



Short communication

# Unprecedented high plasmonic sensitivity of substrates based on gold nanoparticles



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## ARTICLE INFO

## Article history:

Received 2 May 2013

Received in revised form

20 September 2013

Accepted 27 September 2013

Available online 8 October 2013

## Keywords:

Gold nanoparticles

Graphene-based plasmonics

Plasmon resonance

Hybrid devices

Active plasmonic metamaterials

SPR

## ABSTRACT

This work focused on the plasmonic sensitivity of a substrate composed of gold nanoparticles (AuNp) and 3-aminopropyltriethoxysilane (APTES)-modified graphene oxide (GO-APTES). The multilayered system fabricated by dip-coating was analyzed by means of FTIR spectroscopy and scanning electron microscopy (SEM). The localized surface plasmon resonance (LSPR) band of AuNp was shifted from ca. 520 nm (monolayer of AuNp) to ca. 600 nm (multilayered with GO), which was a result of the agglomeration of AuNp. SEM micrographies revealed a system containing agglomerated graphene oxide sheets on the substrate. An unprecedented sensitivity profile, determined in different refractive index (RI) media, was ca. 460 nm/RIU. The bio-sensitivity of the substrate was evaluated by immobilization of BSA on MUA-NHS-treated Au/GO, which presented a shift of 2.07 nm after binding to the protein.

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## 1. Introduction

The absorption of gold nanoparticles (AuNp) is a result of the collective oscillation of electrons in the conduction band of the Nps in response to the electric field from the incident radiation. This so-called localized surface plasmon resonance (LSPR) [1] is sensitive to surrounding changes in the refractive index [2]. Exploiting this characteristic, many studies in literature use AuNp for the development of sensitive sensors to specific molecules, such as casein [3], bovine serum albumin (BSA) [4], and human immunoglobulin (IgG) [5]. In addition, this result can be found in studies on the detection of antigens of cancer such as of the prostate [6,7] and diseases such as Alzheimer's [8,9]. In most of the cases, a monolayer of AuNp is attached to a substrate, usually glass [10]. Research groups have created other strategies to manufacture plasmonic substrates by increasing the Np density, what is commonly done with a three-dimensional assembly (3D) [11]. One of these strategies involves using carbon nanotubes to increase the AuNp density on the substrate and, thus, increasing its sensitivity. Other carbonaceous structures, such as graphene oxide (GO) – atomic layer of  $sp^2$  – hybridized carbon with a honeycomb crystal lattice – have also been used as starting platforms for building new materials with

specific properties and applications [12]. Some authors [13] have already anchored AuNp in GO, demonstrating the potential for its application in surface-enhanced Raman scattering (SERS). In addition, GO enhanced with Nps has been investigated for use in the sensing of  $H_2O_2$  [14], uric acid [15], dopamine [16], trimethylamine [17], and mercury(II) [18].

In this work, we assembled an LSPR substrate containing AuNp and GO via a layer-by-layer technique. The goal of our study was to achieve an unprecedented higher sensitivity, which was ca. 460 nm/RIU (RIU = refractive index unit), as compared to ca. 90 nm/RIU found in the literature in regard to AuNp-based systems.

## 2. Materials and methods

### 2.1. Synthesis of AuNp

The AuNp (ca. 17 nm) was prepared via the Turkevich method [19]. A 0.01%  $H AuCl_4$  solution (Sigma–Aldrich, 99.99%) was refluxed at the boiling point; then 2.5 mL of an 1% sodium citrate solution (Sigma–Aldrich, 99%) was added to the system and left to react for 40 min. The AuNp were characterized by visible absorption spectroscopy (Ocean Optics, USB 2000, with a resolution of 0.3 nm) and by transmission electron microscopy (TEM, Shimadzu JEOL, JEM 1400, and Image-Pro® PLUS version 4.5.0.29 software).

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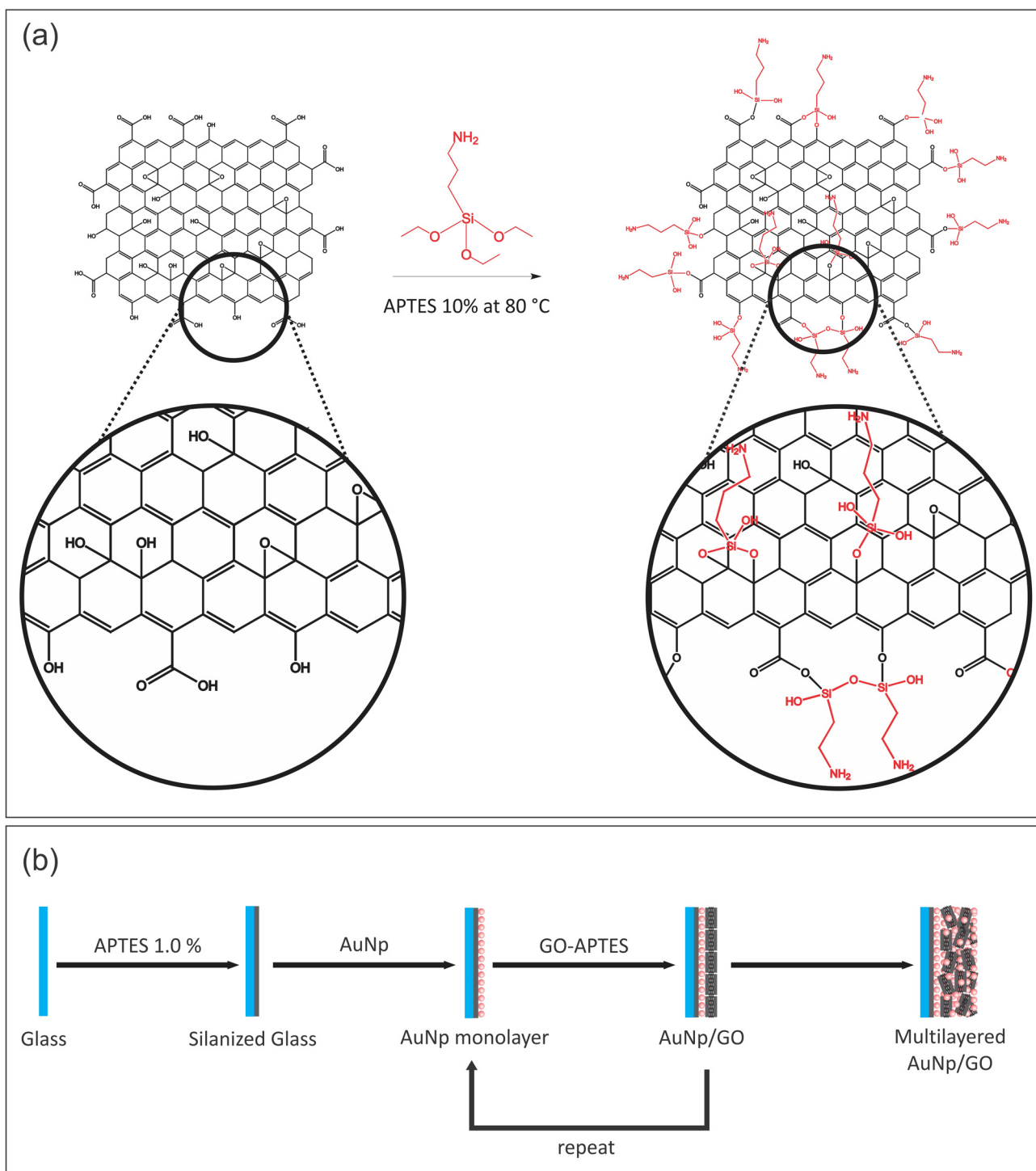


Fig. 1. (a) Graphene oxide modified with APTES. (b) Procedure steps for the preparation of the plasmonic AuNp/GO surface.

## 2.2. APTES-modified graphene oxide (GO-APTES)

Commercially available (CheapTubes, Brattleboro, USA) single-layered graphene oxide (GO) was used as received. An ethanolic solution containing 3 mg of GO and 3 mL of APTES (Sigma–Aldrich,  $\geq 98\%$ ) was stirred at  $50^\circ\text{C}$  for 24 h. The APTES-modified GO (GO-APTES) (Fig. 1a) was centrifuged at 4500 rpm for 40 min, washed with ethanol, and dried at  $80^\circ\text{C}$  overnight. The resulting material was characterized by FTIR spectroscopy (Bomem-Michelson MB-100 with a resolution of  $4\text{ cm}^{-1}$  using a KBr disc method) at between  $4000\text{ cm}^{-1}$  and  $400\text{ cm}^{-1}$ .

## 2.3. Fabrication of the AuNp/GO assembly

Microscope cover slips of  $9\text{ mm} \times 47\text{ mm} \times 1.2\text{ mm}$  were cleaned in a “piranha solution”. The slides were incubated in a 1% APTES ethanolic solution for 4 h. Then the silanized glass was immersed in an AuNp solution overnight. Afterwards, the substrate was immersed in a dimethylformamide (DMF) solution containing 0.1 mg/mL of APTES-GO for 4 h. The substrates were rinsed with deionized water and dried with a nitrogen stream. Soon after, the substrates were again immersed in an AuNp solution for 4 h, rinsed with deionized water, and dried with a nitrogen stream.

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