Evolution of Raman G and G’ (2D) Modes in Folded Graphene Layers

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Abstract

Electronic band structures of graphene layers are remarkably influenced by the ways of these carbon atomic layers stacking themselves. Different electronic band structures could be clearly reflected by different spectral features of Raman G and G’ or 2D modes through strong electron-phonon coupling [1-3]. Such unique optical response promises Raman spectroscopy to be one of the most widely adapted techniques to quickly and precisely identify thickness of pristine Bernal-stacked graphene layers[4]. However, for folded or twisted graphene layers, even their thickness is identical to that of the Bernal-stacked graphene layers, the Raman spectra could be significantly different.

In this work, Bernal- and non-Bernal-stacked graphene layers have been systematically studied by Raman imaging and spectroscopy. It is found that two dominant Raman modes, G and G’ exhibit three types of Raman spectral features when interlayer lattice mismatched, defined by twisting angles varies. Among these folded graphene layers, the most interesting one is the folded graphene layers that present an extremely strong G mode enhanced by a twist-induced Van Hove singularities. The evolution of Raman G and G’ modes of such folded graphene layers are probed by changing the excitation photon energies. For the first time, doublet splitting of the G’ mode in folded double-layer (1 + 1) and the G mode in folded tetra-layer (2 + 2) graphene are clearly observed. The G’ mode splitting in folded double-layer graphene is attributed to the coexistence of inner and outer scattering processes and the trigonal warping effect. While the two peaks of the G mode in folded tetra-layer graphene are assigned to Raman-active mode ($E_{2g}$) and lattice mismatch activated infrared-active mode ($E_{1u}$), which is further verified by the temperature-dependent Raman measurements. Our study provides a summary and thorough understanding of Raman spectra of Bernal- and non-
Bernal-stacked graphene layers and further demonstrates the versatility of Raman spectroscopy for exploiting electronic band structures of graphene layers.

References


Figures