Size-dependent magnetism of patterned MoTe₂ monolayer

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Abstract

The material with highly ordered structure can achieve selective adsorption of molecules and nanoclusters. In this paper, we studied the patterned structures formed by alternating 1 T and 2 H phases in single-layer MoTe₂ based on the first-principles calculations. The results show that the larger structure has the smaller formation energy, and SOC effect has considerable influence on the band structure of the system. The sizes of patterned MoTe₂ monolayer induce magnetic generation which mainly comes from Mo atoms at the interface. The Mo atoms with the largest magnetic moment locate at the lowest symmetry point, the localized magnetism making it possible for this patterned structure to be used for magnetic storage. Systems of different sizes exhibit different magnetic properties: as the size increases gradually, the system will change from ferromagnetism to antiferromagnetism, owing to the changed electronic structure of the Mo-2 atom e₁ and a orbital. Our work could provide a detailed understanding of the patterned structure in TMD films with atomic thickness and lay a foundation for further research on catalysis and adsorption.

1. Introduction

Two-dimensional transition metal chalcogenides (TMDs) have attracted extensive attention in the field of materials science due to their excellent photoelectric properties. With more and more people’s in-depth understanding of two-dimensional materials, many new properties different from bulk materials have been discovered, which is capable of manufacturing new materials and novel devices [1–3]. Therefore, the preparation of monolayer TMDs in large area and the study of their electronic structure, magnetic properties and catalytic properties [4–8] have gradually become a hotspot. MoTe₂ bulk material is an indirect bandgap semiconductor, when the material thickness is reduced to a single layer, the bandgap will change to a direct bandgap of 1.10 eV due to quantum confinement effect [9, 10] and show unusual properties that differ from the bulk counterparts [11–18]. At room temperature, the carrier mobility of the single layer MoTe₂ is larger than that of MoS₂, which broadens the spectral range of the direct bandgap material from visible to near infrared [19]. Previous experimental studies have found that triangular domain boundaries [20] appear in monolayer MoSe₂ grown by MBE method. It was also observed experimentally that different phases coexist stably in monolayer MoS₂ [21]. Domain boundary defects have a great impact on the electronic properties of materials, and the influence is not always negative. For example, the interface between adjacent domains can modulate the electronic properties of materials, and the charge transfer occurring across the location of the domain boundary can enhance catalytic performance [22–34]. So conducting intensive research on understanding how specific architecture between them can be established is necessary for designing and synthesizing such new-type nanostructures. Recently, a single layer PtSe₂ with a highly ordered triangular tiling patterns constructed by 1 H and 1 T phases was prepared experimentally. This patterned monolayer can selectively adsorb molecules as well as nanoclusters and has excellent catalytic performance [35]. To the best of our knowledge, the inherent nature
of this structure has not been elucidated yet. Here, we calculate the patterned structures in single-layer MoTe₂ in order to illustrate the intrinsic properties of this special structure and find the better application possibilities.

In this article, we construct and optimize the single-layer MoTe₂ in the form of a triangular pattern of alternating 2 H and 1 T phases. The electronic property, band structure and magnetism of patterned MoTe₂ monolayer with diverse sizes are studied theoretically by means of the first-principles calculations. Our calculation results provide a deeper understanding of the patterned structure in TMD films with atomic thickness and lay a foundation for further research on catalysis and adsorption.

2. Computational methods

The spin-polarized density functional theory (DFT) calculations were mainly performed with the Vienna Ab initio Simulation Package (VASP) [36, 37]. The structure relaxation and electronic properties calculation were computed, making use of the projector-augmented wave (PAW) method to describe electron-ion interactions [38, 39]. The plane-wave cutoff energy cutoff is 480 eV and the exchange-correlation potentials was described through the Preedew-Burke-Érnzerhof (PBE) functional of generalized gradient approximation (GGA) [40]. A vacuum spacing along z direction is 15 Å and hence the interlayer coupling can be ignored [41], and proper Monkhorst-Pack k-points of gamma-centered in the Brillouin zone are used along periodic directions to ensure energy convergence. Conjugate gradient techniques is used to relax all the structures in this paper. The energy convergence criterion of electron self-consistent iteration is 10⁻⁵ eV per cell, and the atomic force on each ion is smaller than 0.01 eV/Å [42].

3. Results and discussion

3.1. The atomic structure of patterned MoTe₂ monolayer

MoTe₂ monolayer has a sandwich-like structure with a sheet of Mo atom bonded to two adjacent sheets of Te atoms. With different stacking orders, there are different phase structures, i.e., trigonal phase (1 T) and hexagonal phase (2 H). The structure of patterned MoTe₂ monolayer comprising a periodic triangular structure formed by alternating semiconducting 2 H and metallic 1 T phases is plotted in figure 1. The 2 H phase is the most stable case at room temperature but the metastable 1T-MoTe₂ has an uncertain lattice constants. We start our discussion by searching for patterned structure of appropriate lattice constants. We set different lattice constants to calculate the formation energy of the system, and the lattice constant corresponding to the lowest formation energy is the most suitable lattice constant (Table S1). The formation energy of the system is the lowest when the lattice constant is 3.52 Å, so the lattice constants used in the latter calculation are all 3.52 Å.

After optimizing the lattice parameters, we constructed four kinds of patterned MoTe₂ monolayer with different sizes, including 3 × 3, 4 × 4, 5 × 5, 6 × 6 (figure 2). To evaluate the stability of these combinations, the formation energy (E₉) of each [MoTe₂] units was calculated and defined as the following equation:

Figure 1. Top and side views of atomic structures of 3 × 3 size of patterned MoTe₂ monolayer. The blue, yellow, and red balls represent Mo, top layer Te, and bottom layer Te, respectively.
Where $E_{\text{total}}$ and $E_{Te}$ represent total energy of the patterned MoTe$_2$ monolayer and single Te atom, respectively, $n$ is the number of Te atoms lost after patterned structure formation compared with the intrinsic material, and $S$ denotes the number of [MoTe$_2$] units in the patterned system. For these four types of patterned MoTe$_2$ monolayer with different sizes, $n$ is 3, 4, 5, 6, respectively; $S$ is 9, 16, 25, 36, respectively. According to the definition of formation energy, when the value is negative, the material can exist stably, and the lower formation energy indicates the better stability of structures. As shown in figure 3, the formation energy of each [MoTe$_2$] unit decreases with the increase of the system. The formation energy of patterned system with size of 3 $\times$ 3 is the highest, while that of system with size of 6 $\times$ 6 is the lowest. This is because when the T phase and H phase are arranged alternately, the lattice distortion occurs at the interface and the state of atoms at the interface changes. The coordination number of atoms at the interface changes compared with that at the intrinsic state. As the system increases, the proportion of atoms whose coordination number changes becomes smaller and the proportion of atoms that remain in the original state increases, which results in the decrease of the formation energy of the patterned system. It can infer that when the patterned system grows large enough, the whole energy of superlattice will not differ much from the intrinsic material.

### 3.2. The band structure of patterned MoTe$_2$ monolayer

Previous studies have shown that when the atomic number of the material is large, the effect of spin-orbital-coupling (SOC) will have a greater impact on electronic structure. The atomic numbers of molybdenum (Mo) atoms and tellurium (Te) atoms are 42 and 52, respectively. The influence of SOC effect on the simulation of these patterned systems needs to be evaluated in calculation. Therefore, we calculated the band structures based on PBE and PBE + SOC methods, and the results are as follows.

In order to compare the results of calculations, we first calculated band structures of intrinsic 2H-MoTe$_2$ and 1T-MoTe$_2$. Figures 4(a) and (b) are the band structure of intrinsic 2H-MoTe$_2$ calculated by PBE method only and with SOC effect, respectively. When SOC effect is considered, the band near Fermi level in intrinsic 2H-MoTe$_2$ has a slight split, but the position of valence band maximum (VBM) and conduction band minimum
CBM does not shift. The band gap remains unchanged, and the system still behaves as a direct band gap semiconductor. However, in 1T-MoTe2 monolayer, the band structure has changed greatly when we are using PBE + SOC method. The band near the Fermi level splits obviously, but the properties of the system remain unchanged, is still metallic.

In figure 5, the band structures of patterned MoTe2 monolayer based on different simulation methods are given. When considering the SOC effect, the band gap of 3 × 3 system at Fermi level is opened, and the VBM as well as the CBM are separated, leading to a very small band gap of about 0.02 eV. Whether PBE or PBE + SOC methods are used, the band structure of the calculated system is metallic. For 4 × 4 patterned system, the system calculated by PBE method reveals metallicity. When SOC effect is added, the band of system does not split obviously, the CBM at Fermi level bends downward and the lowest is lower than the VBM. However, they are not in the same position and the VBM and the CBM do not coincide. There is no band gap but the density of states of electrons near the Fermi level is close to zero, and system is semi-metal. For 5 × 5 system, it can be seen that the energy band near Fermi level of the superstructure system splits obviously when spin–orbit coupling effect is considered. The system is metallic either by PBE method or by PBE + SOC method. When the size of system increases to 6 × 6, it can be seen that the energy band near the Fermi level of the structure have a distinct splits. The separation of CBM and VBM produces a very small bandgap of about 0.02 eV. The property of the calculated system is still metallic when we are using PBE or PBE + SOC methods.

By comparing the band structures of single-layer patterned MoTe2 with different sizes calculated by PBE and PBE + SOC methods, we can find that the SOC effect has influence on the band structure of the system, the band gap of 3 × 3 and 6 × 6 system is opened of about 0.02 eV and the 5 × 5 system changes from metal to semi-metal.

3.3. The magnetism of patterned MoTe2 monolayer

Next, the magnetic properties of monolayer patterned MoTe2 are studied. We calculated the magnetic moments of patterned MoTe2 monolayer with different sizes by PBE and PBE + SOC methods (table 1). According to the data in the table, four kinds of patterned structures with different sizes have all introduced magnetism compared with the nonmagnetic intrinsic system, the magnetic moment of 3 × 3 system is the largest, and the magnetic moment of 6 × 6 is the smallest. The magnitude of magnetic moment obtained by PBE and PBE + SOC method is not very different, which overall difference is less than 3.28%. In the following work, we use PBE method to study the magnetism of patterned MoTe2 monolayer with different size for saving computing resources. To understand the cause of magnetism in patterned systems, the spin charge density and density of states (DOS) are calculated respectively.

In figure 6, we give spin charge density and PDOS of single Mo atom at different symmetric positions in the patterned MoTe2 monolayer of 3 × 3 size. It is evident that there is no spin polarization near Te atom in the supercell, and Te atom does not contribute to the magnetism of the system. The strong spin polarization of system is localized on the Mo atoms at the interface between T and H phases (Mo-1,2) particularly on the vertices Mo-1 atom, the Mo atoms in the center of T phase (Mo-3) and H phases (Mo-4) also have a little distribution. The magnetic moment of Mo-1 atom is 2.07 μB which accounts for 57% of the total magnetic moment of the
system, and it proves that the atomic with the lowest symmetry has the largest magnetism. The reason is the coordination number of Mo atoms at the interface changes from intrinsic state 6 to 3 (Mo-1) or 5 (Mo-2) when 1 T phase contacts 2 H phase. Although the coordination number of Mo-3 atom and Mo-4 atom is still 6, their electrons are also affected because of their close distance to the boundary. The changing states of these Mo atoms result in an asymmetric spin-up and spin-down DOS near the Fermi energy, which is mainly contributed by the 4d orbital of Mo atom. Every Mo-1 atom at the diamond vertex and its six nearest Mo atoms display ferromagnetic coupling.

The spin charge density as well as the PDOS of 4 × 4 patterned MoTe2 monolayer is shown in figure 7. Spin polarized calculations suggest that patterned structure induces spin polarization and leads to the formation of local moments of about 2.27 μB. The main contribution to the spin polarization results from the Mo-1 atom (1.37 μB) which accounts for 51% of the total magnetic moment, which means that the atomic with the lowest symmetry has the largest spin polarization. The rest of spin polarization distribute around the Mo-2 atom in the boundary and Mo-3 atom in the center of T phase. It can be seen that the coordination number of Mo atoms at the interface changes from 6 to 3 (Mo-1) or 5 (Mo-2, Mo-4) because highly ordered triangular tiling patterns constructed by 1 T and 2 H phases. Although the coordination number of Mo-3 atom and Mo-5 atom is still 6,
their surrounding electrons will be affected and the state has changed due to the distance to the interface. The alter of atomic state changes its electronic structure: the spin-up and spin-down DOS (mainly 4d orbits) near the Fermi level of Mo atom is asymmetric, and the local magnetic moment around the Mo atom results in the magnetism of system. Every Mo-1 atom at the diamond vertex and its six nearest Mo atoms display ferromagnetic coupling.

In figure 8, we give spin charge density and PDOS of single Mo atom at different symmetric positions in the patterned MoTe₂ monolayer of 5 × 5 size. It can be seen that there is no spin polarization near Te atom, and Te atom does not contribute to the magnetism of the system. The polarized charges mainly occurs at Mo-1,2,5 atom at the interface between T and H phases and particularly on the vertices Mo-1 atom, and the Mo-3,4 atom in T phase as well as Mo-6 atom in H phases also have a little distribution. The magnetic moment of Mo-1 atom is 1.20 μB which accounts for 40% of the total magnetic moment of the system, and it proves that the atomic with
the lowest symmetry has the largest magnetism. The reason lies in the coordination number of Mo atoms at the interface has changed from intrinsic state 6 to 3 (Mo-1) or 5 (Mo-2, 5) at edge reconstruction. Although the coordination number of Mo-3,4 atom and Mo-6 atom is still 6, their electrons are also affected because of their close distance to the boundary. The changing states of these Mo atoms result in an asymmetric spin-up and spin-down DOS near the Fermi energy, which is mainly contributed by the $4d$ orbits of Mo atom. Every Mo-1 atom at the diamond vertex and its six nearest Mo atoms display ferromagnetic coupling.

The spin charge density as well as the PDOS of single Mo atom at different symmetric positions in the $6 \times 6$ patterned MoTe$_2$ monolayer is shown in figure 9. It is evident that there is no spin polarization near Te atom in the supercell, and Te atom does not contribute to the magnetism of the system. The strong spin polarization of system is localized on the Mo atoms at the interface formed by alternating 1 T and 2 H MoTe$_2$ areas (Mo-1, 2, 4) particularly on the vertices Mo-1 atom, and the Mo-3 atoms at the T phase also have a little distribution. The
magnetic moment of Mo-1 atom is $1.40 \mu_B$, which accounts for 64% of the total positive magnetic moment of the system, and it proves that the atomic with the lowest symmetry has the largest magnetism. The reason of this is that the coordination number of Mo atoms at the interface changes from intrinsic state 6 to 3 (Mo-1) or 5 (Mo-2, 4) when 1 T phase is contacting with 2 H phase. Although the coordination number of Mo-3 atom is still 6, their electrons are also affected because of their close distance to the boundary. The changing states of these Mo atoms result in an asymmetric spin-up and spin-down DOS near the Fermi energy, which is mainly contributed by the 4d orbital of Mo atom. Every Mo-1 atom at the diamond vertex and its six nearest Mo atoms display antiferromagnetic coupling.

Figure 10. The magnetism change and magnetic moment of patterned MoTe$_2$ monolayer with different sizes in $\mu_B$.

Figure 11. The projected density of states (PDOSs) of Mo-1 and Mo-2 atom of 3 × 3 (a) and 6 × 6 (b) patterned MoTe$_2$ monolayer.
magnetization is antiferromagnetism. To understand the mechanism of the magnetic shift, we take the 

3

associated to the interaction between the images, and the ground state of this patterned structure in 

the atom of 3

monolayer all have magnetism which mainly comes from the 4

In summary, we studied the electronic structure and magnetism of patterned MoTe\(_2\) monolayer based on the 

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Conclusion

By comparing the magnetic properties of single-layer patterned MoTe\(_2\) with different sizes, we can find that 

this patterned structure will introduce strong spin polarization and lead to the formation of magnetic moment 

in the system. The magnetic moments mainly arise from the 4d orbits of Mo atoms at the interface between T 

and H phases. The Mo atoms at the diamond vertex have the largest magnetic moment, making it possible for 

the patterned structure to be used for magnetic point storage. At the same time, we calculated the spin charge 

density of 3 × 3 and 4 × 4 patterned MoS\(_2\) and MoSe\(_2\) monolayer. The results show that the system also 

introduces magnetism and the spin polarization mainly localized on the Mo atoms at the interface (figure S1 is 

available online at stacks.iop.org/MRX/6/126115/mmedia).

It is worth noting that as the structure increasing from 3 × 3 to 6 × 6 or even larger (figure S2), the system 

will change from ferromagnetism to antiferromagnetism (figure 10). For 6 × 6 and larger (7 × 7, 8 × 8) 

patterned structure, the strong spin polarization of system is localized on interface formed by alternating 1 T 

and 2 H MoTe\(_2\) areas particularly on the vertices and its six nearest Mo atoms, and the other atoms do not contribute 

to the magnetism of the system. Therefore the 6 × 6 is the minimal size that the magnetic effect can not be 

associated to the interaction between the images, and the ground state of this patterned structure in 
magnetization is antiferromagnetism. To understand the mechanism of the magnetic shift, we take the 3 × 3 

and 6 × 6 structure as examples for further investigation. From the previous analysis, the magnetism of 

patterned system mainly comes from 4d orbits of Mo atoms at the interface, and hence we focus on the Mo-1 

atom at the diamond vertex and its nearest neighbor Mo-2 atom. Figure 11 shows the PDOSs of these two Mo 

atoms of 3 × 3 and 6 × 6 system. For 3 × 3 system, the asymmetrical spin-up and spin-down channels near 

the Fermi level of five degenerate orbits of Mo-1 and Mo-2 atom all contribute to the total magnetic moment. 

For 6 × 6 system, the electronic structures feature of Mo-1 atom d orbit is similar to that of 3 × 3 system. 

However, the spin-up and spin-down states of Mo-2 atom d orbit are relatively asymmetrical, and the minority 

spin state is obviously more than that of majority spin state. The polarized charges of Mo-2 atom mainly come 

from the e\(_z\) and a orbit and hardly come from the e\(_x\) orbit. In order to explain the contribution of each orbit to 

the atomic magnetic moment more accurately, the difference between spin-up and spin-down of Mo atom 4d, 

d\(_{xy}\), d\(_{yz}\), d\(_{xz}\), d\(_{z}\)\(_{2}\) and d\(_{x}\)\(_{2}\)\(_{y}\)\(_{2}\) orbit was calculated (table 2). It can be seen that the contribution of each orbit of Mo-1 atom 

to the total magnetic moment of the two systems is close to each other. For Mo-2 atom of 6 × 6 system, the spin-up 

charges of d orbits are less than spin-down charges, and the difference between them is negative. This causes 

the atomic magnetic moment direction to be spin-down, forming antiferromagnetic coupling with Mo-1 atom. 

To summarize, a charge transfers from the majority to minority spin state comes up at the Mo-2 atom, thus 

reducing the magnetic localization and increasing the exchange interaction, and lastly the system will change 

from ferromagnetism to antiferromagnetism due to the increase of size.

Table 2. Calculated magnetic moment of Mo atom (M in \(\mu_B\)), difference between spin-up and spin-down of Mo 

atom 4d, d\(_{xy}\), d\(_{yz}\), d\(_{xz}\), d\(_{z}\)\(_{2}\) and d\(_{x}\)\(_{2}\)\(_{y}\)\(_{2}\) orbit (in e\(_{\text{f}}\)).

| Atom  | M      | \(\Delta d\) | \(\Delta d\)\(_{xy}\) | \(\Delta d\)\(_{yz}\) | \(\Delta d\)\(_{xz}\) | \(\Delta d\)\(_{z}\)\(_{2}\) | \(\Delta d\)\(_{x}\)\(_{2}\)\(_{y}\)\(_{2}\) |
|-------|--------|-------------|----------------|----------------|----------------|----------------|----------------|
| 3 × 3 | Mo-1   | 2.072       | 1.736           | 0.391           | 0.331           | 0.345           | 0.272           | 0.397           |
|       | Mo-2   | 0.241       | 0.163           | 0.060           | 0.033           | 0.039           | 0.008           | 0.023           |
| 6 × 6 | Mo-1   | 1.402       | 1.339           | 0.306           | 0.248           | 0.247           | 0.232           | 0.306           |
|       | Mo-2   | −0.127      | −0.130          | −0.006          | −0.009          | −0.002          | −0.035          | −0.078          |

4. Conclusion

In summary, we studied the electronic structure and magnetism of patterned MoTe\(_2\) monolayer based on the 

first-principles calculations. For single-layer patterned MoTe\(_2\) of various sizes, the larger structure has the 

smaller formation energy. When PBE and PBE + SOC are used to calculate the band structure, the SOC effect 

has considerable influence on the band structure of the system. The calculated four sizes of patterned MoTe\(_2\) 

monolayer all have magnetism which mainly comes from the 4d orbits of Mo atoms at the interface formed by 

alternating 1 T and 2 H MoTe\(_2\) areas. The Mo atoms with the largest magnetic moment locate at the lowest 

symmetry point, the localized magnetism making it possible for this patterned structure to be used for magnetic 

storage, and similar features have been found in the calculation of other patterned TMD monolayer. Systems of 

diverse sizes exhibit different magnetic properties: as the structure increases gradually, the system will change 

from ferromagnetism to antiferromagnetism owing to the changed electronic structure of the Mo-2 atom e\(_z\) and 

a orbital. Our calculated result illustrates the intrinsic properties of this special structure and could provide 

useful guidelines for production these patterned systems with new structure. It is believed that the attractive 
predictions will soon be confirmed by further experiments.
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