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### Chemically-Modified, Twisted Bilayer Graphene: A Tunable Platform for Functionalization and Intercalation Studies

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Bilayer graphene is a diverse material system that can be extended by the introduction of an interlayer misorientation angle, the “twist angle” ( $\theta$ ). By varying  $\theta$ , a range of electronic and optical features can be tailored in electronically coupled, twisted bilayer graphene (TBG).<sup>1-2</sup> TBG properties continuously vary from those of Bernal stacked ( $\theta=0^\circ$ ) bilayer graphene as the twist angle is increased. Extending this idea, we demonstrate the influence of  $\theta$  on the chemical properties of TBG. We fabricate large-area, electronically coupled TBG films in a multi-step transfer process starting from graphene grown by low-pressure CVD on Cu foil.<sup>3</sup> Using optical microscopy and Raman spectroscopy, we classify TBG domains into one of five twist angle categories, where individual TBG domains range in size from  $\sim 10 - 100 \mu\text{m}$ . The influence of fluorine and oxygen functional groups on these TBG films is subsequently characterized as a function of twist angle. As a control for non-selective defect introduction, we use ion-implantation to uniformly introduce vacancy-type defects into TBG films.

Following these treatments, Raman spectroscopy is used to probe both the defect structure of TBG and the influence of each process on interlayer coupling, which expresses a unique resonance signature in Raman spectra.<sup>2</sup> Due to the large domain size, together with the optical contrast provided by TBG, optical microscopy is a useful and efficient technique for characterizing the influence of twist angle on each process. Atomic force microscopy (AFM) and X-ray photoelectron spectroscopy (XPS) provide further insight into the structure and bonding configuration of each system. Finally, we take advantage of the optical contrast provided by TBG to explore the intercalation of fluorine into artificial few-layer graphene structures. For a range of conditions, we find fluorination with  $\text{XeF}_2$  at room temperature is restricted to the outermost surface of few-layer graphene, while subsurface layers remain electronically coupled and chemically pristine.

[1] G. Li et al., *Nat. Phys.* **6**, 109-113 (2010)

[2] R.W. Havener et al., *Nano Lett.* **12**, 3162-3167 (2012)

[3] J.T. Robinson, S.W. Schmucker, et al., submitted (2012)