Optical Properties of Twisted Bilayer Graphene

H. Zhuang and R. G. Hennig

Department of Materials Science and Engineering, Cornell University, Ithaca, NY 14850, USA.

Using density functional theory (DFT) and tight binding (TB) calculations, we study the band structure of bilayer graphene as a function of twist angles and compare the results to spectroscopic measurements. To test the accuracy of different exchange-correlation functionals in DFT, we first compare the band structure with DFT and G_0W_0 approximation for bilayer graphene systems with different twist angles. Our DFT results agree with previous DFT calculations for the weak coupling between the two layers and the band structure. Calculations of the quasiparticle dispersion using the G_0W_0 approximation show that DFT underestimates the Fermi velocity of bilayer graphene and the bandgap away from the K point. Scaling of the DFT band structure by an empirical parameter that depends on the twist angle accounts for most of the difference between the DFT and the G_0W_0 approximation. Based on the G_0W_0 calculations, we test a set of tight-binding parameters for the interlayer coupling Hamiltonian. Using this tight-binding model and DFT we study the optical matrix elements M_{op} for the intralayer transition and parallel band transition. Our results reveal that parallel band transition plays an important role in the observed G band resonance in Raman spectroscopy for specific twist angles [1]. We also study the electron-phonon coupling strength using the deformation potential approximation. Combining optical matrix elements with the electron-phonon coupling strength, the Raman intensity curves for various twist bilayer graphene are calculated. The results of these Raman line shapes are consistent with experiment.

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[1] R. W. Havener, H. Zhuang, L. Brown, R. G. Hennig, and J. Park, Nano Letters, submitted.

Contact: hz243@cornell.edu