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Abstract:

The stacking of atomically-thin, or two-dimensional (2D), layered materials into Van der Waals (VdW) films vastly expands the richness of planar material systems. The intrinsic sensitivity of 2D films to their surfaces and interfacial environment yields interlayer interactions within multi-layered films that induce a range of new material characteristics, and that are highly sensitive to perturbations like rotational misorientation[1] and lattice defects. In particular, the study of defects on VdW films is important for understanding their impact on interlayer interactions, and also as a means to modify the constituent layers to engineer band structure, to improve electrical contact, or to synthesize new heterostructures.

Twisted bilayer graphene (TBG), a double-layer of graphene characterized by a non-zero angle of misorientation between layers (the “twist” angle), is one of the simplest VdW structures.[2] By varying the twist angle of an electronically-coupled graphene bilayer, we create a rich, tunable electronic band structure together with singular optical properties; for example, enhanced absorption at a critical wavelength that is twist angle dependent.

In this work, we synthesize TBG and explore the influence of defects on interlayer coupling. Defects are introduced by either ion bombardment (vacancy-type defects) or by chemical functionalization with fluorine or oxygen (sp^3 -defects). We consider defected TBG with uniformly distributed defects throughout both layers, as well as half-defected TBG with defects confined to one layer.

The influence of twist angle on the Raman spectra of TBG is understood,[3] but the interplay between defects and interlayer coupling in TBG is complex and unexplored. In this work, we explore the influence of defects on interlayer coupling with Raman spectroscopy, for a range of twist angles, defect densities, and defect types. We explore the change in Raman spectra as a function of defect density, and facilitate the use of Raman spectra for the characterization of defects in TBG. The influence of defects on both first- and second-order Raman modes is studied, as is the influence of defects on the characteristic optical response of TBG.

Altogether, this work explores the influence of defects on interlayer interactions in TBG, elucidates the Raman response of defected TBG, and provides guidance for the rational integration of defected 2D nanomaterials into VdW films.

[1] T.E. Beechem et al., ACS Nano, doi:10.1021/nn405999z

[2] J.T. Robinson, S.W. Schmucker, et al., ACS Nano 7, 637-644 (2013)

[3] S. Coh et al., Phys. Rev. B 88, 165431 (2013)