Structural Rearrangement of Twisted MoS_2 In Atomic Substitution Nitridation Reaction

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Introduction

- Moiré patterns are formed when two identical patterns are overlaid at an angle mismatch, or "twist"

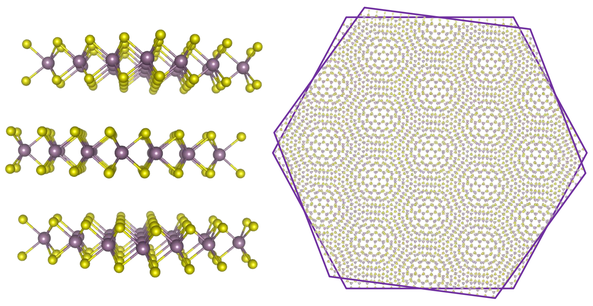
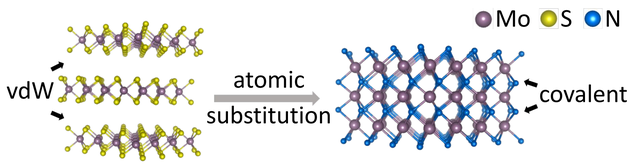


Fig. 1. Lattice structure of MoS_2 and an example of moiré patterns formed in twisted MoS_2 (t-MoS_2).

- 2D materials like MoS_2 are atomically thin materials, layers held by weak van der Waals forces
- Moiré patterns in 2D materials can cause many unique physical and chemical properties[1][2]
- Studies have been done on electron-transfer reactions, but little is known about the effects of atomic substitution on structure

Objective: This study aims to use the nitridation of the 2D material MoS_2 into MoN as a model system to examine structural changes in moiré lattices.

Fig. 2. The nitridation of 2D MoS_2 into MoN, which is naturally of 3D structure.[2]



Methods

- t-MoS_2 samples prepared via gold-assisted mechanical exfoliation onto SiO_2/Si substrate
- Dry transfer process via transfer stage using adhesive poly(bisphenol A carbonate) (PC)

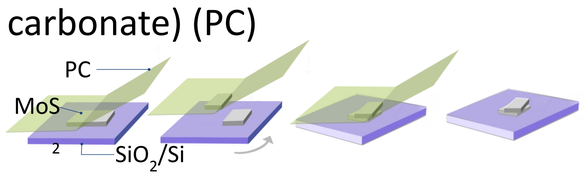


Fig. 3. Diagram of dry transfer process

- Nitridation carried out in a quartz tube furnace with ammonia carried by argon (flow rate of 100 sccm), performed at 850°C for 10 min, with a ramping rate of 45°C/min.

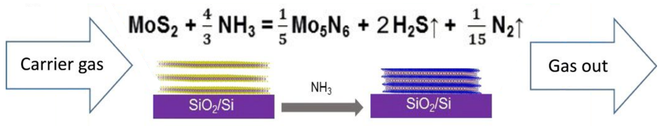


Fig. 4. Nitridation schematic and equation[2]

- Raman spectroscopy (Renishaw inVia Raman microscope at 532 nm, 2.5 mW) and selected area electron diffraction (SAED, via TEM).

Results

Raman spectra of converted "3+2" and "2+2" twisted samples were taken. When compared to regular MoN spectra, we found only peaks characteristic of 5L and 4L layers, instead of peaks from individual 3L and 2L.

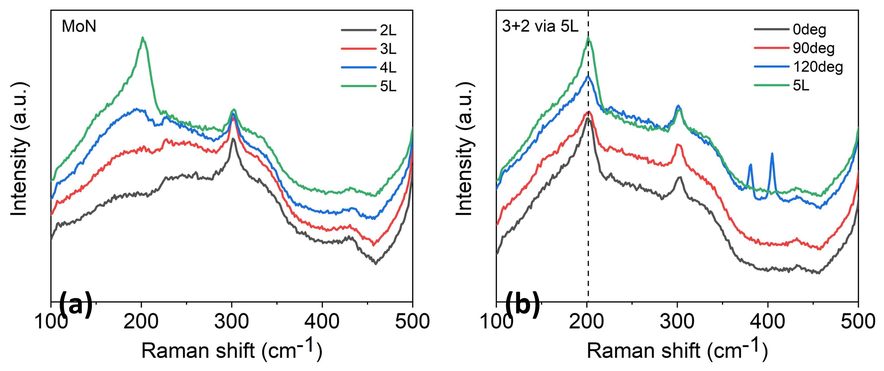


Fig. 5. Raman spectra of MoN layers (a) and converted 3+2 twist samples (b). On all twist angles, 5L peaks were observed.

SAED images of 5°, 15°, and 30° t-MoS_2 samples before and after nitridation, with 1L and 2L on thicker few-layer (FL), were obtained using a transmission electron microscope (TEM). Diffraction spots represent different crystal planes.

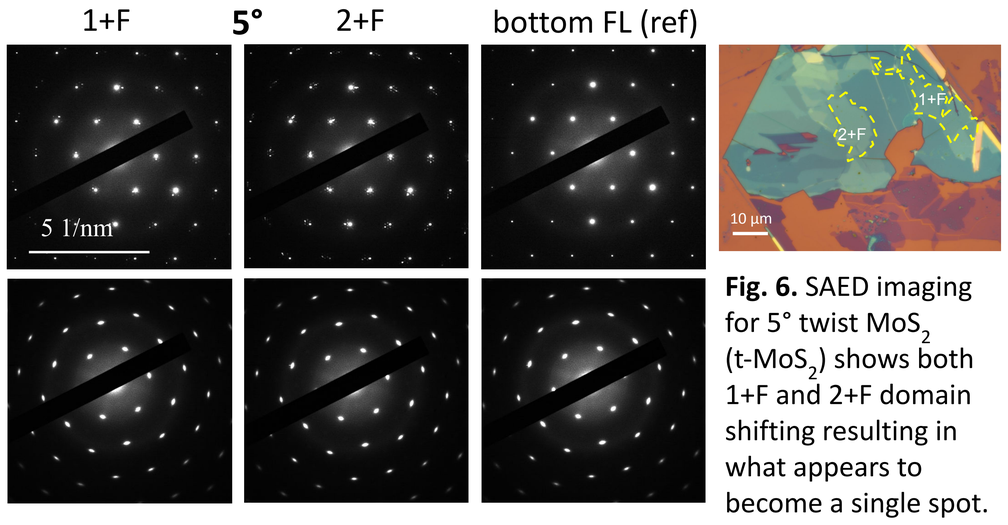


Fig. 6. SAED imaging for 5° twist MoS_2 (t-MoS_2) shows both 1+F and 2+F domain shifting resulting in what appears to become a single spot.

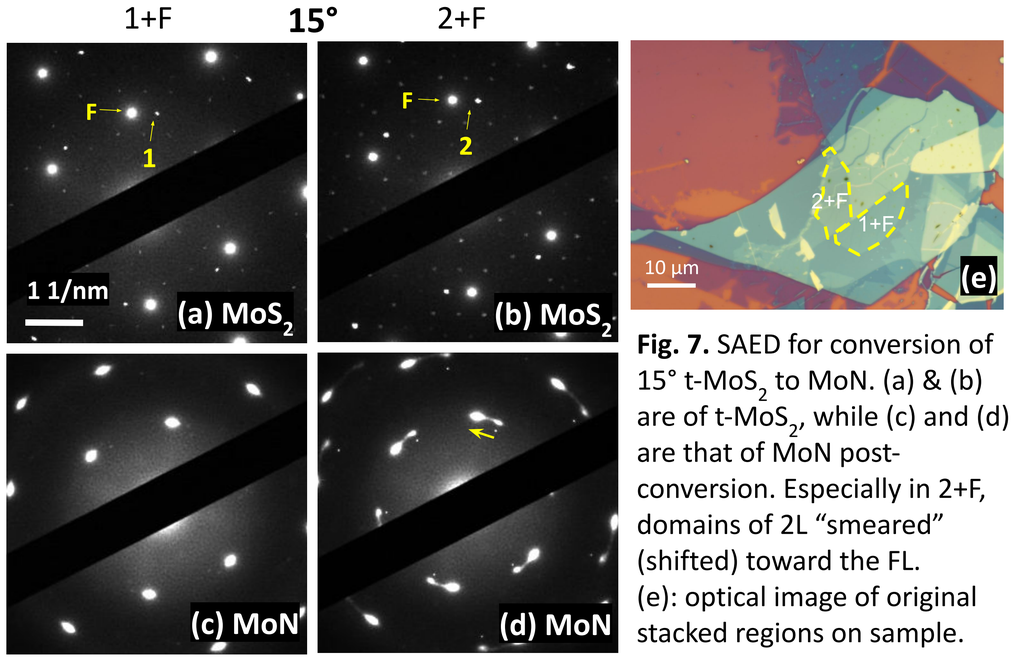


Fig. 7. SAED for conversion of 15° t-MoS_2 to MoN. (a) & (b) are of t-MoS_2, while (c) and (d) are that of MoN post-conversion. Especially in 2+F, domains of 2L "smeared" (shifted) toward the FL. (e): optical image of original stacked regions on sample.

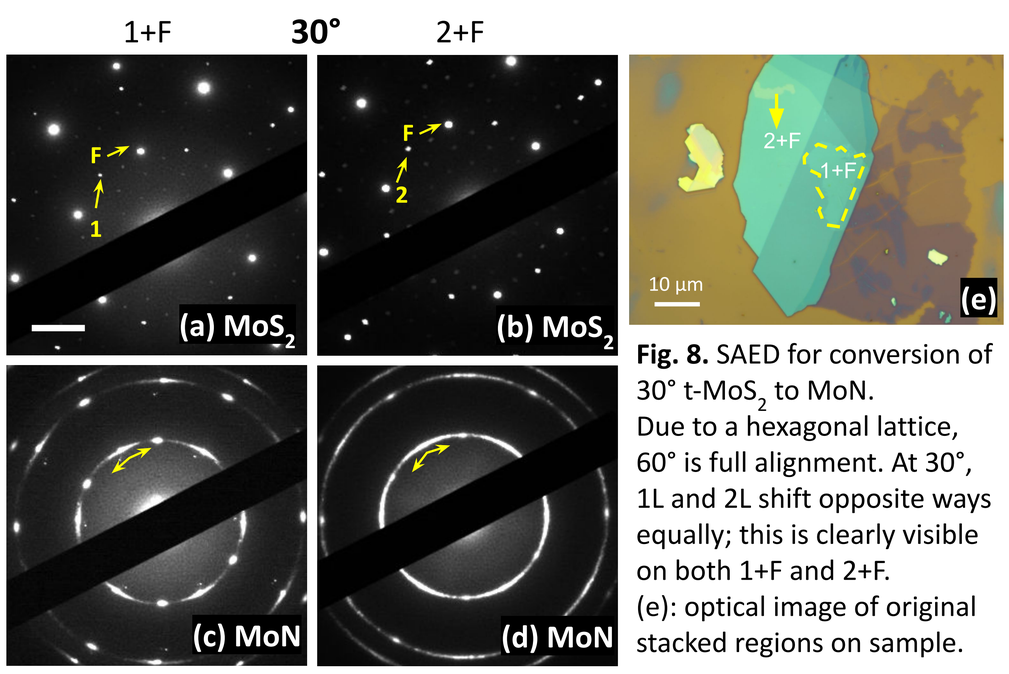


Fig. 8. SAED for conversion of 30° t-MoS_2 to MoN. Due to a hexagonal lattice, 60° is full alignment. At 30°, 1L and 2L shift opposite ways equally; this is clearly visible on both 1+F and 2+F. (e): optical image of original stacked regions on sample.

Discussion

Raman Spectroscopy

- Peaks for 5L and 4L observed after nitridation of layers such as 3+2 and 2+2 indicated moiré lattices may have untwisted/combined. This finding led to SAED imaging for further investigation

SAED

- Lattice responses were found to be dependent on the twist angle; domains in top 1L, 2L shifted toward the closest angle direction needed to align with bottom FL
- In imaging for the 5° sample, domains may have been so close that they fully shifted to align
- For larger angles, domains were likely still in the process of shifting or aligning when nitridation was completed, resulting in the smear
  - Not all atoms shifted; perhaps due to their relative positions in the original moiré lattice

Future Work

- For further investigations into lattice rearrangement, perform SAED / TEM on other stacking configurations after nitridation (angles e.g. 10°, 20°; layering e.g. FL on 1L, etc)
- Create computational models to simulate identify specifics of lattice rearrangement during the reaction

References

[1] Cao, Y., Fatemi, V., Fang, S. *et al.*, Unconventional superconductivity in magic-angle graphene superlattices. *Nature* **556**, 43–50 (2018).

[2] Cao, J. *et al.*, Realization of 2D crystalline metal nitrides via selective atomic substitution. *Sci. Adv.* **6** (2020).

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