Scrutinizing graphene with polarized Raman spectroscopy

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Abstract: The authors report polarized Raman measurement of single-layer graphene. The G peak position shows polarization dependence, because of the stress on the sample. The relation between the local stress and the polarization will be discussed.

Graphene, a two dimensional and one atomic thick flat allotrope of carbon, attracts great attention lately because of its unique structure, great stability, ballistic electron transport [1]. The specific material can be conveniently fabricated by micromechanical cleavage of highly oriented pyrolytic graphite (HOPG) through scotch tape sticking. Graphene layers are optically visible by putting them on some oxide, such as SiO$_2$ and Al$_2$O$_3$, films with specific thicknesses. This characteristic benefits many optical explorations such as Raman measurement [1]. Raman spectroscopy is widely used for graphene study because it reveals defects, strain, ordering through identifying atomic vibration [1]. The G and 2D bands exhibit significant red shifts when the graphene is a strain film. In addition, the G-band splits into 2 distinct sub bands of G' and G' because of the strain-induced symmetry breaking [2-3]. Thus, analyzing the peak shifts could help people to understand the strain in graphene. In this study, a polarized Raman measurement for graphene is performed. The position of the G peak is polarization dependent. The relation between the peak shift and the strain of the sample will be discussed.

Graphene films used in this study were prepared by micromechanical cleavage of HOPG with scotch tape and then transferred to SiO$_2$/Si substrates for optical observation. The SiO$_2$ thickness is 300 nm. A micro-Raman microscope was used to perform Raman polarization-dependence measurements. A He-Ne laser at 632.8 nm and a diode laser at 532 nm serve as the excitation light source. The polarization and power of the incident light were adjusted by a half-wave plate and a polarizer. The laser power was monitored by a power meter and maintained through these measurements. The laser beam was focused by a 50× objective lens (NA = 0.75) to the sample with a focal spot size of ~2 μm, representing the spatial resolution of the Raman system. The scattered radiation was collected backward with the same objective lens and polarization-selected by a polarization analyzer. Finally, the radiation was sent to a 32-cm spectrometer plus a liquid-nitrogen-cooled charge-coupled device for spectral recording.

![Optical image of graphene sample](image1.png)

**Fig. 1.** (a) Optical image of graphene sample. The region enclosed by a dashed line is presented in the right-sided picture. The green circles marked with 1 and 2 represent two graphene edges that are 90° with respect to each other, while the one marked 3 represents the central region. ; (b) Raman spectra recorded at points 1, 2 and 3, marked in (a), with a 532 nm excitation laser. Gray stripe indicate the D-band Raman peak.
Figure 1(a) shows the optical image of a single-layer graphene flake on the SiO₂/Si substrate. The Raman spectrum from the central area (Point 3 in Fig. 1(a)) shows a prominent G-band peak at 1578 cm⁻¹, which agrees with the literature [1]. The 90° corner of the sample indicates the intersection of a zigzag and an armchair edge based on the graphene crystal structure. It is known that the D-band Raman peak at 1338 cm⁻¹ appears in the Raman spectrum of the armchair edge but is absent in that of the zigzag one [4]. The Raman spectra in Fig. 1(b) clearly indicate that points 1 and 2 are the armchair and zigzag edges, respectively.

![Figure 1](image1.png)

Figure 2 shows systematic polarization-dependent Raman measurements. The angle Φ represents the difference between the polarization angle of the analyzer and polarizer. The G peak is a degenerate mode of LO (longitudinal optical) and iTO (in-plane transverse optical) vibrations. As shown in Figs. 2(b), the intensity of the G-band is independent on the angle (Φ), while G-band peak shows a prominent sinusoidal intensity modulation with a period of 180° in Figs. 2(a) and (c). The modulation of the G band peak can be fitted by a function of cos²(θ₁ - θ₂), where θ₁ and θ₂ are the polarization angle of the analyzer and polarizer. According to previous research [2-3], we believe the G peak shift is caused by a local stress on the graphene surface. The relation between the local stress and the polarization dependence will be presented.

**References**


