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Research of detection depth for graphene-based optical sensor

Yong Yang [a,*], Jialve Sun [a], Lu Liu [a], Siwei Zhu [b], Xiaocong Yuan [c,*]

[a] Key Laboratory of Optical Information Science and Technology, Institute of Modern Optics, Nankai University, 300071, Tianjin, China
[b] Institute of Oncology, Tianjin Union Medical Centre, Tianjin, 300121, China
[c] Nanophotonics Research Centre, Shenzhen University, Shenzhen, 518060, China

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ABSTRACT

Graphene-based optical sensors have been developed for research into the biological intercellular refractive index (RI) because they offer greater detection depths than those provided by the surface plasmon resonance technique. In this Letter, we propose an experimental approach for measurement of the detection depth in a graphene-based optical sensor system that uses transparent polydimethylsiloxane layers with different thicknesses. The experimental results show that detection depths of 2.5 μm and 3 μm can be achieved at wavelengths of 532 nm and 633 nm, respectively. These results prove that graphene-based optical sensors can realize long-range RI detection and are thus promising for use as tools in the biological cell detection field. Additionally, we analyze the factors that influence the detection depth and provide a feasible approach for detection depth control based on adjustment of the wavelength and the angle of incidence. We believe that this approach will be useful in RI tomography applications.

1. Introduction

Graphene, with its zero band gap and high carrier mobility, has demonstrated unusual optical properties, including broadband tunable absorption [1] and strong polarization-dependent effects [2]. Graphene-based optical sensors (GOSs) have been developed to detect refractive index (RI) changes with high sensitivity at the graphene sensing layer interface [3–5]. A GOS based on polarization-dependent effects was reported for single cell RI measurements with sensitivity of 10^{-8} RIU (refractive index units), where evanescent waves were generated to detect cell RI changes using a total internal reflection (TIR) structure [6]. The large detection depth of the GOS means that it can be used to measure biological intercellular RI distributions such as that of the cell nucleus [5,6]. Surface plasmon resonance (SPR)-based RI measurement techniques have also been developed over many years and use plasmonic evanescent waves to sense RI changes in the vicinity of metallic surfaces [7–9]. Plasmonic evanescent waves are well known to decay exponentially in a direction perpendicular to the sample interface, and thus the characteristic detection depth for SPR is typically 100–200 nm, while the detection depth for long-range SPR (LRSPR) is of the order of 500 nm–1000 nm [10,11]. However, for cellular RI measurement applications, the typical detection depth for SPR measurements is insufficient to cover cellular dimensions of the order of several micrometers. While a theoretically estimated detection depth of 2.5 μm has been reported for a GOS system [6], no experimental verification was reported. One of our previous works showed that the detection depth of a GOS is greater than that of SPR because some intercellular structures (e.g., the cell nucleus) can be detected [5], but no experimental detection depth results were provided. To the best of our knowledge, no known experimental results related to the detection depth in GOS systems or analyses of the factors that influence the detection depth have been reported in the literature to date.

In this work, we measured the detection depth of a GOS system experimentally using transparent polydimethylsiloxane (PDMS) layers with different thicknesses, and analyzed the effects of both wavelength and angle of incidence. To characterize the penetration depths of evanescent waves, we investigated a typical GOS under TIR structure consisting of a graphene layer located between a high-index medium (BK7 glass) and a low-index medium (PDMS layers of various thicknesses) to analyze the detection depth relationships with different wavelengths and angles of incidence. Experimental results show that the detection depth can reach 2.5 μm at a wavelength of 532 nm and 3 μm at a wavelength of 633 nm. The experimental results here prove that the GOS with TIR structure can realize long-range RI detection and is a promising tool for use in the biological cell detection field.

* Corresponding authors.
E-mail addresses: yangyong@nankai.edu.cn (Y. Yang), xcyuan@szu.edu.cn (X. Yuan).

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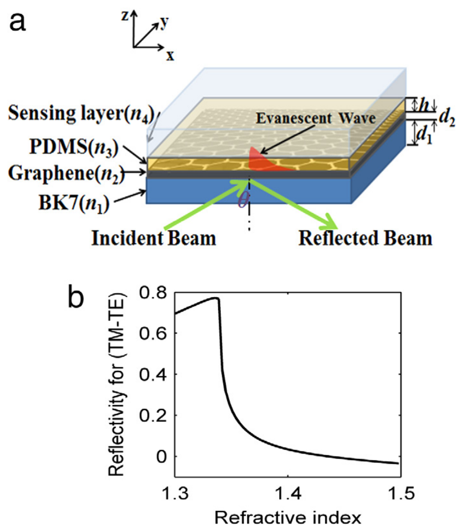

Fig. 1. (a) BK7/graphene/PDMS/medium multilayer structure diagram, where n_1 = 1.517; d_1 is the BK7 glass thickness; n_2 is the RI of graphene, where n_2 = 2.6 + 1.33 i; d_2 is the graphene thickness and is related to the number of graphene layers, where d_2 = 4 nm; n_3 = 1.405 is the RI of PDMS; h is the PDMS thickness; and θ is the angle of incidence. (b) Relationship between the difference in reflectivity (TM-TE) and the RI.

2. Experimental method

Based on polarization-dependent effects under TIR conditions, the typical GOS consists of a graphene layer located between a high-index medium and a low-index medium. A beam incident in the high-index medium, including its transverse electric (TE) mode and its transverse magnetic (TM) mode, strikes the graphene surface at an angle of incidence that is larger than the critical angle with respect to the surface normal. The differences in reflectance between the TE and TM modes are sensitive to the RI of the low-index medium that is in contact with the graphene because the evanescent wave is absorbed by the surrounding medium. To determine the relationship between the reflection difference and the evanescent wave penetration depth, we built a BK7/graphene/PDMS/sensing layer multilayer structure to simulate the detection process using the finite element method, as shown in Fig. 1(a). The evanescent wave beyond the graphene interface propagates along the x direction and its intensity decays exponentially in the z direction. We set a PDMS layer above the graphene interface to generate specific distances between the graphene interface and the sensing layer, and measure the reflection differences between the TE and TM modes with small RI variations in the sensing layer.

The evanescent wave intensity in the z direction can be given as:

$$I\,(z) = I_0 \cdot e^{\left(-\frac{z}{d}\right)} \tag{1}$$

where $I\,(z)$ is represents the intensity at the perpendicular distance z from the graphene interface, and I_0 is the intensity at that graphene interface; d is the characteristic penetration depth, and is given by:

$$d = \frac{\lambda}{4\pi\sqrt{n_1^2 \sin 2(\theta) - n'^2}} \tag{2}$$

where λ is the incident beam wavelength and θ is the angle of incidence; n_1 is the RI of BK7 glass, and n' is the effective RI with a contribution from the graphene layer, the PDMS and the sensing layer. Interpretation of the n' value is complex because of the uncertainties related to detection of the evanescent wave decay in the different layers. Therefore, we used the finite element method to analyze evanescent wave attenuation in the proposed multilayer structure.

Two main methods are used to model multilayer film structures: the transfer matrix method [12,13] and the stationary phase method [14]. In this work, we used the stationary phase method to calculate the relationship between the reflection difference and the evanescent wave penetration depth based on inclusion of graphene layers in the membrane structure, as shown in Fig. 1(a).

Based on Maxwell's electromagnetic wave theory, the reflection coefficient is expressed as follows when using the stationary phase method:

$$r = \frac{r12 + r23exp(2i\beta)}{1 + r12r23exp(2i\beta)} \tag{3}$$

where r is the total reflection coefficient; r_{12} is the reflection coefficient at the interface between BK7 glass and graphene; r_{23} is the reflection coefficient at the interface between graphene and PDMS; and β is a phase transfer factor that can be expressed as

$$\beta = \frac{2\pi}{\lambda} n_2 d_2 \cos\theta_g. \tag{4}$$

Here, θ_g is the incident beam angle relative to the normal at the interface between BK7 and graphene. This incident beam then travels through the graphene layer and strikes the PDMS interface. We define θ_w as the angle of incidence at the interface between graphene and PDMS, at which TIR occurs.

By considering TE mode waves alone, we obtain the following:

$$r_{12} = \frac{n_1 \cos\theta - n_2 \cos\theta_g}{n_1 \cos\theta + n_2 \cos\theta_g} \tag{5}$$

$$r_{23} = \frac{n_2 \cos\theta_g - n_3 \cos\theta_w}{n_2 \cos\theta_g + n_3 \cos\theta_w}. \tag{6}$$

When we consider TM mode waves, we obtain the following:

$$r_{12} = \frac{n_2 \cos\theta - n_1 \cos\theta_g}{n_2 \cos\theta + n_1 \cos\theta_g} \tag{7}$$

$$r_{23} = \frac{n_3 \cos\theta_g - n_2 \cos\theta_w}{n_3 \cos\theta_g + n_2 \cos\theta_w}. \tag{8}$$

Reflectivity R is then expressed as

$$R = |r|^2. \tag{9}$$

TIR only occurs when θ_w is larger than a critical angle or equal to 90°, and an evanescent wave is generated. We calculated the critical angle to be 67.8° based on the RI values of BK7, graphene and PDMS. In a typical GOS, the difference in reflectivity (DR) between the TM and TE modes is calculated to characterize the RI variations within the vicinity of the media. The results of a typical simulation are shown in Fig. 1(b), demonstrating that there is a high sensitivity range when θ_w is close to the critical angle. To characterize the detection depth in a GOS, we monitored DR variations with increasing distance from the graphene surface when a fixed change of RI in the sensing layer is given. If the DR variation is lower than a specific threshold, the corresponding distance from the graphene surface is then defined as the detection depth. Here, the detection depth in a GOS is different from the penetration depth of evanescent wave which is given in Eq. (2).

3. Simulations

Using the finite element method, we calculated the detection depths for different incident beam wavelengths based on the model shown in Fig. 1(a), and the theoretical results are shown in Fig. 2. We selected simulation parameters based on the experimental parameters. Here, the RI value of BK7 is n_1 = 1.517, a typical RI value for graphene is n_2 = 2.6 + 1.3i with d_4 = 4 nm [15], and the RI value of PDMS is n_3 = 1.405 with various values of thickness h. Fig. 2(a)–(d) show detection depths of 2.5 μm, 3 μm, 4 μm and 4.5 μm at incident beam wavelengths of 523 nm, 633 nm, 780 nm and 1064 nm, respectively. There is a linear relationship between detection depth and wavelength, as shown in Fig. 2(e), and this is in accordance with Eq. (2).

We further simulated the changes in detection depth with increasing angle of incidence from the critical angle to 85°. The theoretical