INFLUENCE OF BIAS VOLTAGE ON THE OBSERVED MOIRÉ PATTERNS OF $MoTe_2$/GRAPHENE HETEROSTRUCTURE GROWN BY MOLECULAR BEAM EPITAXY

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In recent years, the importance of the rotation angle between layers of 2D material has come to the forefront of the research on 2D van der Waals heterostructures. Bi-layer graphene with small magic angles is a prime example with its superconducting behavior and the presence of solitons [1, 2]. In heterostructures with different lattice parameters such as $MoTe_2$/graphene, it is possible to observe high-indexed Moiré patterns that are more complex and less studied than the usual low-indexed ones. The STM images have then to be analyzed with great caution because topology and electronic effects are intertwined. These high-indexed patterns imply extra periodic potential modulations in the electronic system, which is at the heart of flat-band formation in van der Waals heterostructures.

In our study, we demonstrate that MBE grown $MoTe_2$ on graphene substrate favors various higher-indexed Moiré patterns for twist angles around 30° (see figure). These patterns have an unusually high degree of complexity, therefore their characterization had to be done rigorously in the full context of Moiré theory. Our analysis provides a useful example to researchers working in the field of how to identify and interpret higher-indexed patterns and also highlights the importance of the electronic effects in $MoTe_2$/graphene heterostructure.

The key findings emerging from our STM investigations combined with DFT calculations and the Moiré-analysis are:

- The Moiré patterns found in our STM measurements are remarkably sensitive to the bias voltage. When changing the bias voltage, the Moiré pattern transforms because of the entanglement with electronic effects, causing the apparent Moiré periodicity to be modified. Although such high-indexed Moiré patterns have been studied in details in theory, their appearance in STM images of real heterostructures is rather new.
- The complexity of the patterns is further increased if commensurate layers are analyzed. Besides the higher-indexed Moiré periodicity, the length of commensurate supercells also shows up with strong contrast in STM images, highlighting the influence of the small atomic deviations on the electronic properties.
- Exceptional variability of the spatial distributions of the charge density in the $MoTe_2$ layer is further investigated by DFT calculations, which also revealed the crucial role of the coupling with the underlying graphene substrate.

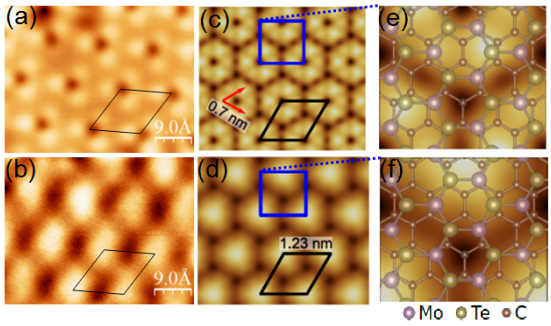

Figure: STM images of the Moiré 2√3R30° 1H-$MoTe_2$/graphene heterostructure recorded at 0.2V/200pA ***(a)*** *and 1.0V/100pA* ***(b)****;* ***(c, d)*** *simulated images calculated for the same +0.2V and +1V bias voltage;* ***(e, f)*** *the smaller spatial dimension zoomed on the blue square as marked in (c) and (d), respectively. C stands for the sp^2-bonded carbon in graphene on terminated 6H-SiC(0001). The Moiré supercell is marked by black rhombus on the corresponding image.*

References

[1] A. C. Gadelha, D. A. A. Ohlberg, C. Rabelo, E. G. S. Neto, T. L. Vasconcelos, J. L. Campos, J. S. Lemos, V. Ornelas, D. Miranda, R. Nadas, et al., Localization of lattice dynamics in low-angle twisted bilayer graphene, *Nature*, 590 (2021), pp. 405-409.

[2] Y. Cao, V. Fatemi, S. Fang, K. Watanabe, T. Taniguchi, E. Kaxiras and P. Jarillo-Herrero, Unconventional superconductivity in magic-angle graphene superlattices, *Nature*, 556 (2018), pp. 43-50.

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