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Strain-tunable magnetic anisotropy in monolayer CrCl_3 , CrBr_3 , and CrI_3

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Recent observation of intrinsic ferromagnetism in two-dimensional (2D) CrI_3 is associated with the large magnetic anisotropy due to strong spin-orbit coupling (SOC) of I. Magnetic anisotropy energy (MAE) defines the stability of magnetization in a specific direction with respect to the crystal lattice and is an important parameter for nanoscale applications. In this work we apply the density functional theory to study the strain dependence of MAE in 2D monolayer chromium trihalides CrX_3 (with $X = \text{Cl}, \text{Br}, \text{and I}$). Detailed calculations of their energetics, atomic structures and electronic structures under the influence of a biaxial strain ε have been carried out. It is found that all three compounds exhibit ferromagnetic ordering at the ground state (with $\varepsilon=0$) and upon applying a compressive strain, phase transition to antiferromagnetic state occurs. Unlike in CrCl_3 and CrBr_3 , the electronic band gap in CrI_3 increases when a tensile strain is applied. The MAE also exhibits a strain dependence in the chromium trihalides: it increases when a compressive strain is applied in CrI_3 , while an opposite trend is observed in the other two compounds. In particular, the MAE of CrI_3 can be increased by 47% with a compressive strain of $\varepsilon = 5\%$.

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I. INTRODUCTION

One of the latest advances in the field of two-dimensional (2D) materials is the observation of intrinsic ferromagnetism in monolayers of CrGeTe_3 ¹ and CrI_3 ². These systems provide an exciting platform for studying the interplay between various competing electronic and magnetic phenomena at nanoscale, when quantum confinement condition is included. These include, for example, magnetoelectric effect³⁻⁶, spin/valley physics⁷ and light-matter interactions under the influence of magnetic ordering⁸.

Unlike in bulk magnetic materials, the long-range magnetic ordering in 2D structures is impossible without magnetic anisotropy, which is required for counteracting thermal fluctuations⁹. Therefore, magnetic anisotropy, which originates mainly from spin-orbit coupling (SOC) effects¹⁰, becomes an important parameter when it comes to 2D magnets as it is qualitatively related to their magnetic stability. Moreover, ferromagnetic 2D materials with large magnetic anisotropy are of great interest for high density magnetic memories and spintronic applications at nanoscale, as in spin valves and magnetic tunnel junctions¹¹⁻¹³.

Strain engineering has been shown to be an effective approach to tune properties of nanomaterials by using the substrate lattice mismatching¹⁴⁻¹⁶. It has been demonstrated that external strain can tune the electronic energy band gap in single layer MoS_2 ¹⁴. In particular, the way in which strain can affect magnetic anisotropy has been subject of several studies involving thin films^{17,18} and 2D materials from density-functional theory (DFT) calculations¹⁹⁻²¹.

Typically 2D crystals can sustain larger strains than their bulk counterpart^{22,23}. Single layer MoS_2 can sustain strains as large as 11%²², and 6% in single layer FeSe ^{15,16}. DFT calculations show that mono-

layer chromium trihalides are soft when compared with other 2D materials. A 2D Young's modulus of 24, 29 and 34 N/m has been reported for CrI_3 , CrBr_3 and CrCl_3 respectively²⁴. This is much smaller than that of graphene (340 N/m)²⁵, monolayer MoS_2 (180 N/m)²² and monolayer FeSe (80 N/m)²⁶.

The softness of single layer chromium trihalides implies that strain modulation of their electronic and magnetic properties can be realized in these systems. Here we provide an extensive study on structural modification at nanoscale resulting from biaxial strain, by means of first-principle calculations. We also investigate the electronic structures, magnetism and magnetic anisotropy of the single layer chromium trihalides under different strains.

The paper is organized as follows. In Section II, the calculation details are given. In Section III, we discuss our results. A brief conclusion will be drawn in Section IV.

II. CALCULATIONAL METHODS

Our DFT calculations were performed using projected augmented wave (PAW) method as implemented in the Vienna ab initio Simulation Package (VASP)^{27,28}. In all of these calculations we adopted the Perdew-Burke-Ernzerhof (PBE)²⁹ flavor for the generalized-gradient exchange-correlation functional. The Brillouin zone was sampled by $8 \times 8 \times 1$ k -point grid mesh³⁰, and a 500 eV plane wave cutoff energy was used. Moreover, a 15 Å vacuum was applied along the z axis to avoid any artificial interactions between images. Relaxations were performed until the Hellmann-Feynman force on each atom becomes smaller than 0.002 eV/Å and the total energy was converged to be within 10^{-8} eV. Spin-polarization had to be taken into account in order to reproduce the semiconducting nature of this system, and the effect of

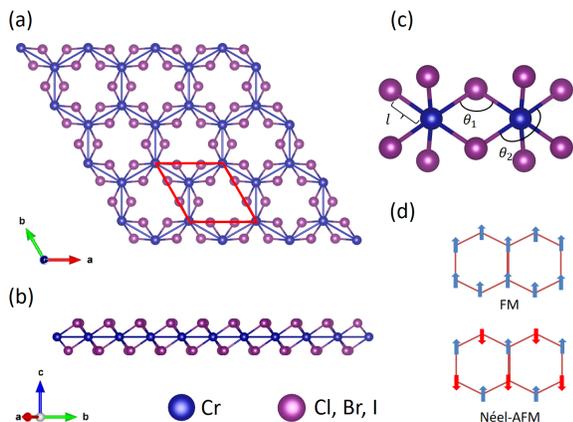


FIG. 1. Atomic structure of monolayer CrI_3 . (a) Top view and (b) side view of a single layer; (c) Bonding between chromium and iodine atoms. The unit cell of CrI_3 which includes two Cr and six I atoms has been indicated in (a). The bond length l between Cr and I atom, the bond angle θ_1 between Cr and two I atoms in the same plane, and the axial angle θ_2 are also shown in (c). The two magnetic orders, namely Néel-antiferromagnetic (AFM) and ferromagnetic (FM) are displayed in (d)

introducing SOC on the electrical and mechanical properties of these materials will be discussed. In addition, two different magnetic configurations were considered to evaluate the magnetic ground state by comparing their total energies. The ferromagnetic (FM) configuration had all magnetic moments initialized in the same direction while in the antiferromagnetic (AFM) configuration the magnetic moments were set to be antiparallel between nearest neighbors. For both cases, spin orientations were initially in the off-plane direction. These two typical magnetic orderings are shown in Fig. 1(d).

Magnetic anisotropy energy (MAE) is defined as the difference between energies corresponding to the magnetization in the in-plane and off-plane directions ($\text{MAE} = E_{\parallel} - E_{\perp}$). Therefore, the positive (negative) value of MAE indicates off-plane (in-plane) easy-axis. To evaluate MAE, one must take SOC effects into account. Thus, non-collinear non-self-consistent calculations were performed to evaluate the total energies E_{\parallel} and E_{\perp} after the self-consistent ground states were achieved.

In this work, only biaxial strain has been studied. For each strain, the lattice constants were changed accordingly and then kept fixed, while the atomic positions were fully optimized. This procedure was repeated for single layer CrCl_3 , CrBr_3 and CrI_3 .

III. RESULTS AND DISCUSSIONS

A. Atomic structures

The bulk chromium trihalides CrX_3 ($X=\text{Cl}$, Br , and I) are layered van der Waals (vdW) materials and the

possibility of mechanically exfoliating CrI_3 to produce 2D monolayers has been demonstrated². These systems exhibit rhombohedral BiI_3 structure (space group $R\bar{3}$) in cryogenic temperatures at which ferromagnetism can be observed. The Curie temperatures for the bulk systems are 27, 47, and 70 K for CrCl_3 , CrBr_3 , and CrI_3 respectively^{31,32}. In the single layer limit, CrI_3 retains its ferromagnetism, and the Curie temperature of the 2D system is found to be $T_c = 45 \text{ K}$ ². Fig. 1(a) shows a schematic plot of the single layer chromium trihalide compound. The chromium ions form a honeycomb network sandwiched by two atomic planes of halide atoms as shown in Figs. 1(a) and 1(b). The parallelogram in Fig. 1(a) represents the unit cell containing two chromium atoms and six halide atoms per layer. Moreover, Cr^{3+} ions are coordinated by edge-sharing octahedra, as shown in Fig. 1(c).

Magnetism in these compounds arises from the partially filled d orbitals, as Cr^{3+} ion has an electronic configuration of $3d^3$. Despite this fact, these materials are found to be electrical insulators, indicating that a Mott-Hubbard mechanism is playing a role in the formation of the band gap. In the octahedral environment, crystal field interaction with the halide ligands results in the quenching of orbital moment ($L = 0$) and splitting of the chromium d orbitals into a set of triply degenerate t_{2g} orbitals (with lower energy) and doubly degenerate e_g orbitals (with higher energy). Furthermore, saturation magnetization measurements give an atomic magnetic moment of $3 \mu_B$ per chromium atom³³. This is consistent with Hund's rule which predicts that the three electrons will occupy the t_{2g} triplet yielding $S = 3/2$.

First, we investigate the structure, magnetism and MAE of the unstrained monolayer systems. Our results from non-collinear self-consistent calculations show that the ground state for the three systems is FM, as indicated in Table 1. Here, the difference in the total energy of the two magnetic phases considered in our study ($E_{FM} - E_{AFM}$) is less than zero for all the chromium trihalides. For this reason, only the optimized structural parameters for the FM ground state are shown in Table 1. For example, for the case of CrI_3 , the lattice constant (a_0) and Cr-I bond length (l) are 7.008 \AA and 2.740 \AA respectively. These results are consistent with previously reported values^{24,31}. The Cr-I-Cr bond angle (θ_1) is 95.2° and is represented in Fig. 1(c). This angle accounts for the ferromagnetic superexchange interaction according to the Goodenough³⁴ and Kanamori³⁵ rules. The axial angle (θ_2) is the angle formed between the chromium ion and two opposing ligands within the same octahedral (e.g. Fig. 1(c)). Therefore, our results suggest some deformation in the octahedral environment as the axial angle is predicted to be slightly smaller than 180° . Interestingly, these angles are nearly independent from the ligands, as they are roughly the same for all three systems. In our previous work, we found that different magnetic orderings (FM or AFM) yield similar results for the structural parameters³⁶. Moreover, these struc-

TABLE I. The calculated lattice constants a_0 , total energy E_t , bond lengths l , bond angles θ_1 and axial bond angles θ_2 for the FM phase of the chromium trihalides. The difference in energy between two magnetic phases $E_{FM}-E_{AFM}$, magnetic anisotropy energy (MAE) and the easy magnetization axis are also listed. SOC has been included in all calculations.

	Lattice Parameters					Magnetic Stability for 1L		
	a_0 (Å)	E_t (eV)	θ_1 ($^\circ$)	θ_2 ($^\circ$)	l (Å)	$E_{FM}-E_{AFM}$ (eV)	MAE ($\mu\text{eV}/\text{Cr}$)	Easy Axis
CrCl ₃	6.056	-38.916	95.8	173.2	2.357	-0.023	24.68	c
CrBr ₃	6.438	-35.334	95.1	173.5	2.518	-0.032	159.54	c
CrI ₃	7.008	-32.318	95.2	173.3	2.740	-0.036	803.65	c

tural parameters are not sensitive to the SOC³⁶, which is essential for investigating the MAE below.

The calculated MAE for each compound is listed in Table 1. The MAE of CrI₃ is about 804 $\mu\text{eV}/\text{Cr}$, surprisingly large when compared to the other compounds in Table 1. Previous studies reported 980 $\mu\text{eV}/\text{Cr}$ ⁵ and 686 $\mu\text{eV}/\text{Cr}$ ²⁴, and these discrepancies might result from different methods adopted. Lado *et al.* suggested that the large MAE in CrI₃ is originated from an anisotropic exchange interaction through a superexchange mechanism, which stems from the strong SOC in the heavier iodine ions¹⁰. In addition, the easy axis for energetically favorable spontaneous magnetization is found to be perpendicular to the basal plane, i.e., along the c direction.

B. Electronic structures

The electronic band structures for CrCl₃, CrBr₃ and CrI₃ under various biaxial strain are shown in Figs. 2(a), 2(b) and 2(c), respectively. In Ref.³⁶, we already show that the band structure of CrI₃ is highly sensitive to the magnetic ordering, the exchange-correlation functional, and SOC. Here we focus on the results calculated from PBE for the FM phase only. The effect of SOC on the electronic band structure can be further elucidated by comparing Fig. 2 with Fig. S1 in the Supplemental Materials³⁷, which shows the collinear results. Unlike CrI₃, the band structures of the other compounds are insensitive to the inclusion of SOC. This provides a further evidence that MAE in these systems is closely related to the SOC in the halide ligands.

Clearly, the band gaps increase from 0.89 eV to 1.58 eV from iodine to chlorine. Both CrCl₃ and CrI₃ exhibit a direct band gap. The band gap character found for unstrained CrI₃ is consistent with previous works, which also included SOC^{5,26}. In addition, from Fig. 2(a) and 2(b), we notice an increase (decrease) in the band gap upon compression (tensile strain) in these systems. However, according to Fig. 2(c), the application of a biaxial strain causes an opposite effect on the band gap of CrI₃. In the three compounds, the VBM remains approximately constant with the application of strain, whereas the CBM is shifted, causing a direct-to-indirect band gap transition in both CrCl₃ and CrI₃. Moreover, a compressive strain will decrease the energy of the valence bands near M and K points in CrI₃. This effect can also be

observed in collinear calculations, however it is more evident when SOC is included.

C. Effect of Strain on Crystal Structure and the Magnetic Order

Next, we investigate the dependence of magnetic properties under different strains. It is mainly Cr atoms that contribute to the magnetic moment, which remains overall constant with 6 μ_B (two Cr³⁺ ions per unit cell) per unit cell under strain (not shown). Fig. 3 shows the energy difference between the two magnetic orderings, namely FM and AFM. A phase transition from FM to AFM is observed in all systems and the AFM region is highlighted in red. Here, the strain ε is defined as following:

$$\varepsilon = \frac{(a - a_0)}{a_0}, \quad (1)$$

where a_0 is the lattice constant for the unstrained system. The energy difference of the two phases is given by (neglecting the MAE since it is relatively small):²⁴

$$E_{FM/AFM} = E_0 - (\pm 3J_1 + 6J_2 \pm 3J_3) \left| \vec{S} \right|^2, \quad (2)$$

where J_1 , J_2 and J_3 are the Heisenberg exchange integrations for the first, second and third nearest neighbors respectively. Neglecting the second and third nearest neighbors and taking the energy difference between FM and AFM phases, we have:

$$E_{FM} - E_{AFM} = -6J \left| \vec{S} \right|^2. \quad (3)$$

Here $\left| \vec{S} \right| = 3/2$. From the energy difference calculated in DFT (as shown in Table I), one can determine the exchange parameter J with Eq. 3. Next, with J available one can roughly estimate the Curie temperature from the mean-field expression:

$$T_c = \frac{3J}{2K_B}. \quad (4)$$

A brief derivation is provided in the Supplemental Materials³⁷. The Heisenberg exchange parameter and

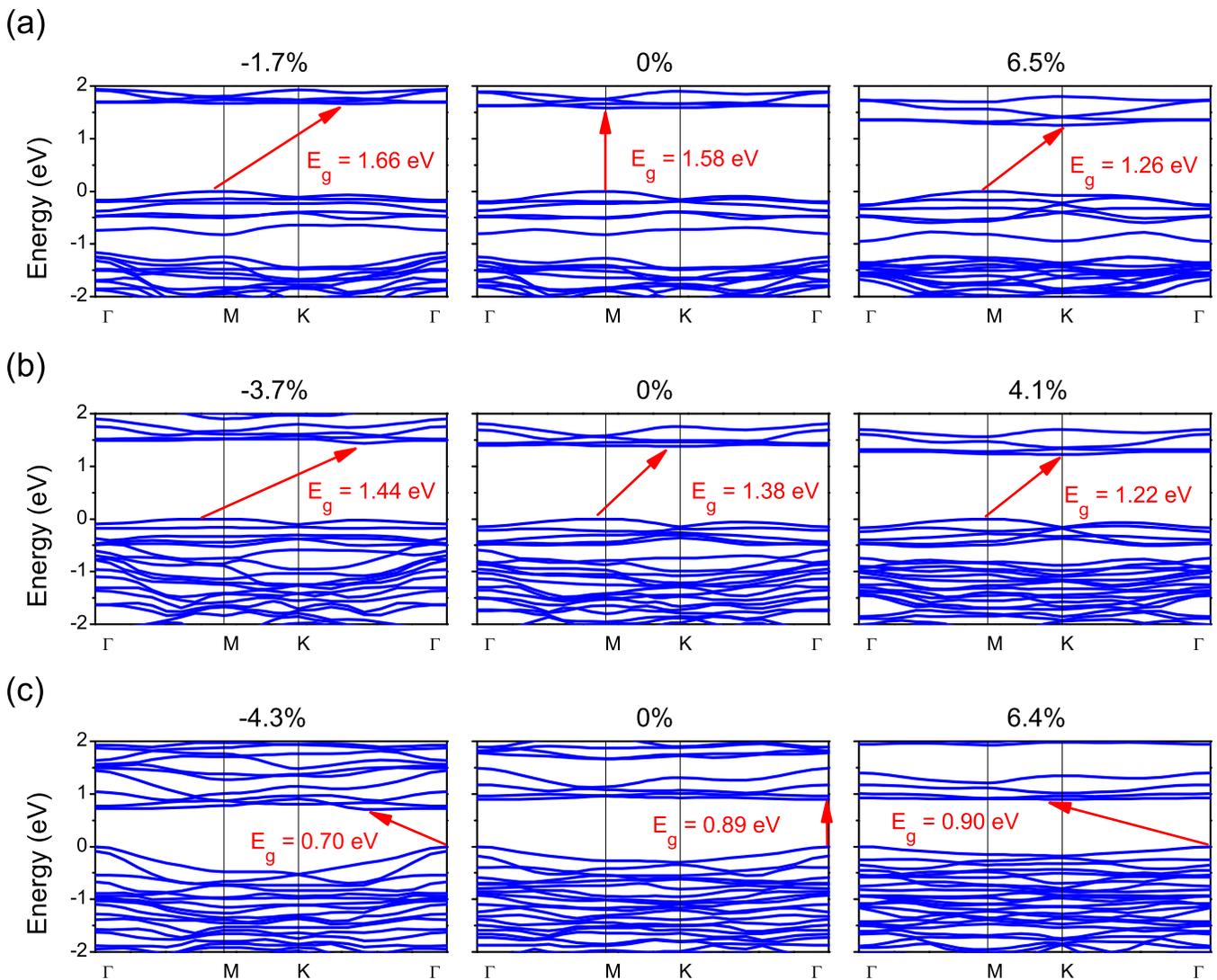


FIG. 2. Non-collinear spin polarized electronic band dispersions for monolayer chromium trihalides. The effect of typical compressive and tensile strain are also shown in (a) CrCl_3 ; (b) CrBr_3 ; (c) CrI_3 . The energy band gaps between conduction band minimum (CBM) and valence band maximum (VBM) have been indicated using red arrows in each case. The VBM has been shifted to zero.

TABLE II. The exchange coupling and Curie temperature of single-layer chromium trihalides.

	J (meV)	Estimated T_c (K)	Experimental T_c (K)
CrCl_3	1.7	29.7	27 (Bulk) ³¹
CrBr_3	2.4	41.3	47 (Bulk) ³¹
CrI_3	2.7	46.4	70 (Bulk) ³¹ , 45 (1L) ²

the estimated Curie temperature for the unstrained systems are listed in Table II, along with the available experimental values for the Curie temperature. The obtained value for J , considering only the first nearest neighbors, agrees with previous DFT calculations²⁴. From Table II, the estimated Curie temperatures for single layer CrCl_3

and CrBr_3 are approximately equal to the experimental values for the bulk systems. Whereas there is a difference between the Curie temperatures for the bulk CrI_3 and monolayer CrI_3 , our estimation is closer to that of monolayer CrI_3 . This is expected, since Eq. 4 is valid for the 2D system, and suggests a relatively large interlayer coupling in bulk CrI_3 .

In Fig. 3, we show the energy difference $E_{FM} - E_{AFM}$ between the two magnetic ordering as a function of ε for CrCl_3 , CrBr_3 and CrI_3 . The evolution of the Curie temperature calculated based on Eq. (4) has been also shown in Fig. 3 (right axis). As ε decreases (i.e., the compressive strain increases), the energy difference increases. There is a phase transition to the AFM phase when the energy difference between FM and AFM ordering becomes greater than zero. This phase transition occurs at -2.5

%, -4.1 % and -5.7 % compressive strain for the CrCl_3 , CrBr_3 and CrI_3 , respectively. Similar result was reported by Zheng *et al.* for CrI_3 under compressive strain²⁶. According to Fig. 3, there is a point in which the Curie temperature is maximum and this point is close to the equilibrium for CrI_3 . However, as shown in Fig. 3(a), this point is slightly shifted from the equilibrium to the region with small tensile strain in CrCl_3 and CrBr_3 , suggesting that in these cases the Curie temperature can be further increased with the introduction of tensile strain. We estimate that a tensile strain of 2.4 % can increase T_c to 39.0 K in CrCl_3 , and a tensile strain of 2.1 % can increase T_c to 44.4 K in CrBr_3 . Beyond this point of local maximum, the Curie temperature tends to decrease and no transition to the AFM phase is observed if we further increase tensile strain within the 10 % range.

We have also calculated the effect of strain on the structural parameters as depicted in Fig. 4. These calculations were repeated for each material with different magnetic ordering (FM and AFM). Since results of AFM are similar, we show only the results for the FM phase. From Fig. 4, one can clearly see that the Cr-X-Cr angle changes linearly with respect to strain. In addition, the angle with $\varepsilon = 0$ is roughly the same (95°) for the three halides. Within the 10 % range from the equilibrium point ($\varepsilon = 0$), the curves display a linear shape with the same slope for the three materials. The axial angle has a similar behavior, but tends to increase upon compression until it saturates near 180° . At this point, with the axial angle fully stretched (around -10 % of compression for all materials) the system achieves a higher degree of symmetry. The Cr-X distance displays a weak strain dependency within the 10 % range.

D. Effect of Strain on the Magnetic Anisotropy Energy

One can also observe how MAE changes with biaxial strain in Fig. 5. In Figs. 5(a)-5(c), the energy was calculated as a function of the angle of magnetization with respect to the basal plane (θ_M), being 0° in-plane and 90° off-plane. This is illustrated in the inset of Fig. 5(f), where E_{\parallel} and E_{\perp} represent the energies calculated when all the spins are parallel and perpendicular to the atomic plane, respectively. These calculations were repeated for different values of strain. If we neglect the higher order terms, the dependency of the energy per chromium atom with respect to θ_M is given by:³⁸

$$E(\theta_M) = E_0 + \lambda_1 \sin^2(\theta_M) + \lambda_2 \sin^4(\theta_M). \quad (5)$$

Here, E_0 is a constant energy shift, λ_1 and λ_2 are respectively the quadratic and quartic contributions to the energy. No substantial difference in energy with respect to the different in-plane directions are observed from DFT calculations, therefore the azimuthal contribution to $E(\theta_M)$ is neglected. From Figs. 5(a)-5(c) we note that $E(\theta_M)$ provides a good fit for the energies. In addition,

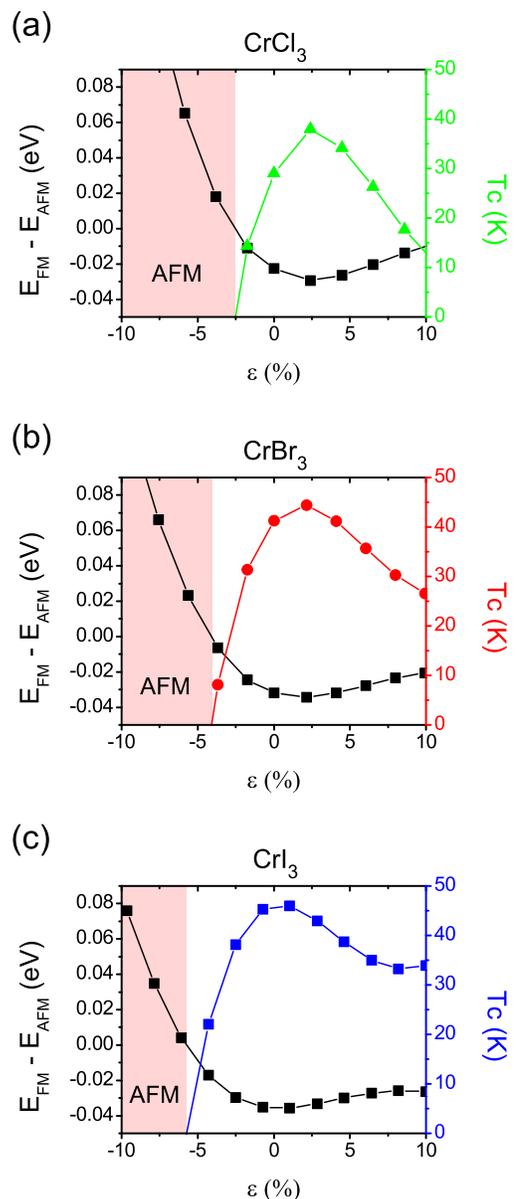


FIG. 3. Energy difference between FM and AFM phases for (a) CrCl_3 ; (b) CrBr_3 ; (c) CrI_3 . The AFM phase region is highlighted in red. The calculated Curie temperature is also shown for each case.

it can be seen from Figs. 5(a)-5(c) that the easy axis remains off-plane for CrI_3 and CrBr_3 even when subject to strain, whereas for CrCl_3 there exists a phase transition to an in-plane easy axis upon compression. This can be further verified by looking at Figs. 5(d)-5(e), in which we take the difference between in-plane (E_{\parallel}) and off-plane (E_{\perp}) energies and plot them with respect to strain. Here, negative values seen in Fig. 5(c) for CrCl_3 represent an in-plane preference for magnetization.

Although bulk CrCl_3 is found to possess an in-plane easy axis³⁹, it should be noted from Fig. 5(c) that the MAE of unstrained monolayer CrCl_3 is positive, indicat-

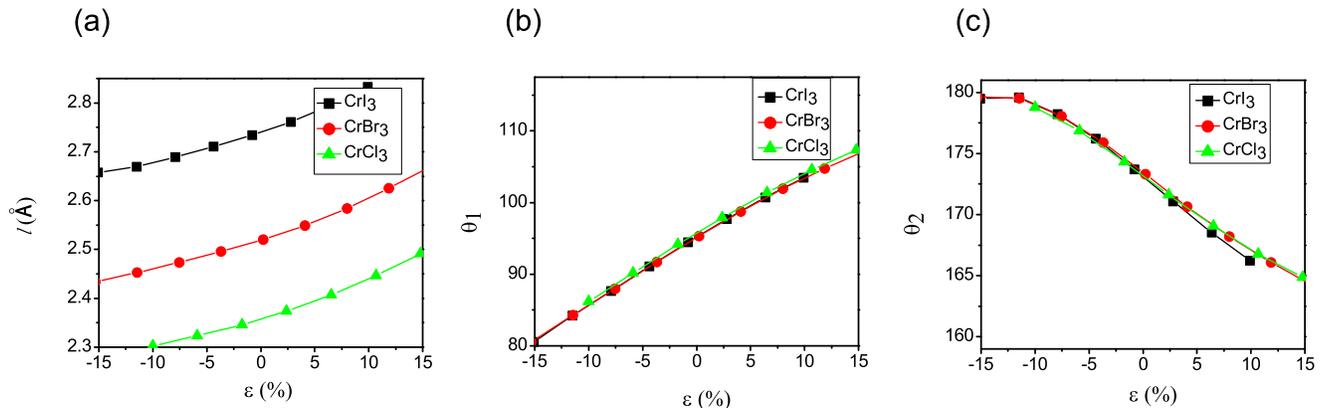


FIG. 4. Dependence of structural parameters on biaxial strain ϵ for the chromium trihalides. (a) Bond length l ; (b) Bond angle θ_1 ; (c) Axial bond angle θ_2 .

ing that the spins of Cr atoms align perpendicular to the basal plane, despite that the MAE is much smaller than that of CrI₃. Similar result was also reported by Zhang *et al.*²⁴. This result suggests a possible transition from an in-plane to an off-plane easy axis for CrCl₃ upon exfoliation. This is not surprising, considering that the MAE of CrCl₃ is much smaller than that of CrI₃. Furthermore, unlike CrBr₃ and CrCl₃ compounds, CrI₃ crystal exhibits much stronger anisotropy when compressed and becomes weaker when stretched. More specifically, a -5% compressive strain will increase 47% of MAE in this system.

Fitting parameters for λ_1 and λ_2 can be found in Figs. 5(g)-5(i). These plots show the strain dependence of these constants for the three monolayer systems, with the λ_2 graph being displayed in the inset. We note that, for all systems the quadratic contribution dominates the MAE, as the change of λ_1 resemble the MAE curves for each compound. However, as tensile strain increases, the quartic contribution gets comparable to the quadratic in magnitude for CrBr₃ and CrI₃.

Finally, we would like to point out that calculations based on LDA yield similar effects on strain on the MAE, although the value is slightly different (see Fig. S3(a) in the Supporting Materials). In addition, the MAE is strongly dependent on the on-site parameter of Hubbard U for Cr. Our PBE+ U calculations show that the MAE increases dramatically when the Hubbard U parameter is increased. The enhancement in the MAE with respect to a 5% compressive strain ranges from 20.7% - 58.1% depending on the value of Hubbard U parameter. All the data has been included in the Supplemental Materials³⁷.

IV. CONCLUSIONS

In summary, we applied density functional theory to investigate the magnetic and electronic properties of a

recently found 2D magnet, monolayer CrI₃ and other compounds from the same family including CrBr₃ and CrCl₃. All three monolayer systems are found to be ferromagnetic in the ground state in accordance to previous work. In addition, CrI₃ exhibits strong magnetic anisotropy (804 $\mu\text{eV}/\text{Cr}$) with an easy axis perpendicular to the basal plane. The MAE decreases dramatically as the atomic number of the halide decreases, with CrBr₃ and CrCl₃ having 160 and 25 $\mu\text{eV}/\text{Cr}$ respectively. We also estimated the Curie temperature from mean field approximation and the calculated energy differences between two different magnetic orders, namely FM and AFM. Our results are in good agreement with experimental data, and suggest a relatively large interlayer coupling in the CrI₃ system. Interestingly, our mean-field estimation predicts that the Curie temperature for CrCl₃ can increase from 29.7 K to 39 K with 2.4 % of tensile strain.

Furthermore, we have studied the effect of biaxial strain and have determined the strain dependence of the atomic structure, electronic and magnetic properties of the chromium trihalides. Strong SOC in the CrI₃ results in a more evident strain dependence of the electronic and magnetic properties as compared with CrBr₃ and CrCl₃. For example, the MAE in CrI₃ increases when compressed and decreases when stretched. A 5% compressive strain will increase MAE by 47% in this system.

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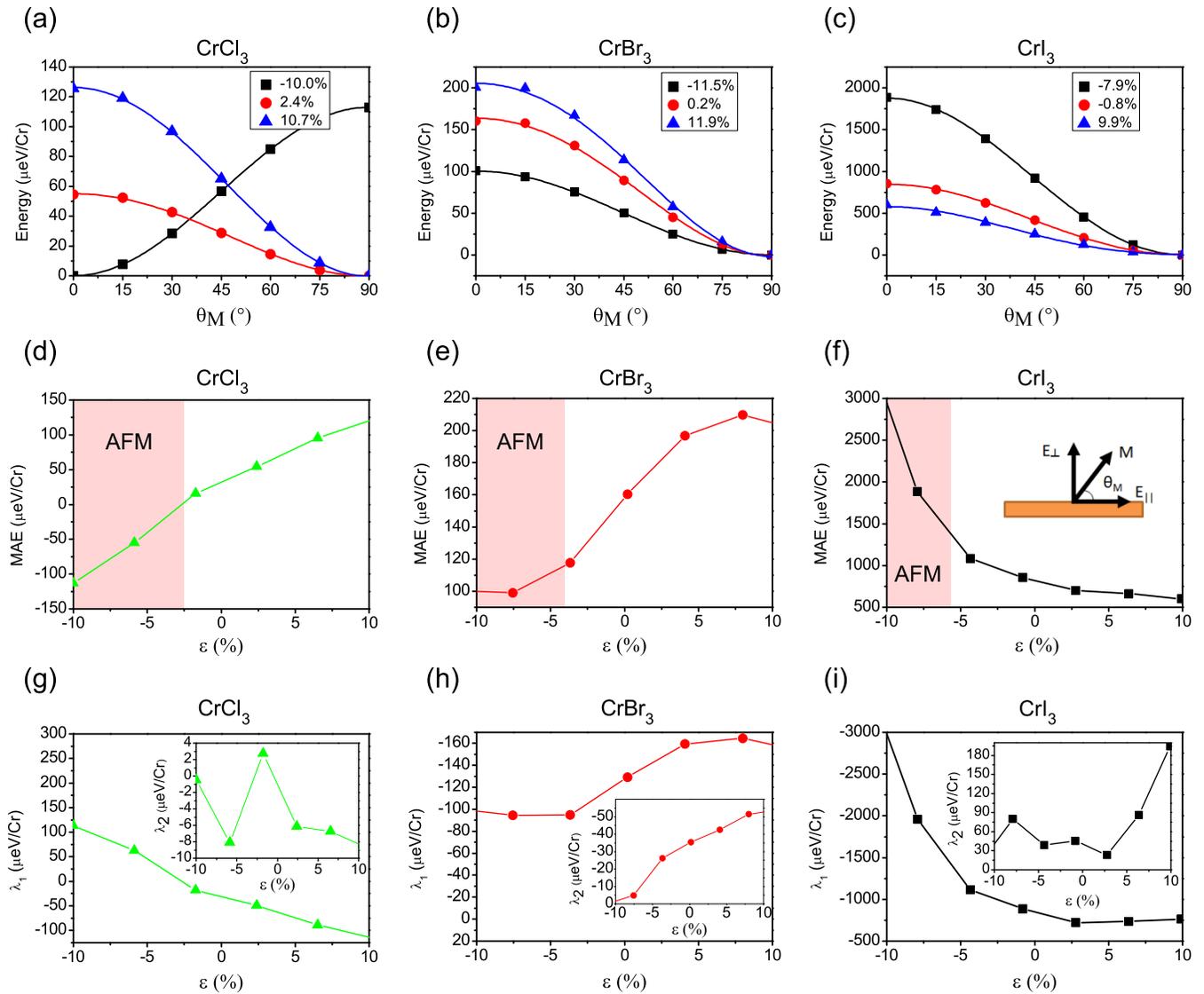


FIG. 5. The effect of strain on the magnetic anisotropy energy. Change of energy with respect to the magnetization angle θ_M for (a) CrCl₃, (b) CrBr₃ and (c) CrI₃. The line is the fitted result. (d)-(f) Change of MAE with respect to strain in (d) CrCl₃, (e) CrBr₃ and (f) CrI₃. The AFM phase region is highlighted in red. The change of the fitted parameters λ_1 and λ_2 are shown in (g)-(i).

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