Two-dimensional (2D) materials, such as graphene, transition metal dichalcogenides and black phosphorus [1–5], have attracted tremendous attention due to their excellent electrical, optical and acoustic properties. Although many efforts have been devoted to investigating 2D materials, the 2D semiconductors with intrinsic magnetism are still rare [6–8]. Recently, the successful synthesis of intrinsic ferromagnetic (FM) semiconductor CrI$_3$, the 2D Janus materials M$_2$Cl$_3$I$_3$ (M=3d transition metals) are studied by the density functional theory (DFT) calculations. A stable 2D (in x-y plane) antiferromagnetic (AFM) Janus semiconductor Fe$_2$Cl$_3$I$_3$ with normal magnetization ($\mathbf{m}||z$) of sublattice is obtained. By applying tensile strain up to about 15%, the following four magnetic states sequentially occur: AFM with $\mathbf{m}||z$ of sublattice, AFM with $\mathbf{m}||xy$ of sublattice, FM with $\mathbf{m}||xy$, and FM with $\mathbf{m}||z$. Such novel magnetic phase diagram driven by strain can be well understood by a Heisenberg Hamiltonian with the single-ion anisotropy term, where the SOC of I atoms is found to play an essential role. In addition, the electric polarization of Fe$_2$Cl$_3$I$_3$ preserves with strain due to the broken inversion symmetry. Our results predict the Janus material Fe$_2$Cl$_3$I$_3$ as a rare example of 2D semiconductors with both spin and charge polarizations, and reveal the highly sensitive strain-controlled magnetic states and magnetization direction, which highlights the 2D magnetic Janus semiconductor as a new platform to design spintronic materials.

I. I. INTRODUCTION

Two-dimensional (2D) materials, such as graphene, transition metal dichalcogenides and black phosphorus [1–5], have attracted a great interest for their potential applications. Inspired by the recently fabricated 2D ferromagnetic (FM) semiconductor CrI$_3$, the 2D Janus materials M$_2$Cl$_3$I$_3$ (M=3d transition metals) are studied by the density functional theory (DFT) calculations. A stable 2D (in x-y plane) antiferromagnetic (AFM) Janus semiconductor Fe$_2$Cl$_3$I$_3$ with normal magnetization ($\mathbf{m}||z$) of sublattice is obtained. By applying tensile strain up to about 15%, the following four magnetic states sequentially occur: AFM with $\mathbf{m}||z$ of sublattice, AFM with $\mathbf{m}||xy$ of sublattice, FM with $\mathbf{m}||xy$, and FM with $\mathbf{m}||z$. Such novel magnetic phase diagram driven by strain can be well understood by a Heisenberg Hamiltonian with the single-ion anisotropy term, where the SOC of I atoms is found to play an essential role. In addition, the electric polarization of Fe$_2$Cl$_3$I$_3$ preserves with strain due to the broken inversion symmetry. Our results predict the Janus material Fe$_2$Cl$_3$I$_3$ as a rare example of 2D semiconductors with both spin and charge polarizations, and reveal the highly sensitive strain-controlled magnetic states and magnetization direction, which highlights the 2D magnetic Janus semiconductor as a new platform to design spintronic materials.

Among various 2D materials, the 2D Janus materials are very attractive. Compared to their prototypes, Janus materials have broken symmetries, and thus can induce many intriguing properties, such as large spin-orbit coupling (SOC), piezoelectricity, polarization, etc. [30–34]. The first graphene-based Janus material was graphene, where the Dirac cone was opened with a small gap and the FM was obtained [35]. Substituting one sulfur layer with selenium in GaS, the piezoelectric coefficient in Ga$_2$Se was enhanced as large as four times [36]. The 2D Janus material MoS$_2$ [37, 38], which has been successfully synthesized recently, not only has a better hydrogen evolution reaction efficiency, but also possesses the topological and ferroelastic properties [39] compared with its prototype MoS$_2$ monolayer [40].

In this work, we propose a stable 2D magnetic Janus semiconductor Fe$_2$Cl$_3$I$_3$. By means of first-principle calculations, Fe$_2$Cl$_3$I$_3$ was found to be a 2D AFM semiconductor with out-of-plane magnetization ($\mathbf{m}||z$) of sublattice. Due to the charge redistribution caused by different electronegativity of Cl and I atoms and the broken inversion sym-
metry, $\text{Fe}_2\text{Cl}_3\text{I}_3$ monolayer possesses electrical polarization of about 0.18 eÅ and piezoelectricity of about 4.48 pm/V. By applying biaxial tensile strain up to about 15% on $\text{Fe}_2\text{Cl}_3\text{I}_3$ monolayer, a novel phase diagram with four magnetic states is found: AFM with out-of-plane magnetization of sublattice, AFM with in-plane magnetization of sublattice, FM with in-plane magnetization, and FM with out-of-plane magnetization. The magnetic phase can be well understood by a Heisenberg model with single-ion anisotropy term, the latter is mainly determined by the spin-orbit coupling of I atoms. Our results demonstrate a strain-controlled magnetic phases of 2D Janus magnetic semiconductors controlled by strain, and thus suggest a promising way to design functional materials.

II. COMPUTATIONAL METHODS

Our first-principles calculations were carried out with the Vienna ab initio simulation package (VASP) based on the density functional theory (DFT) [41, 42]. The interactions between nuclei and electrons were described by the projector augmented wave (PAW) method [43], and the generalized gradient approximation (GGA) in the form proposed by Perdew, Burke, and Ernzerhof (PBE) [44] was used to describe the electron exchange-correlation functional. In order to prevent the unphysical interlayer interactions, we build a 20 Å vacuum. The cutoff energy was set to be 520 eV, and the K-meshes for structure optimization and self-consistent calculations is $9 \times 9 \times 1$ and $15 \times 15 \times 1$ G-centered Monkhorst-Pack grid [45], respectively. The structure optimization of atomic positions and the lattice vectors were done until the maximum force on each atom was less than 0.0001 eV/Å, and the total energy was converged to $10^{-8}$ eV. During the optimization, the conjugate gradient (CG) scheme were employed. The phonon frequencies were obtained by the density functional perturbation theory (DFPT) as implemented in the PHONOPY code [46] using a $2 \times 2 \times 1$ supercell.

III. DFT RESULTS

The crystal structure of 2D Janus $\text{Fe}_2\text{Cl}_3\text{I}_3$ is shown in Fig.1(a), where the Fe atoms are sandwiched by two different halogen atomic layers Cl and I. 2D $\text{Fe}_2\text{Cl}_3\text{I}_3$ with the broken inversion symmetry belongs to the $P31m$ (No.157) space group. Each primitive cell contains one formula units, and the Fe atoms locate in the center of the distorted octahedron consisting of three Cl atoms and three I atoms, and form a honeycomb lattice. To examine the stability of 2D $\text{Fe}_2\text{Cl}_3\text{I}_3$, its formation energy was calculated. The formation energy is defined as $E_f = E_{\text{Fe}_2\text{Cl}_3\text{I}_3} - 2E_{\text{Fe}} - 3/2E_{\text{Cl}_2} - 3/2E_{\text{I}_2}$, where $E_{\text{Fe}_2\text{Cl}_3\text{I}_3}$, $E_{\text{Fe}}$ and $E_{\text{I}_2}$ are energies of corresponding crystals, and $E_{\text{Cl}_2}$ is the total energy of Cl$_2$ molecule.

The negative value $E_f$ of -6.31 eV per formula unit indicates an exothermic reaction. The phonon spectra of $\text{Fe}_2\text{Cl}_3\text{I}_3$ monolayer were calculated as shown in Fig.1(c), where no imaginary frequency mode in the whole Brillouin zone indicates that $\text{Fe}_2\text{Cl}_3\text{I}_3$ monolayer is dynamically stable. The calculated lattice constant is 6.717 Å. As shown in Fig.1(d), the relative energy of $\text{Fe}_2\text{Cl}_3\text{I}_3$ as a function of applied strain changes continuously from compressed strain (-5%) to tensile strain (15%), which indicates the stability of $\text{Fe}_2\text{Cl}_3\text{I}_3$ with the applied strain.

FIG. 1. (a) Top and side views of the 2D Janus material $\text{Fe}_2\text{Cl}_3\text{I}_3$ with two kinds of I atoms labeled. (b) The first Brillouin zone with high symmetry points labeled. (c) Calculated phonon spectra. (d) Strain energy as a function of applied strain.

FIG. 2. (a) Possible spin configurations for Fe atoms: FM, Néel AFM, Stripy AFM and Zigzag AFM. (b) Band structure of $\text{Fe}_2\text{Cl}_3\text{I}_3$ monolayer with SOC.

The magnetic ground state of $\text{Fe}_2\text{Cl}_3\text{I}_3$ was studied by comparing the total energy of different spin configurations: FM, Néel AFM, stripy AFM, zigzag AFM and paramagnetic (PM) configurations. Table I lists the to-
TABLE I. The total energy per unit cell for Fe₂Cl₃I₃ monolayer (in meV, relative to the total energy of zigzag AFM along z-axis magnetization) for several spin configurations of Fe atoms calculated by GGA+SOC+U method.

| Configuration       | MAE (meV) | ∆E (meV) |
|---------------------|-----------|----------|
| Zigzag AFM (m∥z)   | 0.0       | 3.7      |
| Zigzag AFM (m∥x)   | 7.5       |          |
| Zigzag AFM (m∥y)   | 3.7       |          |
| Néel AFM (m∥z)     | 20.1      |          |
| Stripy AFM (m∥z)   | 19.3      |          |
| FM (m∥z)           | 68.9      | 472.1    |
| PM                  |           |          |

The total energy per Fe₂Cl₃I₃ unit cell relative to the ground states. It is clear that the zigzag AFM with out-of-plane magnetization of sublattice possesses the lowest energy, and is the ground state. The energy difference between the ground state and the zigzag AFM with in-plane magnetization of sublattice is about 3.7 meV.

The band structure of Fe₂Cl₃I₃ with SOC is shown in Fig. 2(b). It shows that Fe₂Cl₃I₃ is a semiconductor with a small indirect band gap of about 0.58 eV. Due to the different electronegativity of Cl and I atoms, the charge redistributes. According to the Bader charge analysis, one I atom gains 0.29 e from the Fe atom, and one Cl atom gains 0.55 e from the Fe atom. So a spontaneous electric polarization along the direction perpendicular to the plane with magnitude of 0.18 eÅ was obtained. Thus, the 2D Janus material Fe₂Cl₃I₃ is a rare example of the 2D semiconductors with both spin and charge polarizations.

The effects of biaxial strain from compress 5% to tensile 15% on the properties of 2D Janus material Fe₂Cl₃I₃ are explored. The MAE defined as the energy difference between the states with in-plane and out-of-plane spin configurations, ∆E defined as energy difference between the FM and AFM, and the band gap and electric polarization as a function of the strain were plotted in Fig. 3. From Fig. 3(a), it is noted that, ∆E decreases as the increase of tensile strain, and a phase transition from zigzag AFM to FM occurs with the tensile strain of 7%. Meanwhile, one can observe that MAE changes from positive to negative, and then returns to the positive value, corresponding to the change of magnetization direction from out-of-plane to in-plane, and then back to out-of-plane. The magnetic ground state changed with the strain can be briefly summarized as four steps: zigzag AFM with out-of-plane magnetization of sublattice, zigzag AFM with in-plane magnetization of sublattice, FM with in-plane magnetization, and FM with out-of-plane magnetization. On the other hand, the band gap and the electric polarization preserve with applied strain as shown in Fig. 3(b). It is interesting to note that at tensile strain of 10%, the band gap in Fig. 3(b) shows a discontinuity as a function of strain, while a magnetic phase transition occurs between FM with in-plane magnetization and FM with out-of-plane magnetization.

IV. IV. MODEL ANALYSIS

To better understand the strain-controlled magnetic phase transition in Fe₂Cl₃I₃, the isotropic Heisenberg model including the third nearest-neighbor hoppings is employed. This model was used to explain the AFM to FM transition induced by strain in the 2D magnetic semiconductor CrSiTe₃ [47]. The model Hamiltonian can be written as

$$H_0 = \sum_{\langle i,j \rangle} J_1 \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{\langle\langle i,j \rangle\rangle} J_2 \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{\langle\langle\langle i,j \rangle \rangle\rangle} J_3 \mathbf{S}_i \cdot \mathbf{S}_j,$$

where $J_1$, $J_2$ and $J_3$ represents the nearest-neighbor, next nearest-neighbor and third nearest-neighbor exchange integrals, respectively. By comparing the energies of four different magnetic configurations: FM, Néel AFM, stripy AFM and zigzag AFM, which can be expressed as a function of $J_1$, $J_2$ and $J_3$ based on the classic approximation of spins in Eq. (1), the magnetic phase diagram with respect to $J_1/J_2$ and $J_3/J_2$ can be obtained as shown in Fig. 4.
On the other hand, from the DFT results of Fe$_2$Cl$_3$I$_3$ with different strains, the parameters $J_1$, $J_2$, and $J_3$ can be estimated [47, 48]. The obtained exchange parameters of Fe$_2$Cl$_3$I$_3$ are marked as stars in Fig. 4, where the digital numbers of each star denote the corresponding strain.

![FIG. 4. Magnetic phase diagram of Fe$_2$Cl$_3$I$_3$ as a function of exchange integrals $J_1/J_2$ and $J_3/J_2$. The DFT results with different strains are marked with stars along with the plots of the corresponding magnetic ground states.](image)

As shown in Fig. 4, Fe$_2$Cl$_3$I$_3$ locate at the zigzag AFM and FM phases. In the FM phase, the magnitude of exchange parameter $J_1$ is much larger than that of $J_2$ and $J_3$, and the model Hamiltonian in Eq.(1) is simplified to the Heisenberg model with nearest-neighbor coupling $J_1$. While in the zigzag AFM phase, both $J_1$, $J_2$ and $J_3$ are important. Although the jump of digital numbers in the phase diagram correspond to four different magnetic ground states, the phase diagram is obtained from the isotropic Heisenberg Hamiltonian, and can’t explain the change of the magnetization directions.

In order to understand the change of magnetization direction, i.e. the sign of MAE in Fig. 3(a), we consider the single-ion anisotropy (SIA), which can be written as

$$H_1 = \sum_i A_{xx} S_{i,x}^2 + A_{yy} S_{i,y}^2 + A_{zz} S_{i,z}^2,$$

where $A_{xx/yy/zz}$ represent the magnetic anisotropy strength of $x/y/z$ directions, $S_{i,x/y/z}$ are spin operators. According to the second-order perturbation theory, the MAE caused by SIA can be described by

$$E_{SIA} = \lambda^2 \sum_{o,u} \left| \langle \Psi_u | L_z | \Psi_o \rangle \right|^2 - \left| \langle \Psi_u | L_z | \Psi_u \rangle \right|^2, \varepsilon_o - \varepsilon_u,$$

where $\lambda$ is the SOC constant, $L_z$ represents the angular momentum operator, $\Psi_u$ and $\Psi_o$ are the wavefunctions of unoccupied and occupied states, respectively, and $\varepsilon_u$ and $\varepsilon_o$ are the corresponding energy levels. A positive value of $E_{SIA}$ indicates the out-of-plane magnetization, and negative value indicates the in-plane magnetization.

To unveil the mechanism that leads to the change of $E_{SIA}$ with different strains, we have calculated the orbital-resolved $E_{SIA}$. As an example, the orbital-resolved $E_{SIA}$ of Fe$_2$Cl$_3$I$_3$ with 8% and 15% tensile strains, which correspond to FM with magnetization along the x-axis and FM with magnetization along the z-axis ground states, respectively, was calculated as shown in Fig. 5. By DFT calculations, the main contribution to $E_{SIA}$ is from Fe and I atoms, and I atoms can be classified to two kinds as labeled in Fig. 1(a) according to their different surroundings when the magnetization is along the x-axis. In the case of 8% tensile strain, as shown in Table II, the contributions to $E_{SIA}$ from Fe and two kinds of I atoms are -1.2, 2.5 and -4.1 meV, respectively, giving rise to a total negative $E_{SIA}$ of -2.8 meV. It is consistent with the in-plane magnetization. For the case of 15% tensile strain, a positive value $E_{SIA}$ of 5.3 meV is obtained, which indicates the out-of-plane magnetization. From Fig. 5, one may observe that $E_{SIA}$ mainly originates from $(p_y,p_z)$ and $(p_x,p_z)$ matrix element of I atoms. As the tensile strain increases from 8%
TABLE II. The value of orbital-resolved single-ion anisotropy energy of Fe and two kinds of I atoms (in meV) in 2D Janus Fe$_2$Cl$_3$I$_3$ monolayer with 8% and 15% strain, respectively.

|        | Fe-d | I$_1$-p | I$_2$-p | Total |
|--------|------|---------|---------|-------|
| 8%     | -1.2 | 2.5     | -4.1    | -2.8  |
| 15%    | -2.1 | 6.8     | 0.6     | 4.1   |

To 15%, the sign change of $E_{SIA}$ from I atoms gives rise to the sign change of the total $E_{SIA}$, and thus induces the changes of the magnetization direction.

V. V. EFFECT OF ELECTRONIC CORRELATION

The electronic correlation effect is important for 3$d$ orbitals in transition-metal compounds, so our above DFT calculations are carried out with $U = 4$ eV. In order to examine the influence of different $U$ values on the magnetic ground states, we have studied the magnetic state of Fe$_2$Cl$_3$I$_3$ for the cases without strain and with 15% tensile strain with parameter $U$ from 2 to 5 eV. As shown in Fig. 6, when there is no strain applied, with the increase of $U$ value, although MAE increases and $\Delta E$ decreases, the signs of both preserve, indicating the unchanged AFM ground state with out-of-plane magnetization of sublattice. Similarly, the FM ground state along z-axis magnetization with 15% strain is not changed when different $U$ values are employed. Thus, our results about the magnetic phase transition with respect to the strain are robust against $U$ values.

VI. VI. CONCLUSION

By first-principles calculations, we have proposed a new 2D magnetic Janus semiconductor–Fe$_2$Cl$_3$I$_3$, which was revealed to exhibit the zigzag AFM ground state with out-of-plane magnetic direction. Due to the inversion symmetry breaking and the charge redistribution, Fe$_2$Cl$_3$I$_3$ was found to possess a spontaneous polarization along the z-axis. Furthermore, we have also investigated the effect of biaxial strain on the ground state properties of Fe$_2$Cl$_3$I$_3$, and a magnetic phase transition including the antiferromagnetic-ferromagnetic transition and the change of magnetization direction was obtained. Both magnetic and electric polarization were observed in Fe$_2$Cl$_3$I$_3$ under the biaxial strain. To well understand the magnetic phase transition, a phase diagram based on the Heisenberg model with the single-ion anisotropy term can well interpret the DFT results. Our findings not only expose a new stable 2D magnetic Janus semiconductor, but also reveal the highly sensitive strain-controlled magnetic states, and thus highlight the 2D magnetic Janus semiconductor as a new platform to design spintronic materials.

FIG. 6. Electronic correlation $U$-dependent magnetic anisotropic energy (MAE) and energy difference ($\Delta E$) between antiferromagnetic and ferromagnetic configurations (a) without strain and (b) with 15% tensile strain.

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