



Ultra-sensitive graphene based mid-infrared plasmonic bio-chemical sensing using dielectric beads as a medium



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ABSTRACT

Graphene is moving beyond the realm of simple electronic devices toward areas such as advanced bio-chemical sensing. The infrared (IR) response of graphene, characterized by collective long-lived charge-carrier oscillations, could be applied in IR-absorption spectroscopy, typically used for bio-chemical analysis. However, direct light absorption by propagating plasmons in graphene is forbidden due to the large momentum mismatch. Proposed methods to overcome this bottleneck come at a cost, the use of noble metal particles on graphene reduces the spectral bandwidth and nano-structuring graphene is expensive. Here, we propose a simple and cheap method to fabricate large scale ultra-sensitive graphene based mid-IR biosensors, by introducing dielectric beads to excite mid-IR range plasmons. Interference from waves scattered by the beads excite surface plasmon polaritons, which propagate several micrometers in graphene and enhance the interaction between the molecules and mid-IR light. This method opens an interesting window for the application of graphene in bio-chemical sensing.

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1. Introduction

Mid-infrared (IR) absorption spectroscopy is a powerful technique for bio-chemical sensing as it valuably allows the identification of the biochemical building blocks of life, such as proteins, lipids, and DNA by accessing their vibrational fingerprints [1]. Poor interaction between mid-IR light and nanoscale size biomolecules is a major bottleneck in this field due to the large mismatch between mid-IR wavelengths (2–6 μm) and biomolecular dimensions (<10 nm) [2]. A profusion of approaches have been developed, like the technique of surface enhanced infrared absorption (SEIRA), in attempts to find solutions to this limitation. In SEIRA, surface

plasmon modes of metallic nanomaterials are used to produce an enhanced local electric field in the vicinity of the metallic nanoparticles, which enhances the amplitudes of the vibrational modes of adjacent molecules [3]. There have been many investigations of plasmonic enhanced infrared absorption since Hartstein et al.'s first report in 1980 of enhanced IR absorption by impurities in a solution of p-nitrobenzoic acid using variable thickness (1–6 nm) silver [4]. However, despite the prospect of using plasmonic processes to enhance the IR absorption of molecules, the position of the plasmonic resonances of the metallic nanostructures used typically limits this technique to the near-IR region. To overcome this issue and enhance the mid-IR absorption of molecules or biomolecules, it is crucial that the plasmonic material is tunable and has good light confinement, such as for graphene. Graphene, a single layer of carbon atoms arranged in a honeycomb lattice structure, displays many fascinating properties, such as its unique band structure and very high mobility [5–10]. Its distinctive electronic structure leads to several exceptional optical properties, such as absorption over a wide spectral range, from the far infrared to ultraviolet [11]. In the mid-IR region, the absorption of graphene is mainly due to the

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interband transition [12–14] and its optical response is characterized by plasmons [2,15,16] making it an excellent candidate for mid-IR bio-chemical sensing. However, plasmons in graphene are not only different from those produced by electrons in bulk materials, but also from those in conventional 2D electron gas (2DEG) systems [17–19]. Direct light absorption by the propagation of plasmons in graphene is not allowed due to the large momentum mismatch between photons and plasmons, which currently limits the value of graphene in mid-IR bio-chemical sensing [11]. Numerous methods have been proposed to overcome this limitation, for example growing noble metallic nanostructures on graphene or transferring graphene onto noble metallic nanostructures [20–24]. However, these solutions suffer from the same issue faced by conventional SEIRA, the localized surface plasmon resonances of noble metals typically occur in the visible and short-wavelength IR frequency range. Thus, the use of noble metals reduces the spectral bandwidth and the advantages of graphene are lost due to the relatively poor field confinement of the metals in the mid-IR range [25]. Another technique is to enhance the optical resonances from localized plasmons by patterning graphene into arrays of graphene ribbons [26,27]. But this is too costly and complex to be suitable for large scale applications in devices, as it requires electron beam lithography.

Here, we propose a simple and cheap method to prepare ultra-sensitive graphene-based mid-IR bio-chemical sensors. Dielectric particles are introduced as a medium to excite mid-IR range Surface Plasmon Polaritons (SPPs) waves in graphene through diffraction of the light, which significantly enhances the interaction between mid-IR light and adjacent molecules. Moreover, the excited mid-IR range SPPs have long propagation lengths, up to a few micrometers. The performance of the proposed graphene mid-IR bio-chemical sensor was tested and demonstrated by employing it to detect the vibrational modes of para-aminobenzoic acid (PABA). SPPs enhanced mid-IR absorption devices, based on dielectric beads/graphene structures, open new avenues for the development of

highly selective and sensitive bio-chemical sensors.

2. Experimental

2.1. Device preparation and characterization

Graphene was grown on Cu foil using the well-established chemical vapor deposition growth technique (CVD), as displayed in Fig. 1a [28]. To synthesize large-scale continuous monolayer graphene, the Cu foil was folded into a pocket. Prior to growth, the Cu pocket was annealed at 1035 °C for 40 min with a hydrogen flow rate of 10 sccm. The graphene growth was carried out at 1035 °C for 40 min with a methane flow rate of 10 sccm and a hydrogen flow rate of 10 sccm. The extended preparation and transfer procedure is described in the supporting information (SI). The Raman spectrum of the CVD grown graphene, on a SiO₂ substrate using an excitation wavelength of 532 nm, is displayed in Fig. 1b. The major Raman features are the presence of a strong G band located at 1591 cm⁻¹ and a sharp and symmetric 2D band at 2684.5 cm⁻¹. The sharpness of the peaks and the intensity ratio (2:1) of 2D to G Raman bands well indicate that the graphene is monolayer and of high quality [29]. To measure the mid-IR absorption, the graphene was transferred onto a ZnSe substrate using a graphene/polymethyl methacrylate (PMMA) stamp [30]. Here, ZnSe was chosen as the substrate for its high transmission in the mid-IR wavelength region. Details of the graphene transfer process can be found in the SI. Polystyrene (PS) particles can be deposited on graphene by a variety of low-cost scalable methods, such as transfer from liquid/liquid interfaces [31], dip coating or spray techniques. For the present work, the deposition of PS beads was achieved using dip-coating through a Water/Decane interface or by drop coating, as displayed in Fig. 1c and d. For dip-coating, interactions between beads induce the PS beads to organize into a hexagonal close packed configuration.

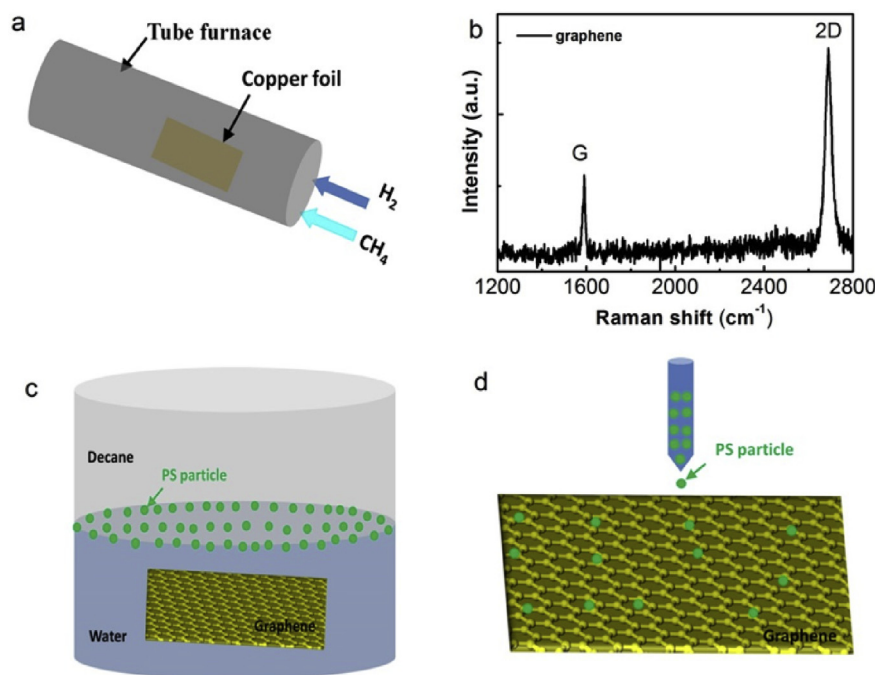


Fig. 1. Sample preparation. (a) Schematic drawing of graphene synthesized by CVD. (b) Raman spectra of monolayer graphene transferred onto a SiO₂ substrate. (c) Schematic drawing of the fabrication of a PS particle monolayer on graphene transferred from a Water-Decane interface. (d) Schematic drawing of drop coating a lower density of PS particles onto graphene using an injector. (A colour version of this figure can be viewed online.)

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