Magnetism and spin exchange coupling in strained monolayer CrOCl†

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The magnetism and spin exchange coupling of monolayer CrOCl with different strains are investigated systematically using first principles. It is found that the magnetic ground state can be changed from ferromagnetic (FM) to antiferromagnetic (AFM), and the Curie temperature (T_C) is enhanced significantly by applying the uniaxial strain along a- or b-axis direction. The variations of spin exchange coupling are explained according to the Goodenough–Kanamori–Anderson (GKA) and Bethe–Slater Interaction (BSI) rules. The strain-dependent magnetic state is mainly attributed to the competition between direct exchange interactions of cation–cation and indirect superexchange ones of cation–anion–cation in monolayer CrOCl. The different competitions in a- and b-axis direction determine the different critical intervals R of magnetic transitions, where R is the distance of the two nearest-neighbor (NN) Cr3+ ions. The AFM–FM transition occurs at R/R_{5a} = 2.9 and 3.75 in a-axis direction, while it happens at R/R_{5a} = 2.65 along b-axis direction. These results indicate that the sensitive relevancy between the external strain and magnetic coupling makes monolayer CrOCl a promising candidate for spintronics.

I. Introduction

The emergence of two-dimensional (2D) materials such as graphene has brought a new opportunity for the development of spintronics.1–4 Ferromagnetism is believed to be very useful for spintronics, however the Mermin–Wagner theorem states that in 2D systems strict long range order at finite temperature is impossible because of the fluctuations. Therefore this has been a widely held belief that ferromagnetism cannot survive in 2D systems until the recent observations of long-range magnetic order in 2D CrI3 and Cr2Ge2Te6.5–7 Moreover, the molecular beam epitaxial growth has been reported to confirm the existence of 2D Cr2Ge2Te6 (20 K).6,7 The development of new ferromagnetic 2D magnetism in Fe3GeTe2, VSe2, MnSe and Cr2Ge2Te6,8–11 has been theoretically predicted, namely MnNX and CrCX (C = S, Se, Te; X = Cl, Br, I), which enlightens a strategy for searching other high T_C FM in polyvalent CrOCl monolayers with two different valence states (of anions: O2– and Cl–).12

Mechanical strain is commonly regarded as an effective route to manipulate the physical properties including the spin order and transition temperature because these properties critically depend on the material’s structure.4,11 For the 2D materials, the small Young’s modulus of elasticity make it possible to withstand large strains in the systems, for example, the monolayer graphene and MoS2可以 undergo about 25% strain,14–16 which is much larger than those of perovskite films. The investigation has demonstrated that strain can not only result in the splitting of certain peaks in the Raman spectrum of graphene,17 but also affect the electronic structure of both monolayer and bilayer MoS2 and modulate optical properties of rippled 2D GaSe.18,19 Moreover, the spin–lattice coupling has been investigated experimentally or predicted theoretically in layered magnets, such as Cr2Si2Te6,20 Cr2Ge2Te6,21 Fe3GeTe2,22 MnPSe3 and CrI3.23,24 Biaxial tensile strain can lead to an AFM–FM transition in 2D MnPSe3,23 and a FM–AFM transition in monolayer CrI3 by spin–lattice coupling effect.24 These investigations indicate that atomic scale in 2D intrinsic magnetic film provide a platform for strain to manipulate electronic structures and spin exchange coupling due to their fascinating high stretchable property.

Different from the bulk AFM state,25 monolayer CrOCl is predicted to be a new type of potential 2D intrinsic ferromagnetic...
materials for its small exfoliation energy, strain-dependent $T_C$, great dynamical and thermal stabilities.\textsuperscript{26} Similarly to the preparation of graphene, the monolayer CrOCl can also be successfully mechanically exfoliated,\textsuperscript{27} however, the magnetic properties of the monolayer CrOCl were theoretically predicted not to follow the GKA rules\textsuperscript{28,29} by which the graphene abide, especially magnetic exchange coupling, even though the GKA rules have been successful in explaining the magnetic properties induced by superexchange interactions on a lot of 2D and bulk materials, e.g. recently-highlighted 2D CrX$_3$ (X = F, Cl, Br, I) materials and bulk ABX$_3$ (A = Mn, Cr; B = P, Si; X = S, Se, Te) etc. Besides the GKA rules, an extended superexchange theory (ESET) is also proposed to explain the magnetic properties in monolayer CrOCl.\textsuperscript{30} However, it remains not clear whether the ESET theory can be applied to the strained monolayer CrOCl, and many relevant properties of monolayer CrOCl are still under investigation, especially effects of strain on electrical, magnetic and optical properties.

In the present paper, using first-principles calculations, we investigated the magnetic phase transition, the spin exchange interaction and mechanism of monolayer CrOCl materials under strain engineering. By applying the 0–16% uniaxial strain, including compression and tensile ones along $a$- and $b$-axis, we find that the uniaxial compressive strain along $a$-axis (15%) or $b$-axis (8%), and tensile strain along $a$-axis (16%) both can lead to FM-AFM transition. The strain-dependent magnetic properties of monolayer CrOCl is mainly attributed to the competition of the direct interaction and indirect superexchange interaction between the two NN Cr$^{3+}$ atoms.

### II. Computation detail

#### II.1. Crystal structure

The bulk transition-metal oxyhalide CrOCl is an AFM semiconductor, which has orthorhombic structure (space group $Pmmn$) with 2D rectangle sublattice in the $a$-$b$ plane, seeing Fig. 1(a–c). The Cr ions bounded by O atoms form alternated pyramids while the planar layers sandwiched by Cl atomic layers are stacked along the $c$-axis with a large van der Waals gap, as shown in Fig. 1(b) and (c). Every Cr ion is coordinated by six covalent bonds including neighboring four O and two Cl ions, and these bonds generate a strong twisted octahedron of CrO$_2$Cl$_2$, as plotted in Fig. 1(d).

#### II.2 Computation method

All calculations were performed based on density functional theory by using the projector-augmented wave (PAW) method as implemented in Vienna $ab$ initio Simulation Package (VASP).\textsuperscript{31,32} The generalized gradient approximation (GGA) with Perdew–Burke–Ernzerhof parametrization (PBE) functional is used for the exchange–correlation functional calculations.\textsuperscript{33} The valence electron configuration is set by: Cr 3d$^{5}$s$^{1}$, O 2s$^{2}$p$^{4}$ and Cl 3s$^{2}$3p$^{5}$ in our calculations. The strong on-site Coulomb interaction for the transition-metal Cr-3d orbital are considered by the GGA+U method following the Dudarev scheme to compensate for the self-interaction errors of GGA.\textsuperscript{34,35} We tested several $U$ values, and the test results showed that when $U = 7.5$ eV and $J = 0.5$ eV, the optimization results were more consistent with the experimental parameters.\textsuperscript{25} For all calculations, we set the plane-wave cutoff energy to 450 eV, the convergence energy to $1.0 \times 10^{-6}$ eV and the residual forces on atoms to 0.001 eV Å$^{-1}$. A $2 \times 2 \times 2$ supercell and one $2 \times 2 \times 1$ supercell are adopted in optimizing the bulk and monolayer structures, respectively. The Brillouin zone integral was generated automatically by the Monkhorst–Pack method, and the $k$-point meshes for bulk and monolayer were $5 \times 6 \times 5$ and $5 \times 6 \times 1$ respectively. For monolayer CrOCl, the vacuum space thickness of 20 Å was set along the $c$-direction, and for bulk, the DFT-D3 method was used to consider van der Waals (vdW) correction.\textsuperscript{36}

### III. Result and discussions

#### Structure optimization

In order to investigate the magnetic coupling in monolayer CrOCl, firstly, four different magnetic configurations, including FM, AFM$_{Neel}$, AFM$_{stripy-1}$, and AFM$_{stripy-2}$ state, are constructed in a $2 \times 2 \times 1$ supercell of monolayer CrOCl, as shown in Fig. 2. When the four magnetic state structures are optimized in unstrained monolayer CrOCl, we find that the energy differences between the three AFM and FM state are 109.11, 175.48, and 101.35 meV per unit cell respectively, as listed in Table 1, which indicates FM order is the ground state (G.S.) in monolayer CrOCl. Moreover, the obtained lattice constants, band gap, and magnetic moment of each metal ion of unstrained monolayer CrOCl at FM ground state is $a = 3.941$ Å, $b = 3.272$ Å, $E_g = 2.35$ eV and $M = 3.235 \mu_B$, respectively, which are consistent with the previous results.\textsuperscript{26} Of course, it should be noted that the non-magnetic (NM) state has not been considered owing to the great energy disparity between the NM state and magnetic states.

To confirm the magnetic order in bulk CrOCl without strain, we combine two FM state layers with the interlayer FM and AFM...
coupling for magnetic calculations, respectively. It shows that the energy of interlayer AFM coupling is favored by 1.68 meV per u.c. lower than the FM state, which indicates that the bulk CrOCl is an intrinsic antiferromagnet. While for the AFM bulk CrOCl, the obtained lattice constants \(a\), \(b\), and \(c\) are respectively 2%, 2.7%, and 0.8% larger, and electronic band gap and magnetic moment are approximately 3.5% less than the experimental values, as illustrated in Table 2.

In the meantime, allowing for the effect of spin–orbit coupling (SOC) on the electronic structure and magnetic properties, we take it into account in the self-consistent calculations to clarify the nature of magnetic anisotropy in CrOCl. Then, we calculate the unit cell total energy \(E_{100}\), \(E_{010}\) and \(E_{001}\) with the magnetic moment along the [100], [010] and [001] direction respectively, as shown in Table 3. It is found that the \(E_{001}\) is 0.74 and 0.72 meV less than \(E_{010}\) and \(E_{100}\), respectively. In other words, monolayer CrOCl has the out-of-plane magnetic anisotropy, but the magnetic anisotropy energy is small. Then, the energies of four magnetic structures are calculated with SOC, respectively. It is found that the unit cell total energy \(E_{FM}\) of the FM state is 116.42, 178.37 and 100.85 meV less than those of the AFM-Neel, AFM-stripy-1 and AFM-stripy-2 state when the easy axis lies along the [001] direction, respectively. It follows that the FM order is still G.S.

Further, to examine the influence of SOC on electronic properties, we also compared the band structure with SOC and without SOC, as shown in Fig. 3. The results show that the inclusion of the SOC in the FM state hardly changes the correspondingly band dispersions near the band edges, and the indirect energy gap with the SOC is just 0.01 eV less than that without SOC. Compared with the band without SOC, The effect of SOC is only to shift the whole bands slightly towards the lower energy region, while the location of the CBM and VBM remain the same. Apparently, distinct from the chromium triiodide CrI\(_3\), the SOC effect is weak in monolayer CrOCl. This findings agrees well with a very recent theoretical results by Wang et al. Therefore, based on this weak influence of the SOC on the energy band, G.S. and magnetism of monolayer CrOCl, we neglect the effect of SOC in the following investigations.

### Magnetic phase transition

To investigate the variations of magnetism and spin exchange coupling of the strained monolayer CrOCl, different tensile and compressive strains along \(a\)- and \(b\)-axis are applied respectively.
for the above four magnetic structures, while the c-direction is completely relaxed. The strain is defined as $\varepsilon = (a - a_0)/a_0$, where $a_0$ and $a$ are the lattice constants of unstrained and strained monolayer CrOCl, $\varepsilon > 0$ and $\varepsilon < 0$ denote the tensile strain and compressive strain, respectively. In our calculation, the strain $\varepsilon$ is varied from $-16\%$ to $16\%$. Then, the calculated total energies of supercell with different magnetic states under uniaxial strain along $a$- and $b$-axis are obtained, as shown in Fig. 4(a) and (b). The parabolic curves indicate that the mono-layer CrOCl is in the range of elastic variation.

From Fig. 4(a), it is found that monolayer CrOCl do not change the FM state until a rather large uniaxial strain $\varepsilon (-15\%$ and $16\%)$ along $a$-axis, where the magnetic transitions between FM and AFM$_{stripy}$ occur obviously, seeing Fig. 4(c). However, there is a little difference under strain along $b$-axis, the FM-AFM$_{stripy}$ magnetic transition will be observed when the strain reach to $\varepsilon = -8\%$, as shown in Fig. 4(b) and (d), but the tensile strain in this direction can’t realize a magnetic transition. Of course, the strain-dependent magnetic transition has also been revealed in many other 2D magnetic materials, such as MnPSe$_3$, CrI$_3$, CrSiTe$_3$ and CrPS$_4$.23,24,39,40

As seen from Fig. 4(c) and (d), the magnetic order of monolayer CrOCl is sensitive to strain and exhibits the obvious anisotropy, however, the magnetic exchange interaction do not conform entirely to the GKA rules, which has revealed that the AFM (FM) order is along the bond angle of $180^\circ$ ($90^\circ$) in cation–anion–cation interaction path, for example MnO.30 In 2D semiconductor MnPSe$_3$,23 FeOCl and CrX$_3$ (X = F, Cl, Br), it is predicted theoretically that the strain-dependent magnetic stability is mainly attributed to the competition of the direct interaction and indirect superexchange interaction between the two NN magnetic atoms. Then, as for whether the similar competition also exists in monolayer CrOCl, it is something that needs to be discussed in depth.

**Exchange coupling**

Both magnetic transition and variation of Curie temperature can be tuned, which may be ascribed to the strain-tuning magnetic exchange coupling in monolayer CrOCl. To clarify the relations among them, it is necessary to estimate the spin exchange coupling of the neighboring level. We considered the first, second and third nearest-neighbor magnetic exchange coupling as: $J_{11}$, $J_{22}$, and $J_{33}$, which are shown in Fig. 2(a). Using the spin Hamiltonian based on the 2D Heisenberg model: $H = -J_{ij}\sum\vec{S}_i \cdot \vec{S}_j$, where $J_{ij}$ is the adjacent spin exchange coupling constants, and mapping the DFT energies of different magnetic states to the Heisenberg model, the four spin configurations energies satisfy the following equations:12,38,42

\[
E_{FM} = E_0 - (4J_{11} + 2J_{22} + 2J_{33})|\vec{S}|^2, \\
E_{AFM-Ne\'el} = E_0 - (-4J_{11} + 2J_{22} + 2J_{33})|\vec{S}|^2, \\
E_{AFM-stripy-1} = E_0 - (2J_{22} - 2J_{33})|\vec{S}|^2, \\
E_{AFM-stripy-2} = E_0 - (-2J_{22} + 2J_{33})|\vec{S}|^2,
\]

where $E_0$ is the nonmagnetic total energy of one magnetic atom, and $|\vec{S}|$ is the magnetic moment of Cr ions that equals to $3\mu_0$.

Firstly, for the unstrained monolayer CrOCl, the calculated results are $J_{11} = 0.189$ meV, $J_{22} = 0.162$ meV and $J_{33} = 0.420$ meV. All of them are positive, indicating their FM interactions. The $J_{11} > J_{22} > J_{33}$ relation suggests the largest spin exchange coupling existed along $a$-axis, which is in agreement with the result of previous calculations.30 According to GKA rules,28–30 a crossover angle of $127 \pm 0.5^\circ$ is proposed to be related to the FM-AFM transition.42,43 The two Cr–O–Cr angles are $95^\circ$, very close to $90^\circ$, so $J_{11}$ is a FM superexchange coupling through O ions, as shown in Fig. 5(a). Similarly, as the Cr–Cl–Cr angle is very close to $90^\circ$, seeing Fig. 5(b), $J_{22}$ is also FM superexchange through Cl$^-$ ion. The Cr–O–Cr angle $154^\circ$ along $a$-axis is much

![Fig. 4](image-url)
larger than $130^\circ$, suggesting that $J_3$ should be AFM coupling through $\text{O}^{2-}$ ion, as shown in Fig. 5(c). However, the actual calculation shows that $J_1$ is a strong FM coupling between two Cr$^{3+}$ ions along $a$-axis, which is obviously incompatible with the GKA rules.

Further, we investigated the variations of adjacent exchange coupling constant $J_1$, $J_2$ and $J_3$ with different strains along $a$- and $b$-axis, as shown in Fig. 6(a) and (b). From Fig. 6(a), it is found that the effects of the strain along $a$-axis on exchange coupling $J_1$ and $J_2$ are weak and insensitive to the strain, while the variation of $J_3$ is very obvious throughout the whole considered strain range. The slight variations and positive values of $J_1$ and $J_2$ indicate that the system in the $c$- and $b$-directions always remain in the FM state. The $J_3$ will become negative when the $-13\%$ compression or $12\%$ tensile strain is applied, and reach the maximum under $-8\%$ compressive strain. However, the strain along $b$-axis acts significantly on the exchange coupling $J_3$, but have negligible influence on the positive $J_1$ and $J_3$, as seen from Fig. 6(b).

Then why don’t the variations of exchange coupling $J_2$ and $J_3$ under different strains completely meet the GKA rules? Firstly, there is no doubt when the tensile strain is beyond $12\%$, we note that the variation of $J_3$ between two NN Cr$^{3+}$ ions along $a$-direction is highly consistent with the Bethe–Slater Interaction (BSI) curve. According to the BSI theory, the direct exchange function $J$ can be positive or negative, depending on the ratio of the interatomic spacing $R$ to the orbital radius $r_{3d}$ of 3$d$ shell. As shown in Fig. 6(c), the curves of $J_2$ and $J_3$ vs. $R/r_{3d}$ are also plotted. It follows that the $J_3$ can be ascribable to the direct exchange coupling, obviously, it cannot be explained ideally by the GKA rules and the extended superexchange theory (ESET). Especially, the transition point of FM-AFM coupling for $J_3$ under compression strain is at $R/r_{3d} = 2.9$ in $J_3 - R/r_{3d}$ curve, which is also good in line with the value in BSI rules and the tiny difference is due to the effect of Cr–O–Cr superexchange coupling. This phenomenon has also been revealed in the monolayer CrPS$_4$ AFM-FM transition, which is driven by the change of distance between Cr–Cr.

In detail, the AFM-FM transition in the monolayer CrOCl with strain can be understood by the intuitive physical picture of Fig. 5(d) and (e), where the bond angles of Cr–O–Cr are given in Fig. 6(d) and (f), where the bond angles of Cr–O–Cr are given for the two cases of $-15\%$ and $16\%$ strain. When the $-15\%$ compression strain along $a$-axis is applied in the system, the Cr–O–Cr angle decreases from the nostrained $154^\circ$ to $126^\circ$ due to the decrease of lattice constant $a$, and the distance $R$ of two NN Cr$^{3+}$ ions in $a$ direction decreases correspondingly from $3.942\:\text{Å}$ ($R/r_{3d} = 3.34 > 2.9$) to $3.31\:\text{Å}$ ($R/r_{3d} = 2.8 < 2.9$). Then, according to the BSI curve, the $J_3$ exhibits the direct AFM exchange coupling due to the Pauli exclusion principle. Furthermore, this...
type of direct exchange coupling between the NN cations through the short distance cation–anion–cation path have been reported in bulk TiOCl and TiOBr.47,48 However, under the 16% tensile strain along a-axis, the Cr–O–Cr bond angle increases from 154° to 162°, and the increase of R makes the direct exchange interaction become weak, while the superexchange effect becomes dominant, as a result, the AFM coupling appears according to the GKA rules.

In addition, it is obvious to deduce that the variation of exchange coupling $J_1$ along b-axis may be ascribable to the competition between the superexchange coupling of Cr–Cl/O–Cr and direct exchange coupling of two NN Cr$^{3+}$ ions. When the tensile or small compression strain is applied, the superexchange interactions between two Cr$^{3+}$ ions by Cl$^-$ and O$^{2-}$ ions play the major roles. According to the GKA rules, two superexchange couplings should be FM due to about 90° bond angle of Cr–Cl/O–Cr, thus, the $J_2$ shows FM coupling, as shown in Fig. 6(b). Likewise, this type of the FM superexchange couplings has also been reported to dominate in many 2D materials.42 On the contrary, if a relatively large compression such as −8% strain is applied, the $J_2$ shows AFM coupling, which is due to the small spacing $R$ ($R_{\text{cr}}$ = 2.55) between two NN Cr$^{3+}$ ions under this strain (see Fig. 5(f)). Since the AFM direct exchange coupling overwhelms gradually the two FM superexchange ones with decreasing the spacing $R$, the AFM interaction plays a major role, just like the phenomenon in MnPSe$_3$.23

Then, we return back to the $J_1$, $J_2$ of the unstrained monolayer CrOCl, it is found that even if the $R_{\text{cr}}$ is smaller than 2.9, the $J_1$ and $J_2$ still show the FM coupling, which indicate that although the AFM direct exchange coupling is strong at the $R_{\text{cr}}$ = 2.63 for $J_1$ and 2.77 for $J_2$ according to the BSI curve, as shown in Fig. 5(a) and (b), the combined interactions of two FM superexchange couplings still prevail over the single AFM coupling. For $J_2$ in b-axis direction, we can obtain that the transition point of competition between AFM direct exchange and two FM superexchange couplings by Cr–O–Cr and Cr–Cl–Cr occurs at $R_{\text{cr}}$ = 2.65 from the Fig. 6(c). Moreover, it can be inferred that the transition point of $J_1$ has a smaller value than 2.63. Obviously, the transition point at $R_{\text{cr}}$ = 2.90 along a-axis direction is largest in three different directions, which may be ascribable to the different competitions in different directions. In a-axis direction, the competition exists between a direct exchange and a superexchange coupling, while in the other two directions, the competition exists between a direct exchange and two superexchange couplings.

**Transition temperature**

In the mean-field approximations, the transition temperature $T_C(T_N)$ of monolayer CrOCl can be evaluated by the following relationship:38,49

$$ T \langle S_i^z \rangle z = \frac{2}{3k_B} \sum_{i\neq j} J_{ij} \langle S_i^z \rangle z, \quad (5) $$

where $k_B$ is the Boltzmann constant. We consider the exchange coupling $J_{ij}$ to the third nearest neighbor. With this method, the calculated $T_C$ for unstrained monolayer CrOCl is 133 K, which is dramatically larger than the values of 2D CrI$_3$ (45 K) and Cr$_2$Ge$_2$Te$_6$ (20 K).6,7 and just a little smaller than that (160 K) calculated by Monte Carlo simulations for monolayer CrOCl,26 where only the NN exchange coupling is considered. However, as the same neighboring exchange interactions are considered, it is found the obtained $T_C$ is higher than the values predicted using Ising model, anisotropic Heisenberg model by Metropolis Monte Carlo simulations.12

To understand the effect of strain on the magnetic transition, we plotted $T_C$ of the strained monolayer CrOCl in Fig. 7, where the uniaxial strains ranging from −16% to 16% along a- and b-axis are considered, respectively. It’s found that the $T_C$ can be effectively enhanced from 133 K to 190 K by applying the compressive strain along a-axis or the tensile strain along b-axis. The similar strain effects have also been reported experimentally in many 2D systems, such as CrX$_3$ and CrSiTe$_3$,42,50,51 and the peak value (190 K) is comparable with the previous record in the diluted magnetic semiconductors GaMnAs.52 Moreover, the variation trend of $T_C$ under small uniaxial strains $\varepsilon$ (from −5% to 5%) are perfectly consistent with that of Monte Carlo simulations.26 It’s worth noting that $T_C$ is not always increased under the compressive strain along a-axis direction, and when $\varepsilon$ is about −8%, there is a turning point of rapid decline in the $T_C$ curve, which also indicates that strain engineering is an effective means of improving $T_C$ of FM monolayer CrOCl to a certain extent.

At the same time, it is not difficult to see that the variation of Curie temperatures $T_C$ tuned by uniaxial strains at FM state are highly consistent with those of exchange coupling in corresponding direction of monolayer CrOCl by comparing the data in Fig. 6 and 7, which confirmed directly that the magnitude of $T_C$ is determined by the corresponding exchange coupling interaction ($J_3$ in a-axis direction, and $J_2$ in b-axis direction).

**IV. Conclusion**

In summary, by using first principles calculations, we find that the magnetic transition FM-AFM in the monolayer CrOCl under uniaxial strain can be manipulated by the strain along either
a- or b-axis direction, and the transition temperature $T_C$ can be effectively increased, which indicate it has potential application prospect in the field of 2D spintronics. To clarify the AFM-FM transition and transition temperature, and thus the magnetic exchange coupling, we consider the possible competition between the significant direct exchange and the indirect superexchange exchange coupling, we consider the possible competition between these conclusions will enable us to better explore and apply 2D magnetic materials.

Conflicts of interest

There are no conflicts to declare.

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