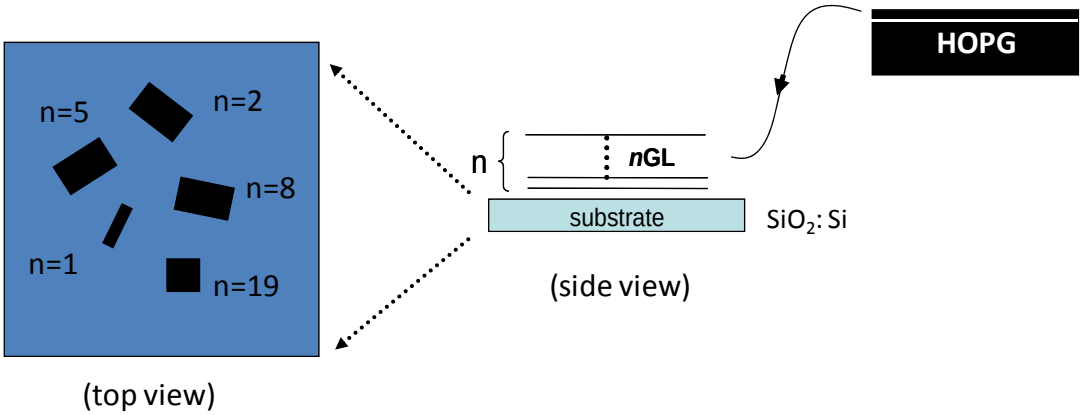
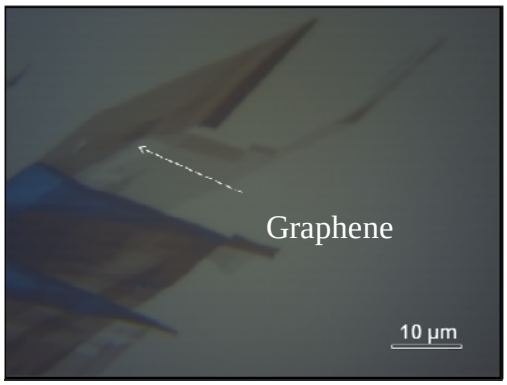
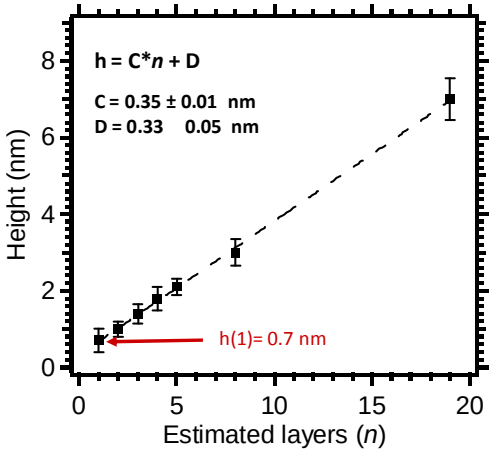
1. Preparation of Graphene – Mechanical Exfoliation

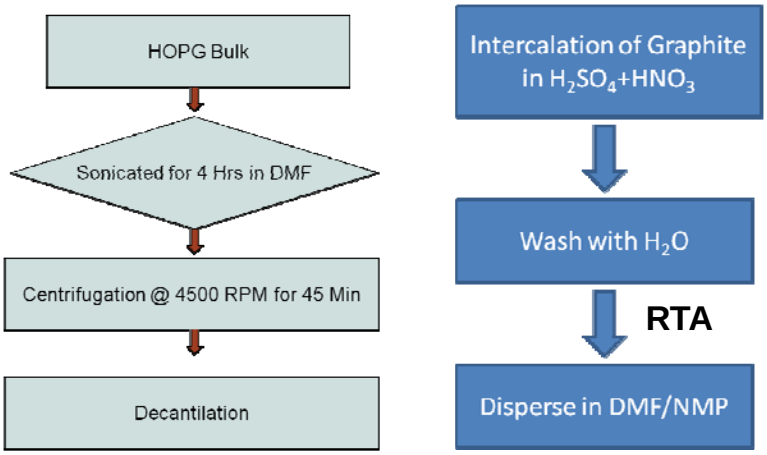
Graphene (n=1) and nGL (n=2-4) samples can be prepared by micro-mechanical cleavage of highly oriented pyrolitic graphite (HOPG). The cleaving process occurred either while transferring a thin film from HOPG (438HP-AB, SPI, Inc.) onto Scotch tape (3M, Inc.), or afterwards, when rubbing the tape against the substrate.

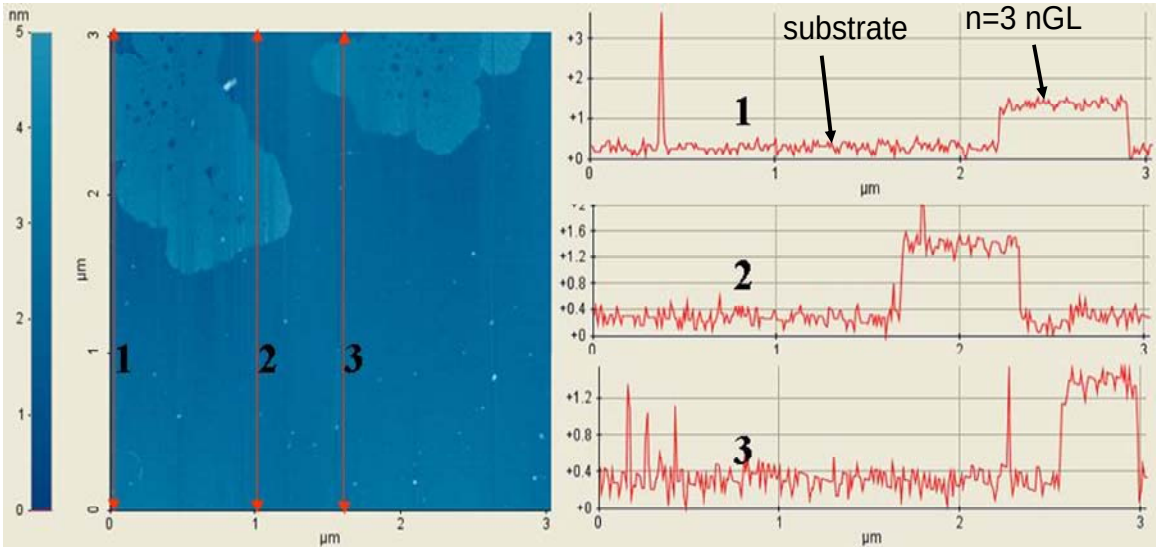






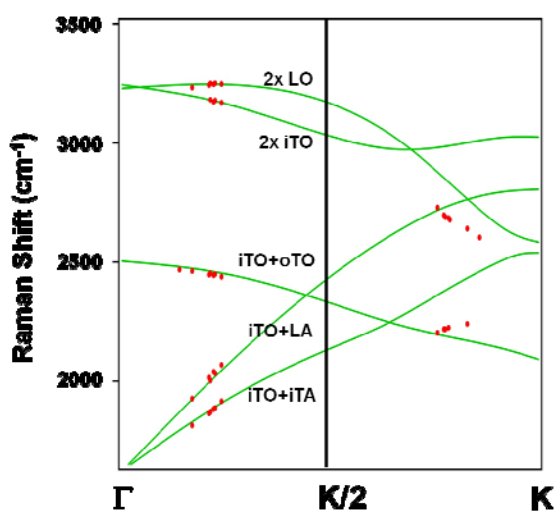
2. Preparation of Graphene- Chemical Method





3. Dispersive Raman Scattering from n-Graphene Layer Films

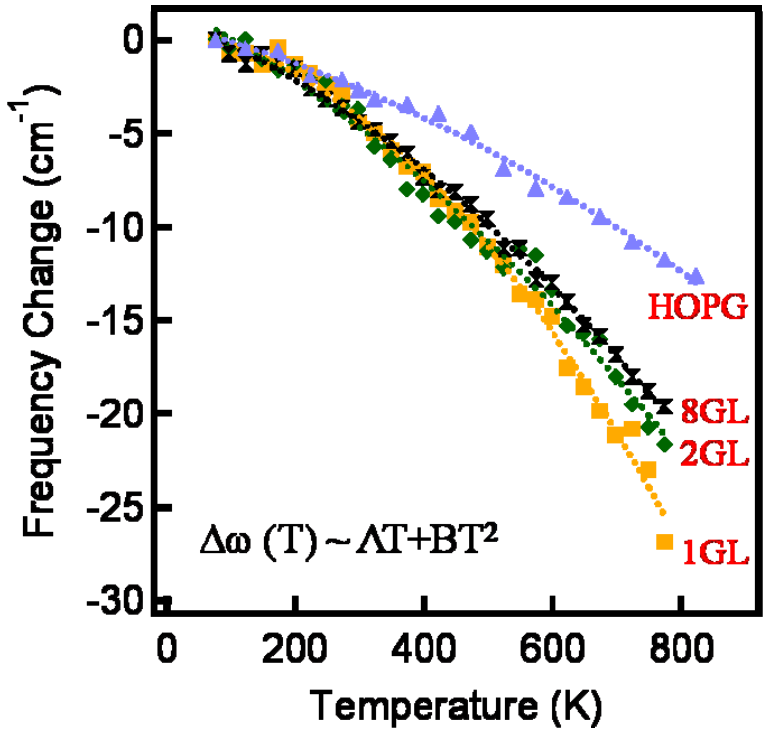
Resonant Raman scattering studies of n-graphene layer films (*n*GLs; n=1-4). We follow the scattering process to sixth order and observe many new Raman peaks which participate in double resonance (DR) Raman scattering. Many of the Raman bands exhibit linear dispersion, i.e., $d\omega/dE$=constant, where ω is the Raman peak frequency and E is the excitation photon frequency. This behavior will be shown to stem from DR, the dispersion of the electronic and phonon states involved, and also on the order of the scattering. For example, we find values in the range of $-20 < d\omega/dE < 170$ cm^{-1}. The band dispersions also exhibit an interesting dependence on the number of layers n and can be used to map out phonon dispersion of *n*GLs for comparison with theoretical calculations



Phonon dispersion (dots) (n=1) based on 2nd order Raman spectra. Theoretical curves (solid lines) are taken from Saito *et. al*., PRL (2002)

4. Temperature Dependent Raman Scattering from n-Graphene Layer Films

Results from a study of Raman G-band ~ 1585 cm-1 as a function temperature T and on (n=1,2,3) were obtained. Data for the frequency and linewidth will be discussed in terms of contributions from the electron-phonon interaction, negative lattice expansion, and multi-phonon processes. The T-behavior depends strongly on whether the nGL is supported on Si substrate or freely suspended. The presumption is that the suspended films exhibit intrinsic behaviour, while the supported films do not. Results for the change in the G-band frequency over the range 80<T<800 K for n=1,2,8 supported on SiO_2/Si and graphite (HOPG) are shown. A noticeable quadratic T-dependence is observed over this wide range of temperature.

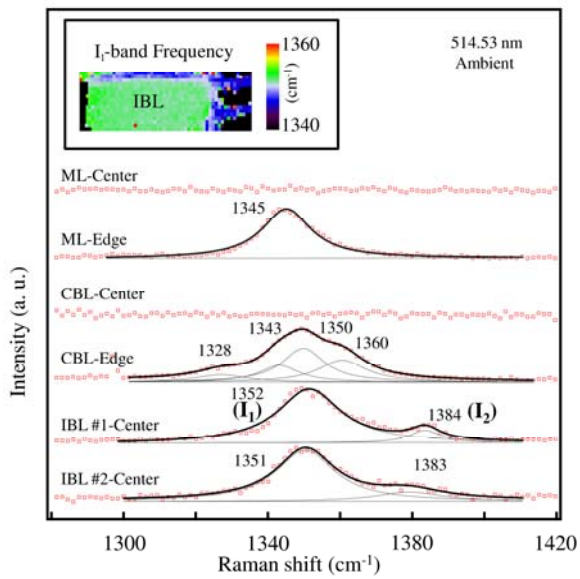


Temperature dependent G-band frequency shift for *n*GLs supported on Si/SiO_2

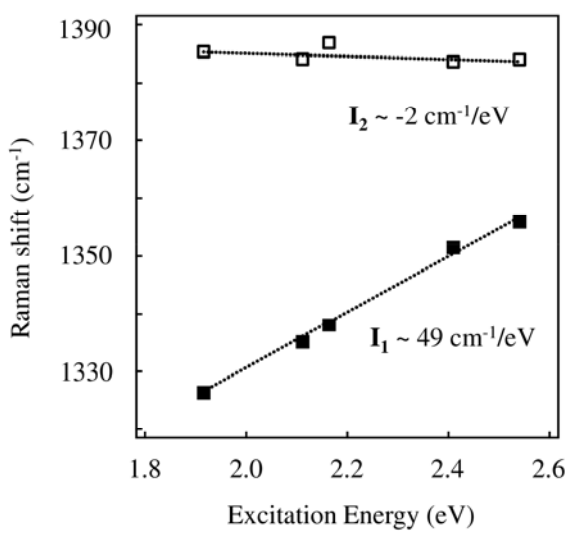
5. Raman Scattering from Incommensurately Stacked Bi-Layer Graphene

Despite the weak interlayer interaction between graphene layers, our Raman scattering studies on incommensurately stacked (IS) bi-layer grapheme (n=2) reveal an altogether different Raman spectrum commensurately stacked (CS) n=2 films as opposed to IS n=2 films. We find that IS layers activate a new band ("I-band") for sp^2 bonded carbon near ~1350 cm^{-1} which has two components I_1 and I_2 (Figure 3): I_1 is dispersive at ~50 cm^{-1}/eV, while I_2 exhibits very weak

dispersion(Figure 4). These Raman peaks are not due to ordinary D-band scattering in sp^2 carbons that is normally associated with defects. From the shape of the 2nd order 2D band at ~2700 cm^{-1}, we see evidence for a large decrease in the electronic subband splitting due to IS layers. A theory is proposed to explain these new results in IS n=2 graphene.



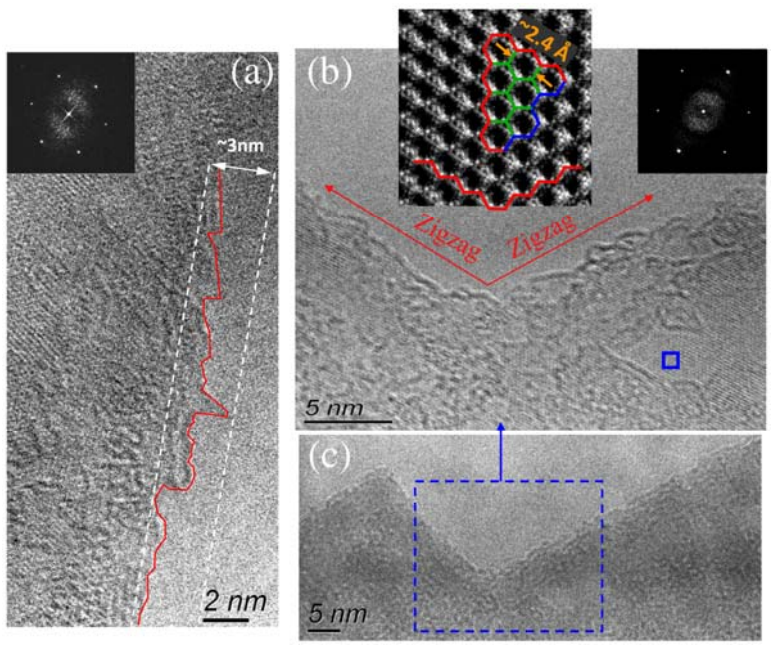
Raman bands near ~1350 cm^{-1} for n=1 monolayer (ML), and n=2 C and I stacked films. The ML-Edge spectrum exhibits normal D-band scattering.



Dispersion of I_1 and I_2 Raman bands

6. Localized Raman Scattering from Graphene Edges

TEM and Raman scattering show that a typical grapheme edge prepared by micromechanical cleavage from HOPG is, on average very straight, but at short range, can be seen meandering by about ~ ± 2 nm and thereby presenting a mixture of zigzag and armchair local symmetries. Nevertheless, we find that these "real" edges exhibit polarized scattering as if the absorption and re-radiation of the photons were made by a line antenna aligned along the average direction of the edge. Scanning the excitation beam from off the sample and then across the edge allows the observation of the onset of the G-band with distance, as well as a D-band localized within w~ 50 nm of the edge. A value for w can be estimated by analyzing the convolution of the Gaussian laser beam profile with the localized scattering which is assumed, for simplicity, to be constant and localized within w of the edge.

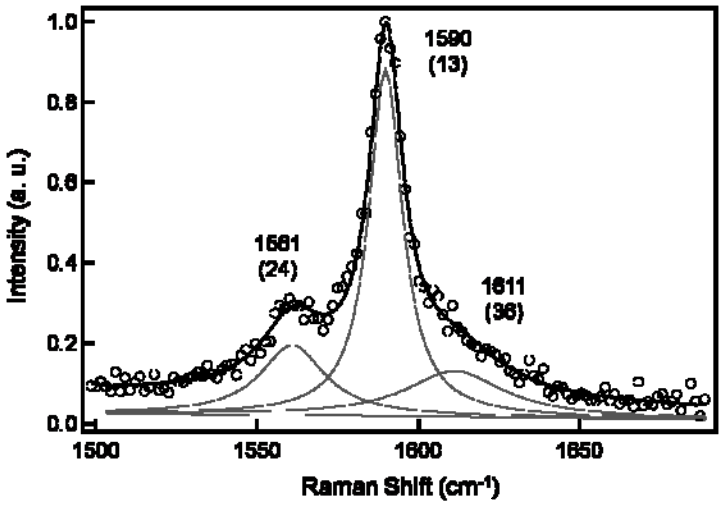


TEM of a graphene edge showing edge roughness ~ 1-3 nm.

7. Raman Scattering from Narrow Graphene Ribbons

Raman data collected on vary narrow (ribbon width ~ 2-3 nm) one- or two-layer graphene nanoribbons (GNRs). New G-bands, more similar to that observed in 1.2-1.6 nm diameter single-walled nanotubes than grapheme are observed . The GNRs were synthesized by H. Dai and co-

workers at Stanford. We discussed the activation of these new Raman modes in narrow GNRs in terms of transverse phonon confinement. Interestingly, several of these GNRs were observed to NOT exhibit a D-band.



Raman spectrum of 3-nm wide GNR showing extra peaks in the wings of the dominant G-band