Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information

Strain Induced Polar Metal Phase in Ferromagnetic Fe_3GeTe_2 Monolayer

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I. Thermodynamic stability of the polarized structure of FGT monolayer under strain.

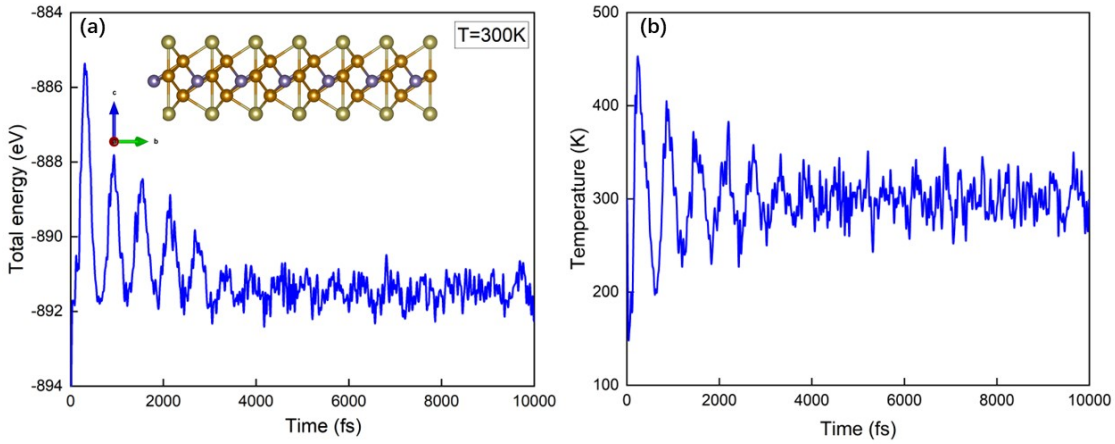

Fig. S1. The total energy of the FGT monolayer in 5x5x1 supercell at 300 K, and the side view of the FGT monolayer with σ=-5% in the inset. (b) The variation of temperature with time.

By means of DFT, we create a $5 \times 5 \times 1$ supercell to take molecular dynamics simulations. From Fig. S1a, the total energy of the FGT monolayer keeps in a stable range at 300K. Meanwhile, the inset of Fig. S1a shows the side view structure of the FGT monolayer under σ=-5%. We can find a clear polarization of the structure, with the Fe_{II} and Ge atoms having a relative displacement along the *z* direction. So, through the analyses, we conclude that the polarization under compressive strain is stable under room temperature.

II. The charge of lost and gain of different atoms under compressive strain.

The quantities of the charge from σ=0% to σ=-7% for Fe_I and Fe_{II} atoms increase 0.007 and 0.140, respectively. In contrast, the quantity of charge in $Fe_{I'}$ decreases 0.134. As for Ge atom, the charge values of Ge have a clearly reduce, ~0.19. When it comes to Te atom, we show that the quantity of the charge in Te_I and $Te_{I'}$ increases 0.25 and decreases 0.06 respectively.

III. The charge density distribution along Z direction.

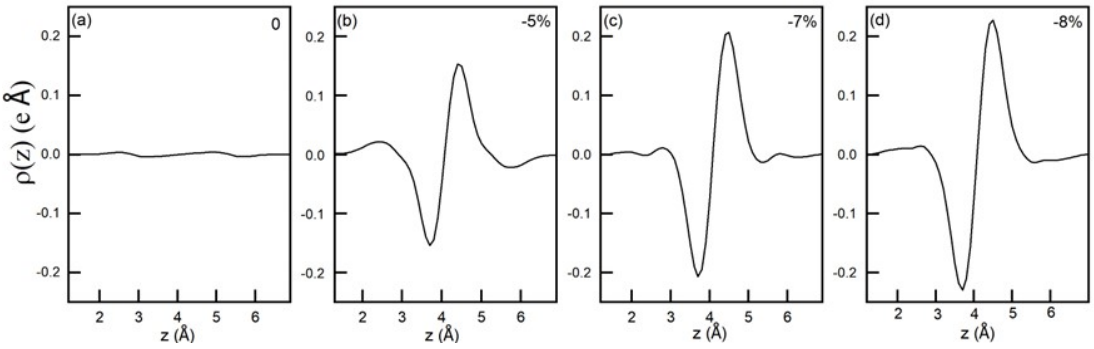

Fig. S2. The charge density distribution of FGT monolayer along *z* direction. (a-d) correspond to σ=0%, -5%, -7%, -8%, respectively.

Fig. S2 represent the charge density distribution of FGT monolayer along z direction under compressive strain. We find that the electronic contribution to the polarization increases with |σ|.

IV. The exchange coupling parameter analysis under strain.

Table. S1. The exchange coupling parameters of FGT and the parameters related to its under strain. The J_1, J_2, J_3 show the different interactions between Fe atoms, first-, second- and third-nearest neighbor interactions, J_1, J_2, J_3 are direct exchange of Fe_I and $Fe_{I'}$, $Fe_{I'}$ and $Fe_{II,}$ and $Fe_{I'}$ and $Fe_{I'}$, respectively.

	σ=0%	σ=-7%
E_0/(eV)	0	0
E_{FM}/(eV)	-1.588	-0.556
E_{AFM1}/(eV)	-0.977	-0.492
E_{AFM2}/(eV)	-1.084	-0.492
J_1 /(meV)	60.55	-3.33
J_2 /(meV)	-39.17	79.44
J_3 /(meV)	0.256	-14.08
Fe_I -$Fe_{I'}$ (Å)	2.467	2.699
Fe_I -Fe_{II} (Å)	2.630	2.402
$Fe_{I'}$ -$Fe_{I'}$ (Å)	4.024	3.743

Through the analysis, we find that when $\sigma = 0\%$, J_1 and J_3 are positive indicating the exchange couplings are FM, while the exchange parameter of J_2 is AFM. J_1 is bigger than J_2 and J_3, showing that the magnetic ground state relies on the magnetic exchange coupling between Fe_I and $Fe_{I'}$. Under compressive strain, the J_1, J_2 and J_3 change distinctly. When $\sigma = -7\%$, J_2 is positive, while J_1 and J_3 becomes negative. Both J_1 and J_3 are smaller than J_2. This shows that the ground state of FGT monolayer is still FM, but it depends on the second-nearest neighbors coupling. According to the above analysis, the second-nearest neighbors and third-nearest neighboring coupling affect the ground state properties of the FGT monolayer and external strain alters the exchange coupling of the FGT monolayer.

V. The position of Fe_{II} and Ge with respect to uniaxial strain.

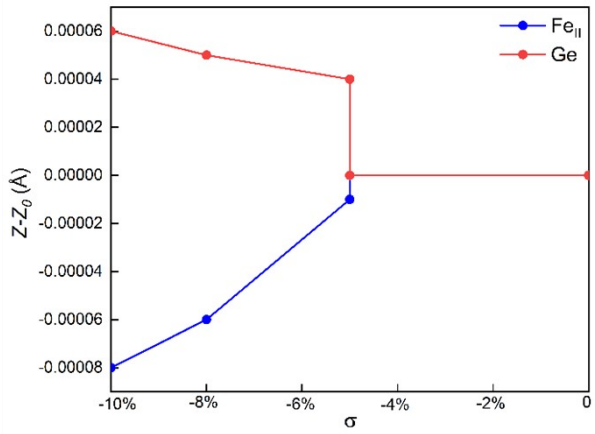

Fig. S3. Uniaxial strain-induced Fe_{II} and Ge atoms deviation from the center along *z* direction. Z_0 is the initial position of Fe_{II} and Ge with $\sigma = 0\%$. Z is the position of Fe_{II} and Ge along the c axis under strain. Z - Z_0 is the value relative to $\sigma = 0\%$.

We calculate the polarization value changes of FGT under uniaxial strain. We find that when the uniaxial strain is -5%, the polarization value of FGT is -2.392 eÅ/per unit cell. When the strain turns to -7%, the polarization value becomes -2.656 eÅ/per unit cell. As the strain increases, the polarization value is also increased. Compared with the biaxial strain, the uniaxial strain results in a significantly larger polarization value. Moreover, we observe that the polarization direction induced by the uniaxial strain is opposite to that of the biaxial strain.