Relationship between Transport Properties and Raman Spectra in Graphene Field Effect Devices

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Abstract

Raman spectroscopy is commonly used to characterize disorder in graphene. Increase of defects leads to raise in the intensity ratio of Raman D to G peaks for low defect densities. Defect also causes the degradation of graphene transport properties. Thus, a certain relationship is expected between the Raman spectra and transport properties in graphene. Here, we investigate Raman spectra and transport properties of graphene as a function of the amount of electron beam irradiation. We show that the carrier mean free path is inversely proportional to square root of the intensity ratio of Raman D to G peaks. Our result may pave the way for evaluating graphene transport properties with Raman spectroscopy.

1. Introduction

Due to high mobility and atomic thickness, graphene is a promising candidate for the next-generation electronics material. Considerable effort has been devoted to industrial production of graphene with higher mobility, i.e., with less defect density. The defect density is commonly evaluated by Raman spectroscopy; The Raman D band is activated in the presence of defects through a double resonance process involving the elastic scattering of electrons by defects. The intensity ratio of Raman D to G band peaks, I_D/I_G , is empirically related to the in-plane correlation length (or domain size) L_D as $I_D/I_G = C(\lambda)/L_D$ for low defect densities, where $C(\lambda) = (2.4 \times 10^{-10} \text{ nm}^{-3}) \times \lambda^4$ is dependent on the Raman excitation wavelength λ [1,2]. Recent work has suggested an alternative relationship, $I_D/I_G = C'(\lambda)/L_D^2$. To our best knowledge, however, the I_D/I_G ratio has not been related to transport parameters such as mobility and carrier mean free path. In this work, we aim to establish a relationship between the Raman signal, I_D/I_G , and transport parameters. The defects are introduced to graphene by electron beam irradiation. Raman spectra and transport properties are investigated as a function of the electron beam dosage in a single graphene film.

2. Experiment

Our graphene sample was fabricated by micromechanical exfoliation [3] of kish graphite onto a highly-doped Si substrate covered with 300-nm SiO₂. A four-terminal field effect device was defined using electron beam lithography followed by oxygen plasma etching. An optical image of the sample is shown in Fig. 1.

The device was exposed to electron beam of 50 keV several times, and after each exposure, Raman and transport measurement were carried out. The Si substrate was used as a back gate. The dosage of e-beam varied from 0 to 60 mC/cm². From the conductivity, we derived the mobility and electron mean free path as a function of the gate-induced carrier density, n.

3. Results and discussion

Figure 2 shows the Raman spectra for several accumulated dosages of electron beam. With the increase of dosage, the Raman D band situated at ~1350 cm⁻¹ develops, as shown in the inset of Fig. 1, indicating the increase in the defect density. Figure 3(a) is the gate-induced carrier density dependence of conductivity for several accumulated dosages of electron beam. The gate-induced carrier density is defined as $n = C_G(V_G - V_D)/e$, where C_G is the gate capacitance per unit area, V_G is the gate voltage, and V_D is the gate voltage corresponding to the minimum conductivity. From this data, we calculate the field effect mobility of hole conduction, μ , and the carrier mean free path, l_{mfp} , as shown in Figs. 3(b) and 3(c), respectively. Here, l_{mfp} is calculated for a particular value of the gate-induced carrier density n = -2.0×10^{16} m⁻². For either μ or l_{mfp} , the value decreases with increasing the dosage, reflecting larger density of defects.

Figure 4(a,b) (4(c,d)) shows the relationship between the mobility μ (carrier mean free path l_{mfp}) and the ratio, I_D/I_G . Figures 4(a) and 4(c) show linear plots, while Figs. 4(b) and 4(d) show log – log plots. In Fig. 4(a), the mobility steeply decreases as I_D/I_G increases. As shown in Fig. 4(b), the mobility is roughly inversely proportional to the I_D/I_G ratio r:

$$\mu = a/r,\tag{1}$$

where the proportionality coefficient $a = 1.3 \times 10^3$ (cm²/Vs) for this particular sample. In the same way, in Fig. 4(c,d), the carrier mean free path is inversely proportional to the square root of *r*,

$$l_{mfp} = b/\sqrt{r}.$$
 (2)

Our detailed study shows that the proportionality coefficient *b* is proportional to square root of *n*: $b = 1.5 \times 10^{-16} \sqrt{n}$ (m²), for large *n* where random potential due to charged impurities is well screened out. Here, we note that since both μ and I_{mfp} take finite values for $r = I_D/I_G = 0$, the ex-

pressions (1) and (2) are not valid for small I_D/I_G values.

The expression (2) is similar to the relationship, $I_D/I_G = C'(\lambda)/L_D^2$, indicating that the so-called in-plane correlation length is proportional to the carrier mean free path. Further studies are needed for a better understanding of this behavior.

4. Summary

We investigate Raman spectroscopy and transport properties in an e-beam irradiated graphene field effect device. We derive a relationship between the Raman I_D/I_G ratio and transport parameters such as mobility and carrier mean free path. We find that the carrier mean free path satisfies an expression similar to the empirical Tuinstra–Koenig relation for in-plane correlation length, L_D .

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References

- [1] F. Tuinstra and J. L. Koenig J. Chem. Phys. 53 (1970) 3.
- [2] L. G. Cancado et al., Appl. Phys. Lett. 88 (2006) 163106.
- [3] K. S. Novoselov et al., Science 306 (2004) 666.

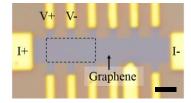


Fig. 1 Optical image of the graphene four-terminal field effect device. A rectangular enclosed by dashed lines is exposed to electron beam. Potential difference between contacts V+ and V- is measured as a result of the current flow from contact I+ to I-. The bar is corresponds to 3 μ m.

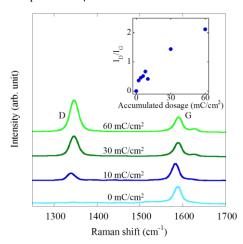


Fig. 2 Raman spectra of for several accumulated dosages of electron beam. Inset is the ratio I_D/I_G as a function of the accumulated dosage.

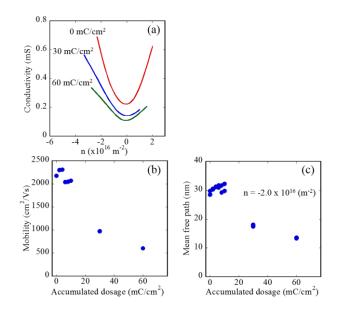


Fig. 3 (a) Gate-induced carrier density dependence of conductivity for several accumulated dosages of electron beam. Field effect mobility of hole conduction (b) and carrier mean free path (c) are shown as a function of the accumulated dosage.

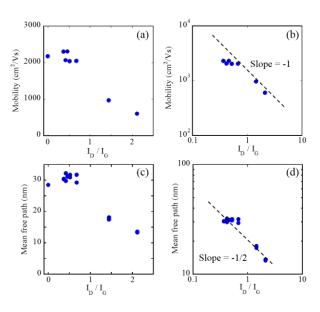


Fig. 4 Mobility as a function of the ratio, I_D/I_G in linear plot (a) and in log – log plot (b). (c, d) The same plots but for the carrier mean free path.