Unravelling Structural Complexity in Twisted Multilayer Graphene: A Microscopy Perspective

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Abstract

Recent advancements in the synthesis of moiré materials have significantly impacted fields such as twistronics, straintronics, and valleytronics, owing to their ability to modulate the electronic band structure without altering the chemical composition[1]. In particular, twisted multilayer graphene (TMLG), which hosts multiple moiré superlattices, has emerged as a promising system exhibiting unique electronic and optical phenomena. In this study, we investigate the structural characteristics of TMLG with a high degree of two-dimensional (2D) crystallinity, synthesized via a simple Joule heating method[2,3]. We focus on unravelling the inherent twist angles, out-of-plane distortions (such as wrinkles and ripples), and other emergent features arising from random stacking that occurred during quench-driven growth. These structural characteristics are systematically characterized using transmission electron microscopy (TEM) and Raman spectroscopy, offering deeper insight into stacking variations, moiré superlattice formation, and an atomic-scale understanding of out-of-plane deformations in tMLG. As a complementary approach, a few-nanometer-thick Ag layer deposited on tMLG was employed as a mapping probe to visualize and correlate local structural variations across the surface.

References

[1] E.Y. Andrei et al., Nat. Rev. Mater. 6 (2021) 201.
[2] U. Mogera et al., J. Phys. Chem. Lett. 6 (2015) 4437.
[3] N. Gupta et al., J. Phys. Chem. Lett. 11 (2020) 2797.

Figures

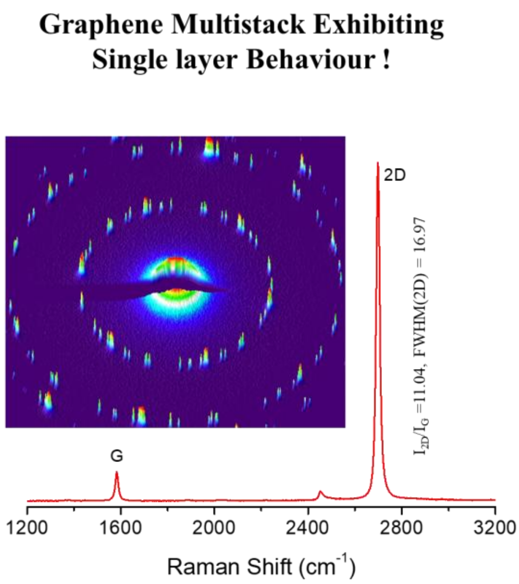

Figure 1: Raman spectrum of twisted multilayer graphene (tMLG), showing the characteristic G and 2D bands. The inset displays the corresponding selected area electron diffraction (SAED) pattern, confirming the presence of rotationally misoriented graphene layers.