NEWS AND VIEWS

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The room temperature ferromagnetism in highly strained two-dimensional magnetic semiconductors

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In spintronics, it is still a challenge in experiments to realize the ferromagnetic semiconductors with Curie temperature T_c above room temperature. In 2017, the successful synthesis of two-dimensional (2D) van der Waals ferromagnetic semiconductors, including the monolayer CrI_3 with T_c = 45 K[1] and the bilayer $Cr_2Ge_2Te_6$ with T_c = 28 K[2] in experiments, has attracted extensive attention in the 2D ferromagnetic semiconductors. One of the key problems is to find suitable 2D magnetic semiconductors, which can have room-temperature operation as required in applications.

The physical properties of 2D materials are sensitive to external regulations, such as electric filed, stacking, heterostructure, strain, etc. The strain engineering is the powerful technique to change the lattice of materials, and thus control the electronic, magnetic, and various other physical properties of 2D materials. In contrast to bulk materials, 2D materials have stronger deformation capacity and thus can withstand greater elastic strain without fracture, which shows great advantages in strain engineering. For example, monolayer MoS_2 can sustain strains as large as 11%[3], monolayer FeSe can sustain strains up to 6%[4, 5], and single-layer graphene can even withstand 25% elastic strain[6]. By density functional theory (DFT) calculations, it was predicted that the application of stain can dramatically change the T_c in various 2D magnetic semiconductors, including the monolayer $Cr_2Ge_2Se_6$[7], the magnetic semiconductor heterostructure $Cr_2Ge_2Te_6/PtSe_2$[8], the 2D magnetic topological insulators $Co_3Sn_3Se_2$, $Co_3Pb_3S_2$, $Co_3Pb_3Se_2$[9], the trilayer magnetic semiconductors CrI_3 with different stacking orders[10], etc.

In a recent paper published in *ACS Nano*, Professor Yuerui Lu's group at Australian National University and Professor Jan Seidel's group at University of New South Wales Sydney have found the experiment evidence of room temperature ferromagnetism in highly strained nanoscale wrinkles in the 2D $Cr_2Ge_2Te_6$[11]. $Cr_2Ge_2Te_6$ is a layered van der Waals material, where the T_c of $Cr_2Ge_2Te_6$ bulk is 61 K and the T_c of bilayer $Cr_2Ge_2Te_6$ is about 28 K. The strained 2D $Cr_2Ge_2Te_6$ is predicted to have ferromagnetism above room temperature according to some DFT calculations[7, 8]. In order to experimentally explore the effect of strain on T_c in 2D $Cr_2Ge_2Te_6$, Professor Lu and Professor Seidel's groups have systematically investigated the wrinkles of the 25 nm thick $Cr_2Ge_2Te_6$ samples (about 36 layers), where the wrinkles are formed during the growth process. These wrinkles induce local strain in the materials, where the strain is dependent on the height and width of the formed wrinkle.

Fig. 1(a) shows the temperature-dependent magnetic force microscopy (MFM) signal of curved wrinkles in the 2D $Cr_2Ge_2Te_6$ with 1.3% strain. The MFM has a magnetically coated tip near the sample surface, which is oscillated at resonance frequency. Because of the magnetic interactions between the tip and the sample, there will be a change in the oscillation phase, and this phase signal is recorded. When the measurement is taken at 4 K, the MFM signals show a stripe domain structure. At 80 K, the stripe pattern vanished in most of the areas, and the magnetic signals are left around the wrinkles. The strongest residual magnetic signal can be seen at the highest points. The higher areas should have higher strain values, because of the increased curvature resulting from the height difference. At 110 K, no more magnetic contrast can be detected, according to the vanish of the long-range magnetic order. Fig. 1(b) shows the temperature-dependent MFM signal of curved wrinkles in the 2D $Cr_2Ge_2Te_6$ with 2.3% strain. The MFM signal persists at room temperature with a clear magnetic signal in wrinkles, leading to the experimental evidence of room temperature ferromagnetism.

In order to investigate the origin of the increased T_c with stain in the 2D $Cr_2Ge_2Te_6$, Professor Bo Gu and Professor Gang Su's group at University of Chinese Academy of Sciences have conducted the DFT calculations and the Monte Carlo simulations to study the T_c of monolayer and bilayer $Cr_2Ge_2Te_6$ with strains[11]. Figs. 1(c) and 1(d) show the T_c and the magnetic anisotropy energy (MAE) as a function of strain for the monolayer and bilayer $Cr_2Ge_2Te_6$, respectively. In both cases, the T_c of $Cr_2Ge_2Te_6$ can be dramatically enhanced by the help of stain. For the bilayer $Cr_2Ge_2Te_6$, the DFT results suggests that the room temperature ferromagnetic order is possible at 6%–8% strain. Since the T_c in strained bilayer $Cr_2Ge_2Te_6$ is higher than T_c in stained monolayer $Cr_2Ge_2Te_6$, it is expected that T_c in strained thick 2D $Cr_2Ge_2Te_6$ can be even higher.

The enhancement of T_c with strain can be understood by the superexchange interaction, as categorized in the earlier reports by Professor Gu and Professor Su's group[7–10]. The strain is expected to decrease the energy difference $|E_p - E_d|$ between the p orbitals of Te and the d orbitals of Cr, and thus increase the antiferromagnetic coupling, J_{pd}, between the p orbitals of Te and the d orbitals of Cr in 2D $Cr_2Ge_2Te_6$. Due to the superexchange interaction, the enhanced J_{pd} can subsequently enhance the indirect ferromagnetic coupling between the d orbitals of Cr atoms, and finally enhance the

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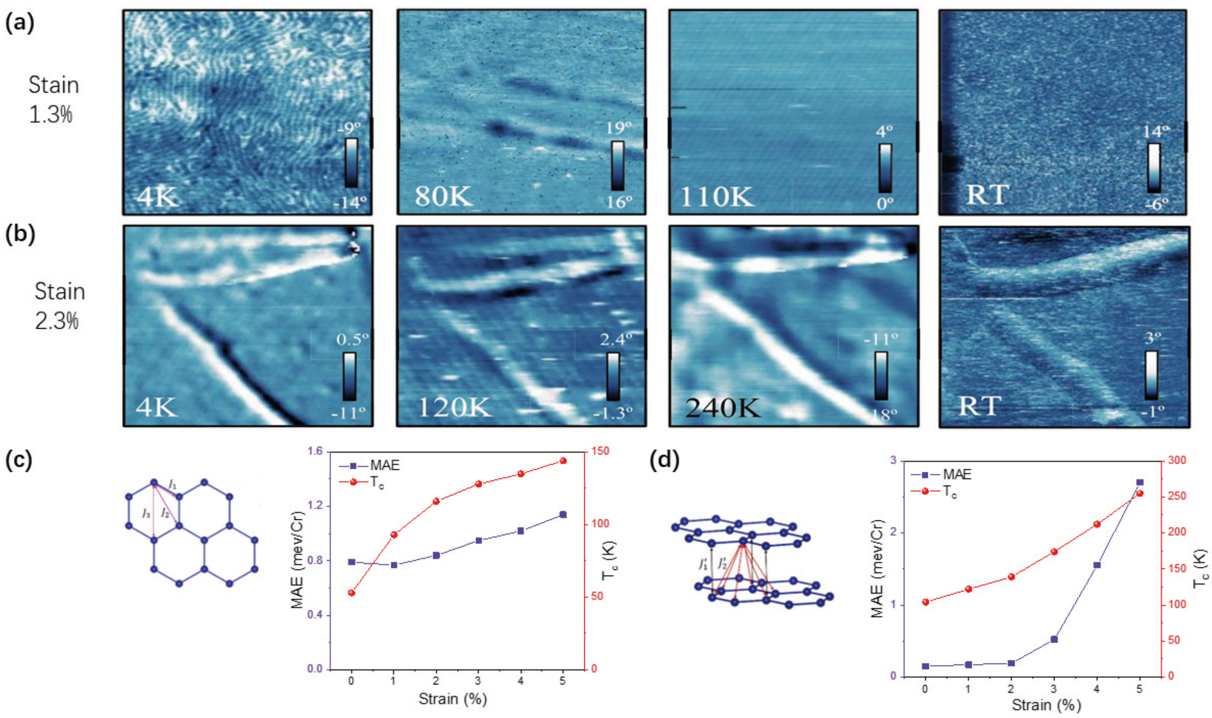

Fig. 1. (Color online) Temperature-dependent magnetic force microscopy (MFM) signal of curved wrinkles in the 2D $Cr_2Ge_2Te_6$ with (a) 1.3% strain and (b) 2.3% stain in experiment. Numerically calculated T_c and the magnetic anisotropy energy (MAE) as a function of strain for (c) monolayer and (d) bilayer $Cr_2Ge_2Te_6$, respectively. Adapted from Ref. [11].

T_c in 2D $Cr_2Ge_2Te_6$.

This work provides experimental evidence of room temperature ferromagnetism in highly strained 2D magnetic semiconductor, and strong insight into the mechanism of the enhanced magnetism of T_c.

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