Families of new Raman peaks in twisted bilayer graphene observed by UV Raman spectroscopy

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In this work we report a UV Raman study of graphene bilayer samples grown by chemical vapor deposition on a copper foil, using different incident laser energies between 2.54 and 4.14 eV. The Raman spectra show a number of extra peaks, classified in different families, and are associated with Moiré patterns of twisted layers of graphene, each family of peaks being associated with a different twist rotational angle. We theoretically analyze these extra peaks considering the set of discrete wavevectors of the Moiré pattern within the interior of the Brillouin zone of graphene, which activate special double-resonance Raman processes. The different peaks within each family correspond to phonons of different branches in graphene (LA, iTO and LO). Our result are ascribed to an umklapp double-resonance process in graphene systems, and allowed us to introduce experimental values of frequencies in the graphene phonon dispersion, which are probed by UV Raman spectroscopy.