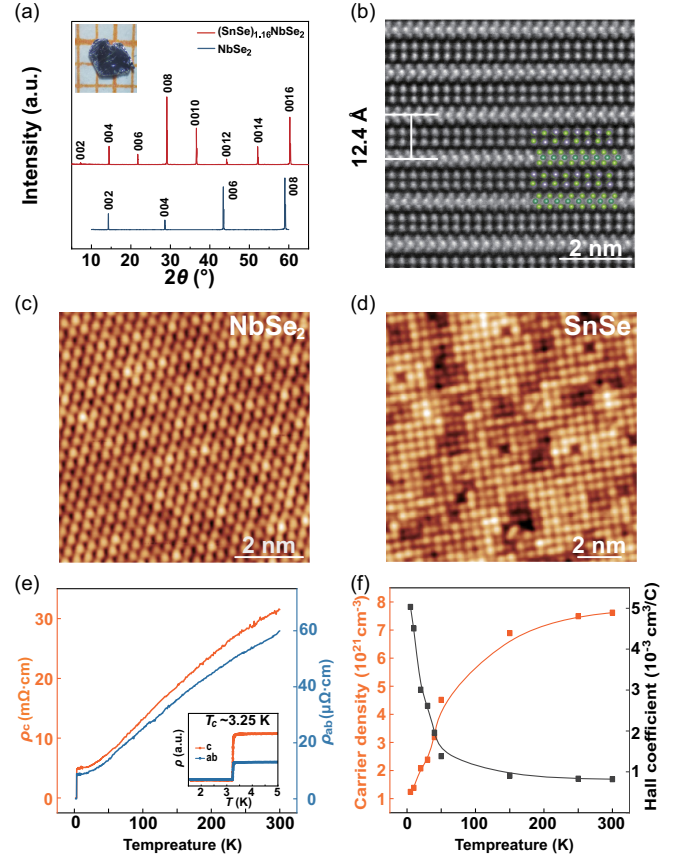


FIG. 1. Crystal structure and metallicity of $(\text{SnSe})_{1.16}(\text{NbSe}_2)$. (a) XRD patterns of $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ and NbSe₂ crystals. The inset shows an optical image of a millimeter-sized $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ crystal. (b) Cross-sectional HAADF-STEM image along the [100] axis. The inset displays the corresponding atomic model structure, where Sn, Nb, and Se atoms are represented by purple, green, and kelly spheres, respectively. (c) and (d) STM images of hexagonal NbSe₂ sublattice ($V = -100$ mV, $I = 100$ pA) and the orthogonal SnSe sublattice ($V = -1$ V, $I = 170$ pA), respectively. (e) Temperature-dependent resistivity along in plane and cross plane. The inset shows the superconducting transition temperature ($T_c = 3.25$ K). (f) Temperature-dependent carrier density measured by Hall effect along in-plane.

along $\mathbf{a}_1$ lattice vector and 4.27 Å along $\mathbf{a}_2$ lattice vector, respectively, agreement with the black phosphoruslike (BP-like) monolayer α -SnSe [44]. The superlattice models of $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ (Fig. S3 [34]), consisting of 2H-NbSe₂ layers separated by α -SnSe layers, were constructed by first-principles calculations. The lattice parameters of two sublattices, matching those of individual monolayers, suggest weak interlayer interactions in this superlattice. Notably, our system cannot be artificially stacked, as isolated α -SnSe monolayers cannot be obtained via mechanical exfoliation.

The resistivity of $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ crystal monotonically increases with temperature [Fig. 1(e)], demonstrating metallic behavior across both room-temperature and

cryogenic regimes. The resistivity is determined to be $59.93 \mu\Omega \cdot \text{cm}$ with hole-type carrier density reaching up to $7.8 \times 10^{21} \text{ cm}^{-3}$ at 300 K [Fig. 1(f)], further underscoring its excellent metallic nature. The anisotropic ρ_c/ρ_{ab} ratio is about 500 at 300 K, indicating a confined conduction along the c axis. The sharp synchronized superconductivity [inset of Fig. 1(e)] in both in-plane and cross-plane directions reveals the unified coherence behavior of electrons, thereby highlighting its electronic-level integrity. Both superconducting transition temperatures (T_c) are observed at 3.25 K, which closely resembles that of monolayer NbSe₂ (~ 3.1 K), rather than bulk NbSe₂ (~ 7 K) [45]. It suggests that interlayer coupling is weak enough to preserve monolayerlike NbSe₂ electronic states, even if c -axis superconduction is present. At the micro-level, the superconducting gap of our crystal is close to that of monolayer 1H-NbSe₂ (Fig. S4 [34]) as revealed by scanning tunneling spectroscopy (STS) [46].

The ferroelectricity and domain engineering of (SnSe)_{1.16}(NbSe₂) sheet were investigated by lateral piezoresponse force microscopy (PFM) at 300 K (Fig. S5 [34]). Figure 2(a) depicts results of local switching spectroscopy PFM (SS-PFM). The amplitude butterfly curves and phase loops, which exhibit a distinct 180° phase contrast, clearly indicate in-plane ferroelectric switching hysteresis behaviors, revealing its bistable polarization states. Polarization switching is observed at voltages of approximately -0.7 and $+0.6$ V. Furthermore, concentric square in-plane phase and amplitude domain patterns [Figs. 2(b) and 2(c)] show antiparallel remnant polarizations with approximate 180° phase contrast, poling at $+5$ V and -5 V by tip bias. The in-plane ferroelectricity has been consistently reproducible in other samples (Fig. S6 [34]). For comparison, the absence of SS-PFM and PFM signals on NbSe₂ sublayer (Fig. S7 [34]) indicates that ferroelectricity is exclusively attributed to SnSe sublayers.

Optical second harmonic generation (SHG), with intensity proportional to the square of second-order polarizability tensor ($\chi^{(2)}$), was conducted via a vertical back-reflection configuration (Fig. S8 [34]). With ultrafast laser excitation, SHG serves as a highly sensitive probe to detect polar point-group symmetries which are minimally influenced by charge carriers transit processes [47]. Strong signals were observed at prominent peaks centered at 532 nm [Fig. 2(d)], corresponding to half-wavelength of incident pump laser. The intensity of SHG signals exhibits a quadratic dependence on excitation power [inset of Fig. 2(d)], indicating typical SHG effects and horizontal symmetry-breaking of (SnSe)_{1.16}(NbSe₂). Figure 2(e) shows highly anisotropic SHG patterns, with maximal intensity along 155°–335° orientation under perpendicular polarization. This observation is similar to SHG patterns along the zigzag direction of ultrathin α -SnX (S, Se) flakes [48,49]. Moreover, SHG mappings (Fig. S9 [34]) indicate significant in-plane anisotropy, further confirming

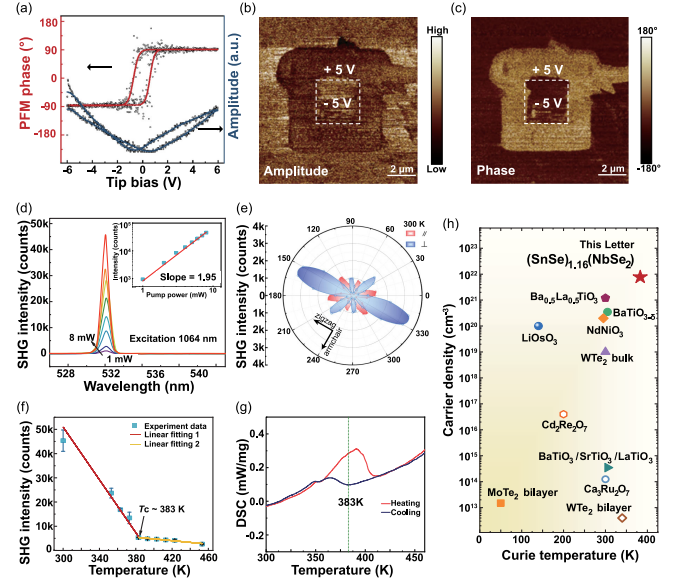


FIG. 2. In-plane ferroelectricity of (SnSe)_{1.16}(NbSe₂). (a) Bias-dependent lateral PFM amplitude and phase hysteretic loops measured at 300 K. (b) and (c) Lateral PFM amplitude and phase domain images after opposite bias poling with box-in-box patterns. (d) SHG spectra of (SnSe)_{1.16}(NbSe₂) sheet under various excitation powers. The inset shows a typical second-order power-dependent SHG intensity with a fitted slope of 1.95. (e) SHG rotational patterns for parallel and perpendicular polarizations, revealing high anisotropy and inversion symmetry-breaking in the in-plane direction of (SnSe)_{1.16}(NbSe₂). The inset axes represent the armchair and zigzag directions of SnSe sublattices. (f) and (g) Temperature-dependent SHG intensity and enthalpy changes of DSC, indicating a second-order phase transition temperature ($T_c \sim 383$ K) from polar to centrosymmetric phase. (h) Comparative survey of various systems with coexisting polarity or ferroelectricity and metallicity, characterized by carrier density and T_c .

spontaneous polarization. Temperature-dependent SHG [Fig. 2(f) and Fig. S10 [34]] and differential scanning calorimetry (DSC) [Fig. 2(g)] jointly reveal a second-order phase transition with Curie temperature (T_c) of 383 K for (SnSe)_{1.16}(NbSe₂). Notably, the T_c is close to that of monolayer ferroelectric α -SnSe [43]. The discussion of phase transition details are in End Matter [50–52]. The heat of phase transition measured by DSC (~ 5 mJ/mg) is much larger than the contribution from the only surface SnSe sublayer, indicating bulk ferroelectric order in (SnSe)_{1.16}(NbSe₂).

Based on above observations, (SnSe)_{1.16}(NbSe₂) crystal demonstrates an integrated bulk from crystallographic and electronics aspects, and exhibits the coexistence of metallicity and ferroelectricity. Notably, SnSe sublattices retain robust polarity even in carrier densities over 10^{21} cm^{-3} . Figure 2(h) presents a comparative survey (Supplemental Material Table S1 for details [34]) of polar metals categorized by their carrier density and Curie or operating temperature [3,8,18,20–22,25,26,29,53].

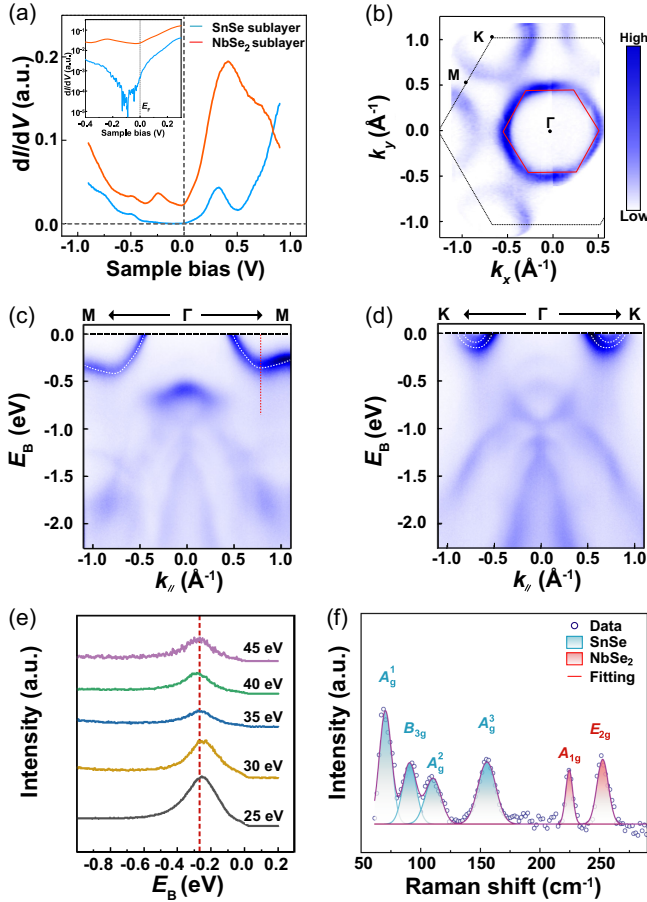


FIG. 3. The decoupled electron behavior of $(\text{SnSe})_{1.16}(\text{NbSe}_2)$. (a) STS spectrum of $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ at 4.2 K, the inset is plotted on a logarithmic scale. (b) Local Fermi surface map of $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ obtained via ARPES measurement at 7.8 K, resembling the monolayer NbSe₂ without observable features of SnSe band. (c) and (d) ARPES band dispersions of $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ measured along the Γ -M (left) and Γ -K (right) directions using 23 eV photons at 7.8 K. The DOS near the E_F is attributed primarily to the Nb-4d electron orbitals. (e) Photon energy dependence of energy distribution curves passing through ARPES cuts along the Γ -M direction at $k_{\parallel} = 0.8 \text{ \AA}^{-1}$. The peaks marked by the red dashed line exhibit no significant dispersion along the k_z direction, indicating quasi-2D electronic behavior in $(\text{SnSe})_{1.16}(\text{NbSe}_2)$. (f) Raman spectra of $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ under 532 nm excitation at 300 K.

Quasi-2D electronic and phononic properties: To explore the physical mechanisms, electronic structures of $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ was investigated by STS, angle-resolved photoemission spectroscopy (ARPES), and first-principles calculations. Figure 3(a) presents dI/dV spectra for NbSe₂ and SnSe sublayers, demonstrating that the conductive ground states of $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ are primarily driven by metallic NbSe₂ sublayers. Logarithmic-scale dI/dV spectra [inset of Fig. 3(a)] show that SnSe sublayer exhibits nonzero density of states (DOS) at the Fermi level (E_F), indicating a metalliclike state.

The phenomenon of the SnSe sublayer may originate from the electronic structure reconstruction which is induced by interlayer coupling mediated by charge transfer. Furthermore, the Fermi surface [Fig. 3(b)] reveals a sixfold-symmetry hole pocket at the Γ point and small pockets at the K point within the hexagonal Brillouin zone. The band dispersions reveal that Nb-4d orbital bands degenerate along the Γ -M direction [Fig. 3(c)] but split along Γ -K direction [Fig. 3(d)]. The calculated band structures of $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ (Fig. S13 [34]) align qualitatively well with ARPES results. Both reveal the distinctively absent high-dispersive bands contributed by the Se- p_z orbital of bulk NbSe₂ [54]. The band structures near E_F differ significantly from the dispersions of bulk NbSe₂, but resemble that of monolayer NbSe₂ [55]. Besides, the band dispersion along the Γ -M direction remains unchanged with varying photon energy from 25 to 45 eV (Fig. S14 [34]). The energy distribution curves corresponding to the band bottom along the Γ -M direction exhibit non-dispersion as a function of photon energy along the k_z direction [Fig. 3(e)], indicating electronic weak coupling and quasi-2D electronic structures of $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ crystal [56].

The active phonon modes of $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ were characterized by Raman spectroscopy [Fig. 3(f)]. The observed Raman peaks include the vibration modes corresponding to SnSe sublattices (blue symbols) and NbSe₂ sublattices (red marks). Notably, the reduction in thickness of α -SnSe from bulk to few layers or monolayer leads to symmetry breaking, thereby resulting in in-plane ferroelectricity [57]. In contrast, Raman spectroscopy was performed of α -SnSe and 2H-NbSe₂ crystals with different thickness. With decreasing α -SnSe single-crystal thickness, B_{3g} and A_g^2 peak clearly shifts toward lower frequencies; whereas A_g^3 mode shifts toward higher frequencies (Fig. S15 [34]). The horizontal B_{3g} and A_g^2 phonon modes of SnSe sublattices tend to evolve into vibration modes of few layers or monolayer α -SnSe crystals [58]. Moreover, the red two peak A_{1g} and E_{2g} modes of NbSe₂ sublattices in the superlattice are well matched with the vibration modes of monolayer 1H-NbSe₂ [59], implying that interlayer vibrational interactions between NbSe₂ sublattices are effectively eliminated. Therefore, the interlayer phonons of $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ decouple, and quasi-2D phonon modes are expected to arise between the sublayers (Fig. S15 [34]). The quasi-2D phonon properties explain T_C of $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ close to that of monolayer α -SnSe, as these phonon models driving structural phase transition are consistent.

In exploring the coexistence of ferroelectricity and metallicity in $(\text{SnSe})_{1.16}(\text{NbSe}_2)$, the DEM theory offers crucial insights. DEM emphasizes that polarity and metallicity have distinct spatial or chemical sublattices and do not interact strongly [2]. In our case, the $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ displays weakly coupled quasi-2D electronic and phononic

properties, and the ferroelectricity and metallicity originate from the α -SnSe and $1H$ -NbSe₂ sublayers by separated vdW gaps, respectively. The quasi-2D properties and vdW interactions ensure weak electron-lattice coupling between two sublayers, thereby the polar order is not annihilated by high-density carriers. This mechanism is analogous to the DEM of three-dimensional polar metals [28,29]. Besides, the interlayer coupling of (SnSe)_{1.16}(NbSe₂) [60] does not dominate and change its quasi-2D properties. The metalization of the SnSe sublayers, driven by this interlayer coupling, facilitates the neutralization of the depolarization field, thereby stabilizing their polarity. The unique coexistence in (SnSe)_{1.16}(NbSe₂) provides an ideal platform for developing nonvolatile electronics and ferroelectric superconductors.

Ferroelectric switching memristor: Ferroelectric memristors are potential hardware solutions for in-memory computing [61]. Figure 4(a) depicts a biological neuron synapse alongside an artificial two-terminal memristor. The barrier height of our device [inset of Fig. 4(b)] can be modulated by reversing in-plane polarization [Fig. S16(a) [34]], leading to OFF and ON states or high- and low-resistance states (HRS and LRS), respectively. Figure 4(b) presents typical hysteresis current-voltage loops measured at 300 K with increasing bias sweep range, obviously demonstrating memristive behaviors. A higher drain bias leads to an expanded hysteresis switching window, with maximum current occurring around ± 0.5 V. The metallic nature of (SnSe)_{1.16}(NbSe₂) memristor enables easy-to-read large current. The current of our device is significantly higher than that of conventional memristors, improving signal reliability and device sensitivity. Moreover, the nonreciprocal current switching ($\sim +35/-5$ mA) driven by positive and negative bias (± 0.3 V) shows repeatable reversible behavior between the LRS and HRS states [Fig. 4(c)]. The coercive field (E_c) of our device is estimated to be ~ 0.1 kV/cm, enabling low-voltage operation and reduced energy consumption. This behavior may originate from quasi-2D polarity coupled with metallic conduction (Fig. S16 [34]). The energy expended for each program or erase operation is determined by the applied switching voltage [62], meanwhile the ultralow resistance reduces energy consumption. Thus, the memristor is advantageous for energy-efficient electronics.

To advance neuromorphic computing, the synaptic behavior can be emulated by tunable conductance. As the strength of regularly varied pulses, the memristive hysteresis loop of our memristor is depicted in Fig. 4(d), showing threshold voltages of approximately -0.25 and $+0.3$ V. The writing and reading process were conducted via a series of small-step voltage pulses in Supplemental Material Fig. S16(b) [34]. Our memristor exhibits bipolar cyclic switching (~ 100) between HRS and LRS at ~ 0.3 V, indicating in-plane bistable and electrically switchable polarization. An incremental pulse spike scheme in

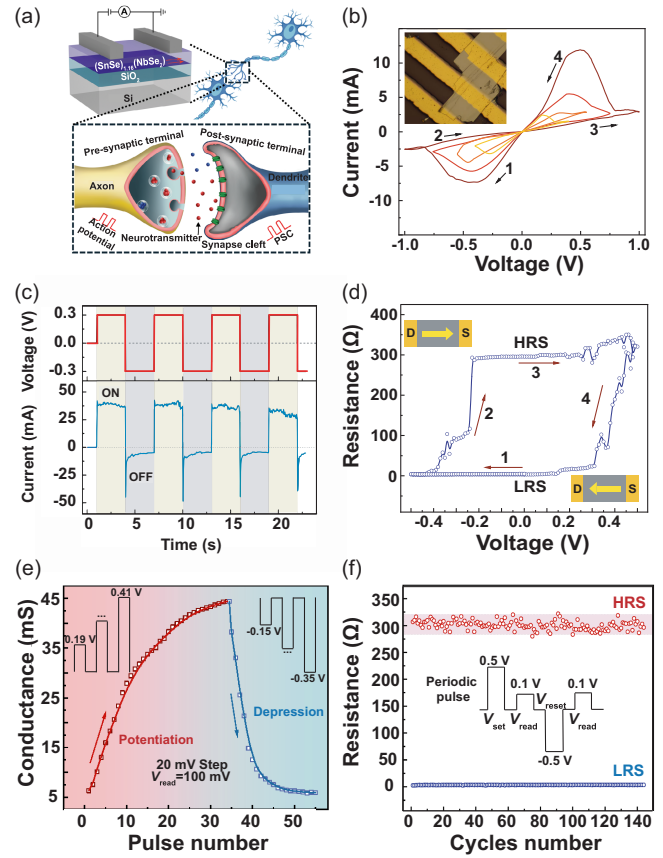


FIG. 4. Room-temperature in-plane ferroelectric memristive behaviors. (a) Schematic illustration of device and neural synapses simulation using a two-terminal ferroelectric memristor based on the (SnSe)_{1.16}(NbSe₂) sheet. Various electrical or chemical signals of different strengths are transmitted from presynaptic terminal to the postsynaptic terminal via voltage pulses or neurotransmitters. (b) Hysteresis I - V loops demonstrating memristive behaviors under different bias sweep ranges. Inset: optical image of a device with ~ 30 μm channel spacing and ~ 350 nm thickness. (c) Reversible current switching driven by applying positive and negative voltage pulses of ± 0.3 V. (d) Memristive resistance loop measured at a read voltage (V_{read}) of ~ 100 mV after applying a stepwise poling voltage pulse. (e) Conductance modulation during potentiation and depression of in-plane ferroelectric memristor via an incremental pulse scheme. (f) Memristive retention characteristics with positive or negative voltage pulses of ± 0.5 V.

Fig. 4(e) was employed to achieve a continuous process of strengthening or weakening conductance in the post-synaptic memristor, demonstrating high-conductance synaptic behaviors characteristic of long-term potentiation or depression. The reliability of the device as a nonvolatile memory was evaluated through retention tests. Reversible switching resistance switching in Fig. 4(f) was consistently maintained over 140 periodic cycles of repeated poling, presenting a reliable HRS/LRS ratio over 100. Besides, these ferroelectric memristive effects suggest the existence of bulk polar order in (SnSe)_{1.16}(NbSe₂), as only the polar

surface SnSe sublayer is scarcely sufficient to dominate potential barrier switching in excellent metallic channels.

Conclusion and outlook—In summary, we demonstrate the coexistence of ferroelectricity and metallicity in a natural bulk vdW superlattice $(\text{SnSe})_{1.16}(\text{NbSe}_2)$ crystal. This system that cannot be achieved by artificial stacking represents a coherently coupled bulk with novel multifunction. The first room-temperature ferroelectric memristor within the metallic channel provides a new probability for low-power-consumption nonvolatile electronics. Besides, our system displays both ferroelectricity and superconductivity [Fig. 1(e) and Supplemental Material, Fig. S4 [34]], while rocksalt SnSe is a topological crystalline insulator [63], suggesting its potential as a ferroelectric topological superconductor. Our Letter not only validates DEM theory but also provides a new strategy to design other vdW quantum systems with coexisting incompatible properties, potentially leading to developing multifunctional electronics and ferroelectric superconductors.

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Data availability—The data that support the findings of this article are openly available at [64].

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End Matter

Details of phase transition—A ferroelectric phase transition occurs when a material shifts from high-symmetry paraelectric phase to low-symmetry ferroelectric phase, resulting in the emergence of spontaneous polarization. Typically, the temperature and electric field represent primary external factors driving such structural phase transitions.

For our crystal, SHG patterns of (SnSe)_{1.16}(NbSe₂) at 453 K and NbSe₂ at 300 K both exhibit sixfold symmetry [see Supplemental Material Fig. S10(b) and S10(c) [34]], consistent with bulk NbSe₂ [50], being a similar structural symmetry. The SHG intensity of (SnSe)_{1.16}(NbSe₂) in Fig. 2(f) decays continuously and smoothly to the baseline near T_C , strongly indicating that the order parameter (spontaneous polarization) driving this ferroelectric phase transition varies continuously. This aligns fully with the definition of a second-order phase transition and forms perfect mutual verification with the DSC results in

Fig. 2(g). Moreover, broad, steplike DSC inflection points confirm continuous small enthalpy evolution instead of abrupt saltation, consistent with the characteristics of a second-order phase transition.

With increasing temperature, the low-symmetric ferroelectric α -phase SnSe sublattices gradually change until it reaches the T_C , at which point it undergoes a transition into a high-symmetric centrosymmetric paraelectric rocksalt phase. This polar-to-centrosymmetric phase transition of the (SnSe)_{1.16}(NbSe₂) superlattice corresponds to the geometric switching pathway of SnSe sublattices from BP-like α -phase to rock-salt phase (Fig. S11 [34]), resembling the pathway of α -SnSe [51,52]. The double-well potential of (SnSe)_{1.16}(NbSe₂) superlattice demonstrates two degenerate energy levels in ferroelectric phase, as revealed by DFT calculations in Supplemental Material Fig. S12 [34], indicating the stability of ferroelectricity and the energetic favorability of switching between two states.