# New second order superlattice-induced Raman peaks in twisted bilayer and defective graphene

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Abstract
Raman spectroscopy is one of the most widely used techniques to characterise carbon materials, such as graphene, carbon nanotubes and amorphous carbons, as it is a fast, non-destructive and easy to handle technique [1].
The second order of the G peak, the 2G peak, cannot be seen in the Raman spectrum of single layer graphene (SLG) [2]. This is because the two-phonon density of states vanishes, as expected from the linear dispersion close to the $E_{2g}$ (G) mode at Γ due to the Kohn anomaly of graphene. However, here we show that this can be detected in twisted bilayer graphene (tBLG) samples, Fig. 1a. We use chemical-vapour-deposition-grown samples where small islands of randomly oriented bilayers are formed close to nucleation points. The 2G detection is explained as follows. The electronic structure of tBLG exhibits angle-dependent van-Hove singularities at the energy where the Dirac cones from each layer intersect [3,4]. The interaction between the two layers opens gaps at the intersection of the cones [5,6]. These gap openings produce big changes in the optical absorption spectrum, and induce a resonance Raman effect whereby the G peak intensity exhibits an important dependence on the twist angle and the excitation energy [7]. We show that close to the M point, i.e. close to the optical resonance, second order processes involving two phonons with momentum q → 0 and energy E → ħωG are in resonance confirmed by simulations. In addition to that we find additional second order Raman features in highly defective graphene that can be explained as superlattice induced peaks.

## References

[1] Ferrari and Basko, Nature Nanotechnol., 8 (2013) 235.
[2] Ferrari, et al., Phys. Rev. Lett., 97 (2006) 187401.
[3] Havener, et al., Nano Lett., 12, (2012) 3162.
[4] Moon, et al. Phys. Rev. B, 87, (2013) 205404.
[5] Carozo, et al., Nano Lett. 11, (2011), 4527.
[6] Ott, et al., to be submitted
[7] Wu, et al. Nature Comm., 5, (2014) 5309.

## Figures

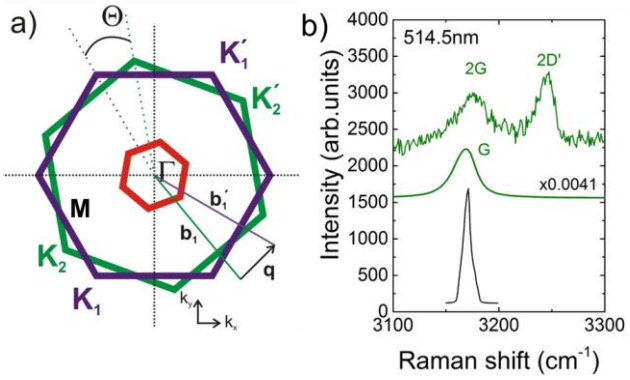


**Figure 1:** a) Structure of twisted bilayer graphene (tBLG). b) New second order Raman peak in tBLG which can be assigned to the second order of the G peak. The black line shows the simulations.