# Tunable Interlayer Excitons in Folded & Twisted Graphene

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We demonstrate how resonantly-excited interlayer electronics states in twisted bilayer graphene (*t*BLG) form a carrier relaxation bottleneck that enables weak photoluminescence (PL) emission and enhanced photocurrent (PC) extraction potential. Similar interlayer electronic properties are also seen in ~50 nm graphene folds that we resolve optically by selective fluorination (see Fig. 1). After resonant two-photon excitation of twisted bilayer graphene (*t*BLG), we show in Fig. 2ab, weak light emission that is spectrally tunable with the stacking angle, θ. These signals are best ascribed to PL associated with stabilized bound interlayer exciton states[1].

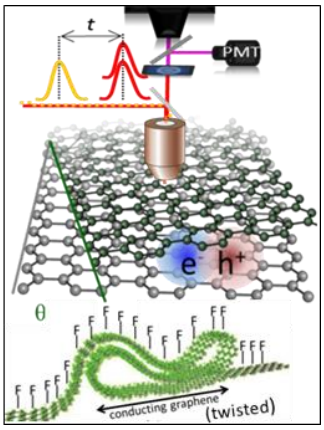


Fig. 1- Resonantly excited interlayer *e-h* pairs are measured in *t*BLG and twisted graphene nanofolds by 2-photon PL & intrabrand TA.

The exciton binding energy and long-lived kinetics of individual *t*BLG domains are estimated through two-photon PL excitation spectra and intraband transient absorption spectra. Spectral peaks from both two-photon photoluminescence and transient spectrum of intraband exciton transition independently suggest interlayer exciton binding energies ranging from ~0.5 to 0.7 eV for stacking angles of 8° to 17°[1,3]. This report of resonant PL from twisted bilayer graphene materials under ambient conditions is best explained by a transient coexistence of strongly-bound interlayer excitons and metallic graphene continuum states. Such θ-tunable interlayer excitons states may permit new optoelectronics by directing energy and charge.

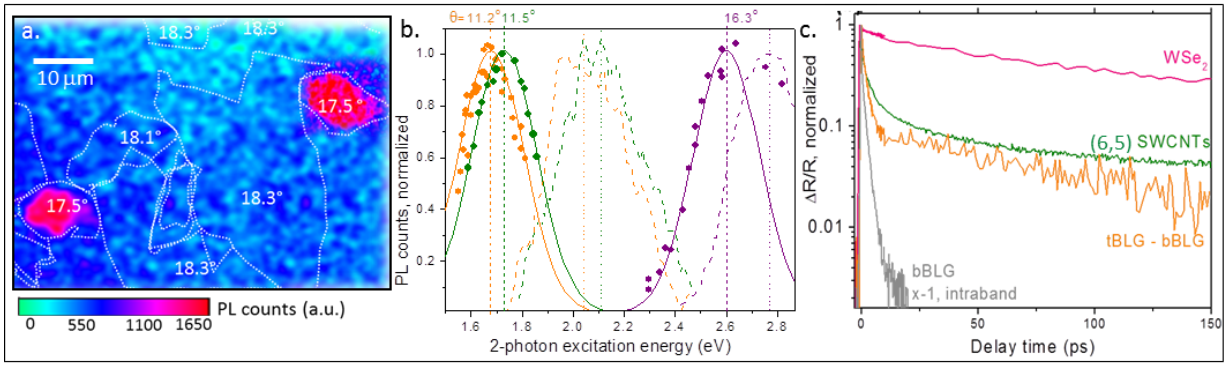


Fig. 2 – **(a)** Spatial PL map of *t*BLG for resonant 2-photon excitation of the 17.5° domains. **(b)** θ-tunable 2-photon PLE spectra (*circles*) for three stacking angles show a dark state lying below the linear absorption spectra (*dashed*, $\sigma_{tBLG}$-$2\sigma_G$). **(c)** Comparative TA relaxation kinetics[2] of resonantly excited *t*BLG, 0° *b*BLG, (6,5) SWCNTs, and few-layer $WSe_2$ show an initial electronic relaxation bottleneck for twisted bilayer materials.

## References

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[2] H. Patel, R. Havener, L. Brown, Y. Liang, L. Yang, J. Park, M.W. Graham, *Nano Letters*, 15, 5932-7 (2015)
[3] Y. Liang *et al.*, *Phys Rev B*, 90, 115418 (2014)