# Strong magnetization and Chern insulators in compressed graphene/CrI<sub>3</sub> van der Waals heterostructures

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(Received 20 October 2017; revised manuscript received 4 January 2018; published 1 February 2018)

Graphene-based heterostructures are a promising material system for designing the topologically nontrivial Chern insulating devices. Recently, a two-dimensional monolayer ferromagnetic insulator  $CrI_3$  was successfully synthesized in experiments [B. Huang *et al.*, Nature (London) **546**, 270 (2017)]. Here, these two interesting materials are proposed to build a heterostructure (Gr/CrI<sub>3</sub>). Our first-principles calculations show that the system forms a van der Waals (vdW) heterostructure, which is relatively facilely fabricated in experiments. A Chern insulating state is acquired in the Gr/CrI<sub>3</sub> heterostructure if the vdW gap is compressed to a distance between about 3.3 and 2.4 Å, corresponding to a required external pressure between about 1.4 and 18.3 GPa. Amazingly, very strong magnetization (about 150 meV) is found in graphene, induced by the substrate  $CrI_3$ , despite the vdW interactions between them. A low-energy effective model is employed to understand the mechanism. The work functions, contact types, and band alignments of the Gr/CrI<sub>3</sub> heterostructure system are also studied. Our work demonstrates that the Gr/CrI<sub>3</sub> heterostructure is a promising system to observe the quantum anomalous Hall effect at high temperatures (up to 45 K) in experiments.

DOI: 10.1103/PhysRevB.97.085401

## I. INTRODUCTION

The Chern insulating state, exhibiting a quantum anomalous Hall (QAH) effect, is a novel two-dimensional (2D) topological quantum state that is insulating in the bulk but hosts robust conducting edge states and exhibits quantized Hall conductivity in the absence of external magnetic field [1–4], which was first theoretically proposed by Haldane [4]. The Chern insulating state has attracted considerable attention in condensed-matter physics and materials science since the realization of the QAH effect in realistic material systems may greatly promote the development of low-power-consumption electronics devices. By now, the Chern insulating state has been theoretically predicted to occur in numerous material systems [5-16], such as mercury-based quantum walls [5], transition-metal (TM)-atom-doped topological insulator thin films [6], graphene- and silicene-based systems [7-13], 2D organic topological insulators [14,15], heavy atomic layers on magnetic insulators [16], and so on. However, the laboratory synthesis of these previously predicted QAH effect material systems is very difficult, making the experimental observations of the QAH effect still full of challenges. Until now, the QAH effect has been merely observed in TM-atom-doped  $(Bi, Sb)_2$ Te<sub>3</sub> thin films [17–20] at an extremely low temperature (such as 30 mK [17]), seriously hindering further development of this important research field. Therefore, predicting a QAH material system that can be facilely fabricated in experiments, with a large band gap and a high Curie temperature  $T_c$ , is of great significant.

Graphene is a unique 2D monolayer material with a honeycomb lattice formed by carbon atoms, which has become an ideal prototype material for engineering the OAH effect [7,8]due to its special linear Dirac band dispersions and relatively mature technologies of sample growth and device fabrication. Theoretical studies showed that the Chern insulating state can be realized in graphene by introducing both magnetic exchange field and Rashba spin-orbit coupling (SOC) [7]. Introducing a long-range ferromagnetism (FM) order in the target materials in experiments is one of the most important and toughest tasks in carrying out the Chern insulating state. In addition, the SOC of graphene must be enhanced since the intrinsic SOC in pristine graphene is extremely weak [21,22]. Previous studies indicated that the magnetic exchange field and the enhanced Rashba SOC can be acquired in graphene by depositing low-concentration TM atoms into graphene [7,9]. The TM atoms tend to, however, gather into clusters [23,24] on graphene surface, causing the experimental realization of the Chern insulating state in graphene based on this tactic hard. In graphene, the Chern insulating state was also predicted through deposition onto a suitable magnetic insulator substrate [12,13], which may overcome the problem of the doping tactics. However, the Chern insulating state proposed in graphene deposited on a bulk magnetic insulator substrate depends quite sensitively on both the cleaved surface of the substrate and

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interface configurations [12], which makes the experimental observation somewhat hard. From this point of view, building graphene-based van der Waals (vdW) heterostructures [25,26] may be a very promising tactics to realize experimentally the Chern insulating state in graphene. To date, various 2D magnets have been proposed, such as hole-doped monolayer GaSe [27], a CrI<sub>3</sub>-type monolayer [28,29], Cr<sub>2</sub>Ge(Si)<sub>2</sub>Te<sub>6</sub> [30,31], etc. With current experimental technologies, vdW heterostructures containing different monolayer materials have been successfully fabricated in experiments, such as WSe<sub>2</sub>/CrI<sub>3</sub> [32], graphene/MoS<sub>2</sub> [33], MoSe<sub>2</sub>/WSe<sub>2</sub> [34], and so on.

In this work, we demonstrate that the compressed vdW heterostructure formed by graphene and a monolayer CrI<sub>3</sub>, denoted as Gr/CrI<sub>3</sub>, is a Chern insulator. The bulk crystalline CrI<sub>3</sub> is a layered FM insulator with the adjacent interlayers joined by vdW interactions [35]. The bulk  $CrI_3$  has a relatively high  $T_c$  of 61 K with an out-of-plane easy axis [35]. Recently, the 2D monolayer CrI<sub>3</sub> was synthesized in experiments with its FM and insulating behaviors preserved well, except for a slightly lower  $T_c$  of 45 K [29], compared to that of the bulk. By depositing graphene on the monolayer CrI<sub>3</sub>, we find that the Dirac points of graphene layer are not located inside the bulk band gap of the 2D monolayer CrI<sub>3</sub> substrate, and the Chern insulating state cannot be realized with the equilibrium interface distance. Fortunately, if the distance between graphene and the monolayer CrI<sub>3</sub> is reduced properly by applying external pressure perpendicular to the  $Gr/CrI_3$ heterostructure plane, the Dirac points of graphene can be tuned flexibly into the bulk band gap of CrI<sub>3</sub>, and the Chern insulator can be carried out. To observe the QAH effect in the Gr/CrI<sub>3</sub> heterostructure in experiments, the interface distance must be compressed by applying an external pressure, different from the previously proposed Gr/Cr<sub>2</sub>Ge<sub>2</sub>Te<sub>6</sub> heterostructure [13], in which the QAH effect can be realized at the equilibrium interface distance. With the decrease of the interface distance, the Fermi level  $E_F$  of the Gr/CrI<sub>3</sub> heterostructure moves from the position near the bottom of the conduction bands to the top of the valence bands, indicating the variation from Ohmic contacts to Schottky contacts (*n* type and *p* type) at the Gr/CrI<sub>3</sub> interface. Also, very interestingly, the substrate CrI<sub>3</sub> can induce a large magnetic exchange field (about 150 meV) in the top graphene layer, much stronger than that (about 70 meV) of the heterostructure of graphene on the (111) surface of BiFeO<sub>3</sub> with chemical bonding at the interface [12]. The substrate can also enhance much of the Rashba SOC in graphene, leading to an achieved QAH gap larger than 10 meV. Our findings demonstrate that the compressed vdW heterostructure of Gr/CrI<sub>3</sub> is an appropriate candidate system to experimentally realize the QAH effect at high temperatures (up to 45 K).

## **II. COMPUTATIONAL METHODS**

The geometry and electronic structures of the vdW heterostructures  $Gr/CrI_3$  are calculated by using the projector augmented-wave formalism (PAW) [36] based on densityfunctional theory (DFT), as implemented in the Vienna Ab initio Simulation Package (VASP) [37]. The exchange and correlation functional is described by using the Perdew-Burke-Ernzerhof generalized gradient approximation (GGA-PBE) [38]. To take into account the correlation effects of Cr 3delectrons, the GGA+U method [39] is adopted, and the value of the on-site Coulomb interaction U and exchange interaction J are set to 3.0 and 0.9 eV, respectively. The plane-wave cutoff energy is set to 500 eV, and a vacuum space larger than 15 Å is adopted to avoid any interaction between two adjacent heterostructure slabs. The convergence criterion for the total energy is set to  $10^{-6}$  eV, and Monkhorst-Pack k-point grids of  $6 \times 6 \times 1$  are adopted. All atoms in the unit cell are allowed to move until the Hellmann-Feynman force on each atom is smaller than 0.01 eV/Å. The vdW interaction functional using the method of Grimme (DFT-D2) [40] is employed in the vdW heterostructure calculations. For the monolaver CrI<sub>3</sub>, the hybrid functional HSE06 [41] is employed to obtain its optimized geometry structures and ground-state electronic structures. The band structure of the vdW heterostructure with the equilibrium interface distance is also tested with the hybrid functional HSE06.

#### **III. RESULTS AND DISCUSSION**

## A. Geometry and electronic structures

Bulk CrI<sub>3</sub> is a layered compound with van der Waals gaps between adjacent 2D monolayers which can be cleaved easily and is stable in air [35]. The experimental work shows that  $CrI_3$  is a FM insulator with a relatively high  $T_c$  of 61 K [35]. Recently, the 2D monolayer CrI<sub>3</sub> was successfully synthesized in experiments [29] and was found to be a FM insulator with an out-of-plane easy axis and a  $T_c$  of 45 K. Thus, the FM and insulating behaviors of the bulk CrI<sub>3</sub> are preserved well even though it is cleaved down to the monolayer limit [29]. In the 2D monolayer CrI<sub>3</sub>, Cr ions are in a honeycomb lattice and coordinated by edge-sharing octahedra with six I ions, as shown in Fig. 1(a). The magnetic properties of the monolayer CrI<sub>3</sub> have been systematically studied using the first-principles calculations in Ref. [28]. The authors considered four possible magnetic configurations: (i) FM ordered, (ii) Néel antiferromagnetic (AF) ordered, (iii) AF-stripy ordered, and (iv) AFzigzag ordered [28]. The calculations show that the most stable magnetic state is the FM state with an out-of-plane easy axis [28], consistent with the experimental results [29]. Because the traditional GGA calculation method cannot precisely describe the band gaps of insulators (or semiconductors), we adopt the hybrid functional HSE06 to investigate the geometry and electronic structures of the 2D monolayer FM insulator CrI<sub>3</sub>. The calculations show that the magnetic moment per unit cell of the monolayer  $CrI_3$  is  $6.0\mu_B$ . As shown in Fig. 1(b), the relaxed lattice constant in the plane of the monolayer CrI<sub>3</sub> obtained using the HSE06 functional is a = 7.00 Å, slightly larger than its bulk lattice constant of a = 6.867 Å measured in experiments [35]. The left panel of Fig. 1(c) displays the calculated spin-polarized band structure of the 2D monolayer  $CrI_3$  using HSE06, indicating that the monolayer  $CrI_3$  is a FM insulator with an indirect band gap of 1.93 eV when the SOC is not included. The top of the valence bands and the bottom of the conduction bands are located at the  $\Gamma$  point and K/K' points, respectively. As shown in the right panel of Fig. 1(c) for the densities of states (DOSs) of the system, the top of the valence bands is mainly contributed by the p orbitals of the I atoms. When the SOC is considered, the band gap



FIG. 1. (a) Top and side views of the monolayer  $CrI_3$ . The unit cell is denoted with solid lines. (b) The calculated total energy of the monolayer  $CrI_3$  as a function of the lattice constant using the HSE06 functional. (c) Left: The spin-polarized band structure of the monolayer  $CrI_3$  calculated with HSE06. The red and blue curves denote the spin-up and spin-down bands, respectively. Right: The densities of states of the monolayer  $CrI_3$  with the optimized lattice constant obtained from HSE06. (d) The corresponding band structure with the SOC included. Note that  $E_F$  in (c) and (d) is set in the middle of the band gap of the monolayer  $CrI_3$ .

of the 2D monolayer  $CrI_3$  becomes small and is reduced to 1.57 eV. In addition, the monolayer  $CrI_3$  with SOC included becomes a direct band gap insulator, with the top of the valence bands and the bottom of the conduction bands both located at the  $\Gamma$  point [Fig. 1(d)]. The band gaps obtained from GGA+*U* calculations without and with SOC are 1.19 and 0.82 eV, respectively, as displayed in Fig. S1 of the Supplemental Material [42]; these values are expectedly smaller than the results obtained from the HSE06 functional.

The 2D monolayer FM insulator CrI<sub>3</sub> has a hexagonal structure in which the magnetic Cr ions form a honeycomb lattice, making the monolayer CrI<sub>3</sub> match very well with the graphene layer. In our calculations, the heterostructure of  $Gr/CrI_3$  is built by depositing a 5  $\times$  5 supercell of graphene on a  $\sqrt{3} \times \sqrt{3}$  supercell of the monolayer CrI<sub>3</sub>, as illustrated in Fig. 2(a). The lattice mismatch between the graphene and the CrI<sub>3</sub> substrate in the constructed heterostructure is only about 1.5%. When building the heterostructure, we considered three representative interface configurations between graphene and the CrI<sub>3</sub> substrate: TCr, TI, and Tno. TCr and TI represent two C atoms of the top graphene layer lying directly above two Cr and I atoms of the monolayer CrI<sub>3</sub> in the unit cell of the built heterostructure, as shown in Fig. 2(a) and Fig. S2(a), respectively, while Tno represents a case where none of the C atoms of the graphene layer lie above any of the Cr or I atoms of the  $CrI_3$  in the unit cell, as shown in Fig. S2(d). For the TCr configuration, there is also one Cr atom located at the hollow site of graphene [Fig. 2(a)]. The binding energy  $E_b$  of the built heterostructure is calculated as  $E_b = (E_{CrI_3} + E_b)$  $E_{\text{graphene}} - E_{\text{Total}})/N$ , where  $E_{\text{CrI}_3}$ ,  $E_{\text{graphene}}$ , and  $E_{\text{Total}}$  are the total energies of the bare CrI<sub>3</sub> monolayer, bare graphene, and the  $Gr/CrI_3$  heterostructure, respectively, and N is the number of C atoms in the built heterostructure supercell. The obtained binding energies of the heterostructures with the three types of interface configurations are all found to be  $E_b = 36$  meV, indicating that graphene can be placed on the monolayer CrI<sub>3</sub> substrate without any site selectivity. The average interlayer distances between graphene and the monolayer CrI<sub>3</sub> in the heterostructure obtained from the LDA, GGA, and GGA with the semiempirical vdW correction of DFT-D2 (labeled "GGA+vdW" in the following) calculations are 3.55, 4.27, and 3.53 Å, respectively. The LDA and GGA+vdW give a similar interlayer distance, while the GGA gives a rather large interlayer distance [43]. Thus, the GGA functional does not result in a stable Gr/CrI<sub>3</sub> heterostructure and gives an unreasonable interlayer distance. According to Ref. [43], we can conclude that the interlayer distance between graphene and the monolayer CrI<sub>3</sub> in the heterostructure obtained from the GGA+vdW calculations is reasonable and reliable. The obtained equilibrium interlayer distance  $d_0 = 3.53$  Å, together with the small binding energy of the heterostructure, means that the graphene bonds to the substrate via the weak vdW



FIG. 2. (a) Top and side views of the built Gr/CrI<sub>3</sub> vdW heterostructure. The distance between graphene and CrI<sub>3</sub> in the heterostructure is denoted as vdW gap. (b) The spin-polarized band structure of the Gr/CrI<sub>3</sub> heterostructure with the equilibrium interface distance ( $\Delta d = 0.0 \text{ Å}$ ) using GGA+U calculations. Inset: The corresponding spatial distribution of the spin-polarized electron density of the graphene layer in the heterostructure. Red and green give the net spin-up and spin-down charge densities, respectively. The isosurface value is set to 0.0002  $e/\text{Å}^3$ . (c) The calculated band structure around the Dirac K point of Gr/CrI<sub>3</sub> in (b) using the HSE06 functional. (d)–(f) The same as (b), except that the interface distance between graphene and the monolayer CrI<sub>3</sub> is compressed by (d)  $\Delta d = -0.5 \text{ Å}$ , (e)  $\Delta d = -0.8 \text{ Å}$ , and (f)  $\Delta d = -1.1 \text{ Å}$ . The red and blue curves denote the spin-up and spin-down bands, respectively.

interaction. Therefore, Gr/CrI<sub>3</sub> belongs to the so-called vdW heterostructures [25,26], relatively easy to fabricate in experiments with current advanced sample synthesis technologies. In addition, we further calculated the interlayer distances and binding energies of the built Gr/CrI<sub>3</sub> heterostructure with various kinds of vdW functionals [44,45] (Table I). The vdW-DF gives the largest interlayer distance ( $d_0 = 3.89$  Å) among all of these vdW functionals. The optPBE-vdW and optB88-vdW functionals give almost the same interlayer distances, which are a bit larger than those obtained with the LDA and DFT-D2 (GGA+vdW) calculations. The vdW-DF2 functional also gives a relatively large interlayer distance ( $d_0 = 3.78$  Å). The interlayer distances obtained with optB86b-vdW and DFT-D2 are close to that obtained with the LDA ( $d_0 = 3.55$  Å) approach. Therefore, DFT-D2 and optB86b-vdW are the better choice to calculate the structural parameters (interlayer distance) of the graphene-based layered vdW heterostructures. In the following, the electronic structures and topological properties

TABLE I. The equilibrium interlayer distances  $d_0$  (Å) and binding energies  $E_b$  (meV) of the Gr/CrI<sub>3</sub> heterostructure calculated with various vdW functionals.

|              | Functional |      |         |         |          |      |  |  |
|--------------|------------|------|---------|---------|----------|------|--|--|
|              | DFT-       | vdW- | optPBE- | optB88- | optB86b- | vdW- |  |  |
|              | D2         | DF   | vdW     | vdW     | vdW      | DF2  |  |  |
| $d_0 \\ E_b$ | 3.53       | 3.89 | 3.65    | 3.63    | 3.57     | 3.78 |  |  |
|              | 36         | 37   | 43      | 45      | 45       | 36   |  |  |

of  $Gr/CrI_3$  with the TCr configuration [as shown in Fig. 2(a)] are systematically investigated, while the calculated results of the heterostructures with the TI and Tno configurations are given in the Supplemental Material, as shown in Figs. S2 and S3. All the calculations are conducted with GGA+U methods, except as specifically indicated.

Figure 2(b) plots the calculated spin-polarized band structure of the built  $Gr/CrI_3$  heterostructure [Fig. 2(a)] with the optimized equilibrium vdW gap ( $d_0 = 3.53$  Å). The inset in Fig. 2(b) shows the corresponding spatial distributions of the spin-polarized electron density around the graphene in the heterostructure. Clearly, the graphene is magnetized by the FM substrate CrI<sub>3</sub>. The Dirac points of graphene are, however, found not to be located inside the band gap of the CrI<sub>3</sub> substrate. To confirm the result, we calculated the band structure of the heterostructure around the K point using the hybrid functional HSE06 method. As shown in Fig. 2(c), the Dirac points of graphene indeed greatly mix with the Cr d bands, indicating no QAH effect exists in this pristine vdW heterostructure. To observe the QAH effect in the heterostructure, the Dirac points of the graphene must be tuned to the bulk band gap of the CrI<sub>3</sub> substrate. Fortunately, our following results show that the Dirac points of graphene can, indeed, be tuned effectively by reducing the vdW gap, which can be easily achieved in experiments by applying external pressure. Also, importantly, reducing the vdW gap can enhance dramatically the magnetization and the Rashba SOC in graphene, which is beneficial for observing the Chern insulating behaviors in the system in experiments.

The calculated spin-polarized band structure of the built vdW heterostructure [Fig. 2(a)] with a reduced vdW gap of  $d = d_0 - 0.5$  Å (namely,  $\Delta d = d - d_0 = -0.5$  Å) is plotted in Fig. 2(d). Amazingly, the Dirac points of graphene in this compressed heterostructure are now located exactly inside the band gap of the monolayer FM insulator substrate CrI<sub>3</sub>, which is needed to acquire the Chern insulating state. The bands in Fig. 2(d) also demonstrate a very large magnetic exchange splitting (of about 80 meV) around the Dirac points of graphene induced by the FM CrI<sub>3</sub> substrate [seen more clearly from the magnified bands plotted in Fig. 5(b) below]. Thus, the slightly reduced vdW gap of the Gr/CrI<sub>3</sub> heterostructure can lead to a very distinct spin splitting in the Dirac bands. Because of the weak vdW interaction in the heterostructure interface, the vdW gap should be easily shrunk in experiments by applying an external vertical compressive pressure. The large magnetic exchange splitting gives rise to the crossing of the spin-up and spin-down bands around the Dirac points [at both K and K' points; Fig. 2(d)], which is essential to achieve the Chern insulating state in graphene-based systems [7]. If the vdW gap is further compressed, the influence from the substrate increases accordingly, inducing stronger magnetization in the graphene layer, as illustrated in the insets in Figs. 2(e) and 2(f), and causing larger magnetic exchange splitting in the Dirac bands of the graphene. The exchange splitting becomes 95 and 150 meV when the vdW gap is compressed by 0.8 Å ( $\Delta d = -0.8$  Å) and 1.1 Å ( $\Delta d = -1.1$  Å), respectively, as seen from the bands in Figs. 2(e) and 2(f). The magnetic exchange splitting in graphene induced by the monolayer CrI<sub>3</sub> (e.g., about 80 meV at  $\Delta d = -0.5$  Å) is much larger than that induced by the monolayer Cr2Ge2Te6 (e.g., about 20 meV at  $\Delta d = -0.5$  Å) [13]. The large exchange splitting (up to 150 meV) in graphene induced by the FM substrate in the compressed heterostructure infers that CrI<sub>3</sub> is a very promising FM insulator substrate for many other 2D target materials.

Application of an external vertical pressure to the Gr/CrI<sub>3</sub> heterostructure can be performed by artificially reducing the interlayer distance *d*. The required effective applied pressure *P* at different interlayer distances  $(d = d_0 - |\Delta d|, \text{ with } d_0 = 3.53 \text{ Å})$  is calculated by  $P(d) = \Delta E/(S|\Delta d|)$ , where



FIG. 3. The calculated effective external vertical pressure as a function of the reduced interlayer distance  $\Delta d$ . Insets: Side views of the Gr/CrI<sub>3</sub> vdW heterostructure with the interlayer distance compressed by 0.0, 0.5, and 1.1 Å, respectively.

TABLE II. The first-principles calculations for the work function of the isolated graphene  $W_{\text{Gr}}$  and the electron affinity  $\chi_{\text{CrI}_3}$ , ionization potential  $\eta_{\text{CrI}_3}$ , and energy band gap  $E_g$  of the isolated monolayer  $\text{CrI}_3$ using GGA, GGA+SOC, HSE06, and HSE06+SOC.

|           | Graphene                | CrI <sub>3</sub>    |                              |            |  |
|-----------|-------------------------|---------------------|------------------------------|------------|--|
|           | $W_{\rm Gr}~({\rm eV})$ | $\chi_{CrI_3}$ (eV) | $\eta_{\mathrm{CrI}_3}$ (eV) | $E_g$ (eV) |  |
| GGA       | 4.21                    | 4.68                | 5.87                         | 1.19       |  |
| GGA+SOC   | 4.21                    | 4.70                | 5.52                         | 0.82       |  |
| HSE06     | 4.29                    | 4.50                | 6.43                         | 1.93       |  |
| HSE06+SOC | 4.29                    | 4.48                | 6.05                         | 1.57       |  |

 $\Delta E = E(d) - E(d_0)$  is the energy difference between the compressed [E(d)] and equilibrium  $[E(d_0)]$  heterostructures,  $\Delta d$  is the distance change, and *S* is the area of the interface. The required effective external vertical pressure as a function of the reduced interlayer distance  $\Delta d$  is plotted in Fig. 3. The calculated effective vertical pressures required to reduce the interlayer distance by 0.5 and 1.1 Å are 4.1 and 17.2 GPa, respectively (Fig. 3).

### B. Work functions, contact types, and band alignments

Figure 4(a) is a schematic of the energy levels for the isolated monolayer CrI<sub>3</sub> and graphene, and the work function (the energy difference between the vacuum level  $E_{vac}$  and  $E_F$ ) of graphene  $W_{\rm Gr}$ , the electron affinity (the energy difference between  $E_{\text{vac}}$  and the conduction band minimum  $E_C$ ), and the ionization potential (the energy difference between  $E_{\rm vac}$ and the valence band maximum  $E_V$ ) of the monolayer CrI<sub>3</sub>  $(\chi CrI_3 \text{ and } \eta CrI_3)$  are labeled. The first-principles calculations of  $W_{\rm Gr}$ ,  $\chi {\rm CrI}_3$ , and  $\eta {\rm CrI}_3$  using the GGA and HSE06 methods are summarized in Table II. The calculated work function of graphene is 4.29 eV (HSE06+SOC), consistent with previous reports [46]. When these two materials are put together to form a heterostructure, the electrons will flow from the graphene to CrI<sub>3</sub> because the electron affinity of the isolated monolayer  $CrI_3$  is larger than the work function of graphene ( $\chi CrI_3 >$  $W_{\rm Gr}$ ) [47]. Figure 4(d) presents the calculated electrostatic potential  $V_{\rm eff}$  for the Gr/CrI<sub>3</sub> heterostructure with  $\Delta d =$ 0.0 Å, from which a potential change  $\Delta V$  (~0.5 eV) can be observed. The corresponding differential charge density of the heterostructure is shown in the inset of Fig. 4(d); it is calculated as  $\Delta \rho = \rho(\text{Gr}/\text{CrI}_3) - \rho(\text{graphene}) - \rho(\text{CrI}_3)$ , where  $\rho(Gr/CrI_3)$ ,  $\rho(graphene)$ , and  $\rho(CrI_3)$  are the charge densities of the Gr/CrI<sub>3</sub> heterostructure, graphene, and monolayer CrI<sub>3</sub>, respectively. In the interface, we can observe charge depletion near graphene and accumulation near CrI<sub>3</sub>, inducing an internal electric field. The direction of the induced internal electric field in the interface is from graphene to  $CrI_3$ , as denoted in Fig. 4(d). Generally, Schottky or Ohmic contacts are formed at the metal/semiconductor interfaces. According to the Schottky-Mott model [48], an *n*-type Schottky barrier is defined as the energy difference between the conduction band minimum  $E_C$ and  $E_F$  ( $\Phi_{Cn} = E_C - E_F$ ), and a *p*-type Schottky barrier is defined as the energy difference between  $E_F$  and the valence band maximum  $E_V$  ( $\Phi_{Vp} = E_F - E_V$ ). Obviously, the sum of the *n*-type and *p*-type Schottky barriers is equal to the



FIG. 4. (a) Schematic of the energy levels for the isolated monolayer CrI<sub>3</sub> and graphene.  $W_{Gr}$  represents the work function of graphene.  $\chi$ CrI<sub>3</sub> and  $\eta$ CrI<sub>3</sub> represent the electron affinity and ionization potential of the monolayer CrI<sub>3</sub>, respectively.  $E_{vac}$  represents the vacuum level. (b) and (c) Schematic drawings of the band alignments for the Gr/CrI<sub>3</sub> heterostructures with the equilibrium interface distance and with the compressed interface distance, respectively.  $W_{CrI_3}$  represents the work function of the monolayer CrI<sub>3</sub> in the heterostructure.  $\Delta V$  represents the potential change generated by the interaction between the monolayer CrI<sub>3</sub> and graphene. (d) The electrostatic potential  $V_{eff}$  for the Gr/CrI<sub>3</sub> heterostructure with the equilibrium interface distance ( $\Delta d = 0.0$  Å). Insets: The corresponding potential change  $\Delta V$  (top) and differential charge density of the Gr/CrI<sub>3</sub> heterostructure (bottom). Yellow and cyan represent the charge accumulation and depletion, respectively. The direction of the induced internal electric field is also denoted. (e) The same as (d), except the interface distance is compressed, with  $\Delta d = -0.8$  Å. The isosurface values in (d) and (e) are set to 0.0003 and 0.0008  $e/Å^3$ , respectively. (f)  $\Phi_{Cn}$ ,  $\Phi_{Vp}$ , and  $\Phi_{Cn} + \Phi_{Vp}$  in the Gr/CrI<sub>3</sub> heterostructure as a function of  $\Delta d$ .

band gap  $E_g$  of the semiconductor in the heterostructure. For the Gr/CrI<sub>3</sub> heterostructure with the equilibrium interface distance, an Ohmic contact (*n* type) is formed at the Gr/CrI<sub>3</sub> interface due to  $W_{\rm Gr}$  being close to  $\chi$ CrI<sub>3</sub> (Table II). Based on the above analysis and calculations, the schematic drawing of the band alignment for the Cr/CrI<sub>3</sub> heterostructure with  $\Delta d = 0.0$  Å is plotted in Fig. 4(b).

Figure 4(e) shows the calculated electrostatic potential  $V_{\rm eff}$  for the compressed Gr/CrI<sub>3</sub> heterostructure with  $\Delta d =$ -0.80 Å [also see Fig. 4(c)]. The potential change  $\Delta V$  and the charge redistribution in the heterostructure interface are both enhanced effectively. Figure 4(f) plots the calculated  $\Phi C_n$ ,  $\Phi V_p$ , and  $\Phi C_n + \Phi V_p$  in the Gr/CrI<sub>3</sub> heterostructure as a function of  $\Delta d$ . With the decrease in the vdW gap, the heterostructure  $E_F$  moves from  $E_C$  to  $E_V$ . Thus, the Ohmic contact at the Gr/CrI<sub>3</sub> interface with the equilibrium distance is tuned first to the *n*-type Schottky contact and then to the *p*-type Schottky contact (with  $|\Delta d| > 0.8$  Å) by reducing the interface distance. In addition, the band gap of the monolayer CrI<sub>3</sub> in the heterostructure becomes smaller with the decrease of the vdW gap due to the stronger interaction with graphene. Therefore,  $E_F$  of the heterostructure and the Dirac points of graphene can be tuned conveniently to the insulating band gap of the CrI<sub>3</sub> substrate by reducing the interface distance.

## C. Topological properties

When the SOC interaction is included in the compressed vdW heterostructure Gr/CrI<sub>3</sub>, an energy gap is opened at the crossing points of the spin-up and spin-down Dirac bands, and  $E_F$  is located just inside the SOC-induced gap, as illustrated in Figs. 5(a) and 5(e). The gap is opened by the enhanced Rashba SOC that arose in the top graphene layer due to the existence of the  $CrI_3$  substrate. Figures 5(c) and 5(d) plot the magnified bands around the K and K' points of the compressed heterostructure when the vdW gap is reduced by 0.5 Å. We find that the SOC-induced local band gaps around the K(5.1 meV) and K' (2.2 meV) points are not equal because of the nonequivalent A and B sublattices of graphene, induced by the CrI<sub>3</sub> substrate. Therefore, a global band gap of about 2.2 meV is opened in this system. If the substrate effect is further enhanced by reducing the vdW gap, the strength of the Rashba SOC will be enlarged accordingly. Figures 5(g)and 5(h) show that if the vdW gap is compressed by 1.1 Å, the enhanced SOC-induced local band gaps around the K and K' points will be enlarged to 12.2 and 25.0 meV, respectively. Therefore, a global SOC-induced band gap larger than 10 meV can be achieved in the compressed vdW heterostructure, which is much larger than the nontrivial gap of pure graphene [21, 22]. The magnitude of this achieved band gap corresponds to a



FIG. 5. (a) The band structure of the Gr/CrI<sub>3</sub> heterostructure with  $\Delta d = -0.50$  Å. The SOC is involved. (b) The spin-polarized bands around the *K* point with  $\Delta d = -0.50$  Å without the SOC. The red and blue curves denote the spin-up and spin-down bands, respectively. (c) and (d) The corresponding SOC bands around the *K* and *K'* points, respectively. (c) is also the close-up of (a) around the Dirac point. (e)–(h) The same as (a)–(d), except  $\Delta d = -1.10$  Å.

temperature higher than 100 K, which is easily accessible under current experimental technologies.

We now adopt the low-energy effective model of graphene around the Dirac points to investigate the mechanism of the gap opening and the topological properties of the compressed vdW heterostructure  $Gr/CrI_3$ . The low-energy continuum model of graphene, including the magnetic exchange field, the Rashba SOC, and the staggered AB-sublattice potential, can be written as [12]

$$H(\mathbf{k}) = -\nu_f (\eta \sigma_x k_x + \sigma_y k_y) \mathbf{I}_s - M \mathbf{I}_\sigma s_z + \frac{\lambda_R}{2} (\eta \sigma_x s_y - \sigma_y s_x) + U \sigma_z \mathbf{I}_s, \qquad (1)$$

where  $v_f = 3t/2$  is the Fermi velocity,  $\eta = \pm 1$  for K and K', respectively, and  $\sigma$  and s are Pauli matrices that act on the sublattices and the spin degrees of freedom. In Eq. (1), the first term represents the nearest-neighbor hopping of graphene with amplitude t = 2.6 eV, and the last three terms describe the magnetic exchange field, the Rashba SOC, and the staggered sublattice potential, respectively. For pristine graphene, perfect linear Dirac bands appear at the K and K'points, respectively. If the staggered potential U is considered, two trivial band gaps are opened around the Dirac points. When a relatively large magnetic exchange field M is included, the spin-up and spin-down Dirac bands cross each other around the K and K' points, and the trivial gaps disappear, as displayed in Figs. 6(a) and 6(c) with U = 2.6 meV and M = 65 meV. When the Rashba SOC  $\lambda_R$  is also included, two band gaps will open at the band crossing points around the Dirac points, as shown in Figs. 6(b) and 6(d), with U = 2.6 meV, M = 65 meV, and  $\lambda_R = 20.8$  meV. Due to the existence of staggered potential,

the SOC-induced local band gaps around Dirac points are not equal. Thus, the physical mechanism of the band evolution around the Dirac points of the compressed heterostructure  $Gr/CrI_3$  can be explained well by this low-energy continuum model of graphene.

The Berry curvatures and Chern number are calculated to identify the topological properties of the above SOC-induced insulating states. The Berry curvature is calculated as [49,50]

$$\Omega(\mathbf{k}) = \sum_{n} f_{n} \Omega_{n}(\mathbf{k}),$$

$$\Omega_{n}(\mathbf{k}) = -2 \operatorname{Im} \sum_{m \neq n} \frac{\hbar^{2} \langle \psi_{n\mathbf{k}} | v_{x} | \psi_{m\mathbf{k}} \rangle \langle \psi_{m\mathbf{k}} | v_{y} | \psi_{n\mathbf{k}} \rangle}{(E_{m} - E_{n})^{2}}, \quad (2)$$

where the summation is over all of the occupied states,  $f_n$  is the Fermi-Dirac distribution function,  $E_n$  is the eigenvalue of the Bloch functions  $|\psi_{n\mathbf{k}}\rangle$ , and  $\upsilon_{x(y)}$  are the velocity operators. The Chern number C can be obtained by integrating the Berry curvatures over the first Brillouin Zone (BZ), as C = $\frac{1}{2\pi}\sum_{n}\int_{BZ} d^{2}k\Omega_{n}$ . The calculated  $\Omega(\mathbf{k})$  around the K and K' points from the low-energy continuum model of graphene are plotted in Figs. 6(b) and 6(d), respectively. The peaks of  $\Omega(\mathbf{k})$ distribute around the SOC-induced band gaps, and they have the same sign near the K and K' points. By integrating the Berry curvatures around the K and K' points in the first BZ, the nonzero Chern number of  $C_K = C_{K'} = 1$  can be obtained, leading to the total Chern number of  $C = C_K + C_{K'} = 2$ . The obtained nonzero integer Chern number indicates that this SOC-induced insulating state is topologically nontrivial and the QAH effect can be realized in this compressed Gr/CrI<sub>3</sub> vdW heterostructure. Figure 6(e) plots the calculated edge



FIG. 6. (a) The spin-polarized band structure around the K point obtained from the low-energy continuum model calculations. The red and blue curves denote the spin-up and spin-down bands, respectively. (b) The corresponding band structure (black curves) around the K point with the SOC included. The red dots give the calculated Berry curvatures for the whole valence bands. (c) and (d) The same as (a) and (b), except around the K' point. (e) The calculated edge density of states of the semi-infinite armchair-edged graphene system. (f) A schematic diagram depicting the observation of the QAH effect in the vdW heterostructure of  $Gr/CrI_3$ . The vertical red arrow denotes the external compression. The small horizontal yellow arrows indicate the two dissipationless edge current channels owned in the heterostructure.

density of states of the semi-infinite armchair-edged graphene system, from which we can observe two distinct chiral edge states in the bulk band gap, consistent with the above-obtained total Chern number C = 2. Figure 6(f) is a schematic diagram depicting the experimental observation of the QAH effect in the built vdW heterostructure of graphene on the monolayer CrI<sub>3</sub> substrate.

In consideration of  $T_c$  of the monolayer CrI<sub>3</sub> substrate (45 K) and the opened Chern insulator gap in the compressed Gr/CrI<sub>3</sub> heterostructure (100 K), we can make the statement that the QAH effect can be observed in this heterostructure at a temperature up to 45 K, which is much higher than that in the magnetic topological insulator thin films (30 mK) [17–20]. The achieved Chern insulating state in the constructed vdW heterostructures also shows strong robustness, as revealed by the band structures of the heterostructures with two other interface configurations, as illustrated in Figs. S2 and S3. Both of the systems also exhibit the Chern insulating state around the Dirac points.

## **IV. CONCLUSIONS**

We systematically investigated the electronic structures and topological properties of the vdW heterostructure of graphene on the 2D monolayer FM insulator  $CrI_3$  from the first-principles calculations and low-energy effective model. The FM  $CrI_3$  substrate induces a very large magnetic exchange field in the graphene layer, which is essential for producing the Chern insulating state, which can be much enhanced by applying vertical external pressure to the heterostructure plane. An Ohmic contact is formed at the heterostructure interface since the work function of graphene is smaller than the electron affinity of the isolated monolayer CrI<sub>3</sub> substrate. With the decrease of the interface distance, the interface varies from an Ohmic contact to an *n*-type Schottky contact (with  $|\Delta d| > 0.2$  Å) and then to a *p*-type Schottky contact (with  $|\Delta d| > 0.8$  Å). Very importantly, the Dirac points of graphene are tuned into the insulating gap of the CrI<sub>3</sub> substrate with a compressive stress to a certain extent, which together with the substrate-enhanced Rashba SOC in graphene leads to a global sizable nontrivial band gap (>10 meV) opened in the heterostructure system. The calculated Berry curvature, Chern number, and edge states indicate that a Chern insulating state can be achieved in the system that is not sensitive to the interface stacking patterns. The experimentally observed temperature of the Chern insulating state in the heterostructure is expected to be up to 45 K, much higher than that in magnetic topological insulator thin films (30 mK). Our findings may greatly push the experimental observations of the QAH effect in graphene-based systems at high temperatures.

## ACKNOWLEDGMENTS

This work was supported by the Natural Science Foundation of Jiangsu Province (China) with Grant No. BK20170376, the Natural Science Foundation of Higher Education Institutions of Jiangsu Province (China) with Grants No. 17KJB140023 and No. 17KJA140001, and the National Natural Science Foundation of China under Grants No. 11574051 and No. 11604134.

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