Intraband Optical Transitions in Graphene

Jason Horng[1], Chi-Fan Chen[1], Baisong Geng[1], Caglar Girit[1], Yuanbo Zhang[2], Zhao Hao[3,4], Hans A. Bechtel[3], Michael Martin[3], Alex Zettl[1,5], Michael F. Crommie[1,5], Y. Ron Shen[1,5] and Feng Wang[†1,5]
[1] Department of Physics, University of California at Berkeley, Berkeley, CA 94720, USA
[2] Department of Physics, Fudan University, Shanghai, 200433, China
[3] Advanced Light Source Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA
[4] Earth Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA
[5] Materials Science Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA
[†] To whom correspondence should be addressed. Email: fengwang76@berkeley.edu

Abstract: We measured tunable interband and intraband transitions in graphene using infrared spectroscopy. Graphene electrons have strong intraband absorption at terahertz frequency range. The absorption spectra are described by a Drude-like frequency dependence.
OCIS codes: (160.4760) Optical properties; (300.6340) Spectroscopy, infrared

1. Introduction

Graphene provides a unique material system to study two-dimensional Dirac electrons. Researchers have demonstrated in graphene fascinating phenomena both for electrical transport [1-3] and interband optical response [4-7]. However, experimental investigation of intraband transitions of the Dirac electrons has been lacking. Such intraband transitions are responsible for high-frequency conductivity of graphene in terahertz (THz) spectral range. Their studies can provide new insight into electrical transport in graphene.

Here we performed spectroscopic measurements of graphene inter- and intra-band transitions across THz to mid-infrared range at different carrier concentrations. We observe a dramatic increase in graphene intraband absorption upon electreical gating. The intraband absorption spectra have Drude-like frequency dependence. Through this frequency dependence we were able to determine independently the oscillator strength and scattering rate of Dirac electrons in graphene.

2. Experimental Approach

Fourier transform infrared spectroscopy (FTIR) was used to measure the transmission spectra of graphene samples in the infrared range. Due to the diffraction limit of the THz radiation, we needed a sample size larger than 300μm. Therefore, we chose the chemical vapor deposition (CVD) method [8] to grow our large-area graphene sample. Raman spectra confirm our samples are monolayer graphene.

We controlled the carrier concentration in graphene using electrostatic gating with the silicon back gate. We measured transmission spectra for different carrier concentrations in reference to that of the charge neutral point (CNP). All optical measurements were performed in vacuum (~ 0.1 mTorr) at 100 K while the conductance of sample was monitored.

3. Gate-induced change in inter- and intra-band transitions.

We show in Fig. 1 a typical transmission change spectrum through the graphene sample due to electrostatic gating. It shows two characteristic features: a decrease in absorption at higher frequency and a corresponding increase in absorption at lower frequencies. The physical mechanisms responsible for these absorption changes are illustrated in the inset of Fig. 1. With hole doping the Fermi surface shifts to a lower energy. Therefore interband transitions with transition energy below $2E_F$ become forbidden, and it leads to a decrease in higher frequency interband

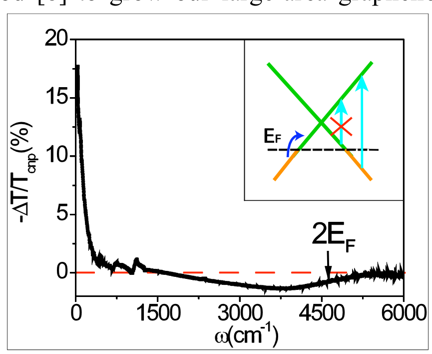

Fig. 1 Gate-induced change of normalized IR transmission –ΔT/T for a hole doped graphene in reference to the CNP. The inset is an illustration of interband (purple arrow) and intraband (blue arrows) transitions in hole-doped graphene. The Fermi surface shifts to a lower energy due to hole doping. It leads to an increase in (lower frequency) intraband absorption and a corresponding decrease in (higher frequency) interband absorption up to $2E_F$.

absorption. At the same time, the lower frequency free carrier absorption (i.e. intraband transition) increases dramatically.

4. Free-carrier response in graphene

The free carrier absorption is directly related to high frequency conductivity of electrons in graphene. Figure 2 shows the real part of the gate-induced change of graphene optical conductivity $\Delta\tilde{\sigma} = \tilde{\sigma} - \tilde{\sigma}_{cnp}$ in the low-wavenumber range (<450 cm^{-1}) for different hole concentrations. Electron-doped graphene also shows a similar change in the optical conductivity. In this spectral range the high frequency conductivity can be described by Drude model $\Delta\tilde{\sigma}(\omega) = iD/\pi(\omega + i\Gamma) - iD_{cnp}/\pi(\omega + i\Gamma_{cnp})$. We have included here a finite carrier response at charge neutral point to account for the presence of inhomogeneous electron and hole puddles. From this fitting, we were able to determine independently the oscillator strength and scattering rate of graphene electrons and their dependence on carrier concentration.

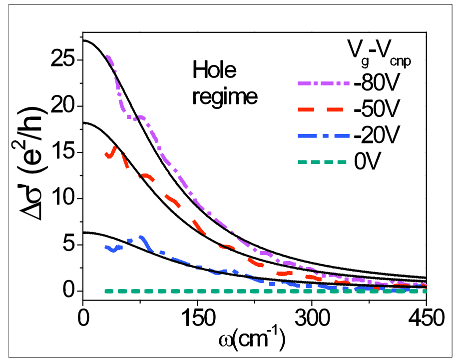

Fig. 2 Gate-induced change of optical conductivity in hole-doped graphene for 30 cm^{-1} < ω < 450 cm^{-1}. (dashed lines). Its frequency dependence can be fit by the Drude model (solid lines)

5. Summary

Our infrared spectroscopy shows that the graphene has strong THz response due to intraband transitions. The intraband absorption can be varied through electrostatic gating and its frequency dependence is described by the Drude model.

6. References

[1] Novoselov, K. S. et al. Two-dimensional gas of massless Dirac fermions in graphene. Nature 438, 197-200 (2005).
[2] Zhang, Y. B., Tan, Y. W., Stormer, H. L. & Kim, P. Experimental observation of the quantum Hall effect and Berry's phase in graphene. Nature 438, 201-204 (2005).
[3] Katsnelson, M. I., Novoselov, K. S. & Geim, A. K. Chiral tunnelling and the Klein paradox in graphene. Nat Phys 2, 620-625 (2006).
[4] Mak, K. F. et al. Measurement of the Optical Conductivity of Graphene. Phys Rev Lett 101, 196405 (2008).
[5] Nair, R. R. et al. Fine structure constant defines visual transparency of graphene. Science 320, 1308-1308 (2008).
[6] Li, Z. Q. et al. Dirac charge dynamics in graphene by infrared spectroscopy. Nat Phys 4, 532-535 (2008).
[7] Wang, F. et al. Gate-variable optical transitions in graphene. Science 320, 206-209 (2008).
[8] Li, X. S. et al. Large-Area Synthesis of High-Quality and Uniform Graphene Films on Copper Foils. Science 324, 1312-1314 (2009).