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Enhanced absorption in a graphene embedded 1D guided-mode-resonance structure without back-reflector and interferometrically written gratings

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A theoretical model based on coupled mode theory is presented to calculate the absorption in a graphene embedded 1D guided-mode-resonance (GMR) structure that does not require a back reflector. The optimized graphene-GMR structure can absorb up to 70% of the incident light which far exceeds the already reported results without using any back-metal reflector or Bragg mirror. The theoretical analysis is valid for binary gratings and pyramidal gratings which are patterned using an interference lithography system. We experimentally validate our theoretical results and analyse the influence of the geometrical parameters to achieve critical coupling for the enhanced absorption.

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Guided Mode Resonance (GMR) structures [1] by virtue of their narrow linewidth [2], high efficiency [3], and high phase sensitivity [4], have been used for many applications such as bio-sensors [4,5], tunable optical filters [6] and electro-optic switches [7]. The electric field corresponding to the resonant mode of the GMR structure is strongly confined within the waveguide that can be used to trap light [8] and hence can enhance the absorption in 2D functional materials such as graphene monolayer. Graphene is an ideal material for optoelectronic devices [9] such as photodetectors [10], absorbers [11] and modulators [12]. However, monolayer graphene lacks the property of spectral selectivity due to its ultra-broadband uniform spectral response from ultraviolet to terahertz [9] range. In addition, its absolute absorption is too low [13] (2.3%) in this wavelength range that results in low photoresponsivity [13]. To overcome these issues, several mechanisms have been proposed to enhance absorption in a graphene layer by embedding it in photonic crystals and subwavelength gratings. Most of the reported works have used critical coupling [14] by guided resonance and achieved more than 70% absorption both theoretically and

experimentally. However, all these reported works have used some sort of back reflectors such as metal films [15-19] and Bragg mirrors [9, 14, 20-22] to achieve perfect absorption. The use of a metallic reflector, however, results in significant suppression of absorption in graphene, due to metallic losses [18]. On the other hand, dielectric Bragg mirrors don't have this kind of loss issue. However, they increase the physical footprint of the device [18] and makes the fabrication process more complicated since it requires material deposition of several layers [23]. In addition, incorporation of these back reflectors makes the device complicated for certain applications. In this context of not using any back reflectors, the previous works [24-27] have reported very low optical absorption in the range of 30-50%. Therefore, absorption enhancement in a monolayer-graphene without using any back reflectors is still rare and in urgent need. In this study, we have reported absorption enhancement ($\approx 70\%$) in monolayer graphene incorporated into a 1D GMR structure without using any back reflector. We have derived the necessary theoretical formulae based on coupled mode theory that can predict and maximize the absorption as described below.

Fig. 1. Schematic of the proposed guided-mode-resonant structure with an embedded graphene monolayer on the silicon nitride coated quartz substrate.

The proposed GMR structure consists of a 1D grating of depth ' h ' and periodicity ' Λ ' coupled with a thin film waveguide (silicon

nitride) of thickness 'd' on a quartz plate as shown in Fig. 1. The refractive index of the material forming the grating is n_g . The grating has a duty cycle (f) defined as w/Λ , where 'w' is the line width of the grating as shown in Fig. 1. An s-polarized broadband light is incident normally on the GMR structure. Since the grating is comprised of high (n_g) and low index (air) materials, the effective refractive index of the grating film for s-polarized light becomes

$$n_{eff} = \sqrt{fn_g^2 + (1-f)n_{air}^2} \quad (1)$$

As shown in Fig. 1, a graphene monolayer is incorporated at the interface of the waveguide and grating layer. For this 1D GMR geometry with the embedded graphene layer, we obtained the amplitude reflection (Γ) and transmission (τ) coefficients from the coupled mode theory [28] which are given below.

$$\Gamma = \frac{r(\omega - \omega_0) + i(r\delta - \gamma)}{(\omega - \omega_0) + i(\gamma + \delta)} \quad (2)$$

$$\tau = t \left\{ \frac{(\omega - \omega_0) + i\delta}{(\omega - \omega_0) + i(\gamma + \delta)} \right\} \quad (3)$$

Here r and t are respectively the off-resonance (Fresnel) amplitude reflection and transmission coefficients when there is no resonance taking place in the structure. ' γ ' is the external leakage rate which is defined as the time rate of amplitude change in the resonator in the absence of input wave. ' δ ' is the intrinsic loss rate due to the embedded graphene layer. The corresponding (intensity) reflectance (R) and transmittance (T) can be written as

$$R = \Gamma \Gamma^* = \frac{r^2(\omega - \omega_0)^2 + (r\delta - \gamma)^2}{(\omega - \omega_0)^2 + (\gamma + \delta)^2} \quad (4)$$

$$T = \tau \tau^* = t^2 \left\{ \frac{(\omega - \omega_0)^2 + \delta^2}{(\omega - \omega_0)^2 + (\gamma + \delta)^2} \right\} \quad (5)$$

Thus, the absorption, $A = 1 - (R + T)$ as a function of incident frequency (ω) can be calculated as

$$A = \frac{(1 - r^2 - t^2)[(\omega - \omega_0)^2 + \delta^2] + 2\gamma\delta(1 + r)}{(\omega - \omega_0)^2 + (\gamma + \delta)^2} \quad (6)$$

This absorption spectrum takes the form of Lorentzian function of frequency whose peak value at the resonant frequency ($\omega = \omega_0$) is given by

$$A = \frac{(1 - r^2 - t^2)\delta^2 + 2\gamma\delta(1 + r)}{(\gamma + \delta)^2} \quad (7)$$

It can be inferred from Eq. (7) that the peak absorbance depends on the off-resonance reflection (r) and transmission (t) coefficients, the external leakage rate (γ) and the loss rate (δ). These parameters greatly depend on the grating parameters, h , f and n_g . To validate this theoretical formula, we analysed the effect of these parameters for a fixed waveguide thickness ($d=100$ nm) and grating period ($\Lambda=500$ nm). For our experiment, we have used negative photoresist NR7-500P (from Futurrex) to form the grating pattern on the silicon nitride coated quartz substrate. The refractive index (n_g) of NR7-500P is taken to be 1.604 from 700 nm - 800 nm wavelength range. First, we fixed $f = 0.8$ and varied h from 100 nm to 600 nm and the obtained FDTD result is shown in Fig. 2(a). The absorbance value varies within the range of 0.4 to 0.7 and attains a maximum value ($A \approx 0.7$) when h is 225 nm and 500 nm. Keeping h value fixed at 225 nm and 500 nm, we analysed the effect of f and the obtained results are shown in Fig. 2(b). As can be seen from this result, the absorbance value is maximum (≈ 0.7) when f is around 0.8 for both 225 nm and 500 nm depth. For our further analysis, we chose the grating depth to be around 225 nm due to the following three reasons. First, because NR7-500P is a thin photoresist, we

could achieve this film thickness easily. Second, photoresist material absorption (if any) will have reduced effect on the sole analysis of the graphene absorption. Thirdly, the results of the coupled mode theory are more accurate when the grating depth is thin [28]. To understand the reason for maximum absorption at $f = 0.8$, we calculated the external leakage rate (γ) and the loss rate (δ) for $f = 0.2$ to 0.9. γ is calculated by Lorentzian fit of the stored energy at the guided resonance. When there is no graphene layer in the GMR, the center frequency of the Lorentzian (fitted as a function of frequency) gives the resonance frequency (ω_0) and the half-width at half maxima provides γ . In presence of the graphene monolayer, we repeat the same procedure to find the half-width at half maxima, from which we calculated ' $\gamma + \delta$ ' and δ . The obtained γ and δ values (Table 1) are plotted in Fig. 2(c).

Fig. 2. (a) Variation of absorbance with grating depth for $f = 0.8$, $n_g = 1.604$, $\Lambda = 500$ nm and $d = 100$ nm. (b) Variation of absorbance with duty cycle for two fixed grating depth, 225 nm and 500 nm (c) Leakage rate (γ) and loss rate (δ) plotted as a function of duty cycle (f) comparison of theoretically calculated absorbance with simulated values as function of duty cycle.

As can be seen from the figure, γ has different values for different duty cycles, whereas δ is almost independent of it. It is interesting to see that both γ and δ have equal values at 0.8. This is the case of critical coupling where the condition for maximum absorption is achieved [14]. We can understand the condition for maximum absorption as follows: when $\gamma > \delta$, photons decay away from the cavity before being absorbed by the graphene layer. On the other hand, when $\gamma < \delta$, photons cannot be coupled into the cavity. Thus, maximum absorption is attained when the critical coupling is achieved when $\gamma = \delta$ at $f = 0.8$. After calculating the γ and δ values, we obtained the off-resonance reflectance (r) and transmittance (t) values.

Table 1. List of calculated parameters; γ , δ , r^2 and t^2 , where γ and δ are in the unit of 10^{-6} nm^{-1} .

f	γ	δ	r^2	t^2
0.3	6.0320	2.4239	0.1359	0.850693
0.4	7.2716	2.4224	0.1392	0.847516

0.5	7.0081	2.3621	0.1504	0.836448
0.6	5.7384	2.2929	0.1621	0.824899
0.7	3.9606	2.2886	0.1732	0.814005
0.8	2.1855	2.1019	0.1815	0.805782

The values of ' r ' and ' t ' are obtained by replacing the grating film with a uniform slab of effective refractive index, n_{eff} and thickness that is same as the grating depth (225 nm). n_{eff} depends on both f and n_g as per Eq. (1). Thus, using Eq. (1), we calculated n_{eff} and used this value to calculate ' r ' and ' t ' (Table 1). After we obtained all these required parameters, we calculated the peak absorbance at the resonance wavelength (λ_0) from Eq. (7). To validate the theoretically calculated absorption values, we simulated the GMR-graphene structure in FDTD and compared the obtained absorption values with the calculated values. The obtained results are plotted in Fig. 2(d), from which we can infer that both the theoretical values and simulated values agrees well with each other.

To validate our theory, we fabricated the proposed GMR structure on a quartz substrate. A 100 nm silicon nitride film was deposited on a quartz plate by plasma enhanced chemical vapor deposition (PECVD) method. Next, monolayer graphene was grown using chemical vapor deposition method (CVD) system at $>1000^\circ\text{C}$ on a copper growth substrate. Then the grown graphene layer is transferred on to the silicon nitride coated substrate using wet chemical etching method [29].

Fig. 3. (a) Measured Raman peak of the transferred graphene (b) Fabricated sample showing the image of transferred graphene on the SiN coated quartz plate. Also shown the interference lithography pattern area (c) AFM image of the grating profile with magnified image in the inset (d) AFM depth profile of the fabricated grating, in the GMR structure.

To confirm the successful transfer of the graphene, we used Raman spectroscopy to characterize the quality of the transferred graphene as shown in Fig. 3(a). The characteristic '2D' peak ($\sim 2650\text{ cm}^{-1}$) is clearly observed which indicates the presence of graphene. The 'G' peak ($\sim 1580\text{ cm}^{-1}$) corresponds to the ordered hexagonal structure of the graphene. Additionally, the number of graphene layers can also be inferred from the relative intensity of the '2D'

peak to the 'G' peak (I_{2D}/I_G). A high I_{2D}/I_G ratio of more than 2 indicates single-layer graphene and a lower ratio corresponds to a greater number of graphene layers. Therefore, from the Raman data, we confirm that high quality, monolayer graphene has indeed been grown and transferred on top of the silicon nitride coated substrate.

To get large area grating patterning, we use 2-beam interference lithography (IL) system to fabricate the required features. Different configurations of IL and their respective benefits and limitations have been well reported by our group in recent years[30, 31]. Here we have used a phase grating (Ibsen Photonics) to split the incident laser beam into multiple sub-beams. The required angle of incidence (θ) to achieve the targeted 500nm periodicity is calculated (from $\Lambda = \lambda / 2\sin\theta$) to be 20.54° for $\lambda = 351\text{ nm}$, the wavelength of our Argon ion laser. A thin layer of NR7-500P resist is spin coated on the graphene coated substrate and exposed using the IL system to record the interference pattern. After developing and drying, the generated pattern area and the graphene layer on the substrate are inspected using a laser confocal microscope (Keyence, VK-X Series) as shown in Fig. 3(b). To infer the grating depth profile, we took atomic force microscopy (AFM) measurement of the patterned area. The top view of the grating (zoomed image in the inset) is as shown in Fig. 3(c). The side wall of the grating depth profile is slanted (as shown in Fig. 3(d)) because of the sinusoidal 2-beam interference pattern. The measured grating parameters are period $\Lambda = 500\text{ nm}$, average depth $d = 240\text{ nm}$, base width 470 nm and top width 100 nm.

Fig. 4. (a) Schematic of the experimental set up used for measurement of resonance spectrum of the guided-mode resonant structure. (b) R, T and A of a graphene monolayer on the quartz substrate without any GMR structure (c) R, T and A of a graphene monolayer when embedded in the GMR structure (d) Comparison of the experimentally obtained absorption spectrum and its Lorentz fit with the simulated spectrum.

The absorption spectrum of the fabricated GMR sample was measured using a spectrometer and a broadband (halogen) light source, as shown in Fig. 4(a). Multimode optical fiber and lenses are used for light coupling collimation purposes. S-polarized light obtained using a polarizer is incident on the sample. We first measured the absorption spectrum for the graphene monolayer (without patterning) on the silicon nitride coated quartz substrate. The obtained transmittance (blue), reflectance (green) and absorbance (red) spectrum are shown in Fig. 4(b). As can be seen from the figure, the graphene monolayer has a uniform absorption of 2.3% throughout the whole spectrum. The absorption spectrum with the grating pattern is shown in Fig. 4(c) that shows resonance at 762.16 nm with peak absorbance value equal to 0.5952. As can

be seen from the figure, the reflectance of the structure at or near resonance is greatly reduced because of the critical coupling condition. To validate the experimentally obtained value with the theoretical formula (Eq. (7)), we need to find the off-resonance reflectance (r) and transmittance (t) values. Since the fabricated grating profile is not binary, Eq. (1) cannot be applied to find the effective index (n_{eff}) of the grating layer. For this purpose, we simulated the experimentally obtained grating profile in FDTD as shown schematically in Fig. 5(a). The effective refractive index (n_{eff}) (in a unit cell) for this grating (pyramidal) profile varies gradually from grating-waveguide interface to grating-air interface. To calculate r and t , this grating is replaced with a uniform slab of thickness equal to the grating depth (240 nm as in experiment) with a gradient refractive index profile (as shown in Fig. 5(b)) that follows an equation, $n_{\text{eff}}(z) = 1.5824 - 0.0018z$.

Fig. 5. (a) Schematic of the simulated (FDTD) grating profile having a slanted side wall to replicate the experimentally obtained grating profile. (b) The corresponding effective gradient refractive index profile calculated in one period of the grating that follows the equation $n_{\text{eff}}(z) = 1.5824 - 0.0018z$, z being along the grating depth.

The reflectance and transmittance values thus obtained are $r^2 = 0.115835$ and $t^2 = 0.870444$, respectively. The calculated leakage rate and loss rate for this geometry are $\gamma = 4.5014 \times 10^{-6}$ and $\delta = 2.2396 \times 10^{-6}$, respectively. Using these values, the absorptance at the resonance wavelength (762.16) is calculated to be 0.5962 which is almost equal to the experimentally obtained value (0.5952), thus validating our theoretical prediction. To further validate the theory, we compared the experimentally obtained absorption spectrum with that of the simulated GMR structure modelled using FDTD. The result is shown in Fig. 4(c), where we have fitted the experimentally obtained absorption spectrum with a Lorentzian function of frequency. Both the results show similar absorption spectrum except that the simulated resonance wavelength is red shifted from the experimental spectrum. The reason for this discrepancy can be due to the deviation in nitride film thickness and/or the change in duty cycle or nonuniform resist thickness throughout the patterned area of the sample. Nevertheless, the simulated absorptance value (0.595179) is well matched with the experiment (0.5952) and theoretical value (0.5962).

In conclusion, the absorption in graphene embedded 1D GMR structure is calculated using the model and formula developed based on coupled mode theory. The proposed model does not require any back reflector such as metal film or Bragg mirror. Using this formula we optimized the GMR parameters to enhance the absorption up to 70% which is higher than the reported values without back reflectors [15, 26, 27] and comparable with results with back reflectors [16]. Our model can be applied for binary as well as other grating profiles such as pyramidal grating profile

obtained using interference lithography. The theoretical, numerical and experimental results correlate well with each other. Finally, this IL system can enable tuning of the operating wavelength easily by changing the grating period.

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