I. INTRODUCTION

The piezoelectric effect is an intrinsic electromechanical coupling in semiconductors with crystal structures lacking inversion symmetry. The reduction in dimensionality of 2D materials often can eliminate inversion symmetry, which allows them to be piezoelectric. Experimentally, the piezoelectricity of MoS$_2$\(^{1,2}\), MoSSe\(^3\) and In$_2$Se$_3$\(^4\) monolayers have been discovered, which pushes the development of piezoelectric properties of 2D materials. In theory, many kinds of 2D materials have been predicted to be piezoelectric by density functional theory (DFT) calculations\(^5\)\textendash\(^12\). The strain-tuned piezoelectric response has also been investigated by DFT calculations, and it is proved that strain can improve the piezoelectric strain coefficients\(^13\)\textendash\(^15\).

Great advances have been made on 2D piezoelectric materials. However, there are two main issues of 2D piezoelectric materials. One is that most 2D materials possess solely piezoelectricity. The multifunctional 2D materials, such as combination of piezoelectricity with topological insulating phase or ferromagnetism, are of particular interest, whose exploitation may promise novel device applications. The coexistence of intrinsic piezoelectricity and ferromagnetism has been predicted in 2D vanadium dichalcogenides and VSi$_2$P$_4$\(^{16,17}\). The piezoelectric quantum spin Hall insulators (PQSHI) have also been achieved in monolayer InXO (X=Se and Te)\(^18\) and Janus monolayer SrAlGaSe$_6$\(^19\). Another is that the out-of-plane piezoelectricity in known 2D materials is absent or weak. The strong out-of-plane piezoelectric effect and its inverse effect are highly desirable for piezoelectric devices, which is compatible with the bottom/top gate technologies. Many strategies have been made for searching 2D piezoelectric materials with large $d_{31}$ or $d_{32}$\(^5\)\textendash\(^11,20\)\textendash\(^22\). A significant improvement is that the piezoelectric strain coefficient $d_{31}$ of Sc$_2$CO$_2$ MXene is up to 0.78 pm/V\(^2\)\textendash\(^22\).

A natural idea is to search for multifunctional 2D piezoelectric materials with large out-of-plane piezoelectricity. A few types of 2D magnetic materials have been studied\(^23\)\textendash\(^31\). For example, the monolayer Cr$_3$Ge$_2$Te$_6$, VS$_2$ and VSe$_2$ have been experimentally proved to magnetic materials\(^25,28\). The Dirac spin-gapless semiconductor (SGS) with 100% spin polarization, high Fermi velocities and high Curie temperatures has been predicted in Mn$_2$C$_6$Se$_{12}$ and Mn$_2$C$_6$S$_6$Se$_6$ monolayers\(^29\). The 2D high-temperature ferromagnetic half-metal (FMHM) can be realized in transition-metal embedded carbon nitride...
FIG. 1. (Color online) The (a) top view and (b) side view of crystal structure of Janus monolayer CrBr$_{1.5}$I$_{1.5}$. The rectangle supercell is marked by black frame, which is used to calculate the piezoelectric stress coefficients. The rectangle’s width and height are defined as x and y directions, respectively.

FIG. 2. (Color online) The four considered magnetic configuration of Janus monolayer CrBr$_{1.5}$I$_{1.5}$: FM (a), AF-Néel (b), AF-zigzag (c), and AF-stripy ordered (d). The crystal cells used in the calculations are marked with red arrows as the spin direction of Cr atoms.

monolayers$^{27}$. The CrI$_3$ monolayer is firstly predicted to be FM order by the first-principle calculations$^{32}$, and then is confirmed experimentally$^{30}$. A series of studies have been carried out to explore the magnetic related properties in CrI$_3$ monolayer$^{33-38}$. These provide many new possibilities to combine the piezoelectricity and magnetism into the same kind of 2D material.

It is noted that the CrI$_3$ monolayer has sandwiched I-Cr-I structure with inversion symmetry, and then possesses no piezoelectricity. However, it is possible to construct Janus structure based on CrI$_3$ monolayer, and then produce piezoelectric effect. Janus monolayer MoSSe has been synthesized experimentally from MoS$_2$ monolayer by breaking the out-of-plane structural symmetry$^{39}$. That is, the Janus monolayer MoSSe can be constructed by replacing one of two S layers with Se atoms in MoS$_2$ monolayer. In this work, Janus monolayer CrBr$_{1.5}$I$_{1.5}$ is constructed from synthesized ferromagnetic CrI$_3$ monolayer by replacing the top I atomic layer with Br atoms, which is dynamically, mechanically and thermally stable. It is found that monolayer CrBr$_{1.5}$I$_{1.5}$ is an intrinsic FM half semiconductor with a sizable MAE. Although the calculated in-plane $d_{22}$ (0.557 pm/V) is small, the out-of-plane $d_{31}$ (1.138 pm/V) is very large, which is obviously higher than ones of other 2D known materials. It is proved that strain engineering can effectively tune piezoelectricity of monolayer CrBr$_{1.5}$I$_{1.5}$. A 2D PAFM can also be achieved in monolayer CrBr$_{1.5}$I$_{1.5}$ by compressive strain, and the calculated results show that magnetic order has important influences on piezoelectricity of monolayer CrBr$_{1.5}$I$_{1.5}$. It is also proved that the PFM can also be achieved in monolayer CrF$_{1.5}$I$_{1.5}$ and monolayer CrCl$_{1.5}$I$_{1.5}$, which show very large $d_{31}$ of 2.578 pm/V and

FIG. 3. (Color online) The temperature and total energy fluctuations of Janus monolayer CrBr$_{1.5}$I$_{1.5}$ with FM magnetic configuration at 300 K.
Within the projector augmented-wave (PAW) method, the energy band structures of CrBr$_{1.5}$I$_{1.5}$ with FM magnetic configuration are obtained. The blue (red) lines represent the band structure in the spin-up (spin-down) direction. The rest of the paper is organized as follows. In the next section, we shall give our computational details and methods. In the next few sections, we shall present structural stabilities, electronic structures, and piezoelectric properties of monolayer CrBr$_{1.5}$I$_{1.5}$, along with strain effects on its piezoelectric properties. Finally, we shall give our discussion and conclusions.

II. COMPUTATIONAL DETAIL

Within DFT$^{40}$, we perform the main calculations with spin-polarization using the the plane-wave code VASP$^{41-43}$ within the projector augmented-wave (PAW) method. The popular generalized gradient approximation of Perdew, Burke and Ernzerhof (GGA-PBE)$^{44}$ is used as the exchange-correlation functional. The kinetic energy cutoff is set to 500 eV with the total energy convergence criterion for $10^{-8}$ eV. All the lattice constants and atomic coordinates are optimized until the force on each atom is less than 0.0001 eV Å$^{-1}$. A vacuum spacing of more than 18 Å is used to avoid interactions between two neighboring images. The elastic stiffness tensor $C_{ij}$ and piezoelectric stress tensor $e_{ij}$ are carried out by using strain-stress relationship (SSR) and density functional perturbation theory (DFPT) method$^{45}$, respectively. A Monkhorst-Pack k-mesh of 8×8×1 is used to sample the Brillouin Zone (BZ) for the calculations of electronic structure and elastic coefficients $C_{ij}$ and density functional perturbation theory (DFPT) method$^{45}$, respectively. A Monkhorst-Pack k-mesh of 8×8×1 is used to sample the Brillouin Zone (BZ) for the calculations of electronic structure and elastic coefficients $C_{ij}$, and a mesh of 4×8×1 k-points for the energy of different magnetic configurations and piezoelectric stress coefficients $e_{ij}$. The 2D elastic coefficients $C_{ij}^{2D}$ and piezoelectric stress coefficients $e_{ij}^{2D}$ have been renormalized by $C_{ij}^{2D} = L_z C_{ij}^{3D}$ and $e_{ij}^{2D} = L_z e_{ij}^{3D}$, where the $L_z$ is the length of unit cell along $z$ direction. By finite displacement method, the interatomic force constants (IFCs) are obtained based on the 4×4×1 supercell with FM ground state. Based on the harmonic IFCs, the phonon dispersions are evaluated using Phonopy code$^{46}$.

III. STRUCTURE AND STABILITY

The structure of Janus monolayer CrBr$_{1.5}$I$_{1.5}$ is similar to monolayer CrI$_3$ monolayer, which contains three atomic sublayers with Cr layer sandwiched between Br and I layers. It is well known that Janus transition metal dichalcogenides (TMD) Monolayer MoSSe has been synthesized by replacing the top S atomic layer in MoS$_2$ with Se atoms$^{39}$. Using the same idea, the Janus monolayer CrBr$_{1.5}$I$_{1.5}$ can be constructed by replacing one of two I layers with Br atoms in monolayer CrI$_3$. The schematic crystal structures of Janus monolayer CrBr$_{1.5}$I$_{1.5}$ are shown in Figure 1. The monolayer CrI$_3$ has centrosymmetry with 3m point-group symmetry (No.162), but monolayer CrBr$_{1.5}$I$_{1.5}$ loses centrosymmetry and horizontal mirror symmetry with 3m point-group symmetry (No.157), which will induce both in-plane and out-of-plane piezoelectricity.
There are twenty-one optical and three acoustical shows the temperature and total energy fluctuations of CrBr$_{1.5}$I$_{1.5}$ monolayer at room temperature. Calculated results show no obvious structural disruption with the temperature and total energy fluctuates being small at the end of the MD simulation at 300 K, which confirms the thermodynamical stability of the CrBr$_{1.5}$I$_{1.5}$ monolayer at room temperature.

The dynamical stability of the CrBr$_{1.5}$I$_{1.5}$ monolayer is analyzed by the phonon spectra, which is plotted in Figure 4. There are twenty-one optical and three acoustical phonon branches with a total of twenty-four branches due to eight atoms per cell. The longitudinal acoustic (LA) and transverse acoustic (TA) modes mean in-plane vibrations, while the ZA branch represents the out-of-plane vibrations. It is clearly seen that the ZA branch is quadratic near the zone center, as typical characteristics of 2D materials. All phonon frequencies are positive, confirming the dynamical stability of CrBr$_{1.5}$I$_{1.5}$ monolayer, which means that it can exist as a free-standing lattice constants with FM state is 6.744 Å, which falls between those of the CrBr$_3$ (6.433 Å) and CrI$_3$ (7.008 Å) monolayers. For monolayer CrBr$_{1.5}$I$_{1.5}$, the difference in atomic sizes and electronegativities of Br and I atoms leads to inequivalent Cr-Br and Cr-I bond lengths (Br-Cr-Br and I-Cr-I bond angles), and they are 2.542 Å and 2.719 Å (92.036 and 89.432), which can induce a built-in electric field.

The ab initio molecular dynamics (AIMD) simulations using NVT ensemble are performed to assess the thermal stability of the monolayer CrBr$_{1.5}$I$_{1.5}$ at room temperature. Figure 3 shows the temperature and total energy fluctuations of CrBr$_{1.5}$I$_{1.5}$ monolayer as a function of the simulation time. Calculated results show no obvious structural disruption with the temperature and total energy fluctuates being small at the end of the MD simulation at 300 K, which confirms the thermodynamical stability of the CrBr$_{1.5}$I$_{1.5}$ monolayer at room temperature.

Four different magnetic configurations (Figure 2) are considered to evaluate the magnetic ground state of monolayer CrBr$_{1.5}$I$_{1.5}$, which are used to investigate the ground state of monolayer CrX$_3$ (X= F, Cl, Br and I). The energy of AF-Néel, AF-zigzag, AF-stripy state and non-magnetic (NM) state with respect to FM state are 56.1 meV, 56.4 meV, 35.2 meV and 6.802 eV with rectangle supercell. Our calculated results show that the FM order is the most stable magnetic state. This means that ferromagnetism in monolayer CrI$_3$ is retained by elements substitution to construct Janus structure. The optimized

![Figure 7](image1)

**FIG. 7.** (Color online) The energy band structures of Janus monolayer CrBr$_{1.5}$I$_{1.5}$ with FM magnetic configuration with a/a$_0$ changing from 0.94 to 1.06.

![Figure 8](image2)

**FIG. 8.** (Color online) The majority-spin gap ($U_p$), the minority-spin gap ($D_n$) and the total gap ($Tot$) of FM CrBr$_{1.5}$I$_{1.5}$ as a function of strain a/a$_0$. 

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...
2D crystal.
It is important to check the mechanical stability of CrBr$_{1.5}$I$_{1.5}$ monolayer for practical application. Therefore, we calculate elastic constants using the SSR method. Using Voigt notation, the elastic tensor with $3n$ point-group symmetry for 2D materials can be expressed as:

$$\mathbf{C} = \begin{pmatrix}
C_{11} & C_{12} & 0 \\
C_{12} & C_{11} & 0 \\
0 & 0 & (C_{11} - C_{12})/2
\end{pmatrix}$$

(1)

The calculated $C_{11}$ and $C_{12}$ are 29.75 Nm$^{-1}$ and 8.26 Nm$^{-1}$, which are between ones of CrBr$_3$ and CrI$_3$ monolayers$^{32}$. The calculated $C_{11}>0$ and $C_{11} - C_{12}>0$ satisfy the Born criteria of mechanical stability$^{49}$, confirming the mechanical stability of CrBr$_{1.5}$I$_{1.5}$ monolayer. We also calculate the Young's moduli $C_{2D}$, shear modulus $G_{2D}$ and Poisson’s ratio $\nu$ using the method suggested by Andrew et al, and they are 27.46 Nm$^{-1}$, 10.75 Nm$^{-1}$ and 0.278, respectively. These indicate that monolayer CrBr$_{1.5}$I$_{1.5}$ can be easily tuned by strain, which is favorable for novel flexible piezotronics and nanoelectronics.

## IV. ELECTRONIC STRUCTURE

To exhibit piezoelectricity, the monolayer CrBr$_{1.5}$I$_{1.5}$ not only should lack inversion symmetry, but also should be a semiconductor. So, we investigate the electronic structures of CrBr$_{1.5}$I$_{1.5}$ monolayer with FM ground state, and the energy bands and atomic partial density of states (DOS) are plotted in Figure 4 and Figure 5, respectively. It is found that CrBr$_{1.5}$I$_{1.5}$ monolayer is an indirect gap semiconductor with gap value of 1.335 eV. Moreover, the valence and conduction bands near the Fermi level are exclusively contributed by the same spin-up component, showing a typical half-semiconductor character. The difference of the band edge energy between the two spin components for the conduction band minimum (CBM) and the valance band maximum (VBM) are 0.884 eV and 0.233 eV, respectively. According to DOS, the occupied Cr-3d orbitals are mainly found in the spin-up direction, and the spin-down Cr-3d states are almost unoccupied. For both spin directions, the conduction band is dominated by Cr-3d states, which are weakly hybridized with the Br-3p and I-3p states. In the spin-down direction, the valence band are almost pure Br-3p and I-3p character. For the spin-up direction of the valence band, the states are contributed by the Br-3p and I-3p states with a mixture of Cr-3d states. In fact, many electronic properties of monolayer CrBr$_{1.5}$I$_{1.5}$ are similar to ones of CrI$_3$ monolayer$^{32}$.

The magnetic moment of primitive cell is equal to 6 $\mu_B$ accurately, which is consistent with its semiconducting property. The local magnetic moments of Cr is 2.985 $\mu_B$, which suggests that monolayer CrBr$_{1.5}$I$_{1.5}$ is robust intrinsic ferromagnetic 2D semiconductor with large magnetic moments. MAE is an important parameter to confirm ferromagnetic behavior of monolayer CrBr$_{1.5}$I$_{1.5}$. The small MAE will result in superparamagnetic rather than ferromagnetic behavior. By using GGA+spin orbital coupling (SOC), it is found that an easy axis is...
FIG. 11. (Color online) For Janus monolayer CrBr$_{1.5}$I$_{1.5}$ with FM state, the piezoelectric stress coefficients ($e_{22}$ and $e_{31}$) and the piezoelectric strain coefficients ($d_{22}$ and $d_{31}$) with the application of biaxial strain (0.94 to 1.06). The black marks mean AF-Néel results at 0.94 strain.

along the c-direction for monolayer CrBr$_{1.5}$I$_{1.5}$, and the corresponding MAE is 356 µeV per Cr atom. For CrBr$_3$ and CrI$_3$ monolayers, the easy axis is also along the c-direction, and the MAE of monolayer CrBr$_{1.5}$I$_{1.5}$ is between ones of them (185.5 µeV per Cr atom for CrBr$_3$ and 685.5 µeV per Cr atom for CrI$_3$)$^{32}$.

V. PIEZOELECTRIC PROPERTIES

The CrI$_3$ monolayer with $3m$ point-group symmetry are centrosymmetric, showing no piezoelectricity. The CrBr$_{1.5}$I$_{1.5}$ monolayer with $3m$ point-group symmetry lacks both inversion symmetry and reflectional symmetry across the xy plane, which means that both $e_{22}/d_{22}$ and $e_{31}/d_{31}$ with defined x and y direction in Figure 1 are nonzero. For 2D materials, only the in-plane strain and stress are taken into account$^{5-12}$, and the piezoelectric stress and strain tensors by using Voigt notation can become:

$$
e = \begin{pmatrix}
0 & 0 & -e_{22} \\
-e_{22} & e_{22} & 0 \\
e_{31} & e_{31} & 0
\end{pmatrix}
$$

(2)

$$
d = \begin{pmatrix}
0 & 0 & -2d_{22} \\
d_{22} & d_{22} & 0 \\
d_{31} & d_{31} & 0
\end{pmatrix}
$$

(3)

With a applied uniaxial in-plane strain, both in-plane and vertical piezoelectric polarization ($e_{22}/d_{22} \neq 0$ and $e_{31}/d_{31} \neq 0$) can be produced. However, by imposing bi-axial in-plane strain, the out-of-plane one still will remain, while the in-plane piezoelectric response will be suppressed($e_{11}/d_{11}=0$ and $e_{31}/d_{31} \neq 0$). The independent $d_{22}$ and $d_{31}$ are can be attained by $e_{ik} = d_{ij}C_{jk}$:

$$
d_{22} = \frac{e_{22}}{C_{11} - C_{12}} \quad \text{and} \quad d_{31} = \frac{e_{31}}{C_{11} + C_{12}}
$$

(4)

The orthorhombic supercell is used as the computational unit cell (in Figure 1) to calculate the
TABLE I. For CrX_{1.5}I_{1.5} (X=F, Cl and Br) monolayers, the lattice constants a_{0} (Å), the elastic constants C_{ij} in N m^{-1}, the piezoelectric stress coefficients e_{ij} in 10^{-10} C m^{-1}, the piezoelectric strain coefficients d_{ij} in pm/V, MAE in µeV/Cr and easy axis (EA).

| Name            | a_{0}  | C_{11} | C_{12} | e_{22} | e_{31} | d_{22} | d_{31} | MAE   | EA   |
|-----------------|--------|--------|--------|--------|--------|--------|--------|-------|------|
| CrF_{1.5}I_{1.5}| 6.250  | 49.97  | 16.34  | 1.339  | 1.710  | 3.983  | 2.578  | 2151  | ab   |
| CrCl_{1.5}I_{1.5}| 6.590  | 34.88  | 9.94   | 0.238  | 0.809  | 0.956  | 1.804  | 110   | c    |
| CrBr_{1.5}I_{1.5}| 6.744  | 29.75  | 8.26   | 0.119  | 0.432  | 0.557  | 1.138  | 356   | c    |

VI. STRAIN EFFECTS

The strain can effectively tune the electronic structures and piezoelectric properties of 2D materials. Here, we use a/a_{0} to simulate the biaxial strain, where a and a_{0} are the strained and unstrained lattice constants, respectively. To determine the ground state of strained CrF_{1.5}I_{1.5} monolayer, four different initial magnetic configurations (Figure 2) are considered. The energy differences of AF-Néel, AF-zigzag and AF-stripy with respect to FM state as a function of strain with rectangle supercell shown in Figure 6. It is found that a magnetic phase transition can be induced by compressive strain with the critical point being about 0.95, which implies the robustness of the intrinsic ferromagnetism in CrF_{1.5}I_{1.5} monolayer. Calculated results show that CrF_{1.5}I_{1.5} monolayer prefers FM ground state with a/a_{0} being greater than about 0.95 in considered strain range, and the AF-Néel become ground state with a/a_{0} being less than about 0.95. Similar phenomenon can also be found in CrI_{3} monolayer, and the AF-Néel phase becomes the most stable phase at 0.92 strain.

The energy band structures of FM CrF_{1.5}I_{1.5} monolayer with strain from 0.94 to 1.06 except 1.00 are plotted in Figure 7, and the minority-spin, minority-spin and total gaps are shown in Figure 8. It is clearly seen that strained CrF_{1.5}I_{1.5} monolayer are all indirect gap semiconductors in considered strain range. It is found that the majority-spin and total gaps coincide except 0.94 strain, which means that CrF_{1.5}I_{1.5} monolayer holds half- semiconductor character. At 0.94 strain, the VBM is at minority-spin channel from previous majority-spin one. From 1.06 to 0.94 strain, strain makes both conduction and valence bands of minority-spin channel move toward Fermi level, which leads to the reduced minority-spin gap. The majority-spin gap shows a nonmonotonic behavior, which is mainly due to change of CBM. In fact, at 0.94 strain, the AF-Néel becomes ground state, and we plot the energy bands along with FM states in Figure 9 using orthorhombic supercell. The AF-Néel state still is an indirect gap semiconductor with the gap value of 1.282 eV, and the local magnetic moments of Cr is 2.893 µB.

The strain engineering has been proved to be an very effective way to enhance piezoelectric properties of 2D materials, and then the strain effects on piezoelectric properties of CrBr_{1.5}I_{1.5} monolayer are performed. The elastic constants including C_{11}, C_{12}, C_{11}-C_{12} and
C$_{11}$+C$_{12}$ of CrBr$_{1.5}$I$_{1.5}$ monolayer with FM state as a function of biaxial strain are plotted in Figure 10, along with ones of AF-Néel state at 0.94 strain. It is clearly seen that C$_{11}$, C$_{12}$, C$_{11}$-C$_{12}$ and C$_{11}$+C$_{12}$ are all decreases with increasing strain from 0.94 to 1.06 strain. It is found that C$_{11}$, C$_{12}$, C$_{11}$-C$_{12}$ and C$_{11}$+C$_{12}$ with AF-Néel state are lower than ones with FM state at 0.94 strain. So, it is important to consider the magnetic configurations for calculating elastic constants. Calculated results show that the CrBr$_{1.5}$I$_{1.5}$ monolayer is mechanically stable in the considered strain range, since the calculated elastic constants satisfy the mechanical stability criteria. The piezoelectric stress coefficients (e$_{22}$ and e$_{31}$) along the ionic and electronic contributions and piezoelectric strain coefficients (d$_{22}$ and d$_{31}$) of CrBr$_{1.5}$I$_{1.5}$ monolayer with FM state as a function of biaxial strain are plotted in Figure 11 and Figure 12, along with ones of AF-Néel state at 0.94 strain. It is found that the compressive strain can enhance the d$_{22}$ due to improved e$_{22}$ based on Equation 4, and the d$_{22}$ improves to 0.993 pm/V at 0.94 strain from unstrained 0.557 pm/V. The tensile strain can decrease the d$_{22}$, and the d$_{22}$ at 1.06 strain reduces to 0.039 pm/V due to very small e$_{22}$ (0.0066×10$^{-10}$ C/m). For d$_{31}$, the opposite strain dependence is observed, and the tensile strain can improve d$_{31}$ due to reduced C$_{11}$+C$_{12}$. At 1.06 strain, the d$_{31}$ of CrBr$_{1.5}$I$_{1.5}$ monolayer is 1.545 pm/V, increased by 36% with respect to unstrained one. In considered strain range, the electronic and ionic parts have opposite signs for both e$_{22}$ and e$_{31}$, and they (absolute value) all decreases with strain from 0.94 to 1.06.

The magnetic configuration may have important effects on piezoelectric coefficients, and a magnetic phase transition may induce the jump of piezoelectric coefficients. We recalculate the e$_{22}$ and e$_{31}$ along the ionic and electronic contributions and d$_{22}$ and d$_{31}$ of CrBr$_{1.5}$I$_{1.5}$ monolayer with AF-Néel state at 0.94 strain. It is found that magnetic configuration has important effect on e$_{22}$ from 0.285×10$^{-10}$ C/m of FM state to 0.181×10$^{-10}$ C/m of AF-Néel state, and has little influence on e$_{31}$ (0.502×10$^{-10}$ C/m for FM and 0.495×10$^{-10}$ C/m for AF-Néel). The similar effects on d$_{22}$ and d$_{31}$ also can be found, and the d$_{22}$ (d$_{31}$) changes from 0.993 pm/V (0.933 pm/V) of FM state to 0.677 pm/V (0.999 pm/V) of AF-Néel state. It is also found that magnetic configuration has important effects on both the electronic and ionic parts of e$_{22}$, and has neglectful influences on ones of e$_{31}$. So, it is very important to consider magnetic order for piezoelectric coefficients. It is interesting that the PAFM can be induced by compressive strain, which may open up potential opportunities for intriguing physics and novel devices.

VII. DISCUSSION AND CONCLUSION

In fact, one of two I layers of monolayer CrI$_3$ can also be replaced by F or Cl atoms, namely monolayer CrF$_{1.5}$I$_{1.5}$ and CrCl$_{1.5}$I$_{1.5}$. For CrCl$_{1.5}$I$_{1.5}$ monolayer, the FM order still is the ground state by comparing energy difference of four different initial magnetic configurations (Figure 2). However, for monolayer CrF$_{1.5}$I$_{1.5}$, the FM and AF-Néel orders have almost the same energy, and the difference is only 0.48 meV/CrF$_{1.5}$I$_{1.5}$ formula. So, we focus on the FM state of both monolayer CrF$_{1.5}$I$_{1.5}$ and CrCl$_{1.5}$I$_{1.5}$ for a better comparison. For monolayer CrF$_{1.5}$I$_{1.5}$ (CrCl$_{1.5}$I$_{1.5}$), the optimized lattice constants is 6.250 (6.590) Å, and the calculated C$_{11}$ and C$_{12}$ are 49.97 (34.88) Nm$^{-1}$ and 16.34 (9.94) Nm$^{-1}$, which satisfy the Born criteria of mechanical stability. From Figure 13, it is proved that monolayer CrF$_{1.5}$I$_{1.5}$ (CrCl$_{1.5}$I$_{1.5}$) is dynamically stable. The side views of crystal structures of CrX$_{1.5}$I$_{1.5}$ (X=F, Cl and Br) are plotted in Figure 14, and it is clearly seen that the distortions of octahedral environment located by Cr atoms become more and more severe with X from Br to Cl to F due to the more difference in atomic sizes and electronegativities of X and I atoms. It is found that an easy axis of monolayer CrCl$_{1.5}$I$_{1.5}$ is along the c-direction, and the corresponding MAE is 110 µeV per Cr atom. However, for monolayer CrF$_{1.5}$I$_{1.5}$, an easy axis is along the in-plane direction, and the MAE is up to 2151 µeV per Cr atom. Finally, the piezoelectric properties of monolayer CrF$_{1.5}$I$_{1.5}$ and CrCl$_{1.5}$I$_{1.5}$ are investigated, and their d$_{31}$ is up to 2.578 pm/V and 1.804 pm/V, respectively. The related data are summarized in Table I. In fact, many PFMs can be achieved in 2D CrX$_3$ (X=F, Cl, Br and I) family by using the same design principle of mono-
layer CrBr$_{1.5}$I$_{1.5}$, for example Janus monolayer monolayer CrCl$_{1.5}$F$_{1.5}$, CrCl$_{1.5}$Br$_{1.5}$, CrBr$_{1.5}$F$_{1.5}$ and so on.

In summary, our theoretical calculations demonstrate that the PFM can occur in Janus CrBr$_{1.5}$I$_{1.5}$ monolayer with dynamic, mechanical and thermal stabilities, which possesses a sizable MAE. By breaking the inversion and mirror symmetry, both in-plane and out-of-plane piezoelectric polarizations can be induced by a uniaxial in-plane strain. Amazingly, the out-of-plane $d_{31}$ (1.138 pm/V) is obviously higher compared with ones of many familiar 2D materials. It is proved that strain engineering can effectively tune piezoelectricity of monolayer CrBr$_{1.5}$I$_{1.5}$. The PAFM can also be realized by compressive strain, and $d_{32}$ ($d_{31}$) is 0.677 pm/V (0.999 pm/V) at 0.94 strain. Finally, similar to CrBr$_{1.5}$I$_{1.5}$, the PFM can also be achieved in the monolayer CrF$_{1.5}$I$_{1.5}$ and CrCl$_{1.5}$I$_{1.5}$ with very large $d_{31}$ being 2.578 pm/V and 1.804 pm/V. Our works supply an experimental proposal to achieve large out-of-plane piezoelectric response in PFM, and hope that the work can stimulate further experimental effort on 2D PFM.

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