Intrinsic ferromagnetism with high Curie temperature and strong anisotropy in a ferroelastic V*X* monolayer (*X* = P, As)

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ABSTRACT

Two-dimensional (2D) multiferroic materials, with combined large magnetic anisotropic energy (MAE) and high Curie temperature (T_C), have great potential in emerging electronic device applications, such as high-density multistate data storage and fast operation. However, various 2D spintronic materials fabricated in experiment, so far have small MAE and low T_C. Using first-principles calculations, we proposed two VX (X = P, As) monolayers, which are identified to possess robust stability dynamically, thermally, and mechanically. VX monolayers are half-metals with coexisting ferroelastic and ferromagnetic properties. Moreover, their ground states exhibit significant in-plane MAEs of 101.48 and 262.68 μeV per V atom for VP and VAs monolayers, respectively. Additionality, the Curie temperatures of VP and VAs monolayers predicted from Heisenberg model are as high as 545 and 890 K, respectively, and the ferromagnetism could survive at room temperature. Our results are beneficial for further research on 2D multiferroics and provide the possibility for future nanodevices.

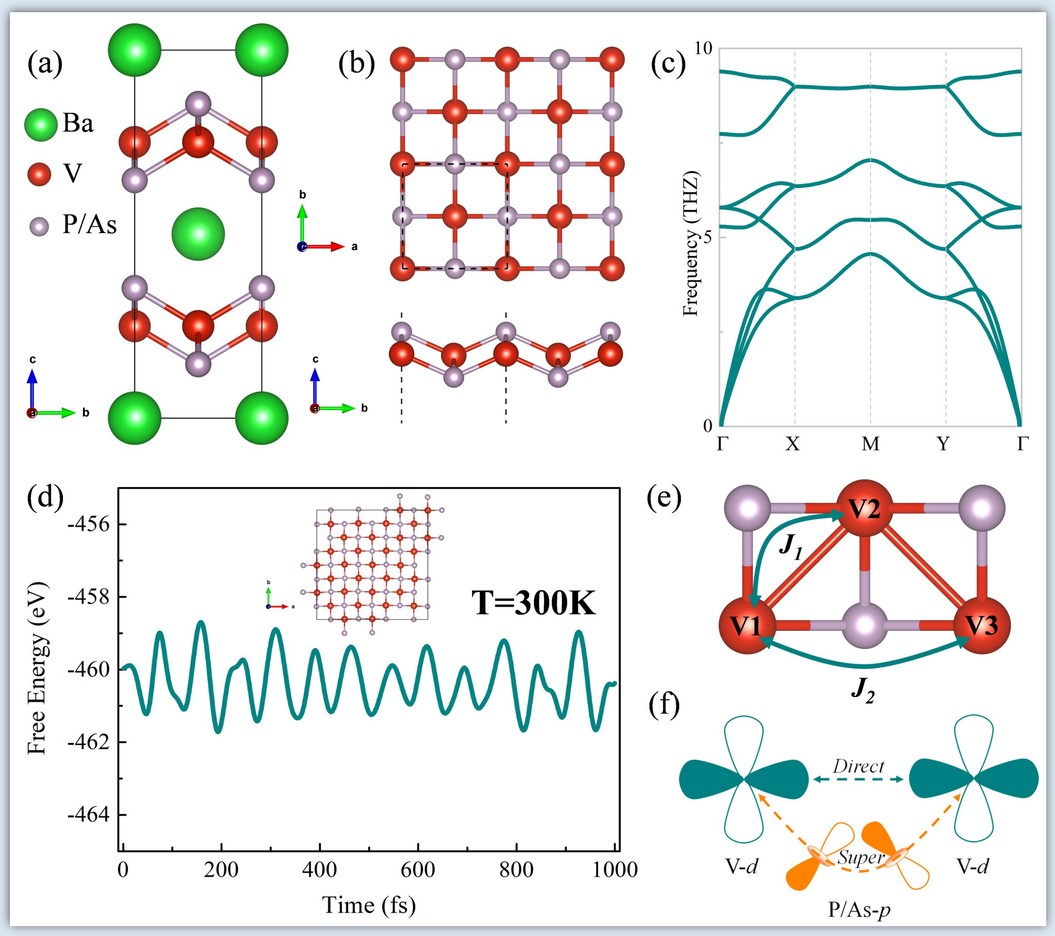

FIG. 1. (a) Crystal structure of layered ternary compounds BaV_2X_2 (X = P, As). (b) The top and side views of the VP monolayer. The dashed rectangle indicates the primitive cell. (c) The phonon spectra of the VP monolayer. (d) The evolution of energy over time at T = 300 K from molecular dynamics simulations of VP monolayer. (e) The exchange constants $\boldsymbol{J}_1$ and $\boldsymbol{J}_2$ are the NN and NNN interaction parameters, respectively. (f) Illustration of the direct-exchange and super-exchange mechanisms.

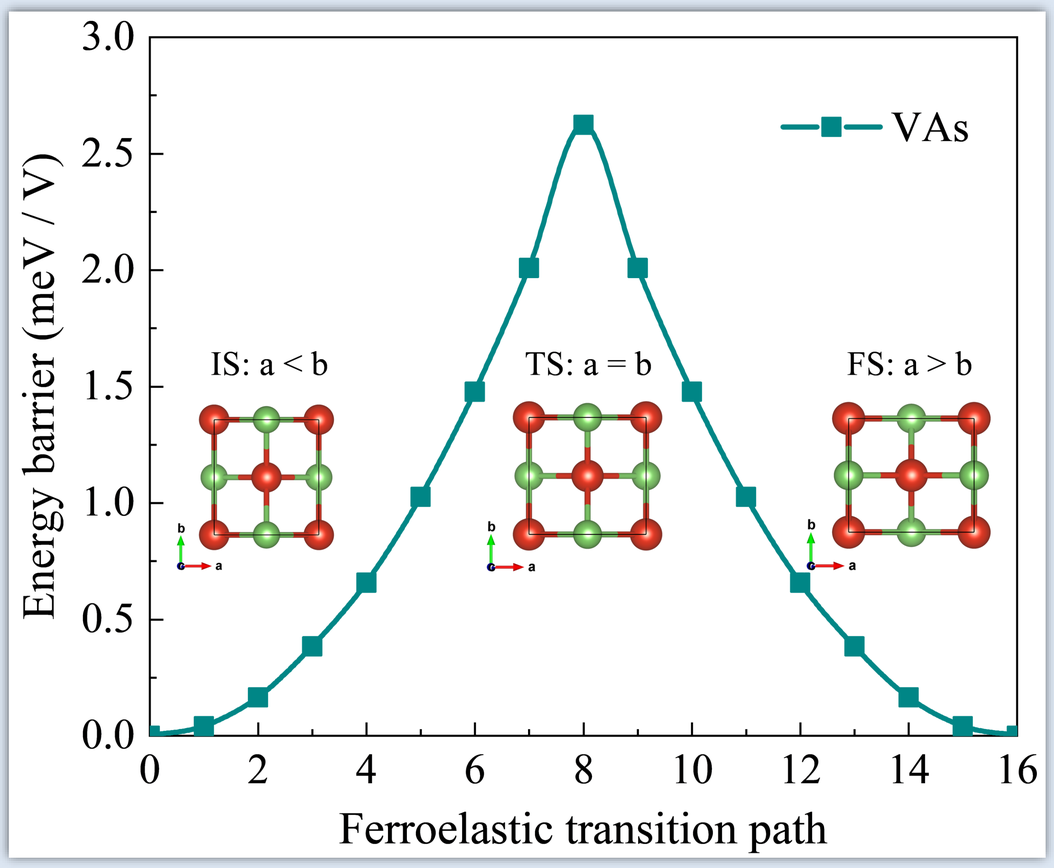

FIG. 3. The energy barrier for ferroelastic switching of VAs monolayer. The insets are schematic diagrams of the initial state (IS), transition state (TS) and final state (FS).

Computational Details:

VASP Pseudo-Potentials: PAW; Energy cut-off: 600 eV;

K-mesh in the Brillouin zone: 8×8×1 Γ centered Monkhorst-Pack

Functional approximation: PBE, and an additional on-site interaction *U*; HSE06

Curie temperature simulation: *Ising* model and anisotropic *Heisenberg* model

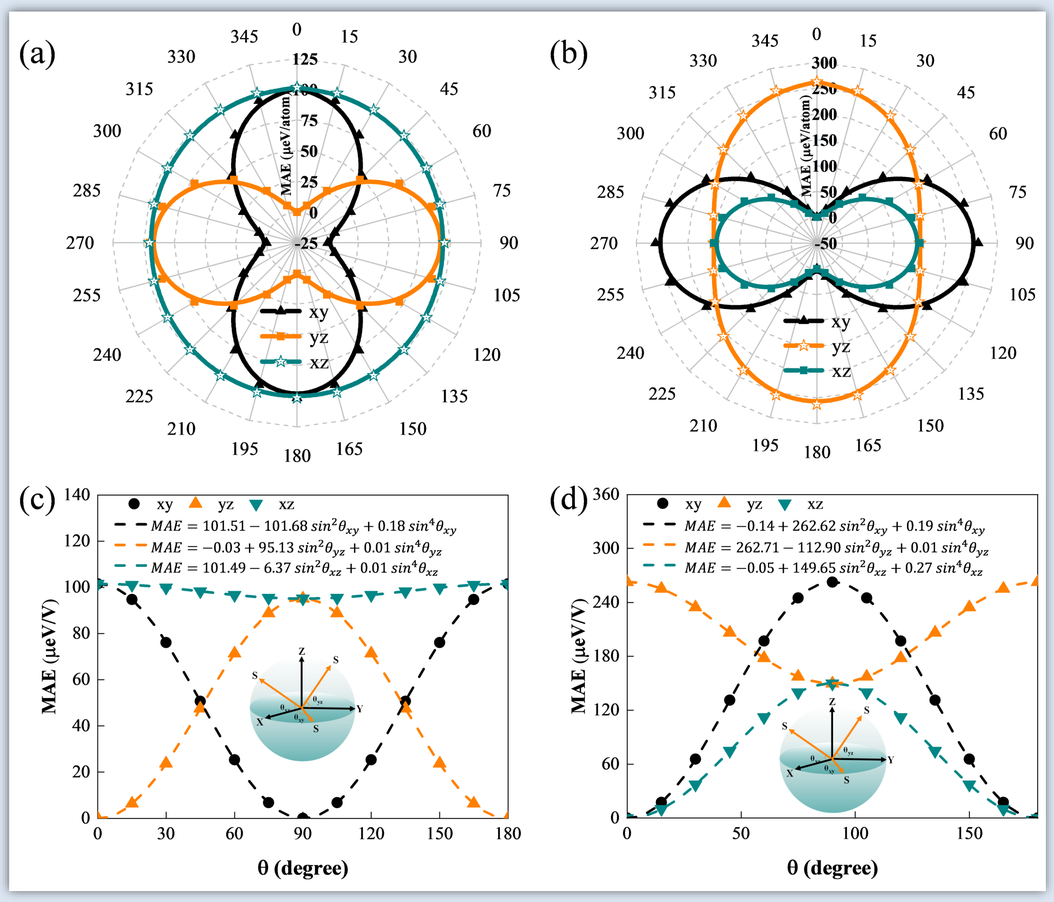

FIG. 2. Angular dependences of the MAEs in the polar and Cartesian coordinate systems for (a), (c) VP monolayer and (b), (d) VAs monolayer with the direction of magnetization lying on three different planes. The inset illustrates that the spin vector ***S*** on the *xy*, *yz*, and *xz* planes is rotated with an angle θ about the *x*, *y*, and *z* axes, respectively, $\theta xz = 0^\circ$ represents the moment lying in the *z* axis.

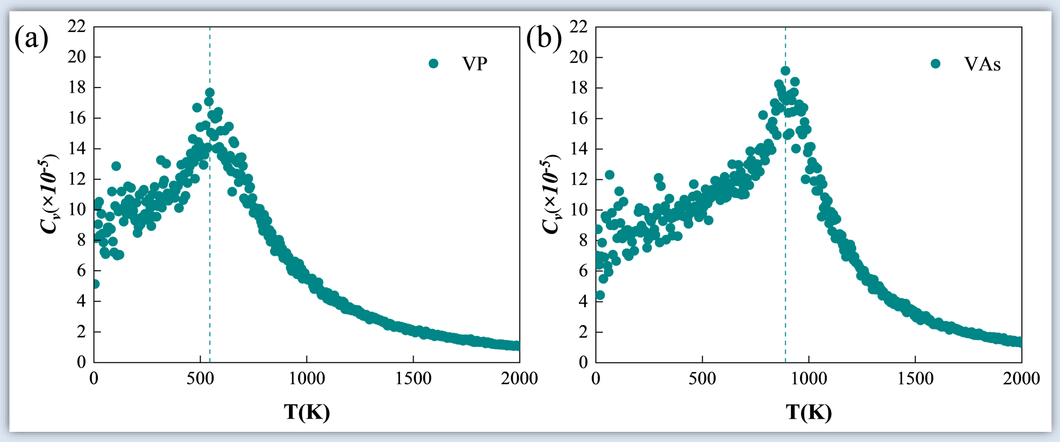

FIG. 4. The Monte Carlo simulated specific heat C*v* as a function of temperature using the Heisenberg model for (a) VP monolayer and (b) VAs monolayer.

TABLE I. The magnetic coupling parameters $\boldsymbol{J}_1$, $\boldsymbol{J}_2$, and $\boldsymbol{J}_3$ (meV), magnetic anisotropy energies (μeV per V) in different directions, $\boldsymbol{T}_C$ (K) from Ising and anisotropic Heisenberg (AH) models for VX (X = P, As) monolayers, and the magnetic moment M (μB) per V atom.

System	J_1	J_2	J_3	E(100)	E(010)	E(001)	$T_{C\text{-Ising}}$	$T_{C\text{-AH}}$	M
VP	26.30	10.41	-2.21	101.48	0	95.14	1850	545	2.17
VAs	28.87	3.55	12.40	0	262.68	149.82	2840	890	2.23

Conclusions

In summary, based on first-principles calculations, we reveal two stable 2D multiferroic materials VP and VAs monolayers with coexisting ferromagnetism and ferroelasticity. Both systems show dynamic, thermal, and mechanical stability and they can easily be fabricated from peeling the bulk hanks to the low exfoliation energies. Furthermore, VP and VAs monolayers have high Curie temperatures (545 K for VP and 890 K for VAs) and large MAE (101.48 μeV/V atom for VP and 262.68 μeV/V atom for VAs). Our discovery provides an ideal platform for exploring 2D multiferroic materials and spintronic devices in practical applications.

[1] X. Cheng, S. Xu, F. Jia, G. Zhao, M. Hu, W. Wu, and W. Ren, Phys. Rev. B. **104**, 104417 (2021). DOI: 10.1103/PhysRevB.104.104417