Laser Induced Structural Modification of Single Layer and Bilayer Graphene

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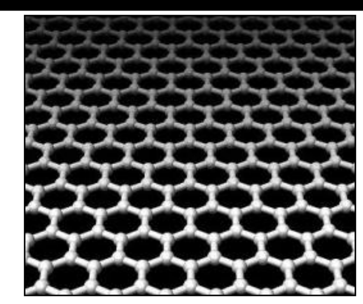
Graphene, the next Silicon?

Graphene, a single layer carbon honeycomb lattice, is a novel electronic material which was experimentally observed for the first time in 2004. Since then, graphene has attracted much attention from many research groups, in industry and academia. Some of graphene's characteristics include:



single layer graphene

- ***Record high Electron Mobility (200,000 cm^{2}V^{-1}s^{-1})***
- ***High crystal quality***
- ***Possibility of ballistic transport at room temperature (Allows for possiblity of creating ballistic transistors)***

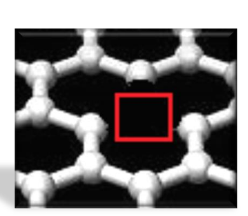
These characterisics have led to the exploration of graphene as a replacement for Silicon in the fabrication of electronic devices.

Defects

There are several motivations for studying defects on graphene.

- ***Fabrication/Metrology techniques are likely to Introduce defects***
- **Defects are desirable in certain electronic devices**

We are specifically interested in how graphene undergoes changes under laser irradiation. This is because Raman Spectroscopy is extensively used to characterize graphene.



- ***A defect on graphene comprises of a 'vacancy', or a missing Carbon atom from a lattice point.***
- ***Defects sites alter electronic properties of graphene.***

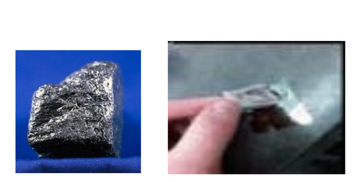
Goals and Methods

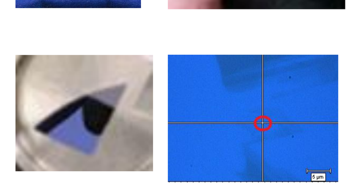
- ***Irradiate single layer graphene flakes with laser radiation at 514nm wavelength at varying power levels (4-18mW)***
- ***Perform time resolved Raman Spectroscopy to observe:***
  - ***Defect Creation***
  - ***Charge Carrier Concentration Changes***
- ***Study graphene flakes with a wide range of initial doping states.***
- ***Observe the effect of long term (~minutes) laser exposure as well as short (~seconds) laser pulses on graphene.***

Fabricating Graphene

We have systematically prepared and characterized graphene in the following steps :

1. Micromechanical Cleavage of Graphite
2. Deposit graphene on SiO_2/Si substrate
3. Look for graphene flakes using optical microscopy
4. Use Raman Spectroscopy to characterize flakes



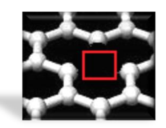


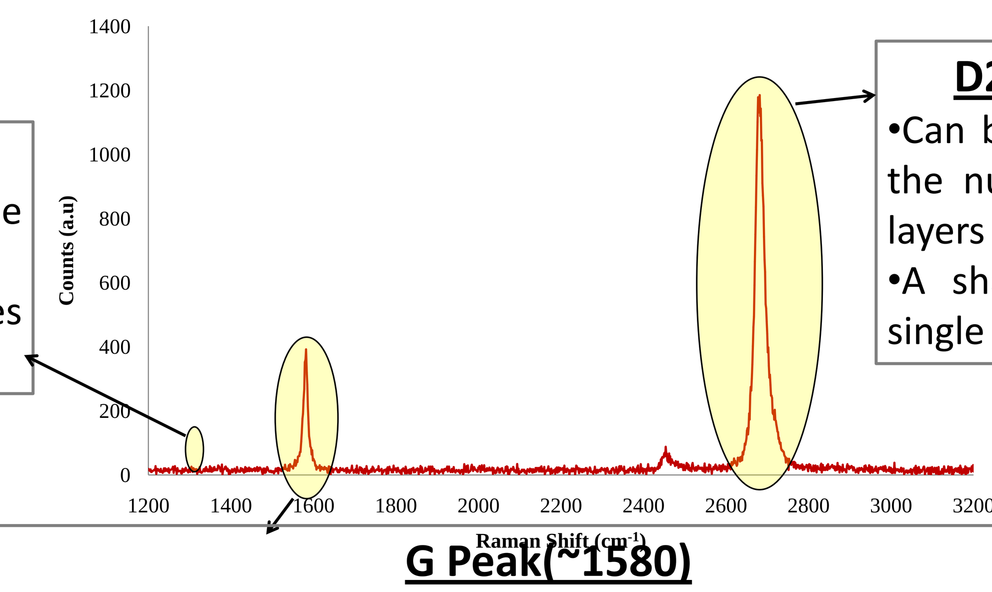
Probing Graphene: Raman Spectroscopy

Raman Spectroscopy

Raman Spectroscopy is an optical probing technique which detects inelastic light scattering from the material under observation. Raman spectroscopy can be used to '*fingerprint*' materials.

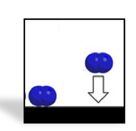
Raman Spectrum of Graphene





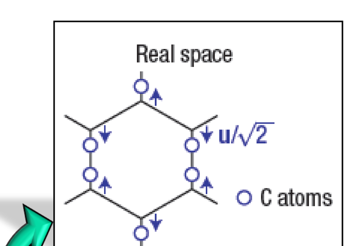
D Peak(~1350)
- Can be used to identify the number of graphene layers
- A sharp peak indicates single layer

D2 Peak(~2700)
- Can be used to identify the number of graphene layers
- A sharp peak indicates single layer



G Peak(~1580)
- FWHM and Peak Center are related to charge carrier concentration
- I2D/IG ratio gives initial doping state
- The peak originates from the E_{2g} phonon (see below)

The E_{2g} Phonon



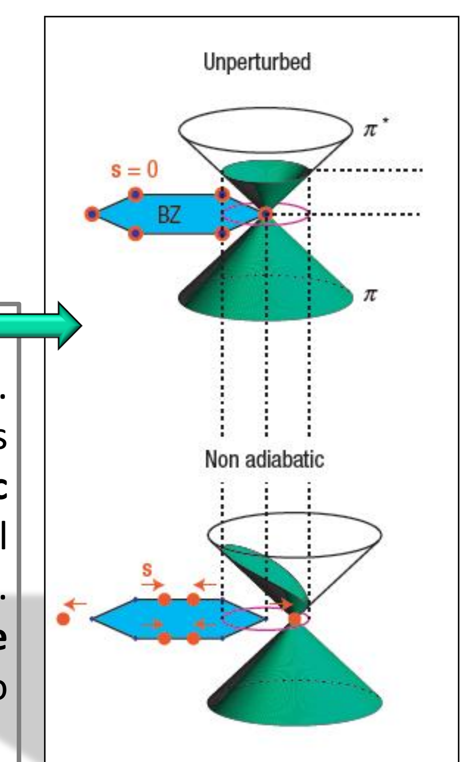
E_{2g} phonon mode /Source: Ref 3

Band Structure and Doping/Source: Ref 4

Phonons are vibrational modes of a crystal. The **E_{2g} phonon** is the mode where C atoms facing each other have opposite phase.

- Graphene's electronic structure consists of two Dirac cones with zero gap.
- In pristine graphene, energy levels up to Fermi Level are occupied. In doped graphene the occupation differs from the pristine case.

The E2g phonon perturbs the electronic structure. Hence, the phonon is **sensitive to electronic occupation**. Doping will change phonon properties. In addition, the **size of the graphene crystal** will also change phonon properties.



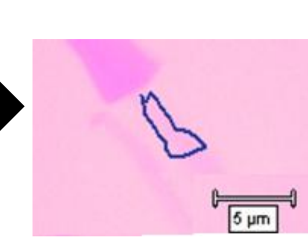
Perturbed band structure/Source: Ref 3

Time Resolved Raman Spectroscopy

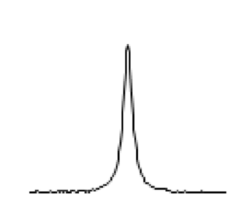
- From the explanation above, the G-Peak parameters can be used to determine system dynamics
- We perform time resolved Raman Spectroscopy to monitor system dynamics under irradiation.
- We specifically monitor changes in the E2g phonon by observing G-Peak parameters.
  - A **rising G-Peak center** indicates **defect creation**.
  - A **dropping G-Peak center** along with a **related rising G-Peak FWHM** indicates a **drop in charge carrier concentration**.

Experimental Methods

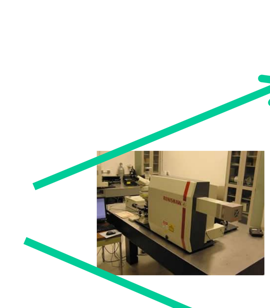
Fabricate Graphene



Optically Identify



Identify SLG using Raman Spectroscopy



Sequential

Pulse laser to high power and repeat measurement

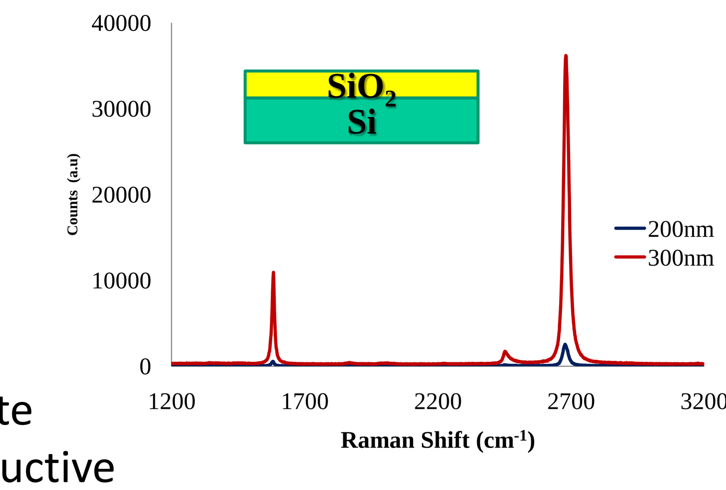
Continuous

Measure spectra back to back at high power

- All samples were irradiated with a 514.5nm laser

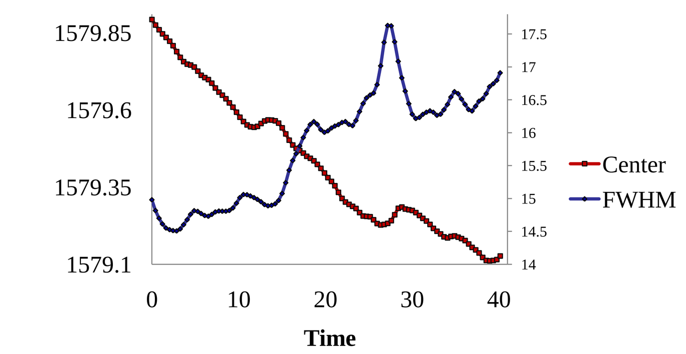
Results : Substrate Effects

- We observed strength of the Raman signal to be dependent on substrate thickness
- 300nm substrate gives a Raman signal that is several orders of magnitude greater when compared to 200nm substrate
- Prevailing explanation is that 300nm substrate gives theoptical path length that gives constructive interference.



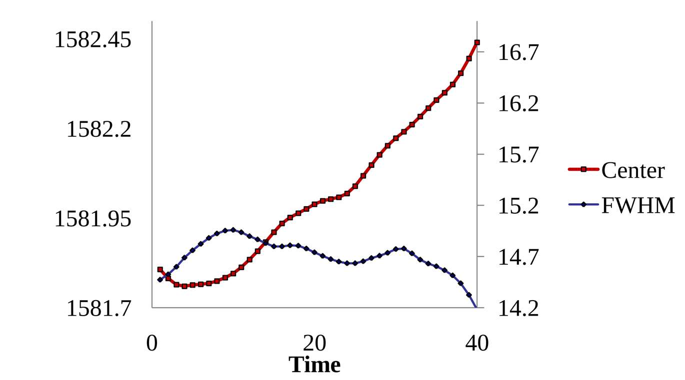
Results : Continuous Irradiation/High Doping

- Initial I2D/IG ration = 1.7 (High Doping)
- Irradiated at 4.5 mW
- Peak Center drops and FWHM rises
- Indicates drop in charge carrier density

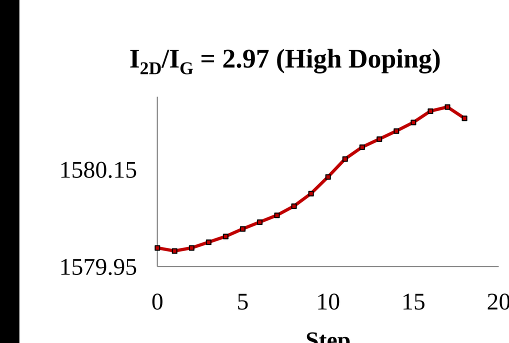


Results : Continuous Irradiation/Low Doping

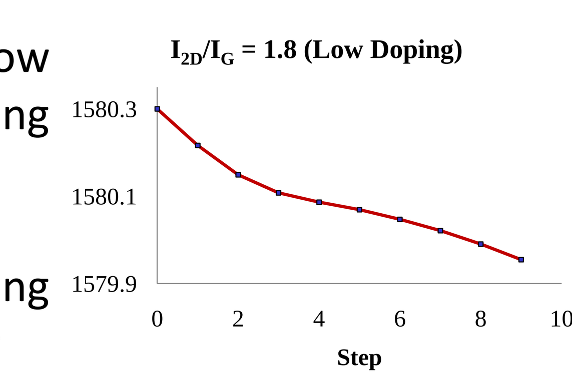
- Initial I2D/IG ration = 3.5 (Low Doping)
- Irradiated at 4.5 mW
- Peak Center rises with relatively small changes in FWHM
- Indicates defect creation with minor changes in charge carrier density



Results : Sequential Irradiation



- Flakes with low doping show rising peak center, indicating defect creation.
- Flakes with high doping show dropping peak center



Conclusions and Future Work

- **We have demonstrated that single layer graphene flakes with high doping levels respond differently to laser radiation than flakes with low doping levels.**
  - **Flakes with low doping levels develop defects as soon as they are exposed to laser radiation.**
  - **Flakes with high doping levels show changes in charge carrier concentration under laser irradiation.**
- **We have demonstrated that it might be possible to change structural and electronic properties of graphene using laser pulses.**

Future Work

- **We propose to study how graphene flakes respond to short laser exposure at higher power.**

Acknowledgements
- Prof. Yuri Gogotski, Materials Science and Engineering, Drexel University
- Dr. Zhorro Nikolov, Centralized Research Facilities, Drexel University

Key References

1. Raman Spectrum of Graphene and Graphene Layers, Ferrari et al, PRL (2006)
2. The rise of Graphene, Geim and Novoselov, Nature Materials (2007)
3. Breakdown of the adiabatic Born-Oppenheimer Approximation in Graphene, Pisana et al, Nature Materials (2007)
4. Laser Induced Disassembly of a Graphene Single Crystal into a Nanocrystalline Network, Krauss et al, PRB (2009)