

# Strain effects on the magnon-magnon interaction and magnon relaxation time in ferromagnetic CrGeTe<sub>3</sub>

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In the synthesis of two-dimensional (2D) materials and the fabrication of 2D devices, the strain introduced by lattice mismatch is inevitable. Meanwhile, strain is also a frequently used strategy to improve the Curie temperature of 2D magnets. However, the impact of strain on the magnon relaxation time is unclear. In this work, we investigated the impacts of strain on the magnon-magnon interaction and magnon relaxation time in the ferromagnetic CrGeTe<sub>3</sub> monolayer by combining first-principles calculations and theoretical analysis. We find the magnon relaxation times in strained and unstrained CrGeTe<sub>3</sub> monolayers have similar dependences on temperature, wave vectors, and magnetic fields. However, the magnon relaxation time is remarkably reduced by tensile strain. When the external magnetic field is 0.1 T, the maximum shortened ratio of the magnon relaxation time can reach 49.3%, revealing an enhancement of the magnon-magnon interaction. With the magnetic field strengthening, the shortened ratio of the magnon relaxation time decreases, but it still reaches up to 19.4% under a 5-T magnetic field. This shortening of magnon relaxation time is undesired for applications in spintronics. Our work provides physical insights into the impacts of strain on 2D magnets.

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## I. INTRODUCTION

In the past two decades, two-dimensional (2D) materials have been widely applied in various fields, including electronics [1,2], energy [3,4], and biomedicine [5,6]. The 2D magnet is an important member of the family of 2D materials. Compared to nonmagnetic 2D materials, 2D magnets can be applied in data storage [7,8], signal transfer [9,10], and spincaloritronics [11,12], due to their controllable magnetoresistance [13], large spin Seebeck coefficients [14], and the quantized anomalous Hall effect [15]. In magnetic materials, the quantization of the spin wave is defined as a magnon. In signal transfer using spin as a carrier, the transmission distance is strongly related to the magnon lifetime, and a long magnon lifetime is preferred [16,17]. In 2D magnets, the magnon lifetime is determined by the magnon-phonon, magnon-magnon, and magnon-carrier interactions. In recent years, Wang *et al.* studied the magnon lifetime governed by the magnon-phonon interaction in 2D magnets [18,19]. It was found that the acoustic and optical magnon lifetimes governed by the magnon-phonon interaction showed linear and quadratic dependences on the wave vector, respectively, and the magnon lifetime governed by the magnon-phonon

interaction depended heavily on the temperature. In 2017, few-layer ferromagnetic CrI<sub>3</sub> [20] and CrGeTe<sub>3</sub> [21] were synthesized successfully by mechanical exfoliation. Subsequently, various 2D magnets were unveiled and studied, such as ferromagnetic Fe<sub>5</sub>GeTe<sub>2</sub> [22], antiferromagnetic MnPS<sub>3</sub> [23], conductive Fe<sub>3</sub>GeTe<sub>2</sub> [24], room-temperature magnetic CrTe<sub>2</sub> [25], and MnBi<sub>2</sub>Te<sub>4</sub> with the quantized anomalous Hall effect [26,27].

In practical applications, the spintronics device performance is strongly dependent on the quality of 2D magnetic materials. Thus, the controllable synthesis of 2D magnetic materials with high quality is essential for their large-scale applications. Mechanical exfoliation and chemical vapor deposition are two frequently used synthesis methods for 2D magnetic materials [28,29]. Furthermore, the synthesized 2D magnets need to be transferred to the SiO<sub>2</sub>/Si substrate for the detection of magnetic orders and the measurement of physical properties in experiments. During both the mechanical exfoliation and transfer processes, it is inevitable that strain is introduced into 2D magnets, due to the lattice mismatch between 2D magnets and the substrate. Meanwhile, strain is also an effective and frequently used strategy to tailor the physical and chemical properties of 2D materials. Thus, several researchers have investigated the influences of strain on the electronic, phononic, and thermal properties of 2D magnetic materials [30–34], which can provide theoretical guidance and support for

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the manufacturing of nanosized spintronic devices. For instance, Liu *et al.* [35] improved the strength of magnetic exchange coupling and the Curie temperature of CrGeTe<sub>3</sub> significantly by the biaxial in-plane strain, while Webster and Yan [36] manipulated the magnetic phase between ferromagnetism and antiferromagnetism, as well as the out-of-plane magnetic anisotropy energy of the CrI<sub>3</sub> monolayer by in-plane biaxial strain. However, the impact of strain on the magnon lifetime is still unclear, which is adverse for the applications of 2D magnets in information technology.

In this paper, we took the CrGeTe<sub>3</sub> monolayer as an example to explore the effect of strain on the magnon lifetime dominated by the magnon-magnon interaction. Because the CrGeTe<sub>3</sub> monolayer is a magnetic semiconductor with an indirect band gap, it has an ultralow carrier concentration near the Fermi level and a weak magnon-carrier interaction. Additionally, the appearance of out-of-plane magnetic anisotropy in 2D magnets increases the difficulty of theoretical studies and analysis for magnon lifetimes. In the CrI<sub>3</sub> monolayer, the magnetic anisotropy is remarkable and up to about 29.3% of the nearest-neighbor magnetic exchange constant [37]. Unlike CrI<sub>3</sub>, the magnetic anisotropy in the CrGeTe<sub>3</sub> monolayer is only 2% of the magnetic exchange constant and can be ignored [38,39], which simplifies the calculation of magnon lifetime and has little effect on the Curie temperature of the CrGeTe<sub>3</sub> monolayer [40–42]. Here, we combined first-principles calculations and theoretical analysis to obtain the magnon lifetime governed by the magnon-magnon interaction in strained and unstrained CrGeTe<sub>3</sub> monolayers, for a comprehensive insight into the effect of strain on magnon-magnon interactions and the magnon relaxation time. We find that, although strain has little influence on the dependence of the magnon relaxation time with temperature, wave vector, and external magnetic field, strain can reduce the magnon relaxation time significantly. These findings are of significance for designing 2D electromechanical devices, spintronics devices, and nanosized strain sensors.

## II. METHODS

### A. Computational details

In this work, the Vienna *ab initio* simulation package [43,44] was used to implement all first-principles calculations with the exchange-correlation functional of the Perdew-Burke-Ernzerhof method of the generalized gradient approximation [45,46]. Meanwhile, the GGA + *U* approach introduced by Dudarev *et al.* [47] was used to describe the *d*-orbital electrons in Cr<sup>3+</sup> ions, and the effective Hubbard “ $U_{\text{eff}} = U - J$ ” was set as 1 eV, as used previously in the literature [38,48]. We select this value because Gong *et al.* [21] and Kang *et al.* [49] reported that the value of “ $U_{\text{eff}}$ ” for the Cr<sup>3+</sup> ions in CrGeTe<sub>3</sub> should be between

0.2 and 1.7 eV; otherwise, the calculated band gap and magnetic ground state may be contrary to the experimental results. In establishing the 2D model of the CrGeTe<sub>3</sub> monolayer, we imposed a 20-Å vacuum space along the *c* axis to weaken the possible interaction between adjacent layers. In the optimization of the geometrical structure, the CrGeTe<sub>3</sub> monolayer was relaxed with the 500-eV cut-off energy and the  $5 \times 5 \times 1$  [(MP) Monkhorst-Pack] grid until the energy and atom force converged to  $10^{-8}$  eV and 0.001 eV/Å, respectively. In self-consistent calculations, a denser MP grid of  $9 \times 9 \times 1$  was set to calculate the energy difference between possible magnetic configurations. Monte Carlo simulations based on the Heisenberg model were implemented by the MCSOLVER code to obtain the Curie temperature of the CrGeTe<sub>3</sub> monolayer with and without strain [50,51], which was commonly used in previous studies [52–54]. Besides, the density-functional perturbation theory was used to calculate phonon dispersions [55], as implemented by the PHONOPY code.

### B. Magnon dispersion and spin autocorrelation function at finite temperature

Before discussing magnon dispersion and magnon-magnon interactions, it is necessary to determine the magnetic ground state of 2D magnets. The diagram of four possible magnetic configurations is presented in Fig. S1 within the Supplemental Material [56]. For the CrGeTe<sub>3</sub> monolayer, the total energies of Néel-, stripy-, and zigzag-antiferromagnetic (AFM) configurations are 93.22, 54.54, and 31.24 meV per unit cell higher than the ferromagnetic (FM) phase, respectively. Therefore, the magnetic ground state of the CrGeTe<sub>3</sub> monolayer is ferromagnetic and can be identified by the spin-density distribution in Fig. S1(b) within the Supplemental Material [56]. Meanwhile, these energy difference between Néel-, stripy-, and zigzag-AFM and FM states can also be used to calculate the magnetic exchange constants [57,58]. Due to the small second-nearest- and third-nearest-neighbor magnetic exchange constants in the CrGeTe<sub>3</sub> monolayer, we only consider the contribution of nearest-neighbor magnetic exchange coupling ( $J_1$ ) in this paper. The value of  $J_1$  can be obtained from Eq. (S7) within the Supplemental Material [56], which is strongly related to the distance between two magnetic sites [59,60]. Therefore, strain can tailor the value of  $J_1$  effectively.

After obtaining the magnetic exchange constant, the magnon dispersion can be calculated by diagonalizing the Hamiltonian. In 2020, Wang *et al.* [61] calculated the magnon dispersion at finite temperature by

$$\hbar\omega_k^\pm(T) = 3J_1S_0(1 \pm \gamma_k) - 3J_1S_0(1 \pm \gamma_k) \times \frac{1}{NS_0} \sum_{k'} (1 - \gamma_{k'}) \langle a_{k'}^+ a_{k'} \rangle_T + 2g\mu_B B, \quad (1)$$

with  $\gamma_k = 1/3 \sum_{i,j \in N} e^{ik(r_j - r_i)}$ .  $\omega^+$  and  $\omega^-$  represent the magnon frequencies for optical and acoustic magnon branches, respectively. Details of Eq. (1) can be found in Part 2 and Part 3 of Ref. [56]. In the derivation, the Holstein-Primakoff approximation [62,63] and Wick theory [64] were used. Furthermore, the contribution from the temperature-dependent magnon operators, dominated by the “dynamical magnon-magnon interaction,” was considered, due to the failure of the low-temperature approximation at nonzero temperature [65,66]. This contribution can be gained by a self-consistent procedure with a convergence limit of  $10^{-5}$  eV. The first term is the static magnon energy without temperature dependence, while the last term is the Zeeman term induced by an external magnetic field. In this work, we also use this method to calculate the magnon dispersions at finite temperatures for pristine and strained CrGeTe<sub>3</sub> monolayers. It is worth emphasizing that the magnetic exchange constant,  $J_1$ , of the CrGeTe<sub>3</sub> monolayer is insensitive to the increase of temperature [18]. Thus, in this work, we neglect the temperature dependence of  $J_1$  and only consider the magnon redistribution induced by finite temperature to calculate the magnon dispersion.

### III. RESULTS AND DISCUSSION

#### A. Stability of pristine and strained CrGeTe<sub>3</sub> monolayers

The lattice parameters of the optimized CrGeTe<sub>3</sub> monolayer are  $a = b = 6.91$  Å, which are slightly larger than the experimental values ( $a = b = 6.83$  Å) but consistent with previous theoretical studies [38,67,68]. The top and side views of the optimized CrGeTe<sub>3</sub> monolayer are presented in Fig. 1(a), where each Cr and Ge atom is surrounded by six Te atoms. The high-symmetry path in the irreducible Brillouin zone of the CrGeTe<sub>3</sub> monolayer is shown in Fig. 1(b), which is used in plotting the phonon and magnon dispersions. Here, the phonon dispersion was calculated

to identify the thermodynamic stability of our optimized CrGeTe<sub>3</sub> monolayer, and the calculated phonon dispersion is presented in Fig. S2(a) within the Supplemental Material [56]. There is no obvious imaginary frequency in the phonon dispersion, indicating the thermodynamic stability of the optimized CrGeTe<sub>3</sub> monolayer. Besides, we also calculated the stiffness tensor of the optimized CrGeTe<sub>3</sub> monolayer, as shown in Table S1 within the Supplemental Material [56]. In Table S1 within the Supplemental Material [56], we find that the values of C<sub>11</sub>, C<sub>12</sub>, and C<sub>66</sub> are 56.098, 56.098, and 22.425 N/m, respectively, and all of them satisfy the Born-Huang stability criteria [69], revealing the mechanical stability of the optimized CrGeTe<sub>3</sub> monolayer. The stress-strain relationship of the optimized CrGeTe<sub>3</sub> monolayer was calculated by using the strain-energy method [70], as presented in Fig. S3 within the Supplemental Material [56]. In Fig. S3 within the Supplemental Material [56], it is found that the maximum stress and strain that the optimized CrGeTe<sub>3</sub> monolayer can withstand are 20.6 GPa and 21%, respectively. In this paper, the maximum biaxial strain applied to the CrGeTe<sub>3</sub> monolayer is 10%. In 2020, Yin *et al.* [71] disclosed the impressive impact of strain on the phononic properties and thermal conductivity of graphene-like borophene, which may destroy the stability of 2D materials. Here, we also calculated the phonon dispersion of the 10% strained CrGeTe<sub>3</sub> monolayer, as shown in Fig. S2(b) within the Supplemental Material [56]. It is noted that, although strain decreases the highest phonon frequency significantly, there is still no obvious imaginary frequency, indicating the thermodynamics of the 10% strained CrGeTe<sub>3</sub> monolayer.

#### B. Magnon dispersion of pristine and strained CrGeTe<sub>3</sub>

After structural optimization, we implemented self-consistent calculations of the FM and AFM configurations to obtain the magnetic exchange constants of pristine and

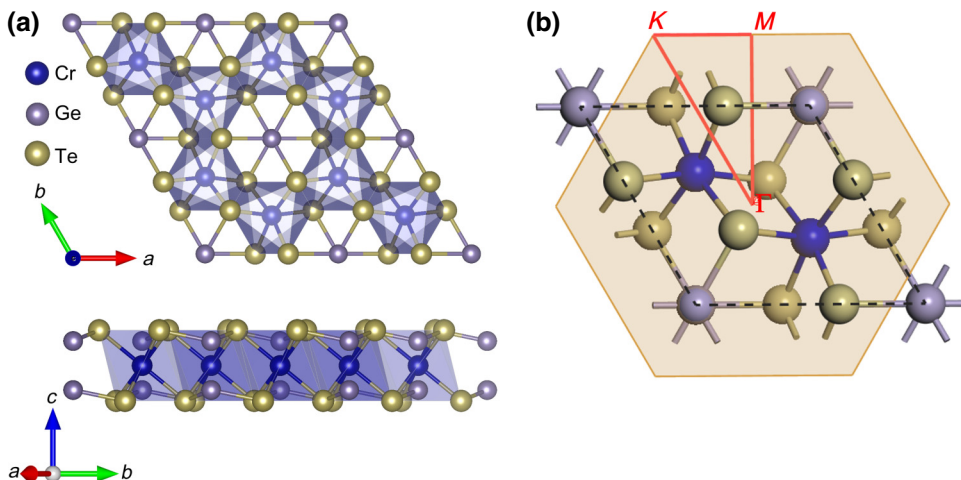


FIG. 1. Geometrical structure (a) and high-symmetry path in the irreducible Brillouin zone (b) of the CrGeTe<sub>3</sub> monolayer.

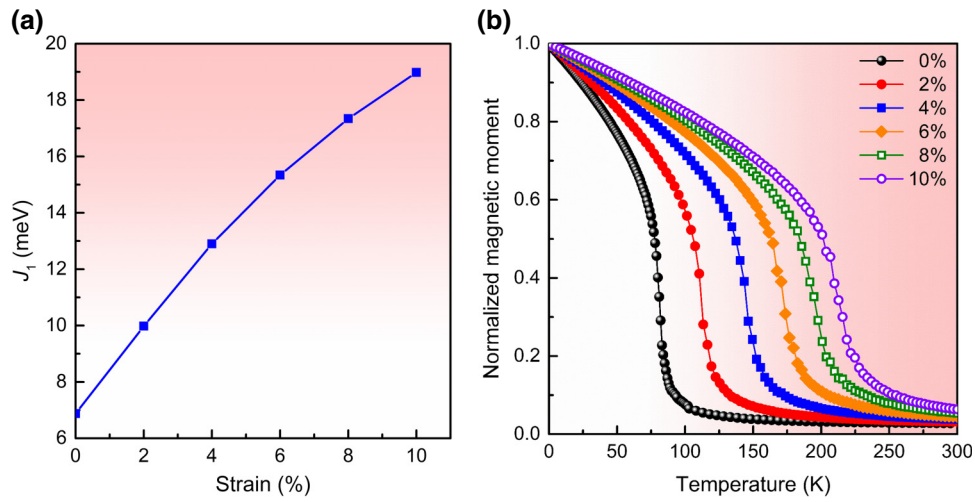


FIG. 2. Magnetic exchange constant (a) and Curie temperature (b) of pristine and strained CrGeTe<sub>3</sub> monolayers.

strained CrGeTe<sub>3</sub> monolayers from Eq. (7). The results are shown in Fig. 2(a). It can be found that the magnetic exchange constant increases with biaxial tensile strain monotonously, and the enhancement can be up to about 176%, which is in agreement with the enhancement of 191% reported in Ref. [35]. Based on these magnetic exchange constants and the Heisenberg model, we employed Monte Carlo simulations and calculated the temperature dependence of the magnetic moment, as shown in Fig. 2(b). The Curie temperature is elevated from 85 to 215 K, corresponding to an enhancement ratio of about 153%. These results suggest that tensile strain is an effective strategy to improve the low Curie temperature of 2D magnets and promote their application in spintronics. In the CrGeTe<sub>3</sub> monolayer, the strength of magnetic exchange coupling is dominated by the competition between AFM direct- and FM superexchange couplings [72,73]. The AFM direct-exchange coupling depends heavily on the distance between two magnetic Cr<sup>3+</sup> ions in the CrGeTe<sub>3</sub> monolayer. According to the Goodenough-Kanamori rule [74,75], the superexchange coupling depends strongly on the Cr-Te-Cr angle. Once tensile strain is applied, the distance between two magnetic atoms increases significantly from 3.94 Å (pristine) to 4.34 Å (10% strain), suppressing the AFM direct-exchange coupling. Meantime, the Cr-Te-Cr angle changes slightly. Hence, the difference between FM super- and AFM direct-exchange couplings increases with tensile strain, and the magnetic exchange constant is enhanced by strain remarkably.

Subsequently, the obtained magnetic exchange constants were substituted into Eq. (12), and then the magnon dispersions, considering the contribution from dynamic magnon-magnon interactions, were calculated, as shown in Fig. 3. In Eq. (12), there are two factors affecting the

magnon frequency: the temperature ( $T$ ) and the external magnetic field ( $B$ ). In Figs. 3(a) and 3(b), we plot the magnon dispersions of pristine CrGeTe<sub>3</sub> monolayers at finite temperatures and different external magnetic fields, where the insets are the enlarged dispersion curves near the center point ( $\Gamma$  point) of the irreducible Brillouin zone. Interestingly, the external magnetic field can open a gap between the acoustic branch and zero frequency at the  $\Gamma$  point, and the gap becomes large as the external magnetic field increases, as shown in the inset of Fig. 3(b) (in the purple rectangle). Meantime, we find that the magnon frequency also increases with the external magnetic field but decreases with temperature, as shown in the insets of Figs. 3(a) and 3(b) (in the green rectangle). To highlight the temperature dependence of magnon frequency, we analyze the changes to acoustic and optical magnon frequencies with temperature at several wave vectors, as shown in Fig. S4 within the Supplemental Material [56]. Obviously, all magnon frequencies reduce with temperature monotonously, and the reduction of the optical magnon is more remarkable than that of the acoustic magnon. These results reveal that the contribution of dynamic magnon-magnon interactions is to decrease the magnon frequency, and this reduction is positively correlated with the static magnon energy. This finding is consistent with the results and physical model reported in Ref. [76].

In Fig. 3(c), the magnon dispersions of the strained CrGeTe<sub>3</sub> monolayer are presented. It is clear that the frequency of the optical magnon increases with tensile strain significantly; this is caused by the enhancement of the magnetic exchange constant. To observe the influence of strain on the magnon frequency quantitatively, we plot the temperature-dependent magnon frequencies of acoustic and optical magnons of the strained CrGeTe<sub>3</sub> monolayer in Fig. 4. For the 4% strained CrGeTe<sub>3</sub> monolayer,

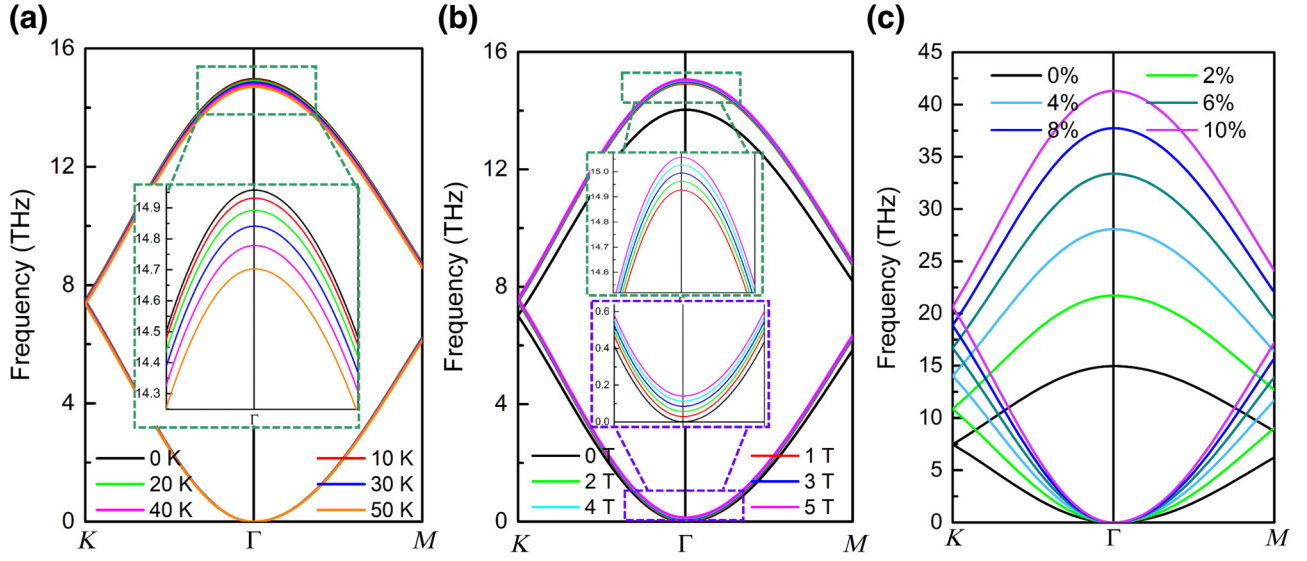


FIG. 3. Magnon dispersions of the pristine CrGeTe<sub>3</sub> monolayer at different temperatures (a) and external magnetic fields (b), and magnon dispersion of the strained CrGeTe<sub>3</sub> monolayer (c). Insets in (a),(b) are enlarged dispersion curves near the center point of the irreducible Brillouin zone.

all frequencies of the acoustic and optical magnons at wave vectors ( $k$ ) of  $9.0 \times 10^5 \text{ cm}^{-1}$  ( $0.02\pi/a$ ),  $4.5 \times 10^6 \text{ cm}^{-1}$  ( $0.10\pi/a$ ),  $1.35 \times 10^7 \text{ cm}^{-1}$  ( $0.30\pi/a$ ),  $2.25 \times 10^7 \text{ cm}^{-1}$  ( $0.50\pi/a$ ),  $3.15 \times 10^7 \text{ cm}^{-1}$  ( $0.70\pi/a$ ), and  $4.5 \times 10^7 \text{ cm}^{-1}$  ( $1.0\pi/a$ ) decrease with temperature, as shown in Figs. 4(a) and 4(b). Meanwhile, the decrease relies on the static magnon energy, which agrees well with that in the pristine CrGeTe<sub>3</sub> monolayer. However, for strained CrGeTe<sub>3</sub> monolayers, the decrease in magnon frequency induced by finite temperature is smaller than that of the pristine CrGeTe<sub>3</sub> monolayer, as shown in Fig. 4(c). The reason is that tensile strain enhances the strength of magnetic exchange coupling and elevates the magnon

frequency significantly. Therefore, the magnons become difficult to excite.

### C. Magnon relaxation time of pristine and strained CrGeTe<sub>3</sub>

To evaluate the strength of the dynamic magnon-magnon interaction and the effects of strain on the magnon-magnon interaction, the spin autocorrelation functions [ $\langle S_k^+(t)S_{-k}^-(0) \rangle$ ,  $\langle S_k^-(t)S_{-k}^+(0) \rangle$ , and  $\langle S_k^z(t)S_{-k}^z(0) \rangle$ ] were employed. In this paper, the external magnetic field is applied along the  $z$  axis, so  $\langle S_k^z(t)S_{-k}^z(0) \rangle$  is particularly emphasized:

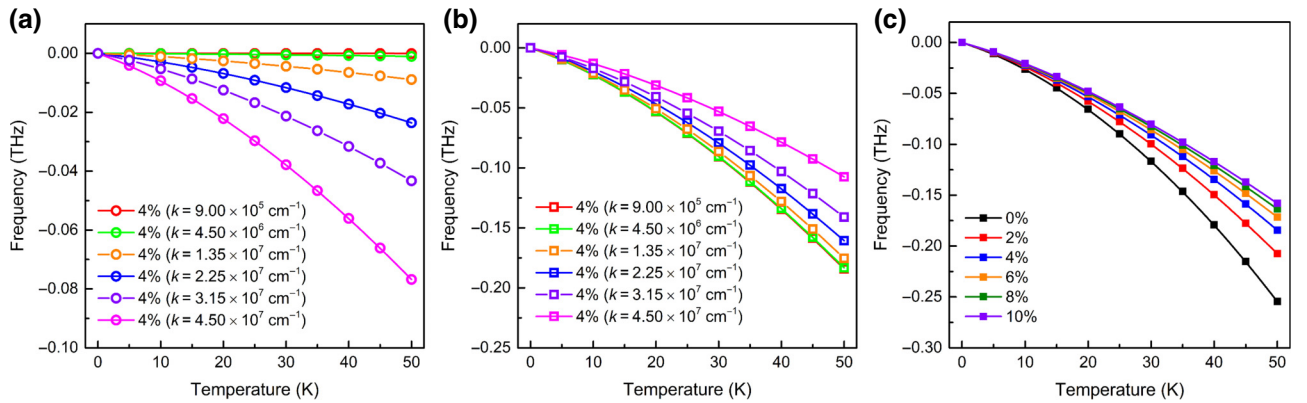


FIG. 4. Sampled contribution of dynamic magnon-magnon coupling for the acoustic (a) and optical (b) magnon frequencies of 4% strained CrGeTe<sub>3</sub> monolayer, and (c) temperature dependence of optical magnon frequencies at the  $\Gamma$  point of strained CrGeTe<sub>3</sub> monolayers.

$$\begin{aligned} \langle S_k^z(t) S_{-k}^z(0) \rangle &= \frac{\text{Tr} \left\{ e^{-\theta H} \left( (N^{1/2} S_0 \delta_{k,0} - N^{-1/2} \sum_{k'} a_{k'}^+(t) a_{k'+k}(t)) (N^{1/2} S_0 \delta_{-k,0} - N^{-1/2} \sum_{k''} a_{k''}^+(0) a_{k''-k}(0)) \right) \right\}}{\text{Tr} \{ e^{-\theta H} \}}, \\ &= N S_0^2 \delta_{k,0} - 2 S_0 \delta_{k,0} \sum_{k'} \langle a_{k'}^+(0) a_{k'+k}(0) \rangle + N^{-1} \sum_{k'} e^{-i(\omega_{k'+k} - \omega_{k'})t} \langle a_{k'}^+(0) a_{k'+k}(0) a_{k'+k}^+(0) a_{k'}(0) \rangle. \end{aligned} \quad (2)$$

Obviously, there is a four-magnon term, which is difficult to solve numerically. In Ref. [61], Wick's theorem was employed to decouple the four-magnon term into numerous two-magnon terms, and then  $\langle S_k^z(t) S_{-k}^z(0) \rangle$  could be rewritten as follows:

$$\langle S_k^z(t) S_{-k}^z(0) \rangle = \begin{cases} N S_0^2 \delta_{k,0} - 2 S_0 \delta_{k,0} \sum_{k'} \langle a_{k'}^+ a_{k'} \rangle + N^{-1} \sum_{k'} \left( 2 \langle a_{k'}^+ a_{k'} \rangle^2 + \langle a_{k'}^+ a_{k'} \rangle \right) & (k = 0), \\ N^{-1} \sum_{k'} e^{-i(\omega_{k'+k} - \omega_{k'})t} \langle a_{k'}^+ a_{k'} \rangle \langle a_{k'+k}^+ a_{k'+k} + 1 \rangle & (k \neq 0). \end{cases} \quad (3)$$

First, we calculated the spin autocorrelation functions of the pristine CrGeTe<sub>3</sub> monolayer under finite temperature and nonzero external magnetic field at  $k = 9.0 \times 10^5 \text{ cm}^{-1}$  ( $0.02\pi/a$ ), as shown in Fig. S5 within the Supplemental Material [56]. It can be observed that the

amplitude of  $\langle S_k^z(0) S_{-k}^z(0) \rangle$  grows with temperature but decreases with the magnetic field, because the amplitude of  $\langle S_k^z(0) S_{-k}^z(0) \rangle$  in Eq. (3) is determined by the magnon density  $\langle a_k^+ a_k \rangle$ . As the temperature increases, more magnons are excited, and the value of  $\langle a_k^+ a_k \rangle$

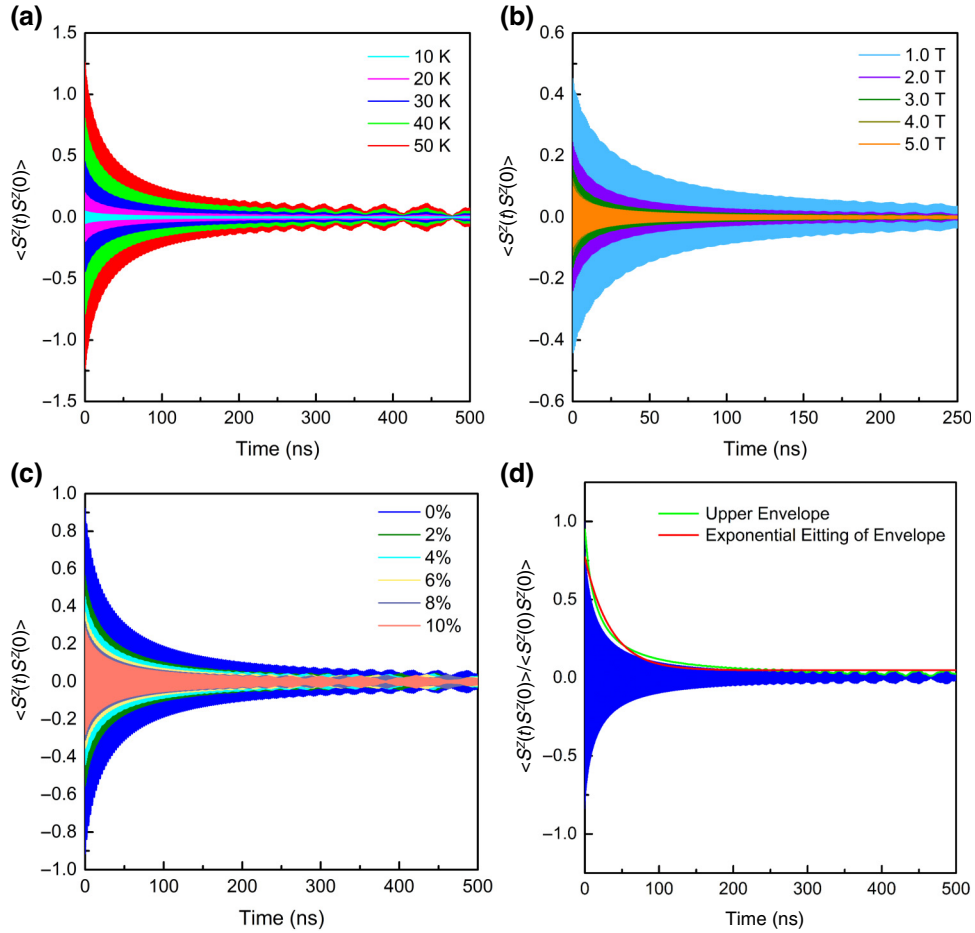


FIG. 5. Spin autocorrelation functions of 4% strained CrGeTe<sub>3</sub> monolayer at  $k = 9.0 \times 10^5 \text{ cm}^{-1}$  ( $0.02\pi/a$ ) under different temperatures ( $T$ ) (a) and external magnetic fields ( $B$ ) (b), spin autocorrelation functions of unstrained and strained CrGeTe<sub>3</sub> monolayers at  $k = 0.02\pi/a$  under  $T = 30 \text{ K}$  and  $B = 1 \text{ T}$  (c), and the diagram of normalized spin autocorrelation function (d).

increases. Meanwhile, a stronger external magnetic field results in higher magnon frequencies and leads to a reduction of the excited magnon density. To identify this analysis, we present the magnetic moment per  $\text{Cr}^{3+}$  ion  $\left[ M(B, T) = M_0 \left( 1 - (1/NS_0) \sum_k \langle a_k^+ a_k \rangle_{B,T} \right) \right]$  versus magnetic field in the pristine  $\text{CrGeTe}_3$  monolayer in Fig. S6 within the Supplemental Material [56], where the temperature grows from 0 to 50 K. In Fig. S6 within the Supplemental Material [56], the magnetic moment increases with strengthening of the magnetic field and tends to a specific value, but it is reduced close to zero as the temperature increases. These results are consistent with our analysis of the influences of temperature and external magnetic field on the magnon density  $\langle a_k^+ a_k \rangle$ . Similar phenomena can also be found in the 4% strained  $\text{CrGeTe}_3$  monolayer, as shown in Figs. 5(a), 5(b), and 6(a).

In Fig. 5(c), the spin autocorrelation functions of unstrained and strained  $\text{CrGeTe}_3$  monolayers at  $k = 0.02\pi/a$  at 30 K and under a magnetic field of 1 T are presented. Obviously, the amplitude of  $\langle S_k^z(0) S_{-k}^z(0) \rangle$  decreases with the applied strain monotonically. This is due to the enhancement of magnetic exchange coupling and magnon frequency by strain, as shown in Figs. 2(a) and 3(c). The enhancement of the magnon frequency makes the excitation of magnons difficult, and then the magnon density  $\langle a_k^+ a_k \rangle$  is reduced at a wave vector of  $0.02\pi/a$ . The magnetic moments per  $\text{Cr}^{3+}$  ion in unstrained and strained  $\text{CrGeTe}_3$  monolayers versus the external magnetic field are shown in Fig. 6(b), where the temperature is set to 30 K. In Fig. 6(b), we find that the magnetic moment increases with the applied strain at a specific magnetic field, suggesting a reduction of the magnon density with strain. This agrees with the results in Figs. 4(c) and 5(c).

We extracted the magnon relaxation time by fitting the upper envelope of the normalized spin autocorrelation function exponentially with a decay function  $[f(t) = A \exp(-t/\tau_{\text{MM}})]$ , as shown in Fig. 5(d). The blue, green, and red lines represent the normalized spin autocorrelation function, the upper envelope, and the exponential fitting of the upper envelope, respectively. The relaxation time,  $\tau_{\text{MM}}$ , of the spin autocorrelation function can reflect the strength of the magnon-magnon interaction, and a shorter  $\tau_{\text{MM}}$  corresponds to a stronger magnon-magnon interaction. The extracted magnon relaxation times of pristine and strained  $\text{CrGeTe}_3$  monolayers are presented in Fig. 7. In Fig. 7(a), the magnon relaxation times of both pristine and strained  $\text{CrGeTe}_3$  monolayers are significantly reduced with an increase of the wave vector from  $0.02\pi/a$  to  $0.3\pi/a$ , and then gradually decrease with the wave vector, when the temperature and external magnetic field are 30 K and 1 T, respectively. Furthermore, strain has little effect on this downward trend, and only decreases the magnon relaxation time. The maximum shortened ratio induced by 10% strain is about 51.22% at  $k = 0.2\pi/a$ ,

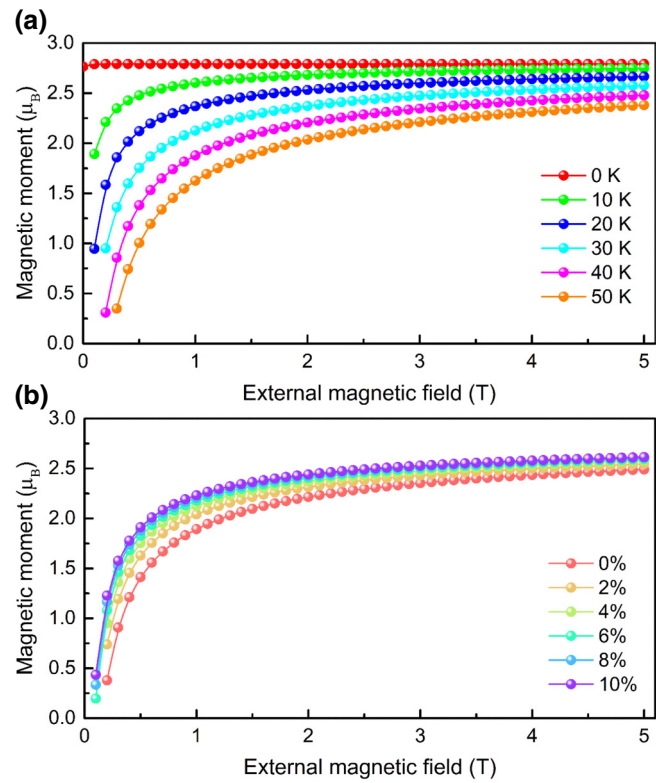


FIG. 6. (a) Magnetic moment per  $\text{Cr}^{3+}$  ion in 4% strained  $\text{CrGeTe}_3$  monolayer versus magnetic field. In (a), the temperature changes from 0 to 50 K. (b) Magnetic moment per  $\text{Cr}^{3+}$  ion in unstrained and strained  $\text{CrGeTe}_3$  monolayers versus magnetic field at 30 K.

revealing the remarkable enhancement of the magnon-magnon interaction by strain. At  $k = 0.02\pi/a$ , the shortened ratio induced by 10% strain is also up to about 13.3%. In Fig. 7(b), the magnon relaxation times of both pristine and strained  $\text{CrGeTe}_3$  monolayers are reduced significantly with temperature from 1 to 10 K, and then change slightly as the temperature increases from 10 to 55 K, when the wave vector and the external magnetic field are  $0.02\pi/a$  and 1 T, respectively. This trend is also not affected by strain. When the temperature is close to 0 K, the shortened ratio induced by 10% strain is 8.3%, while it is around 13% as the temperature increases.

As shown in Fig. 7(c), the magnon relaxation times of both pristine and strained  $\text{CrGeTe}_3$  monolayers are reduced continuously as the external magnetic field increases from 0.1 to 5 T, when the wave vector and temperature are  $0.02\pi/a$  and 30 K, respectively. Meanwhile, the application of strain also reduces the magnon relaxation time, and the maximum shortened ratio of about 49.3% occurs at a strain and magnetic field of 10% and 0.1 T, respectively. As the external magnetic field increases, the shortened ratio decreases, but is still up to 19.4% at 5 T. These results reveal that the dependence of magnon relaxation

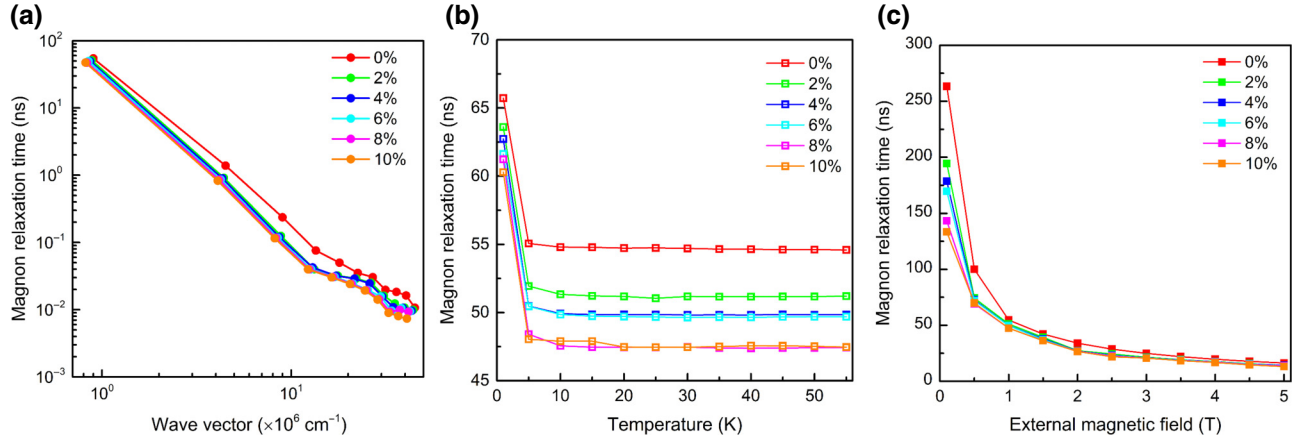


FIG. 7. (a) Dependences of magnon relaxation times in unstrained and strained CrGeTe<sub>3</sub> monolayers on wave vector at a temperature of 30 K and external magnetic field of 1 T. (b) Dependences of magnon relaxation times in unstrained and strained CrGeTe<sub>3</sub> monolayers on temperature under an external magnetic field of 1 T and wave vector  $k = 0.02\pi/a$ . (c) Dependences of magnon relaxation times in unstrained and strained CrGeTe<sub>3</sub> monolayers on external magnetic field at a temperature of 30 K and wave vector  $k = 0.02\pi/a$ .

time on wave vector, temperature, and external magnetic field changes slightly with strain, but the value of the magnon relaxation time decreases significantly as the strain increases.

Why does strain suppress the excitation of magnons but reduce the magnon relaxation time? To answer this question, we calculated the values of the magnon group velocity by

$$v_k = \frac{d\omega_k}{dk}. \quad (4)$$

According to Eq. (4), it is clear that the decay rate of the spin autocorrelation function depends on both the magnon density and the frequency difference between the  $k'$  and  $k' + k$  points, while the frequency difference between the  $k'$  and  $k' + k$  points near the center of the Brillouin zone can

be described by the value of the magnon group velocity. A larger absolute value of group velocity suggests a quicker decay of spin autocorrelation and a shorter magnon relaxation time. The values of magnon group velocity obtained at 30 K are plotted in Fig. 8. At  $k = 0.02\pi/a$  (near the center of the Brillouin zone), the enhanced ratios of the magnon group velocity are about 57%, 107%, 152%, 190%, and 223% for 2%, 4%, 6%, 8%, and 10% strained CrGeTe<sub>3</sub> monolayers, respectively. Hence, the decay of the spin autocorrelation function quickens with the applied strain, leading to a shortening of the magnon relaxation time, as shown in Fig. 7.

#### IV. CONCLUSIONS

In this paper, we explored the impact of strain on the magnon relaxation time dominated by magnon-magnon

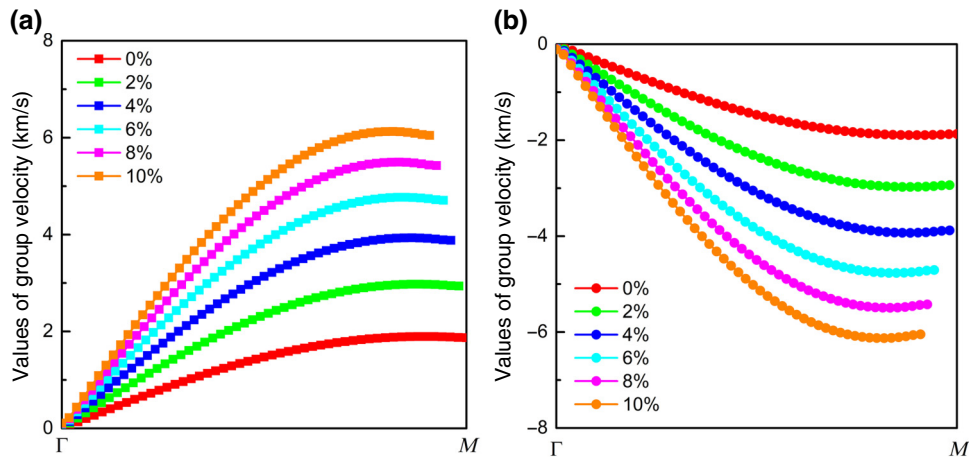


FIG. 8. Values of group velocities for acoustic (a) and optical (b) magnons along the  $\Gamma$ - $M$  path in the irreducible Brillouin zone of unstrained and strained CrGeTe<sub>3</sub> monolayers.



interactions in the ferromagnetic CrGeTe<sub>3</sub> monolayer by first-principles calculations and theoretical analysis. We find that the in-plane biaxial strain can enhance the magnetic exchange coupling remarkably, improving the Curie temperature of 2D CrGeTe<sub>3</sub>. The enhancements of the magnetic exchange constant and Curie temperature are up to 176% and 153%, respectively. Meanwhile, the applied strain decreases the magnon relaxation time significantly. At a low magnetic field, the maximum shortened ratio of the magnon relaxation time at  $k=0.02\pi/a$  can reach 49.3%. To elucidate this shortening of the magnon relaxation time induced by strain, we calculated the magnon group velocity and observed that the applied strain could enhance the magnon group velocity significantly. The enhancements of the value of magnon group velocity are about 57%, 107%, 152%, 190%, and 223% for 2%, 4%, 6%, 8%, and 10% strained CrGeTe<sub>3</sub> monolayers, respectively, resulting in rapid decay of the spin autocorrelation function and a shortening of the magnon relaxation time.

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