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 \( \Gamma \) 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 \( q \rightarrow 0 \) and energy \( E \rightarrow \hbar \omega G \) 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
[6] Ott, et al., to be submitted

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.