Molecular Beam Epitaxy of 2D Vanadium Diselenide on Molybdenum Disulfide

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Introduction

In the era of 2D materials, heterostructures are the next step in producing novel effects and devices for the future [1]. In particular, a stacked system of a ferromagnetic and a semiconducting material has applications in spin-charge conversion effect [2], valleytronics [3], etc. This study will make use of a heterostructure which comprises of VSe2, a ferromagnetic material [4], and MoS2, a widely used semiconducting material. Molecular beam epitaxy (MBE) is the choice fabrication method of VSe2, of high quality, for its good controllability in film thickness and other parameters (e.g., flux and temperature). VSe2 was grown on MoS2 in a home-made MBE ultrahigh-vacuum system of base pressure of 2 × 10−9 mbar, where V and Se were simultaneously evaporated by an electron-beam evaporator and a Knudsen cell, respectively, onto a substrate maintained at 360°C. The growth morphology and electronic properties of VSe2 on a MoS2 substrate are studied in detail by STM/STS. Interestingly, phase transition occurs upon thermal annealing of the 2D VSe2 crystals. These give insight on the tunability of the crystalline phases and hence the electronic and ferromagnetic properties of VSe2.

Results & Discussion

1 Structural properties of 1L-VSe2/MoS2 heterostructure

(a) STM image (70x70nm2; I = 9 pA, V = 1.8 V) of monolayer VSe2 grown on MoS2 by MBE, and the height of 1L-VSe2 is ~6.0 Å as depicted by white arrow that gives the line profile in the inset. (b) Atomic structures of 1T-VSe2 (top) and 2H-MoS2 (bottom). (c) Moiré superstructure labelled by a black diamond, which corresponds to experimental observation seen in (a). Due to the different lattice constant values of VSe2 and MoS2, the stacking of these two materials gives rise to a hexagonal Moiré superstructure with a periodicity of (16√2) VSe2 on (17×17) MoS2.

2 1st layer VSe2: CDW and lattice distortions

(a) Atomic resolution image (10x10nm2; I = 180 pA, V = 10 mV) of 1L-VSe2, and its (b) 2D-FFT image generated by WSXM software6. The peaks circled by the pink, white, and yellow circles represent the Bravais lattice points, Moiré superstructure, and charge density wave (CDW) structure, respectively. The grey, blue, and green circles highlight the peaks associated with the lattice distortions of 1L-VSe2, as seen in the (c) Fourier-filtered image (10x10nm2) which is obtained by selectively retaining the grey, blue, green and pink circles in (b). The dotted lines show the directions of the lattice distortions, which correspond to the circles in (b) by their colour, and they form the superstructure marked by the black lines in (a). (d) Fourier-filtered image (10x10nm2), which is obtained by selectively retaining the yellow and pink circles in (b), shows that CDW in 1L-VSe2 is unidirectional. The periodicity of CDW is observed to be 4a from the line profile marked by the white arrow.

3 2nd layer VSe2: CDW

(a) STM image (77x77nm2; I = 10 pA, V = 2.0 V) of 1st and 2nd layer VSe2 grown on MoS2, (b) Atomic resolution image (10x10nm2; I = 180 pA, V = 5 mV) of 2L-VSe2, which is the region marked by the white box in (a), and its (c) 2D-FFT image. (d) Fourier-filtered image (10x10nm2), which is obtained by selectively retaining the pink circles in (c), shows the lattice points of VSe2. (e) Fourier-filtered image (10x10nm2), which is obtained by selectively retaining the pink and white circles in (c), shows the Moiré superstructure from the lattice mismatch of VSe2 and MoS2. (f) Fourier-filtered image (10x10nm2), which is obtained by selectively retaining the pink and yellow circles in (c), shows that CDW in the 2nd layer has a (4 x 4) hexagonal superlattice. The unit cell of each structure is marked by a grey diamond.

4 Vacuum annealing of VSe2

(a) Large-scale STM image (250x250nm2; I = 20 pA, V = 1.8 V) of 1L-VSe2/MoS2 before high temperature vacuum annealing. (b) STM image (250x250nm2; I = 3 pA, V = 2.0 V) of 1L-VSe2/MoS2 after vacuum annealing of 336°C for 12h. (c) STM image (50x50nm2; I = 30 pA, V = 1.6 V) of the line defects formed on VSe2, which corresponds to the region marked by the white box in (b).

Conclusion

VSe2 grows epitaxially on MoS2, and forms a Moiré superstructure due to lattice mismatch. The Moiré pattern can be seen up to the 2nd layer of VSe2. The atomic resolution STM images of VSe2 reveals that 1L-VSe2 has CDW and lattice distortions, while 2L-VSe2 only has CDW. Interestingly, the CDW in 1L-VSe2 is unidirectional, while the CDW in 2L-VSe2 has a hexagonal superlattice. High temperature vacuum annealing of VSe2 produces line defects that forms 1D ribbons of VSe2 and small triangular domains.