NOVEL SOURCES OF RAMAN D-BANDS IN NANOCARBONS: INCOMMENSURATE STACKINGS AND FOLDS

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Introduction

Raman measurements provide a unique window into the microstructure of sp2 carbon systems. The graphene family of materials provides a particularly rich are of investigation on the interplay of geometry and Raman response in carbon. For example, the Raman D-band in carbons is generally taken to be indicative of disorder, since the mode in question is not at the zone center and hence does not participate in Raman scattering at lowest order is momentum conservation is respected [1,2,3]. However, all that is required for a D-band response is to break translational symmetry, and disorder per se is not strictly required. Monolayer graphene provides several unique mechanisms for breaking translational symmetry and thereby activating the D-band without disorder.

Skew-stacked Graphene Bilayers

For example, if a single layer of graphene is folded back onto itself at a skewed angle, then the two layers in contact will rest in a skewed orientation. Treating one layer as a perturbation on the other, it imposes an ordered scattering potential, but one that is skewed at an angle to the crystalline lattice of the other sp2 layer. Depending on the precise skew angle, there may exist reciprocal lattice vectors of the perturbing potential that can facilitate momentum conservation for the double-resonant D-band process, playing the role that is played by the disorder potential in disordered carbons, but at a discrete set of scattering wavevectors rather

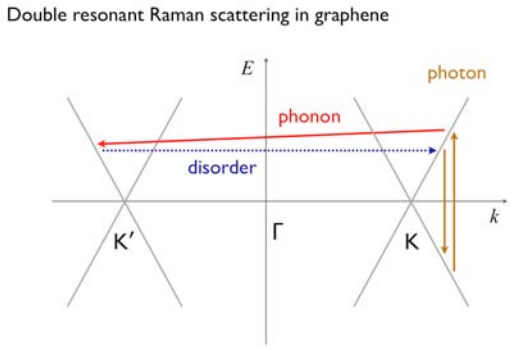

Fig. 1: The double-resonant Raman scattering process is normally facilitated by disorder, which compensates for the finite wavevector of the phonon involved.

than the Fouirer continuum of a localized scatterer. This pinning of the double resonant process in momentum space implies that the resulting D-band peak will not disperse (i.e. change in frequency) as the laser energy is varied, a result at variance with previous observations of D-band scattering in carbons.

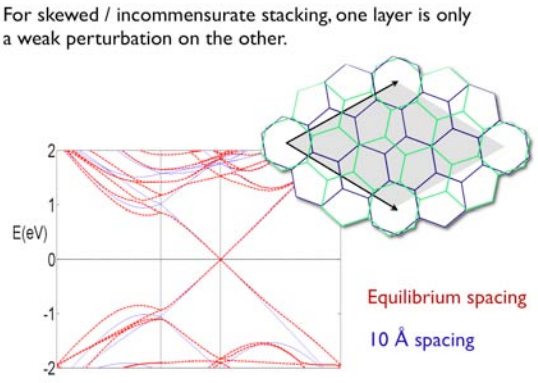

Fig. 2: The electronic structure of skew-stacked graphene is closely approximated by that of two independent monolayers, indicating that a perturbative treatment of interlayer scattering is viable.

Folds in Monolayer Graphene

In addition to disorder, the edges of a graphene sheet can also activate the D-band. A fold in a graphene monolayer is not an edge, since the graphene sheet is actually continuous, but it does provide a deviation from translational symmetry due to *e.g.* the spatially inhomogeneous curvature around the fold. Raman scattering from folds in single-layer graphene sheets can show a D-band at the fold for both incommensurate and commensurate folding, although the parent single-layer graphene lacks a D-band.

Acknowledgments: VHC, AG, PEL, CN and YT acknowledge the friendship and scientific acumen of Peter Eklund, who passed away as this work was in progress.

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