Exploring the Cryogenic Phase Changes within 2D MoTe₂ via TEM, 4DSTEM and Electron Spectroscopy Techniques

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Research in the field of atomically thin 2D materials has grown remarkably since the discovery and isolation of graphene, the 2010 Nobel Prize 2D material, due to its structural flexibility and tunable electronic properties[1] [2]. Graphene has been one of the most well-researched 2D materials, but being a quasi-metal limits its applications in electronic devices [3][4]. This has fueled research into two-dimensional transition metal dichalcogenides (2D TMDs) which can exhibit more than one phase with distinct structures and physical properties such as the metallic distorted octahedral (1T') phase and the semiconducting trigonal prismatic hexagonal (2H) phase [5]. Among them, MoTe₂ is a topological Weyl semimetal (TWS) and is the most promising TMD material for phase engineering at the atomic level [6]. Since TWSs are highly conducting at low frequencies, they challenge the classical idea of polarization and metallicity, this phenomenon has gathered a lot of attention in the ferroelectric community [7]. The possibility of controllable TMDs phase engineering has attracted tremendous attention due to its appeal in both fundamental physics and potential applications beyond graphene [8].

MoTe₂ has a smaller phase energy difference ($\Delta E < 50 \text{ meV}$) between the 1T' and the 2H phase which can make phase transitions possible compared to other TMDs such as MoS₂ ($\Delta E > 0.8 \text{ eV}$) [9]. Recently, MoTe₂ phase changes have been achieved by strain engineering [10], controlled chemical vapor deposition (CVD) [11], electrostatic doping[12] and laser irradiation [13]. The underlying mechanism of these phase changes has been proposed to be heating, Te defects, or strain effects [9][14][15]. However, a microscopic understanding at the atomic level is still lacking and more experimental evidence is required to further explore the phase change phenomenon.

Our preliminary results show a clear change in the phase structures between room and cryogenic temperatures, which is an important feature, since the electronic properties of 2D materials are highly dependent on their precise structural arrangement and chemical composition[16]. This temperature-driven study focuses on atomic-scale observations of local phase transitions in MoTe₂ in cryogenic environments by using in-situ cryogenic transmission electron microscopy (TEM) and 4D scanning transmission electron microscopy (4DSTEM) while monitoring electronic band structure and chemical composition changes via monochromated high-resolution electron energy loss spectroscopy (EELS). In this presentation, we will detail the benefits of using low temperature atomic resolution electron microscopy and spectroscopy to explore the exotic phase changes of topological semimetals (18).



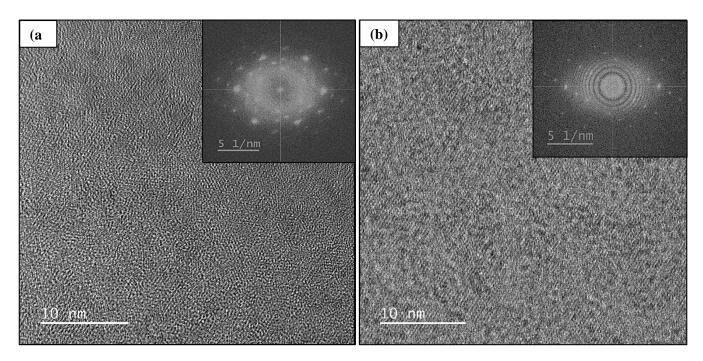


Figure 1 TEM images of few layer 1T' MoTe₂ at room temperature (RT) and cryogenic temperatures with Fast Fourier transform (FFT) insets respectively. (a)The TEM image taken at RT shows evidence of a multiple phases of MoTe₂ in the field of view with circularly distributed spots shown in the FFT inset. (b) The TEM image taken at cryogenic temperatures shows a more uniform phase with linearly distributed spots in the FFT inset. The difference in focus between two images might have an effect on the shape of FFT spots but the distribution of spots in each image shows difference in homogeneity.

References:

- [1] MH Kang et al., in "Comprehensive Nanoscience and Nanotechnology, vol. 1–5", (Elsevier), p. 55.
- [2] AK Geim, Science **324**(5934) (2009), p. 1530.
- [3] R Vargas- Bernal in "Two-dimensional Materials", eds. PK Nayak, E Rijeka, (IntechOpen).
- [4] GR Bhimanapati et al., ACS Nano. 9(12) (2015), p. 11509.
- [5] D Mackenzie, Engineering (2020).
- [6] Y Tan et al., Nanoscale **10**(42) (2018), p. 19964.
- [7] F-T Huang et al., Nat. Commun. **10**(1) (2019), p. 4211.
- [8] Y-C Lin et al., Nat. Nanotechnol. 9 (2014), p. 391.
- [9] KAN Duerloo, Y Li and EJ Reed, Nat. Commun. **5**(1) (2014), p. 4214.
- [10] S Song et al., Nano Lett. **16**(1) (2016), p. 188.
- [11] TA Empante et al., ACS Nano. **11**(1) (2017), p. 900.
- [12] Y Wang et al., Nature **550**(7677) (2017), p. 487.
- [13] C Suyeon et al., Science, **349**(6248) (2015), p. 625.
- [14] S Yuan et al., Nat. Commun., **10**(1) (2019), p. 1775.
- [15] MB Vellinga, R de Jonge and C Haas, J. Solid State Chem., **2**(2) (1970), p. 299.
- [16] MC Michael et al., Wiley-VCH GmbH. **2170142**(17) (2021).
- [17] S Fang et al., Nat. Commun. **10**(1) (2019), p. 1127.
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