An Analytical Model for Optimizing the Optical Absorption of Graphene-Based Two-Dimensional Multilayer Structure

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Abstract

Two-dimensional (2D) materials are promising but remain to be further investigated, with respect to their interesting usage in optoelectronic devices. These materials have far less than ideal absorption due to their thin thickness, limiting their deployment in practical optoelectronic applications. Graphene is a 2D material with honeycomb structure. Its unique and fantastic mechanical, physical electrical and optical properties make it to be an important industrial and economical material. In this work, a simple analysis is performed for the reflectance, transmittance, and absorption properties of multilayer thin film structures with graphene sandwiched in dielectric layers. Based on Maxwell’s electromagnetic wave theory and coupled Fresnel equations, we investigate how to get maximum absorption for a proper choice of media and graphene layers. Numerical results show this absorption is controlled with matching thicknesses of layers, number of graphene layers, wavelength and angle of incident electromagnetic wave.

Keywords Reflectance · Transmittance · Absorption · Multilayer thin film structure · Graphene · Maxwell’s electromagnetic wave theory · Fresnel equations

1 Introduction

Graphene, a one-atom thick sheet of carbon atoms, is one of the most promising nanomaterials and has interesting features and applications in optics, mechanics, and electronics [1–5]. Single-layer graphene with optical absorption of approximately 2.3% for incident light, is adequate for some photovoltaic devices [6–11]. Of course, further applications need stronger absorption with enhanced interactions between graphene
and light. Thus, there is an urgent need for research and investigations to improve and develop absorption mechanisms. A most common way to control absorption through intense resonances is surface plasmon polaritons (SPPs) [12–15]. Crucial for the optical performance of small nano particles and ultra-thin structures is frequently that relevant surface plasmon excitations are available. Graphene can form strong SPPs with closely 100% absorption in the mid-infrared to terahertz regions with micro-nano structures or via controlling the chemical potential by an external gate or doping [12, 16–19]. An optical switching mechanism by gated graphene layer coupling to external radiation through SPPs has been described in Ref. [20]. Furthermore, enhanced absorption of the graphene layer can be realized by guided modes [21–25] and meta-surfaces [26–29]. Other ways to enhance the absorption of a graphene sheet in the visible spectrum have been reported. In Ref [30], a maximum absorption of 60% was reported for a transverse magnetic (TM) wave with a monolayer resonant grating in the visible region. A broadband absorption enhancement (> 75%) was observed in a multilayer structure [31] and nearly 100% visible absorption for single layer graphene was demonstrated in a multilayer, film-based, attenuated total reflectance configuration [32]. Perfect absorption (100%) in the visible and infrared regions was exhibited in an absorption-cavity of graphene sandwiched in dielectric layers. High-sensitivity sensing is realized when the symmetrical structure is slightly broken [33].

Graphene-based sensors are another important area, where it has been suggested [34] that graphene ribbons can convert molecular signatures to electrical signals based on sensitive graphene plasmons to the molecular analytes. We have the possibility of controlling “optical” properties in graphene with a proper gate voltage [35] and/or doping, thus a multitude of possible mechanisms are available for sensing and tunable optics over a broad frequency range [36].

When we were to freely control the optical properties of a thin film structure with the thickness of a monolayer graphene, the maximum attainable light absorption could be dictated with the contrast of the surrounding media [37–39].

By tuning the geometrical properties and the effective dielectric function of the nanocomposite structure, the impedance of the system is matched to maximize the absorption [40–43].

Here we study another line of approach, not invoking surface plasmons and collective excitations, to determine optimal conditions for light absorption, i.e., the possibility to tune the optical properties of graphene layers by an appropriate choice of the dielectric environment. The proposed structure is simple and thus has the advantage of being non-polluting and reusable for experimental investigations. The simulated results have shown that the absorption is a function of incident angle and wavelength, dielectric thicknesses, and the number of graphene layers. We hope these results will provide useful suggestions for potential applications of graphene layers in the visible spectrum for optoelectronic devices.
2 Theoretical Model

This section summarizes our formalism for modeling multilayer structures. This calculation provides us with an effective approach for studying the transport of electromagnetic waves in an anisotropic layered device.

To calculate the reflectance, we apply Maxwell’s equation

$$\nabla(\nabla E) - \nabla^2 E = \epsilon_0 \frac{\alpha^2}{c^2} E$$

(1)

where $E = E_0 e^{i(\omega t - k \cdot r)}$ is the electric field with amplitude $E_0$, $c$ is the speed of light and $\epsilon_0$ is dielectric constant. We first consider a plane wave propagating through a nonabsorbing medium with refractive index $N_2$, which is incident on a nano medium with refractive index $N_1$. All plane waves that normal incident on a plane boundary between two semi-infinite material regions, are reflected and transmitted independently of their state of polarization. The amplitudes of normally incident, transmission and reflection electric fields are, respectively, $E_i$, $E_t$, and $E_r$ (Fig 1). The tangential electric and magnetic fields must be continuous across the boundary.

The Fresnel formulas for reflection and transmission of a light obliquely incident on a plane boundary are [44]

$$r_\parallel = \frac{E_\parallel}{E_\parallel} = \frac{\cos \theta_i - m \cos \theta_r}{\cos \theta_i + m \cos \theta_r}, \quad t_\parallel = \frac{E_\parallel}{E_\parallel} = \frac{2 \cos \theta_i}{\cos \theta_i + m \cos \theta_r}$$

$$r_\perp = \frac{E_\perp}{E_\perp} = \frac{\cos \theta_i - m \cos \theta_r}{\cos \theta_i + m \cos \theta_r}, \quad t_\perp = \frac{E_\perp}{E_\perp} = \frac{2 \cos \theta_i}{\cos \theta_i + m \cos \theta_r}$$

(2)

Where $E_\parallel$ and $E_\perp$ are the electric vectors parallel and perpendicular to the plane of incidence, $\theta_i$ and $\theta_r$ are incidence and transmittance angles and $m$ is the ratio of the refractive index of medium 1 to medium 2 ($m = N_1/N_2$).

Fig. 1 Reflection and transmission of obliquely incident light. Wave is incident at an angle $\theta_i$ with respect to the surface normal of the nano film.
Now, we consider our multilayer nano structure (Fig. 2). It was made of FTO glass, Silicon dioxide (SiO\(_2\)), graphene (Gr), silver (Ag) and gold (Au) layers. Based on Maxwell’s electromagnetic wave theory and coupled Fresnel equations \([45]\), we can express the total reflection coefficient of a TM wave as \([33, 44]\):

\[
r_{01234} = \frac{r_{01} + r_{1234}\exp(2i\delta_1)}{1 + r_{01}r_{12}r_{23}r_{34}\exp(2i\delta_1)}
\]

and the transmission coefficient as \([46]\)

\[
t_{01234} = \frac{(1 + r_{01})(1 + r_{1234}\exp(i\delta_1))}{1 + r_{01}r_{12}r_{23}r_{34}\exp(2i\delta_1)}
\]

where \(r_{01}\) is the reflection coefficient at FTO/SiO\(_2\) interface, \(r_{12}\) is the reflection coefficient at SiO\(_2\)/Gr interface, \(r_{23}\) is the reflection coefficient at Gr/Ag interface, \(r_{34}\) is the reflection coefficient at the Ag/Au interface, and \(r_{1234}\) is the total reflection coefficient above the SiO\(_2\) layer and \(k_i\) is the longitudinal wave vector.

Using Snell’s law—invariance \((N_i \sin \theta_i = N_{i+1} \sin \theta_{i+1})\) and Eq. (2), the coefficients \(r_{i,i+1}\) can be obtained from \([46]\):

\[
r_{i,i+1} = \frac{\varepsilon_i k_{i+1} - \varepsilon_{i+1} k_i}{\varepsilon_i k_{i+1} + \varepsilon_{i+1} k_i} \quad (i = 0, 1, 2, 3)
\]

where

\[
\varepsilon_i = n_i^2, \quad k_i = \frac{2\pi}{\lambda} \sqrt{\varepsilon_i - \varepsilon_0 \sin^2 \theta}, \quad \delta_i = k_i d_i \quad (i = 1, 2, 3)
\]

Here \(k_i\) is the wave number component perpendicular to the interface of p-polarized light in medium \(i\). For different layers, the parameters \(\varepsilon_i\), \(n_i\) and \(d_i\) are the relative dielectric constant, the refractive index and the thickness of different layers, respectively. \(\lambda\) is the incident wavelength and \(\theta\) is the angle between the incident electromagnetic wave and the z direction.

**Fig. 2** Schematic cross-sectional view of the FTO/SiO\(_2\)/Gr/Ag/Au. A p-polarized electromagnetic wave with an oblique angle \(\theta\) is incident from SiO\(_2\) and has its field vectors oriented. The question addressed here is under what conditions does this nano structure obtain the optimum absorption?
The total reflectivity $R$ and transmittance $T$ are \([44, 46]\):

$$R = \left| r_{01234} \right|^2, \quad T = \text{Re} \left( k_1 \epsilon_1 \right) / \left( k_4 \epsilon_4 \right) \left| t_{01234} \right|^2 \quad (7)$$

and the thin film absorption $A$ is then $1 - R - T[47]$.

3 Results and Discussion

Using a theoretical approach, we study optical transport through multilayered structures. Three types of samples were studied using different combinations; (a) FTO/SiO$_2$/Gr/Ag/Air, (b) FTO/SiO$_2$/Gr/Au/Air and (c) FTO/SiO$_2$/Gr/Ag/Au. The graphene thickness is determined by the number of graphene layers $L$ as $d_2 = 0.34 \text{ nm} \times L \ [4]$. Here, we assume the refractive index of the glass is $n_{\text{FTO}} = 1.518$ and $n_{\text{SiO}_2} = 1.46, n_{\text{air}} = 1, n_{\text{Au}} = 0.17 - 4.86i, n_{\text{Ag}} = 0.43 + 2.455i$. The refractive index of graphene is not a simple constant in different conditions such as graphene layers, incident wavelengths, substrates, doping density and so on. Here, we employ the refractive index of $2.6 + 1.3i$ in our calculation and simulation which is basic value for undoped and un-patterned graphite cited by researchers in the exploration of graphene properties \([33, 48–52]\).

To realize how the optimal conditions can optimize the absorptance in ultrathin films, we summarize below our important results.

3.1 FTO/SiO$_2$/Gr/Ag/Air

Calculations and simulations for the reflectivity as a function of incidence angle are shown in Fig. 3. This figure shows that the reflectivity by theory calculation decreased by increasing SiO$_2$ thickness and number of graphene layers. As the incidence angle increases from 0 to 90, the reflectivity curve decreases first to reach its minimum value at $\theta \approx 80^\circ$, then increases ($\theta_1 \to 90, \cos \theta_1 \to 0, r \to 1$) when the thickness of SiO$_2$ is decreased from 1000 to 200 nm and the number of graphene layers decreases from 50 to 10. For higher incident angles, the electromagnetic wave is reflected back, so reflectivity increases.

The basic principle to enhance absorption in nano films is to reduce reflection and transmission of incident light from the thin absorption layer \([53]\). The simulation results of transmission, reflection and absorption curves can be found in Fig. 4, which exhibits what happens to the values of $T, R$ and $A$ when $\theta$ increases from zero to $\pi/2$. A strong absorption peak occurs where $\theta = 73$. We see that at $\theta < 70$ absorption increases due to reduced reflection, then at higher angles, reflectivity increases and thus we have a weak absorption.

In such a way, we plot reflectivity vs the thickness of SiO$_2$ and Ag layers (Fig. 5). This result shows a general trend of increasing reflectivity as the thickness of $d_1$ layer is increased up to 300 nm and the thickness of $d_3$ layer is increased up to 70 nm. As shown in Fig. 3, the calculated reflectance decreases as the thickness of the SiO$_2$ ($d_1$) and graphene layer ($d_2$) increase. Therefore, while $R$ depends
on the thickness of layers, \( d_3 = d_{Ag} \) plays a more important role and increases the reflection of incident light.
3.2 FTO/SiO$_2$/Gr/Au/Air

To enhance the light absorption and emission of 2D materials, a variety of optical structures are designed, for example, distributed Bragg reflector microcavities, metallic reflectors, dielectric super-absorbing, photonic crystal nanocavities, and plasmonic nanostructures. [47, 54–58]. Investigating and designing multilayer nanostructured materials is necessary to confine light within 2D materials, increase light absorption and improve the performance of optoelectronic devices.

Now we replace the silver layer in the structure (a) with a gold layer [57–60]. Therefore, we consider a multilayered structure FTO/SiO$_2$/Gr/Au/Air and carry out measurements on this new structure. In the near-infrared -to-visible range there is a necessity for broadband absorption enhancement.

As indicated in Eqs. 2–5, reflection coefficients $r_{i,i+1}$ are not dependent on wavelength, but both the face and the dominator of the total reflection and transmission coefficients $r_{01234}$ and $t_{01234}$ have an exponential relation with the inverse of
wavelength. Therefore, the behavior of reflectivity and transmittance coefficients and consequently absorbance depends critically on the wavelength range. Figure 6 shows that the absorbance value decreases approximately linearly with increasing incident wavelength and decreasing the incident angle. Instead of utilizing critical coupling as the absorption enhancement mechanism [47, 61], we propose the use of suitable structure parameters such as incident angle $\theta$ which results in broadband absorption enhancement. The experimental total absorption in the graphene structures shows a large dependence on the incident angle and wavelength [47].

In order to begin the comparison between the absorbance of these structures, we plot $A$ as a function of structure parameters in the following. As shown in Fig. 7, the absorption conditions are examined in terms of incident angle and incident wavelength for two structures, Au-coated (Gr/Au) and Ag-coated (Gr/Ag). Firstly, the comparison exhibits the role of layers in absorbance of these structures in which Au/air creates an enhanced and somewhat broader absorption than Ag/air structure. Secondly, the absorbance decreases as wavelength increases while the absorbance peak appears at $\theta = 73$ for both structures.

Fig. 7 Absorbance for two structures Gr/Au/Air and Gr/Ag/Air as a function of a incident angle with $L=60$, $d_1=200$ nm, $d_3=300$ nm, $\lambda=532$ nm and b wavelength with $L=60$, $d_1=200$ nm, $d_3=300$ nm, $\theta $ = $\frac{\pi}{4}$. (Color figure online)
We continue our discussion of the theoretical absorption data for FTO/SiO₂/Gr/Ag/Au structure with the investigation of the structure parameter effect provided by the thickness of layers, number of graphene layers and incident angle.

In Fig. 8, we observe that the absorbance peaks appear and vary with increase in the SiO₂ thickness and number of graphene layers. As shown, enhanced broad-band absorption is demonstrated in larger thicknesses and more layers of graphene for \( d_1 = 500 \text{ nm} \) and \( L = 58 \).

As shown in Fig. 4, transmittance decreases as the incident angle increases. In such a way, we plot transmittance as a function of the incident angle, Au layer thickness and Ag layer thickness. Transmittance decreases by increasing the magnitude of the incident angle up to \( 90^\circ \) and Au layer thickness up to 80 nm, but \( \theta \) plays a lesser role than \( d_{Au} \) (Fig. 9a). At the same time, that \( T \) depends on the thickness of layers, \( d_4 ( = d_{Au} ) \) plays an important role (Fig. 9b) and can effectively manage the transmittance values. Thus, Au layer plays a more significant character than the incident angle and the Ag layer and controls the outside fields.

These results can provide guidance for potential applications of graphene in optoelectronic devices, photoacoustic imaging, and biosensing [62–65].

Finally, our calculations have shown that the wavelength and angle of the incident electromagnetic wave, relative dielectric constants \( n_1, n_2, n_3 \), thickness \( d_1 \),

![Fig. 8 Absorbance of FTO/SiO₂/Gr/Ag/Au as a function of incident angle with \( d_3 = 300 \text{ nm}, d_4 = 50 \text{ nm} \) \( \lambda = 532 \text{ nm} \), a \( L = 60 \), and b \( d_1 = 300 \text{ nm} \).](image-url)
Fig. 9 Transmittance as a function of $\theta$ incident angle and $d_1(=d_{Au}), \ b \ d_3(=d_{Ag})$ and $d_4(=d_{Au})$ for $L=40, \ d_1=300$ nm, $\lambda=532$ nm

Fig. 10 Absorbance as a function of $d_1(=d_{SiO_2})$, for $L=50, \ a \ \lambda=532$ nm, $b \ \theta = \pi/3$
and number of graphene layers can effectively control the absorption of graphene (Fig. 10).

It should be mentioned that Van der Waals interaction between neighboring layers depends strongly on the number of layers. Understanding the interlayer coupling and its correlation effect can be paramount for designing novel graphene-based heterostructures with interesting physical properties [56]. In addition, research results have shown the important design parameters such as graphene plasmons [61], antimonene/graphene structures [55] and stacked period number of dielectric materials [53] can be effective strategies for devising to manipulate light and enhance absorption in multilayer heterostructures with various applications.

4 Conclusion

In summary, a multilayer graphene-based structure is proposed to achieve absorption enhancement. Large enhancement and tunability of light absorption in 2D materials is promising for ultra-thin optoelectronic devices that interact with light. We have shown the absorption \( A \) as a function of the relevant physical parameters. Through Maxwell’s electromagnetic wave theory and coupled Fresnel equations, relationships are demonstrated, and three types of samples were studied with different combinations. To achieve continuous control of transmission, reflection and consequently absorption, the effects of wavelength and angle of the incident light, number of graphene layers and thickness of different layers are investigated. We found that absorption increases with the decrement of SiO\(_2\) layer thickness and increment of Ag layer thickness. As expected, absorption is enhanced in the large number of graphene layers. In addition, Au coated structure creates an enhanced and broadband absorption than the Ag coated structure. Meanwhile, the wavelength and angle of the incident electromagnetic wave can be effectively controlled the light absorption and thus for a situation with an appropriate media one can approach higher absorption.

Author’s Contribution K. H carried out the calculation and wrote the manuscript.

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Declarations

Conflict of interest The author declares no competing interest.

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