Multiferroic van der Waals heterostructure FeCl₂/Sc₂CO₂: Nonvolatile electrically switchable electronic and spintronic properties

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Multiferroic van der Waals (vdW) heterostructures offer an exciting route toward high-performance nanoelectronics and spintronics device technology. Here we investigate the electronic and transport properties of multiferroic vdW heterostructures composed of a ferromagnetic FeCl₂ monolayer and a ferroelectric Sc₂CO₂ monolayer using first-principles density functional theory and quantum transport simulations. We show that FeCl₂/Sc₂CO₂ heterostructure can be reversibly switched from semiconducting to half-metallic behavior by electrically modulating the ferroelectric polarization states of Sc₂CO₂. Intriguingly, the half-metallic phase exhibits a type-III broken gap-band alignment, which can be beneficial for tunneling field-effect transistor applications. We perform a quantum transport simulation based on a *proof-of-concept* two-terminal nanodevice to demonstrate all-electriccontrolled valving effects uniquely enabled by the nonvolatile ferroelectric switching of the heterostructure. These findings unravel the potential of FeCl₂/Sc₂CO₂ vdW heterostructures as a building block for designing the next generation of ultimately compact information processing, data storage, and spintronics devices.

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I. INTRODUCTION

The continuous miniaturization of electronic products posses huge technological challenges on the design of nanoelectronic devices and functional materials. Spintronics and two-dimensional (2D) materials provide opportunities for solving this problem. Compared to the conventional spintronics based on three-dimensional (3D) bulk magnetic materials, 2D-material-based spintronics offers enormous opportunities for the post Moore's law era due to its low energy consumption, fast device operation, mechanical flexibility, high storage density, and gate-controllable operation [1,2]. The recent advancements of van der Waals (vdW) heterostructures, obtained via the vertical stacking of different 2D materials, further expands the design flexibility and the functionality of 2D-material-based nanoelectronic, optoelectronics, and spintronics devices [3–20].

2D materials with intrinsic magnetism are uncommon and often required complex nanofabrication techniques [21–23]. Nonetheless, recent theoretical [24] and experimental [25,26] advancements in 2D materials with intrinsic magnetism have revitalized the exploration of intrinsic magnetism in 2D layered materials for spintronic applications [8,27], such as the

very large negative magnetoresistance ratio of over 10 000% in the vdW tunnel junction made of ultrathin magnetic semiconductor CrI₃ [28,29], 100% spin polarization in tunnel junction composed of CrI₃ tunnel barrier [30], valley splitting, and valley-polarized electroluminescence in ultrathin CrI₃ vdW heterostructures [31,32]. Beyond 2D magnetic materials, the 2D ferroelectric monolayer has also received intensive research attention in recent years [33], primarily because of their huge potential in memory and neuromorphic devices [34] as enabled by the presence of electrically switchable spontaneous ferroelectric polarization. The recent exploration of intrinsic ferroelectric properties in 2D monolayers, such as In_2Se_3 [35], 1T-phase MoS₂ [36], MX (M=Ge, Sn; X=S, Se, Te) [37,38], bilayer BN [39], and CuInP₂S₂ [40], have shed much light on the physics and device application potential of 2D ferroelectric materials.

The union of 2D ferroelectric and ferromagnetic materials leads to the emergence of *multiferroic vdW heterostructures and devices*, in which the all-electric switching of the ferroelectric sublayer and the magnetic properties of the ferromagnetic sublayer can be simultaneously combined to render unique material properties and device functionalities [41–45]. Such a *magnetoelectric effect* represents a promising route toward the development of ultracompact and multifunctional nanodevices. The simultaneous presence of two tuning knobs, i.e., the electric fields and the magnetic fields, offers enormous

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design flexibility in engineering the physical properties of vdW heterostructures for electronic, magnetic and spintronic devices with electrically switchable operations.

In this paper, we investigate the electronic and the transport properties of a multiferroic vdW heterostructures composed of a ferromagnetic FeCl₂ monolayer and ferroelectric monolayer Sc₂CO₂ by using first-principle density function theory (DFT) simulations. Here Sc₂CO₂ serves as a ferroelectric monolayer with excellent lattice matching with the ferromagnetic FeCl₂ monolayer—highly beneficial for the computational design and simulation of heterostructures and devices. We show that the electronic band structures of the FeCl₂/Sc₂CO₂ heterostructure can be flexibly tuned between a ferromagnetic and half-metallic phase by changing the ferroelectric polarization of Sc₂CO₂, and such switching is nonvolatile and reversible. The half-metallic FeCl₂/Sc₂CO₂ heterostructure exhibits a broken-gap type-III band alignment which can be useful for tunneling-based electronic device applications. Based on nonequilibrium Green's function (NEGF) transport simulation, we design a proof-of-concept two-terminal nanodevice based on the FeCl₂/Sc₂CO₂ heterostructure and concretely demonstrate the presence of valving effects. These findings reveal the potential of multiferroic FeCl₂/Sc₂CO₂ heterostructures as a building block in designing future nanoelectronics and spintronics applications. Although multiferroic heterostructures have received increasing research attention recently, the progress of 2D vdW heterostructure multiferroic heterostructures are still in the very early stages. In particular, there are very few heterostructures that exhibit being reversibly switched from semiconducting to half-metallic behavior by electrically modulating the ferroelectric polarization states. As experimental studies and device fabrications typically prefer to have a larger variety of candidate systems, the results presented in this paper offer an important addition to the multiferroic family that can be useful for future studies.

II. COMPUTATIONAL METHODS

The geometrical optimization and the spin-polarized DFT computations are performed using the VIENNA AB INITIO SIMULATION PACKAGE [46,47] with the generalized gradient approximation in the Perdew-Burke-Ernzerhof scheme for the exchange-correlation energy functional. The electron-ion interaction is described by the projector augmented wave [48]. The kinetic energy cutoff is set to be 500 eV. The first Brillouin zone integration is sampled with $15 \times 15 \times 1$ k-point meshes. To avoid any artificial interactions between the adjacent slabs, a 20 Å vacuum distance is set along the direction vertical to the contact interface. Based on the D3 method of Grimme for describing the interlayer vdW interaction [49], the forces on all atoms less than 0.01 eV/Å are achieved for the structural relaxation. To understand the reliability of DFT-D3 in comparison with other methods, DFT-D2 and vdW-DF are also employed to test the systems studied in this paper. Although the interlayer distance varies slightly under different methods, the relative stability of the structures and the calculated band structures are nearly identical to those obtained using DFT-D3 [50], thus confirming the reliability of DFT-D3 method. The energy convergence accuracy is set to

 10^{-6} eV per atom. Dipole corrections are included in all calculations. The results considering the effective Hubbard U (with a semiempirical value of U = 3 eV [51]) and the spin orbit coupling (SOC) are used for comparison, which are presented in the Supplemental Material [50]. Finally, the transport properties for a two-probe device systems are calculated using the Atomistix Toolkit 2018 package based on the DFT-LCAO and the NEGF method [52-55]. We use the periodic-type, Neumann-type, and Dirichlet-type boundary conditions [56] in the x (orthogonal to the transport direction), y (perpendicular to the device plane), z (direction of electron transport) directions of the device, respectively. For the x directions, the device is two-dimensionally periodic and infinitely wide. For the y directions, we chose the Neumann boundary condition, which corresponds to a zero electric field at the boundary of the computational box. In the z direction, the boundary condition for the electrostatic potential is determined by the electrostatic potential in the electrodes, corresponding to a Dirichlet boundary condition.

III. RESULTS AND DISCUSSIONS

A. Structural and electronic properties

The lattice structures of free-standing ferroelectric monolayer FeCl₂ and ferromagnetic monolayer Sc₂CO₂ are 3.366 Å and 3.41 Å, respectively, which agree with previously reported values [44,57,58] [see Figs. 1(a) and 1(b)]. The lattice mismatch is less than 1% when forming the two monolayers stacked vertically to form a 2D/2D vdW multiferroic heterostructure. Although the lattice optimization of both submonolayers can be implemented in the calculation of the 2D/2D heterostructure, such a relaxation scheme will lead to an additional straining effect that modifies the electronic properties of the heterostructures in addition to the weak vdW interaction between the 2D submonolayers. The lattice vectors of FeCl2 are thus fixed in our calculations to preserve its physical properties and to minimize strain-induced modifications. Three typical stacking configurations, denoted AA, BB, and CC, are considered. Structurally, the three stacking configurations can be described based on the relative location of the O atom: (i) AA stacking configuration-the O atom in contact with $FeCl_2$ is directly under the Cl atom, (ii) BB stacking configuration-the O atom at the contact interface is under an empty space of the FeCl₂, and (iii) CC stacking configuration-the O atom is directly under the Fe atom. Since the ferroelectric Sc₂CO₂ has bistable *P*-polarization states, i.e., $P \uparrow$ and $P \downarrow$, six different heterostructures were considered [see Figs. 1(c) to 1(h)]. Here, when the carbon atom is closer to the top (bottom) layer, the polarization states becomes $P \uparrow (P \downarrow)$. The detailed structural parameters of the six optimized heterostructures are presented in Table I. The different stacking configurations produce different optimized interlayer distances when the heterostructures are at equilibrium. The minimum distance between the atoms at the interface (Cl and O atoms) are much larger than the sum of the covalent radii of Cl and O atoms, thus indicating that vdW interactions are the dominant coupling mechanism between the two monolayers.



FIG. 1. Crystal structures of the 2D ferroelectric FeCl₂, the 2D ferromagnetic Sc₂CO₂, and their 2D/2D vdW heterostructures. Top view and side view of the structures of single layers of (a) FeCl₂ and (b) Sc₂CO₂. The blue line represents the unit cell. (c)–(h) FeCl₂/Sc₂CO₂ vdW heterostructures with different stacking configurations under various polarization states. The $P \uparrow$ and the $P \downarrow$ states are shown in (c) (e), (g) and (d), (f), (h) P \downarrow , respectively, for *AA*, *BB*, and *CC* stacking configurations. The Fe, Cl, Sc, C, and O atoms are denoted by dark cyan, green, white, gray, and red spheres, respectively.

The binding energy, E_b , of the FeCl₂/Sc₂CO₂ vdW heterostructure is calculated, which is defined as $E_b = (E_{\text{FeCl}_2/\text{Sc}_2\text{CO}_2} - E_{\text{FeCl}_2} - E_{\text{Sc}_2\text{CO}_2})/A$, where $E_{\text{FeCl}_2/\text{Sc}_2\text{CO}_2}$ is the total energy of FeCl₂/Sc₂CO₂ heterostructure, E_{FeCl_2} and $E_{\text{Sc}_2\text{CO}_2}$ represent the energy of the isolated monolayer FeCl₂ and Sc₂CO₂, respectively, and A is the surface area at the equilibrium state. The binding energy of the combined systems for all stacking modes are summarized in Table I. Many studies have shown that the FeCl2 and Sc2CO2 are stable [57–59]. The calculated binding energies are all negative and are smaller than those of other typical vdW heterostructures [60,61], thus confirming that the proposed 2D/2D heterostructures structures simulated in this paper are energetically stable and feasible to explore experimentally. The projected band structures of the multiferroic heterostructures are shown in Fig. 2. Here green, blue, and red symbols denote the contributions from Sc_2CO_2 , spin-up and spin-down electronic states of FeCl₂, respectively. The band structures of isolated Sc_2CO_2 and FeCl₂ are shown in Fig. S1 of the Supplemental Material [50]. In the isolated form, monolayer Sc_2CO_2 is a semiconductor with a indirect band gap of 1.8 eV [57,62], while monolayer FeCl₂ is a ferromagnetic semiconductor with the spin-up and spin-down energy gaps of 4.16 and 0.41 eV, respectively. When forming vdW heterostructures, the electronic band structures of the FeCl₂/Sc₂CO₂ heterostructures largely exhibit a superposition of the electronic states from both materials due to the weak vdW coupling between the two monolayers. The

TABLE I. Calculated parameters of the FeCl₂/Sc₂CO₂ multiferroic vdW heterostructures. *d* is the interlayer distance. d_{\min} is the minimum distance between Cl and O atoms. E_b is the binding energy. ΔV is potential step, defined as $\Delta V = W_{\text{FeCl}_2} - W_{\text{Sc}_2\text{CO}_2}$. W_{FeCl_2} is the work function of the FeCl₂

Stacking	$AA - P \uparrow$	$AA - P \downarrow$	$BB - P \uparrow$	$BB - P \downarrow$	$CC - P \uparrow$	$CC - P \downarrow$
d (Å)	3.248	3.119	2.725	2.46	2.856	2.677
d_{\min} (Å)	3.248	3.119	3.35	3.13	3.45	3.31
$E_b (meV/Å^2)$	-75.1	-82.2	-82.1	-95.5	-80.3	-89.3
Gap (eV)	0.376	0	0.264	0	0.379	0
ΔV (eV)	2.062	-0.387	2.072	-0.656	2.004	-0.488
$W_{\rm FeCl_2}$ (eV)	5.625	5.21	5.55	5.371	5.613	5.307



FIG. 2. Electronic band structures of (a)–(d) AA-stacked heterostructure, (e)–(h) BB-stacked heterostructure, and (i)–(l) CC-stacked heterostructure under $P \uparrow$ (top panel) and $P \downarrow$ (bottom panel) polarized states. The Fermi level is set to zero. Here green, blue, and red symbols denote the contributions from Sc₂CO₂, spin-up and spin-down electronic states of FeCl₂, respectively.

retaining of individual electronic bands structures upon forming a vdW heterostructure is a typical feature of 2D/2D vdW heterostructures.

The electronic bands corresponding to the FeCl₂ monolayer are sensitively energetically shifted depending on the *P*-polarization states of the Sc_2CO_2 monolayer. In the case of $P \uparrow$ -polarized Sc₂CO₂, the electronic states of Sc₂CO₂ are upshifted energetically when compared to that of the independent Sc_2CO_2 monolayer. In this case, a common band gap is retained and the heterostructure behaves as a semiconductor [see Fig. 2, top panel]. In contrast, the electronic properties are more dramatically modified when the P-polarization states of the Sc_2CO_2 monolayer are switched to the $P \downarrow$ -polarization state. In this case, the electronic bands of FeCl₂ are energetically downshifted and cross the Fermi level, thus transforming the FeCl₂ into a half metal. Furthermore, the spin-down bands of the $FeCl_2$ monolayer (denoted by red symbols in Fig. 2) overlap with the spin-degenerate valence bands (denoted by green symbols in Fig. 2) of the Sc_2CO_2 monolayer. The heterostructure is thus a half-metallic semimetal with zero common band gap when the ferroelectric polarization of the Sc_2CO_2 monolayer is switched to the $P \downarrow$ state. Such contrasting electronic properties are particularly evident in the BB stacking configurations due to the small interlayer distance that strongly couples the two monolayers. In terms of heterostructure band alignment, FeCl₂/Sc₂CO₂ in the halfmetallic state exhibits a broken-gap type-III band alignment under all stacking configurations, i.e., when material A and material B merge, the resulting heterostructure is type I if the VBM and CBM are contributed by the same monolayer, type II if the VBM and CBM are contributed by two different

monolayers, and type III if the VBM and CBM are contributed by two different monolayers and overlap with each other. Such band alignment type reveals the potential of $FeCl_2/Sc_2CO_2$ in tunneling field-effect transistor applications.

We further note that when an $U_{eff} = 3 \text{ eV}$ is applied to account for a strong correlated effect of the electrons in a 3d shell of Fe atoms, the characteristic transformation of the FeCl₂ monolayer from the ferromagnetic semiconductor into half-metallic semimetal remains robust (see Fig. S2 of the Supplemental Material [50]). Furthermore, when the SOC is included in the simulations, as shown in Fig. S3 of the Supplemental Material [50], the electronic band structures of the heterostructure exhibit no obvious change with only a slight decrease of the energy band gap. We further note that the observed transformation of the FeCl₂ from semiconducting to semimetallic behaviors are also consistent with other 2D/2D multiferroic vdW heterostructures, such as MnCl₃/CuInP₂S₆ [39], Cr₂Ge₂Te₆/In₂Se₃ [41], FeI₂/In₂Se₃ [43], and CrI₃/Sc₂CO₂ [44].

B. Mechanism of interfacial interaction and charge redistribution

We attribute the switching behavior discussed above to the electrical interactions mediated by the charge transfer across the FeCl₂/Sc₂CO₂ contact interfaces, which is appreciably modified between the $P \uparrow$ and the $P \downarrow$ polarization states. When FeCl₂ and Sc₂CO₂ is constructed into the multiferroic system, the spatial inversion symmetry is broken due to the presence of ferroelectricity, and the charge balance of FeCl₂ is perturbed. As a result, the charges redistribute to allow the



FIG. 3. Plane-averaged electrostatic potential of FeCl₂/Sc₂CO₂ (a) $P \uparrow$ and (f) $P \downarrow$ along the *Z* direction; the plane-averaged differential charge density $\Delta \rho$ of FeCl₂/Sc₂CO₂ (b) $P \uparrow$ and (g) $P \downarrow$; (c)–(e) and (h)–(j) the three-dimensional isosurface of the electron density difference (isosurface value is $0.005 e Å^{-3}$) of the multiferroic system with $P \uparrow$ and $P \downarrow$ states, respectively, where the blue and red areas represent electron depletion and accumulation, respectively. Band alignments for FeCl₂/Sc₂CO₂ with ferroelectric polarization of (k) $P \uparrow$ and (l) $P \downarrow$, respectively, of isolated layers (left panel) and that after forming heterostructure (right panel). The energies are displayed in the unit of eV. The blue horizontal dashed lines denote the work functions of the systems.

system to reach a new equilibrium. The Sc₂CO₂ monolayer with different polarization states exhibit very different charge transfer characteristics when forming the vdW heterostructure as illustrated in Figs. 3(a) to 3(j). Since the Sc₂CO₂ is an out-of-plane ferroelectric material, the intrinsic dipole and the in-plane average electrostatic potential differ significantly along the out-of-plane direction for the two P-polarization states. When FeCl₂ is interfaced with $P \uparrow Sc_2CO_2$, the conduction bands of FeCl₂ for spin-up and spin-down subbands are all energetically higher than the valence bands of Sc₂CO₂, while the valence bands of FeCl₂ are lower than that of Sc₂CO₂. Such energy band structures are typical of type-II band alignment [Fig. 3(k)], which hinders the electron transfer between FeCl₂ and Sc₂CO₂. Nonetheless, the built-in electric field with a direction pointing from the Sc_2CO_2 to the FeCl₂ results in a weak electron transfer. Therefore, a small number of electrons accumulates around the FeCl2 as shown in Figs. 3(c) to 3(e). Such interfacial charge transfer process enhances the original internal electric field and leads to a potential step of 2.062 eV across the contact interface. However, such effect is still insufficient to close the common band gap in the $FeCl_2/Sc_2CO_2$ heterostructure.

In contrast, when the polarity state of Sc_2CO_2 is switched to $P \downarrow$, a larger charge transfer changes occurs as the valence band edge of Sc_2CO_2 becomes energetically higher than the spin-down conduction band of FeCl₂ [Fig. 3(1)]. The electrons in the valence bands of Sc_2CO_2 are sufficiently energetic to transit to the spin-up conduction bands of FeCl₂. As shown in the plane-averaged differential charge density $\Delta\rho$ and the 3D isosurface of the electron density difference of FeCl₂/Sc₂CO₂ heterostructures in Fig. 3(g) and Figs. 3(h)–3(j), a large amount of charge transfers occurs from Sc_2CO_2 to FeCl₂. The partial electron filling of the spin-down subbands of FeCl₂ from the Sc_2CO_2 valence bands causes the conduction bands to shift energetically downward toward the Fermi level. The overlap between the spin-down conduction band of $FeCl_2$ and the valence band of Sc_2CO_2 leads to the disappearance of the common gap, and the heterostructure is transformed into a semimetal with type-III band alignment.

C. Structural and electronic properties of the multiferroic heterostructure with the bilayer ferroelectric materials

Studies have shown that increasing the number of polar unit cells can make the free carrier density gradually approach the bound charge density. Therefore, the bilayer Sc_2CO_2 exhibits metallicity due to the strong covalent interaction between carbon and oxygen sublattices, facilitating the development of the necessary potential difference [57], as shown in the Supplemental Material [50]. The potential difference is twice that of a monolayer, which may cause more charge transfer when contacts the other materials. The results show that the nature of the electronic structure of FeCl₂ to be flexibly switched from semiconductor to half-metallic remains by changing the ferroelectric polarization of bilayer Sc₂CO₂, as shown in Figs. 4(b) and 4(c). The half-metal phase is more pronounced because of more electron transfer. Three more sandwich configurations were constructed, where the monolayer FeCl₂ is BB stacked vertically between two monolayer Sc₂CO₂: $P \uparrow$ FeCl₂ $P \uparrow$, $P \downarrow$ FeCl₂ $P \uparrow$, and $P \uparrow$ FeCl₂ $P \downarrow$, as shown in Fig. 4(a). $P \downarrow \text{FeCl}_2 P \downarrow$ and $P \uparrow \text{FeCl}_2 P \uparrow$ are mirror symmetric, which have the same electronic properties. The conduction subband of the spin down of FeCl₂ in the $P \downarrow$ polarization state will downshift and cross the Fermi level. This property does not change when adding another layer of Sc_2CO_2 with the same polarity, as can be seen from Fig. 4(d). However, when a layer of Sc₂CO₂ with opposite polarity is added, the overall polarity disappears. While the internal



FIG. 4. (a) Optimized geometries of four configurations of bilayer $Sc_2CO_2/FeCl_2$ or $Sc_2CO_2/FeCl_2/Sc_2CO_2$ vdW heterostructures; (b)–(f) electronic band structures of bilayer $Sc_2CO_2/FeCl_2$ or $Sc_2CO_2/FeCl_2/Sc_2CO_2$ vdW heterostructures under different polarized states, respectively.

polarity still exists, the built-in electric field will redistribute the spatial charge at the interface. Therefore, the $P \downarrow \text{FeCl}_2P \uparrow$ heterostructure exhibits half-metal properties in the spin-down state, and the $P \uparrow \text{FeCl}_2P \downarrow$ heterostructure is semiconducting, which is similar to monolayer ferroelectric materials.

D. Electrically switchable transport of a two-channel nanodevice: Quantum transport simulation

The two polarization states of $P \uparrow$ and $P \downarrow$ in Sc₂CO₂ can be reversibly switched by applying an external electric field to the heterostructure, and the polarization state is retained even after the removal of the external electric field. The energy barrier for switching polarization from the polar to nonpolar configuration is 0.52 eV per formula unit and proceeds via a multistep process of oxygen atom displacements [57]. The energy barrier is higher than that of the well-known perovskite ferroelectric LiNbO₃(0.26 eV) [63]. The switching of the FeCl₂/Sc₂CO₂ vdW heterostructure between the semiconducting and half-metallic behavior is thus nonvolatile. Importantly, due to the atomic thickness of the $FeCl_2/Sc_2CO_2$ vdW heterostructure, an external electric field generated by an external gate electrode can readily penetrate across the monolayers, thus allowing such ferroelectric-based polarity switching to be straightforwardly and efficiently achieved allelectrically [64].

Although memory devices based on multiferroic vdW heterostructures have been previously proposed by Zhao *et al.* [44] and Li and Zhao [45], the transport properties and the nanodevice application capabilities of multiferroic vdW heterostructures remain largely unexplored thus far. Here we construct a two-probe source-channel-drain proof-of-concept device to demonstrate how the vastly contrasting behaviors in $P \uparrow$ and $P \downarrow$ polarization states of the Sc₂CO₂ monolayer can be harnessed for valving applications. In Fig. 5(a), we present the simulated I - V curves at room temperature, where the devices exhibit an obvious valving effect with an on-off ratio of 3200. Moreover, the magnitude of the current can reach the order of μ A in the on state. The spin-up and spin-down currents exhibit different behaviors with the applied bias voltage in the $P \downarrow$ polarization states [see Fig. 5(b)]. The spin-up current decreases when the external bias exceeds 0.23 V, while the spin down increases monotonously with the external bias. Such contrasting behaviors arise from the spin-dependent band structures of the heterostructures [see Figs. 2(g) and 2(h)]. The spin-up bands have a large band gap above the Fermi level that suppresses electron transport [Fig. 2(g)]. This is in stark contrast to the case of spin-down bands in which the energy regime around the Fermi level is populated by the spin-down bands contributed by FeCl2 sublayer [Fig. 2(h)]. Such spincontrasting band structures directly lead to contrasting spin-up and spin-down currents as observed in Fig. 5(b). We calculate the transmission spectrum of the proposed FeCl₂/Sc₂CO₂ multiferroic vdW heterostructure device at zero bias [65–70]. The Fermi level is set to the zero-energy location. For the P \uparrow state, a large zero transport gap exists around the Fermi level for both spin-up and spin-down channels, which essentially switches off the electrical current in the device [Figs. 5(c)and 5(e)], thus providing a valving effect. Such a transport



FIG. 5. Simulated transport current as a function of the applied bias for FeCl₂/Sc₂CO₂ *BB* stacking with (a) different ferroelectric polarization states. (b) The spin-up and spin-down currents when the heterostructure is in the $P \downarrow$ state. The spin-dependent transmission spectrum of the device in (c) $P \uparrow$ and (d) $P \downarrow$ polarization states at zero bias. (e) and (f) show the schematic diagrams of a FeCl₂/Sc₂CO₂ multiferroic all-electric-controlled device based on the nonvolatile switching of the $P \uparrow$ and the $P \downarrow$ polarization states of the Sc₂CO₂ monolayer.

gap arises directly from the semiconducting nature of the the $FeCl_2/Sc_2CO_2$ heterostructure when the Sc_2CO_2 monolayer is electrically switched to $P \uparrow polarization$. On the other hand, when the Sc_2CO_2 monolayer is electrically switched to the $P \downarrow$ polarization, the heterostructure becomes semimetallic, which opens the transport channel with a large transmission coefficient around the Fermi level, thus allowing an electrical current to flow in the device [Figs. 5(d) and 5(f)].

IV. CONCLUSIONS

In summary, we investigated the electronic properties of the FeCl₂/Sc₂CO₂ multiferroic vdW heterostructure by using first-principles DFT and nonequilibrium Green's function transport simulations. We show that the electronic properties of the FeCl₂/Sc₂CO₂ heterostructures can be reversibly controlled by electrically switching the ferroelectric polarization state of the Sc₂co₂ monolayer, thus enabling a great wealth of functionalities to be derived from FeCl₂/Sc₂CO₂. The half-metallic state of FeCl₂/Sc₂CO₂ exhibits type-III band alignment, thus suggesting potential in tunneling field-effect transistor applications. The quantum transport simulation of a proof-of-concept two-terminal device demonstrated the capability of $FeCl_2/Sc_2CO_2$ heterostructures for valving applications. These findings reveal the potential of $FeCl_2/Sc_2CO_2$ heterostructures as a building block for designing the next generation of ultracompact and all-electric-controlled spintronics nanodevices.

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