Optical probe of the electronic excitations in pristine and heavily-doped multi-layer graphenes

Ping-Heng Tan
(谭平恒)

Institute of Semiconductors, Chinese Academy of Sciences
(中科院半导体所)

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What’s so special about graphene?

Graphene: Truely 2D system
Peculiar physical properties
- Massless Dirac Fermion
- Ballistic transport at LT
- Quantum hall effect at RT
- High mobility and high thermo-conductivity
- Nanoribbons with confinement gap

Observable with naked eye!

Optical contrast
Raman

Dimensionality of carbon

Diamond, graphite
3D

GRAPHENE, realized in 2004
(Novoselov, Science, 2004)

Carbon nanotubes
2D

1D

Fullerenes
0D
**Opto-electronic applications**

Spintronic device

Transparent electrode

FET

Battery
- Elad Pollak, *Nano Lett.* 2010

Touch Screen
- Ahn, *Nat Nanotech* 2010

Photodetector
- Bonaccorso, *Nature photonics* 2010

**Graphene Multi-layers**

Latil, *PRL* 2006

Ohia, *PRL* 2007

**Probing electronic excitations in graphene layers via phonons?**

Phonon could interact with electronic excitation.

Monolayer graphene

Multilayer graphene

**High carrier density/mobility in graphene layers is important for applications!**

- Distinctive band structure
- 

\[ E_{2g} \]

\[ 1582 \text{ cm}^{-1} \]

\[ 44 \text{ cm}^{-1} \]
Outline

- Probing Fermi level of heavily-doped graphene via 2D and G modes
- Probing low-energy electronic excitations near Dirac point via shear modes
- Conclusion

Probing electronic excitations in graphene layers via phonons G, 2D

GIC and its characterization

FeCl3-GIC

Pos(G) is very sensitive to the charge transfer and doping level in GIC

Raman spectra of FeCl3-intercalated FLG

613K, 6h
Probe Fermi energy of FeCl$_3$-intercalated GIC

\[ 2E_F = E_i - \hbar \omega_{ij} \]

\[ E_F \approx 0.85\text{eV} \]

In agreement with EELS: 0.9eV

WJ Zhao, PH Tan*, JACS 2011

The way to probe low-energy excitations in graphene layers?

Low-energy excitation near Dirac point in graphene layers

~< 10 meV

Ohta, PRL 2007

Raman modes in graphene and graphite

C mode: sensitive to interlayer coupling!

A challenge to detect this mode!
VBG technique for low frequency measurement

Single-stage spectrograph + Notch filter of Volume Bragg Grating

3.0 cm\(^{-1}\) Ge/Si SL

Linear chain model for the shear mode

Displacement:

\[ \omega_{\text{bLG}} = \sqrt{2} \sqrt{1 + \cos(\pi)\omega_{\text{b}}} \]

\[ \omega_{\text{bLG}} = \sqrt{2} \omega_{\text{b}} \]

Interlayer force constant per unit area:

\[ \alpha \sim 12.8 \times 10^{18} \text{ N m}^{-3} \]

Shear modulus:

\[ C_{\text{sh}} = 4.3 \text{ GPa} \]

PH Tan*, Nature materials 2012

Breit-Wagner-Fano lineshape of the C mode

Breit-Wagner-Fano lineshape:

\[ I(\omega) = \frac{I_0}{1 + \frac{2(\omega - \omega_0)}{\Gamma}} \frac{1 + \frac{1}{2q^2}}{1 + \frac{1}{2q^2}} \]

\[ I(\omega) = \frac{I_0}{1 + \frac{2(\omega - \omega_0)}{\Gamma}} \frac{1 + \frac{1}{2q^2}}{1 + \frac{1}{2q^2}} \]

\[ \omega_0 \] Uncoupled mode frequency

\[ \Gamma \] Broadening parameter

\[ q \] Coupling coefficient

FWHM = \( \Gamma (q^2 + 1)/(q^2 - 1) \)

Scott, RMP 1974

PH Tan*, Nature materials 2012
Probe the low energy transition near Dirac point in graphene layers

Raman spectroscopy is a versatile tool for probing electronic excitation in graphene layers.

1). Multi-wavelength Raman spectroscopy allows a direct measurement of the Fermi level for stage-1 FeCl$_3$-GIC.

2). The low energy of the C mode makes it a probe of electronic transitions near Dirac point, resulting in a Breit-Wigner-Fano lineshape due to resonance between the shear mode and electronic transitions.

Conclusion

Thanks for your attention.

Email: phtan@semi.ac.cn
Mobile: 13511036486
Website: http://skbms.semi.ac.cn

Any comments?