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Graphene-polymer Multilayer Heterostructure for Terahertz Metamaterials

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Abstract

The optical response and plasmon coupling between graphene sheets for graphene/polymer multilayer heterostructures with controlled separation were systematically investigated. Anomalous transmission of light was experimentally observed in mid-infrared range. The position of the broad passband in the transmission spectra was observed to red-shift with the increase of the number of layers.

Introduction

The surface plasmon in graphene has attracted increasing research interests because of many desirable properties such as strong confinement and low loss.¹⁻³ The wavelength of graphene plasmons was experimentally observed at the order of 200 nanometres, more than 40 times smaller than the wavelength of illumination which is in the terahertz (THz) regime.^{4, 5} Patterning graphene into micro-ribbons⁶ or photonic-crystal-like structures⁷ could induce plasmon resonance in THz range, which affords a new type of THz metamaterials. Graphene plasmonics is very promising to make transformation optics⁸ due to its tunability via chemical modification or electrical gating. The activities of surface plasmons in graphene multilayers can be controlled more flexibly than graphene monolayers because of the vertical interlayer coupling or in-plane lateral coupling. It has been theoretically predicted that the plasmon resonance can be controlled by the coupling effect among adjacent layers, whereas less attention has been paid to the experimental demonstration.^{9,10}

Here we fabricate graphene/polymer multilayer heterostructure and systematically investigate the optical response as well as plasmon coupling between graphene sheets. Anomalous transmission of light was experimentally observed in THz range. The graphene/polymer multilayers could be a promising platform to filter infrared and terahertz waves and to tailor the propagation and diffraction of light along the graphene sheets deeply below the sub-diffraction-limit.

Experiments

Large area monolayer graphene films used in the work were grown by chemical vapour deposition (CVD) on 25 μm thick Cu foil (Alfa Aesar AA13382RG). Poly(methyl methacrylate) (PMMA) was spin-coated on top of the graphene, the resulting thickness was characterized by variable angle spectrometric ellipsometer (VASE). The copper used to grow graphene was etched away and monolayer graphene-PMMA (G-PMMA) heterostructure was wet-transferred onto intrinsic silicon substrates after doping with HNO_3 . Multilayer heterostructure was formed by repeatedly transferring one layer of G-PMMA on top of the other, as shown in Fig. 1. THz transmittance measurements were carried out on Bruker Vertex 80V Fourier Transform Infrared (FTIR) with a mercury lamp as THz source and DTGS as THz detector.

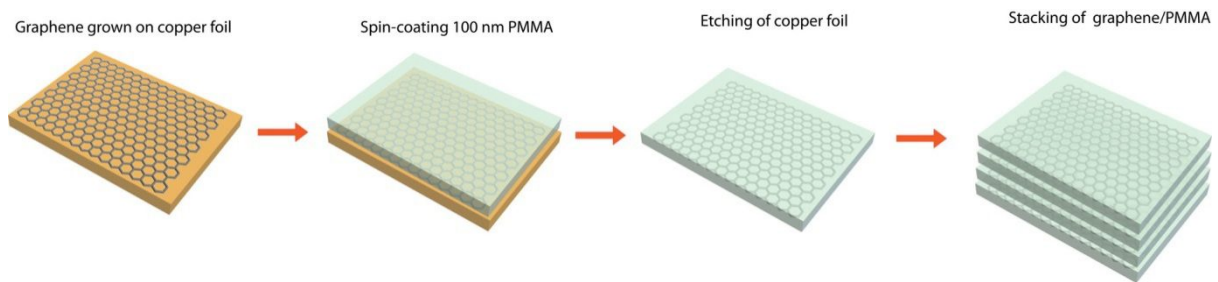


Figure 1. Schematic diagram of fabrication procedure for graphene/PMMA heterostructures.

Experimental and simulation results

The Raman spectra and images were measured on WITEC Alpha 300 confocal micro-Raman system equipped with a 532 nm laser source and 100 \times objective lens. The representative Raman spectrum from monolayer graphene transferred onto intrinsic silicon is shown in Fig. 2. It is found that the Raman 2D band is much stronger than G band with a 2D/G ratio of 1.5, indicating the nature of one atomic layer. The G and 2D band can be fitted well by single Lorentzian peak with full-width at half-maximum (FWHM) of 26 and 38 cm^{-1} respectively, which suggested that the graphene used was of high crystalline quality. These are key Raman characteristics for monolayer graphene.

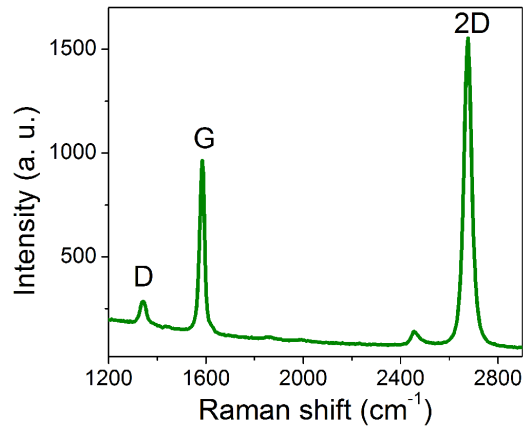
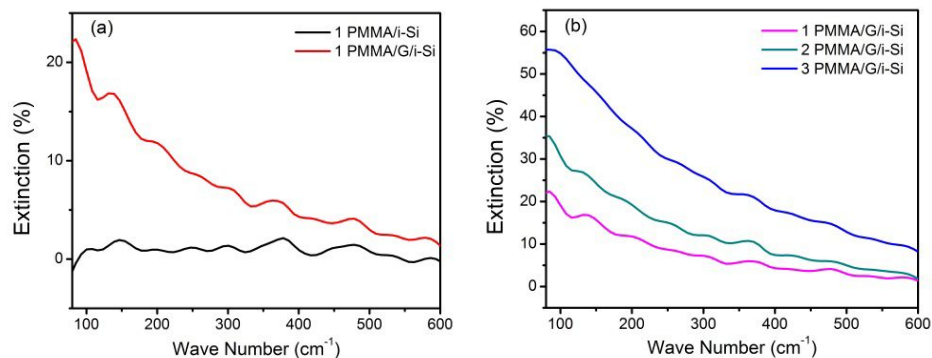


Figure 2. Raman spectrum of monolayer graphene.

The extinction spectra measured on as-prepared PMMA/Graphene (PMMA/G) heterostructures with one, two and three graphene layers are shown in Fig. 3a and Fig. 3b. It is found that the number of graphene layers plays an important role to determine the extinction ratio. The introduction of monolayer graphene between the polymer and intrinsic silicon (i-Si) leads to an increase of the extinction ratio up to 22.5% at 2.55 THz due to free carrier absorption.⁷ The extinction ratio increases with the number of graphene sheets, i.e., an extinction ratio of 55.6% is achieved for a graphene/polymer heterostructure with three graphene layers.

The transmission properties of multilayer graphene sheets and in particular, the influence of the number of graphene sheets are studied as well, as shown in Fig. 3c. A passband can be observed for the samples with two, three, four, five graphene sheets. It is very interesting to find that the peaks of the passband evolve with the number of graphene sheets, i.e., the larger the number of graphene sheets, the sharper the edges of the passband and the lower the transmission peak intensity. The transmission peak tends to shift towards longer wavelength for larger number of graphene sheets, due to the lateral boundary surface plasmon polariton (SPP) coupling between graphene sheets.¹⁰



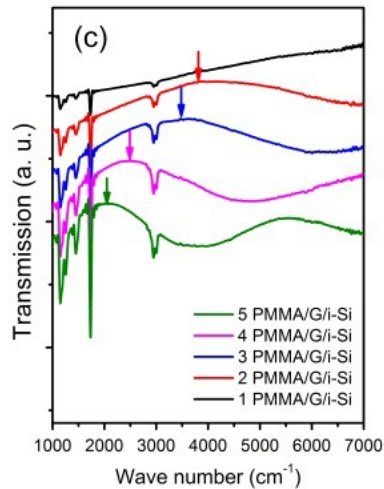


Figure 3. (a) The extinction spectra of monolayer PMMA/G stack and reference sample (PMMA only on intrinsic silicon substrate). Inset shows the schematic of PMMA/G stack. (b) The extinction spectra of the PMMA/G stacks with different number of layers; (c) The transmission spectra as a function of the number of the doped graphene sheets.

Conclusion and outlook

In summary, we have studied light transmitting properties of the graphene multilayers composed of arbitrary layers of graphene sheets. The extinction co-efficiency resulting from monolayer graphene/polymer stack due to the free carrier absorption is $\sim 22.5\%$ and it increases with the number of layers. A broad passband was observed in the transmission spectrum and it can be tuned by the number of layers. Future efforts will be put on tuning the broadband transmission by electrically gating the heterostructures.

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