Intrinsic multiferroicity in two-dimensional VOCl₂ monolayers†

Haoqiang Ai, Xiaohan Song, Siyun Qi, Weifeng Li ‡ and Mingwen Zhao †* 

The coexistence of ferroelectricity and magnetism in two-dimensional (2D) multiferroic materials with the thickness of few atomic layers offers a tantalizing potential for high-density multistate data storage but has been rarely verified in experiments. Herein, we propose a realistic 2D multiferroic material, VOCl₂ monolayer, which is mechanically strippable from the bulk material. It has a large intrinsic in-plane spontaneous electric polarization of 312 pC m⁻² and stable antiferromagnetism with the Néel temperature of 177 K. The off-center displacement of V ions that contributes to the ferroelectricity can be ascribed to the pseudo Jahn–Teller distortion. The energy barrier (0.18 eV) between two ferroelectric states with opposite electronic polarization renders the thermodynamic stability of the ferroelectricity and the switchability of the electric polarizations. The interplay between electric polarization and magnetism would lead to tunable ferroelectricity. Our findings are expected to offer a realistic platform for the study of 2D multiferroic materials as well as their applications in miniaturized memory devices.

Introduction

The spontaneous electric polarization in ferroelectrics, which is switchable by applying an electric field, is the basis of ferroelectric tunnel junctions and non-volatile memories in which information is encoded through the ferroelectric polarization.¹–⁵ Ferroelectric perovskite oxide materials, such as PbTiO₃ and BaTiO₃,⁴,⁵ with switchable electric polarization, have offered a tantalizing potential for memory storage and integrated microelectronics. Electronic devices demand a greater decrease in switching time and length scale, approaching the level of individual electrons and atoms, so ultrathin ferroelectric (2D) materials with intrinsic in-plane or out-of-plane ferroelectricity.⁶–⁹ However, when three-dimensional (3D) ferroelectric perovskite oxides are thinned down, a critical thickness for sustaining the FE states emerges due to the effects of surface energy, depolarizing electrostatic field and electron screening.⁵–¹¹ These ultrathin ferroelectric films are also detrimentally impacted by dangling bonds and dead layers at the ferroelectric/metal interfaces.¹²,¹³ A promising approach to solve this problem is to adopt two-dimensional (2D) materials with intrinsic in-plane or out-of-plane ferroelectricity.⁴–⁷,¹⁰,¹³–¹⁶ A series of 2D materials have been revealed or predicted to have stable ferroelectricity, such as SnTe,¹⁰ CuInP₂Se₆,⁴ α-In₃Se₃,¹³,¹⁸ 1T-MoS₂,¹⁴ phosphorene analogues⁷,¹⁵,¹⁷ and Sc₂CO₂.¹⁶ The thickness of these 2D materials is only a few atomic layers and thus quite promising for minimizing the length-scale of ferroelectric devices.

Multiferroics, which combine ferroelectricity and magnetism in a single phase, have been attracting significant attention¹⁹,²¹–²⁴ due to their fascinating physics and applicable magnetoelectric functionalities. However, very few multiferroics are reported²⁵ because of the difficulty in balancing the energy-lowering covalent bond formation and energy-raising electronic Coulomb repulsion.²³,²⁶ Ferroelectricity and magnetic order in type-I multiferroics have different sources and couple weakly, whereas the magnetic order induces the ferroelectricity in type-II multiferroics.¹⁹,²³ The interplay between ferroelectricity and magnetic order is crucial for tuning the magnetic state by an electric field, and vice versa. For example, the coexistence of large ferroelectric polarization and antiferromagnetism with high transition temperatures in multiferroic BiFO₃ makes it appealing for applications in nonvolatile logic and memory devices.²⁷–³⁰

Combining ferroelectricity and magnetism in a few-atomic layers (2D multiferroics) is quite promising for miniaturization of nonvolatile logic and memory devices. Very recently, first-principles calculations have demonstrated the coexistence of ferroelectricity and ferromagnetism in the electron-doped CrBr₃ monolayer¹⁹ and chemically functionalized phosphorene,³¹ offering an avenue for the design of 2D multiferroics. However, the intrinsic multiferroicity in 2D materials without the need for doping or surface treatment has been rarely reported.

In this study, we proposed a realistic 2D multiferroic material, VOCl₂ monolayer, with intrinsic ferroelectricity (FE)
and antiferromagnetism (AFM) through mining the 2D van der Waals (vdW) materials.\textsuperscript{11} It can be exfoliated from the layered bulk crystal.\textsuperscript{33,34} The in-plane spontaneous electric polarization 312 pC m\textsuperscript{−1} is comparable to that of 3D multiferroics.\textsuperscript{21} The energy barrier of 0.18 eV between two ferroelectric (FE) phases with opposite electronic polarization implies that the ferroelectricity is thermodynamically stable at room temperature and switchable by applying a moderate external electric field.\textsuperscript{7,20} Meanwhile, this 2D material has an AFM ground state with anisotropic exchange interaction\textsuperscript{35} and semiconducting features. The Neel temperature evaluated from the Monte Carlo simulations based on the Ising model is about 177 K. The magnetism-dependent ferroelectricity implies the possibility of tuning the ferroelectricity by magnetic field. Our theoretical study offers a promising realistic platform for investigating the interplay of ferroelectricity and anti-ferromagnetism in 2D materials, which is crucial for the miniaturization of nonvolatile logic and memory devices.

**Methods**

Our first-principles calculations were performed on the basis of spin-polarized density functional theory (DFT) implemented in the plane wave basis Vienna \textit{ab initio} simulation package (VASP) code.\textsuperscript{36–38} The electron–ion interactions were described by the projector augmented wave (PAW) method.\textsuperscript{39} The electron–electron interaction was treated within a generalized gradient approximation (GGA) in the form proposed by Perdew, Burke and Ernzerhof (PBE) for the exchange–correlation functional.\textsuperscript{40} We also employed the GGA+U strategy\textsuperscript{19,41,42} to check the dependence of ferroelectricity and magnetism on the U value due to the localized d electrons (see the ESIF). The supercell was repeated periodically along the \textit{a}- and \textit{b}-directions (the two in-plane basis vectors), while a vacuum region of about 20 Å was applied in the \textit{c}-direction to avoid the spurious interactions with its periodic images. The energy cutoff employed for plane-wave expansion of electron wave functions was set to be 520 eV. A \(2 \times 2 \times 1\) supercell was adopted to search for the stable magnetic order and the ground state structure of the monolayer. Meanwhile, the Brillouin zone (BZ) integration was sampled with a \(k\)-grid density of \(7 \times 7 \times 1\) using the Monkhorst-Pack \(k\)-points scheme.\textsuperscript{41} The structures were fully optimized until the maximal forces were less than 0.005 eV Å\textsuperscript{−1} and the convergence criteria for energy was set to be \(10^{-6}\) eV. The convergence of this strategy has been verified. All the atomic positions and the lattice constants were fully optimized using a conjugate gradient (CG) algorithm. van der Waals (vdW) correction was included by using the D2 method.\textsuperscript{44} The phonon spectra were calculated using the finite displacement method implemented in Phonopy.\textsuperscript{45} Ferroelectric spontaneous polarization was calculated using the Berry phase approach,\textsuperscript{46,47} in which both electronic and ionic contributions were taken into account. The minimum energy pathways of ferroelectric transitions were determined through the climbing image nudged elastic band method (CINEB)\textsuperscript{48} based on the interatomic forces and total energies acquired from the DFT calculations.

**Results and discussion**

**Geometry and stability**

We started from the ground-state atomic structures of VOCl\textsubscript{2} monolayer. A bulk VOCl\textsubscript{2} crystal with a layered structure (\textit{Immm} symmetric group) has been synthesized experimentally.\textsuperscript{13,34,49} The weak vdW interaction facilitates the exfoliation processes that produce 2D VOCl\textsubscript{2} monolayers from the bulk crystal. As a VOCl\textsubscript{2} monolayer is exfoliated from the bulk counterpart, severe surface reconstruction may take place. To determine the energetically most favorable structure of the VOCl\textsubscript{2} monolayer, we constructed four configurations, namely \(\alpha\), \(\beta\), \(\gamma\) and \(\delta\)-configurations, as shown in Fig. S1 (ESIF) and then relaxed them without any symmetry constraints. Electron spin-polarization was also involved in these calculations. We found that the noncentrosymmetrical \(\alpha\)-VOCl\textsubscript{2} monolayer is the energetically most favorable followed by \(\beta\)-VOCl\textsubscript{2} monolayer, which is less stable by about 9.15 meV per formula unit. The centrosymmetrical \(\gamma\) and \(\delta\)-configurations eventually convert to the \(\alpha\)-configuration after relaxation. We thus focus on the \(\alpha\)-VOCl\textsubscript{2} monolayer in investigating the electronic and magnetic properties in the following parts.

The optimized structure of \(\alpha\)-VOCl\textsubscript{2} monolayer is depicted in Fig. 1. The unit cell illustrated by the dashed black line belongs to a space group of \(Pmm2\). Each V atom connects to four halogen atoms and two O atoms and fills the octahedral holes defined by halogen and oxygen framework, as shown in

![Image](https://example.com/image.png)
Fig. 1(b). It is interesting to see that the central inversion symmetry is absent in the VOCl$_2$ monolayer. The V atom shifts toward one of the bridging O atoms along the basis vector $a$ (taken as the $z$-direction), leaving the octahedral center by about 0.248 Å. The two V–O bonds are no longer equivalent in the VOCl$_2$ monolayer, similar to the case of bulk VOCl$_2$ crystal.$^{33,34}$ The optimized lattice constants and bond lengths are listed in Table 1, which agree well with the previous study.$^{49}$ To provide more reference information to the experimental observation, the simulated STM image of VOCl$_2$ monolayer is also displayed in Fig. 1(c).

We then verify the stability and plausibility of the VOCl$_2$ monolayers. From the phonon spectra shown in Fig. 1(d), we do not observe the imaginary-frequency modes and thus believe that the VOCl$_2$ monolayers are dynamically stable. Mechanical cleavage and liquid exfoliation are the most popular and feasible ways of fabricating 2D monolayers from their parent bulks with layered structures.$^{50–52}$ To extract large area free-standing nanosheets by exfoliation, a small cleavage energy is needed.$^{53}$ Herein, we model the exfoliation process by introducing a fracture in the bulk and performing a scan on the separation distance $d$ between two fractured parts, as adopted in many previous theoretical studies.$^{20,53,54}$ The calculated cleavage energy $E_{cl}$ and cleavage strength $\sigma$ as functions of separation distance are illustrated in Fig. 1(e). The cleavage strength is defined as $\sigma = \frac{\partial E_{cl}}{\partial d}$, as implemented in previous studies.$^{54}$ The ideal cleavage energy and the maximum cleavage strength are about 0.16 J m$^{-2}$ and 0.72 GPa, much smaller than those of graphite (0.37 J m$^{-2}$ and 2.10 GPa),$^{54,55}$ suggesting that VOCl$_2$ monolayers can be easily prepared from their bulk via mechanical exfoliation in experiments like graphene, due to their weak interlayer vdW interactions.

**Ferroelectricity**

Having demonstrated the stable configuration of the VOCl$_2$ monolayer, we then turned to the intrinsic ferroelectricity of the 2D system. Evidently, the off-centering displacement of the V ion in an octahedron breaks the inversion symmetry of the VOCl$_2$ monolayer. The centers of positive and negative charges no longer coincide. Consequently, a sizable spontaneous polarization is most likely to occur. To evaluate the spontaneous polarization of the VOCl$_2$ monolayer, we choose an adiabatic pathway from FE phase ($\alpha$-configuration) to paraelectric (PE) phase ($\delta$-configuration) by using the modern theory of polarization based on the Berry phase method.$^{46,47}$ Unsurprisingly, our calculations reveal a spontaneous in-plane electric polarization along the $a$-direction. The total polarization of the VOCl$_2$ monolayer as a function of normalized displacement with respect to the PE phase is plotted in Fig. 2(a). As a marker of spontaneous electric polarization, the double well potential along the adiabatic path is also plotted in

**Table 1** Structural parameters of the VOCl$_2$ monolayer. Calculated total energies $E$ (meV) relative to the energy of AFM-3 configuration ($E_{AFM-3}$) of four magnetic orders (FM, AFM-1, AFM-2 and AFM-3) in a $2 \times 2 \times 1$ supercell of the VOCl$_2$ monolayer. Nearest-neighbor exchange coupling parameters ($J_a$ and $J_b$) along $a$- and $b$-direction, and next-nearest-neighbor exchange coupling parameter ($J_{ab}$) (in meV)

<table>
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<tr>
<th>Lattice constants</th>
<th>Bond length</th>
<th>Bond angles</th>
<th>$E_{FM}$</th>
<th>$E_{AFM-1}$</th>
<th>$E_{AFM-2}$</th>
<th>$E_{AFM-3}$</th>
<th>$J_a$</th>
<th>$J_b$</th>
<th>$J_{ab}$</th>
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<td>$a = 3.791$ Å</td>
<td>2.382 Å (V–Cl)</td>
<td>120° (O–V–O)</td>
<td>171.4</td>
<td>184.5</td>
<td>6.7</td>
<td>0</td>
<td>0.39</td>
<td>−21.82</td>
<td>0.62</td>
</tr>
<tr>
<td>$b = 3.367$ Å</td>
<td>2.143 Å (V–O$_1$)</td>
<td>89.9/88.4° (Cl–V–Cl)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td></td>
<td>1.648 Å (V–O$_2$)</td>
<td></td>
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Magnetic properties

The unfilled d-orbitals of V ions in the VOCl₂ monolayer may lead to electron spin-polarization and magnetism. We determined the energetically preferred magnetic order in VOCl₂ monolayer by using a 2 × 2 × 1 supercell. Starting from different initial spin alignments of the V ions, self-consistent DFT calculations converged to a ferromagnetic (FM) order and three “antiferromagnetic (AFM)” orders (AFM-1, AFM-2 and AFM-3), as shown in Fig. 3(a). The spin charge density distribution was calculated from Δρ = ρ↑ − ρ↓, where ρ↑ and ρ↓ represent the charge density of spin-up and spin-down, respectively. We can see that the local magnetic moments originate predominantly from the V atoms, while O atoms carry only tiny local magnetic moments. The local magnetic moment of the V ion is about 1.0μB in consistency with the V⁴⁺ charge state in the VOCl₂ monolayer. Our calculations show that the AFM-3 order is energetically most favourable. In this magnetic order, the nearest-neighbour magnetic moments are aligned in an antiparallel way along both a- and b-direction. It is energetically more stable than the FM order by about 171.4 meV. We also employed the GGA+U strategy with different (U-J) values in determining the magnetic orders and found that the VOCl₂ monolayer always had an AFM ground state independent of the (U-J) values, as shown in Table S1.† The electronic band structure of the AFM-3 VOCl₂ monolayer is plotted in Fig. 3(b). The partial electron density of states (PDOS) projected onto to V, O and Cl atoms are presented in the right panel of this figure. Clearly, VOCl₂ monolayer has an indirect band gap of about 0.81 eV with the valence band maximum (VBM) and conduction band minimum (CBM) residing at R (1/2, 1/2, 0) and Γ (0, 0, 0), respectively. Both the valence band and conduction band nearest to the Fermi level are predominantly contributed by the V atoms.

Magnetic anisotropy is crucial for stabilizing long-range magnetic order in 2D materials because it lifts the so-called Mermin–Wagner restriction.⁴¹,⁵⁷,⁵⁸ We evaluated the magnetic anisotropy of the VOCl₂ monolayer by taking the spin–orbit coupling (SOC) into account.⁴³ Three in-plane magnetizations along the [100], [010], and [110] directions, and two out-of-plane magnetizations along the [001] and [111] directions were considered. The calculated magnetic anisotropy energies (MAEs) for these magnetizations are in the range of 3.3–16.6 μeV per atom (see Table S4 of the ESI†), which are larger than those of cubic Fe (1.4 μeV per atom) and Ni (2.7 μeV per atom).⁴¹,⁵⁹ The easy axis of VOCl₂ monolayer is along the out-of-plane [001] direction, which is perpendicular...
to the ferroelectric polarization direction. We also employed Monte Carlo (MC) simulations based on the Ising model to reveal the temperature-dependent magnetism. This strategy has been widely adopted in previous studies.\textsuperscript{41,60} For simplification, we only considered the nearest-neighbour (NN) and next-nearest-neighbour (NNN) exchange interactions. The NN exchange coupling parameters along the a- and b-directions are respectively \(J_a\) and \(J_b\) and the NNN exchange coupling parameter is \(J_{ab}\),\textsuperscript{35,60} as indicated in the insets of Fig. 4.

The Hamiltonian is written as

\[
H = H_0 - \sum_{\langle ij \rangle_a} J_a M_i M_j - \sum_{\langle\langle j \rangle \rangle_a} J_b M_i M_j - \sum_{\langle\langle k \rangle \rangle} J_{ab} M_i M_j
\]  

where \(H_0\) is the nonmagnetic Hamiltonian; \(M_i\) is the net magnetic moment at site \(i\); \(\langle ij \rangle_a\) corresponds to the nearest V atoms along the a-direction; \(\langle\langle j \rangle \rangle\) stands for the NNN V atoms. The total energies of the four magnetic orders are given as:

\[
E_{\text{FM}} = E_0 - 4M^2 (J_a + J_b + 2J_{ab})
\]

\[
E_{\text{AFM-1}} = E_0 - 4M^2 (-J_a + J_b - 2J_{ab})
\]

\[
E_{\text{AFM-2}} = E_0 - 4M^2 (J_a - J_b - 2J_{ab})
\]

\[
E_{\text{AFM-3}} = E_0 - 4M^2 (-J_a - J_b + 2J_{ab})
\]

Here, \(E_0\) represents the energy for the nonmagnetic part. By solving these equations, we obtained the three exchange coupling parameters \((J_a, J_b\) and \(J_{ab})\), as listed in Table 1, which are implementable in the MC simulations.

In our MC simulations, a 50 \(\times\) 50 square supercell containing 2500 local magnetic moments was adopted and the calculations lasted for \(2 \times 10^9\) loops at each temperature. The Néel temperature \(T_N\) can be estimated from the temperature-dependent specific heat curves,\textsuperscript{41,55,60} corresponding to the abrupt change in the specific heat, as shown in Fig. 4. We calculated \(T_N\) for the VOCl\(_2\) monolayer is 177 K. Although the Néel temperature is much lower than room temperature, it exceeds the liquid nitrogen temperature (77 K) that can be easily achieved in experiments.

To understand the anisotropic magnetic coupling, we plotted the orbital-resolved electron density of states (DOS) of the VOCl\(_2\) monolayer in Fig. 5(a). We projected the DOS onto the 3d orbitals of V and 2p (3p) orbital of O(Cl) atoms. The d-orbitals of the V ion in the distorted octahedral crystal field split into \(d_{xy}\), \(d_{yz}\), \(d_{xz}\), and \(d_{xy}\) orbitals and a doubly-degenerate \(d_z^2\) orbital. From Fig. 5(a), we can see that the valence states nearest to the Fermi level, which are spin-polarized, arise mainly from the \(d_{xy}\) orbital of the V atom. According to Goodenough–Kanamori-Anderson rules,\textsuperscript{41,62} the super-exchange interaction is very weak both in a- and b-directions because electron transfer is forbidden by symmetry in 90° V–Cl–V configuration and there are no visible states of the O atom overlapping with the \(d_{xy}\) state of the V atom in energy. The relatively strong AFM coupling between V ions in the b-direction can be attributed to the direct-exchange interaction.\textsuperscript{63} It is noteworthy that there is much overlap between the \(d_{xy}\) states of the nearest-neighbour (NN) V atoms along the b-direction owing to the short distance (3.367 Å) between them. On the contrary, in the a-direction, a larger distance (3.791 Å) between NN V atoms results in the weak direct exchange interaction indicated by the small \(J_a\) and \(J_{ab}\).

Interestingly, both \(J_a\) and \(J_{ab}\) have positive values, suggesting that the magnetic moments prefer FM order in these directions. However, the strength of the FM coupling between NNN V ions is stronger than that between NN V ions along the a-direction, leading to the AFM-3 order shown in Fig. 3(a).

**Origin of multiferroicity**

It is noteworthy that a ferroelectric displacement of the B cation in perovskite ABO\(_3\) with magnetism is inhibited due to the d electrons (known as the "d\(_0\)" rule), meaning that magnetism is often incompatible with ferroelectricity.\textsuperscript{21,25} However, this is not the case for the VOCl\(_2\) monolayer. To reveal the origins of the ferroelectric displacement of V in the AFM-3 VOCl\(_2\) monolayer, we compared the orbital-resolved electron density of states projected onto the 3d orbitals of V ion of FE (Pmm2) and PE (Pmmm) states, as shown in Fig. 5. For the centrosymmetric PE state, the halogen-oxygen octahedron breaks the degeneracy of the \(t_{2g}\) orbital of an ideal regular octahedral crystal field, but the energy gap between d\(_{xy}\) and d\(_z^2\) orbitals in the proximity of the Fermi level is very small, as shown in Fig. 5(b). The off-centre displacement of V ions along the a-direction in the FE state, however, pushes the DOSs of d\(_{xy}\) and d\(_z^2\) orbitals downwards and upwards, respectively, widening the energy gap between them. As a consequence, the total energy of the FE state is lower than that of the PE state. The ferroelectric displacement of V atoms could be understood in terms of the Pseudo Jahn–Teller distortion (PJTE) mechanism.\textsuperscript{21,26,64} For the d\(_1\) configuration of the V ion in the FE state, the highest occupied molecular-orbital electronic configuration is \((t_{2u})^6(d_{xy})^1\) and the lowest unoccupied molecular-orbital configuration is \((t_{1u})^5(d_{xy})^1(d_{xz} + d_{yz})^1\).
where the arrows indicate the two spin states. As these electronic configurations possess the same spin multiplicity, they could mix under nuclear displacements with a small energy gap to produce the PJTE dipolar distortion. Furthermore, considering that the ferroelectric displacement is inhibited in the perovskite ABO$_3$ with the $d^{1}$ configuration, the multiferroicity found in the VOCl$_2$ monolayer could offer a novel strategy to design new multiferroics.

Another interesting issue is the interplay between ferroelectricity and magnetic order. As discussed above, the easy axis of magnetization of the VOCl$_2$ monolayer is perpendicular to the ferroelectric polarization direction, which would preclude direct electric-field control of the magnetization like in BiFeO$_3$. However, we compared the ferroelectricity of the AFM-3 and FM states. We found that the ferroelectricity of the FM state is suppressed in comparison with the AFM-3 state. The in-plane spontaneous electric polarization of the FM state is 171 pC m$^{-1}$, which is only about 1/2 of that of the AFM-3 state. The ferroelectric displacements of V ion with respect to the center of octahedron are 0.238 Å for the FM state, close to that of the AFM-3 state, 0.248 Å. This implies that the change of ferroelectricity from AFM-3 to FM state arises mainly from the electronic contributions rather than the ionic displacement. A similar scenario has also been reported in LuFe$_2$O$_4$, an electronic ferroelectric, in which charge structures can be affected by an external magnetic field. Thus, it would be possible to tune the ferroelectricity by an external magnetic field in the VOCl$_2$ monolayer.

In addition, the coexistence of ferroelectricity and antiferromagnetism was also found in VOBr$_2$ monolayers. Both the results and mechanisms involved in the VOBr$_2$ monolayer are quite similar to those of the VOCl$_2$ monolayers (shown in the ESI†).

**Conclusions**

In summary, we demonstrate from first-principles calculations the coexistence of ferroelectricity and antiferromagnetism in the VOCl$_2$ monolayer, which is exfoliated from the layered bulk material. The absence of central inversion symmetry due to the off-center displacement of V ions in the octahedral site leads to a high in-plane spontaneous polarization of 302 pC m$^{-1}$. The energy barrier of 0.18 eV between two ferroelectric states with opposite electronic polarization ensures the stability of the ferroelectric state at room temperature and the switchability of polarization under an external electric field. Meanwhile, the VOCl$_2$ monolayer has stable antiferromagnetic order with highly-anisotropic magnetic coupling. The Néel temperature evaluated from Monte Carlo simulations based on the Ising model is 177 K. The magnetism-dependent ferroelectricity implies the possibility of tuning the ferroelectricity by...
magnetic field. Compared with recently-proposed 2D ferroelectrics such as SnTe and α-In₅Se₃ and 2D magnetic semiconductors such as CrI₃, the integration of AFM and FE in VOCl₂ monolayer is quite promising for 2D multiferroic-based nanoscale memory devices.

Conflicts of interest

There are no conflicts to declare.

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Notes and references
