The 2G peak in twisted bilayer graphene

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The 2G peak cannot be seen in the Raman spectrum of single layer graphene (SLG) [1]. This follows from the backscattering condition of the real-space picture, which states that phonons that scatter electrons and holes backwards contribute the most to the two phonon Raman spectrum [2]. Here we show that a peak close to double the G peak position can be detected in twisted bilayer graphene (tBLG) due to its peculiar electronic band structure. This is distinct from the double resonant 2D', which is also observed. We consider chemical vapor deposited samples where small islands of randomly oriented bilayers are formed close to nucleation points [3]. Figure 1b plots the measured Raman spectra [4] of three representative tBLG samples with different twist angles θ = 9.6°, 12.9° and 14.9°, and their corresponding 2G features compared to SLG.

The 2G detection is explained as follows. The electronic structure of tBLG exhibits angle-dependent van Hove singularities vHS(θ) at momentum **k≈M** where the Dirac cones from each layer intersect [5,6] (see Fig.2a and b). The interaction between the two layers opens gaps at the intersection [5,6] (see Fig.2c). The optical absorption is thus dominated by vHS(θ) and induces a resonance Raman effect whereby the G peak intensity exhibits a dependence on the twist angle and the excitation energy [7]. The electrons and holes excited close to the **M** point can undergo a second order scattering process with two phonons with momentum ±**q** close to 0 and energy close to $\hbar\omega_G$ involving real electronic states. A 2G Raman feature is seen because the process fulfils both the electronic resonance condition and the momentum requirement ±**q** imposed by the backscattering condition. On this basis, 2G features are expected in graphene related systems with vHs and inter-band gaps where electronic resonance and backscattering is possible (e.g., twisted trilayers, MWNTs).

References

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Figures

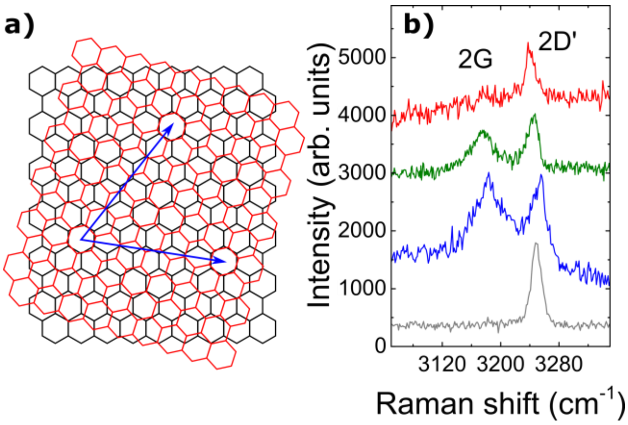

Figure 1: a) Schematic representation of a tBLG. b) Measured Raman spectra of tBLG with twist angles red: θ=9.6°, green: θ=12.9°, blue θ=14.9° compared with graphene (grey) showing the new feature close to 2G [3].

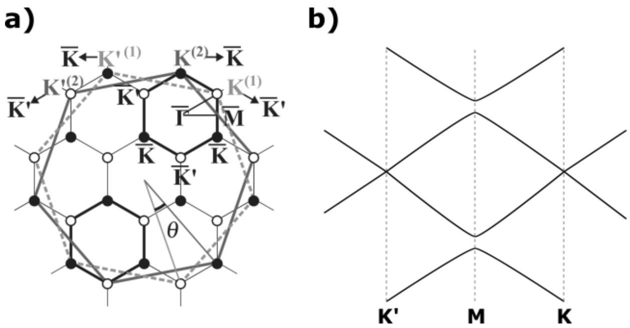

Figure 2: a) Corresponding Brillouin zone with the equivalence of **K** and **K'** from each layer [6]. b) Corresponding π-π* band structure.