Graphene: A Dynamic Platform for Electrical Control of Plasmonic Resonance

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1 Introduction

Nanophotonics is a thriving research field, which holds tremendous promise for optical information processing at the nanoscale. One of the most promising directions in nanophotonics is to use free charge oscillations in metal nanostructures known as surface plasmons (SPs) for control and manipulation of optical signals [1, 2]. Surface plasmons exhibit a resonant behavior at a particular wavelength, when the electric restoring force equals the Lorentz force caused by an incident field. At such a resonance the electromagnetic energy is confined to subwavelength length scales in the close vicinity of the nanostructures. This unprecedented ability to confine energy in small volumes has resulted in a number of novel applications like surface plasmon assisted lithography [3], data storage [4], enhanced chemical reaction rates [5, 6], improved photovoltaic devices [7], particle trapping [8, 9], and biosensing [10].

At present, there are two main challenges in realizing the full promise of plasmonics. First, the traditional noble metals (gold and silver) used in plasmonics exhibit relatively high intrinsic optical loss. They are also not fully CMOS compatible and hence cannot be integrated with electronic components. In recent years a number of significant breakthroughs were made in identifying alternative plasmonic materials [11]. Naik et al. demonstrated that transparent conducting oxides like aluminum doped zinc oxide, and ceramic nitrides like TiN can be used to lower losses and thereby improve the performance of plasmonic devices [12, 13]. The second major challenge in plasmonics is the lack of post fabrication dynamic control of plasmonic resonance. Currently the resonant wavelength of plasmonic nanostructures is dependent on their...
size, shape, and material properties [1, 14]. In the literature we find that a number of phenomena like phase transitions [15], electrical carrier injection in semiconductors [16, 17], ultrafast optical pumping [18] and mechanical stretching of elastic membranes [19, 20] have been used to tune plasmonic resonances. The magnitude and speed of change in resonance are important criteria in evaluating the merits of these approaches for specific applications.

Recently the synergy between the exceptional optoelectrical properties of graphene and plasmon resonances in metallic nanostructures led to the development of electrically modulated plasmonic resonance. These devices can potentially lead to applications like modulators, photodetectors, and sensors in near-infrared (NIR) to mid-infrared (MIR) wavelength ranges. In this review we begin with a discussion of the fundamental optical properties of graphene, which makes it very attractive for the dynamic control of plasmon resonance. We also describe numerical approaches to model graphene using full wave electromagnetic simulations. We then survey the current status of graphene-hybrid devices in MIR and NIR wavelength ranges. We then identify current difficulties, and present our perspective of this exciting new research direction.

2 Optical properties of graphene

2.1 Band structure of graphene

Graphene is a hexagonal lattice of sp² hybridized carbon atoms arranged in a single atomic layer. In the crystalline phase, the electronic configuration of carbon is 1s², 2s², 2p². The 2s and 2p orbitals can mix with each other in three possible ways giving rise to sp, sp², and sp³ hybridizations forming different carbon-based molecules [21]. In the reciprocal space, the unit cell can be described by two basis vectors a₁ and a₂ as shown in Fig. 1. Starting from these basis vectors and using the tight binding approximation [21] it can be shown that the band structure can be described using the equation

\[ E(k) = \pm \gamma_0 \sqrt{1 + 4 \cos \frac{\sqrt{3} k_x a_0}{2} \cos \frac{k_y a_0}{2} + 4 \cos^2 \frac{k_y a_0}{2}} \]

where, \( a_0 \) is the lattice constant, \( k_x \) and \( k_y \) are reciprocal lattice vectors, and \( \gamma_0 \) is the nearest neighbor hopping energy (typically in the range 2.6–3.1 eV).

The conduction and valance bands touch each other at the six corners of the hexagonal 1st Brillouin zone known as the K points as shown in Fig. 1b. Close to the K points the dispersion relation for graphene is linear \( (E = h \nu_F k) \).

The Fermi energy \( (E_F) \) is given by

\[ E_F = h \nu_F k_F = h \nu_F \sqrt{\pi n_{g,2D}} \]

where, \( n_{g,2D} \) is the carrier concentration in the two-dimensional graphene sheet, \( \nu_F \) is the Fermi velocity \( (=1 \times 10^6 \text{ m/s}) \). This linear dispersion implies that charge carriers in graphene behave as massless Dirac fermions. This is the underlying reason for many novel electronic and optical properties of graphene like large carrier mobility (and hence low DC scattering rate) and allowed interband transitions from terahertz to visible frequencies [22, 23]. It can easily be shown that this linear dispersion results in a linear dependence of the electronic density of states (DOS) on the Fermi wavevector \((k_F)\) around Dirac point which is given by

\[ \text{DOS}(k_F) = \frac{k_F}{\pi \nu_F} = \frac{\omega}{\pi \nu_F^2} \]

Therefore, undoped graphene is extremely sensitive to small external perturbation especially at IR frequencies. This also leads to highly nonlinear current change close to
the Dirac point [24]. This is clearly different from a conventional 2D electron gas, with parabolic dispersion, where the density of states is constant \((m^*/\hbar^2)\). Further, the sheet carrier density can be electrically tuned by electrostatic gating using a field effect transistor (FET) structure. Due to these fascinating electrical properties, graphene was initially considered as an alternative to silicon for nanoelectronics.

2.2 Intraband and interband transitions in graphene

The electronic properties and optical properties of graphene are very closely related. In undoped graphene at 0 K, optical absorption is frequency independent—determined only by universal absorbance, \(A(\omega) = \pi e^2/\hbar c = \pi a = 2.3\% [23]\). This is due to the cancellation of frequency dependent terms in the three important parameters determining optical absorption in Fermi golden rule—the square of transition matrix element \((\propto v_F^2/\omega^2)\), the joint density of states \((\propto \omega/\hbar v_F^2)\), and the photon energy \((\propto \omega) [25]\).

The optical absorption in doped graphene depends strongly on the sheet carrier density. A photon incident on a graphene sheet can excite two types of possible transitions—an intraband transition where the initial and final states of the electron are within the same band, and an interband transition where the incident photon excites an electron in valence band into the conduction band (see Fig. 1d). Further, an interband transition can generate an e-h pair only if the energy of the incident photon is greater than \(2E_F\). These transitions can be analytically expressed in terms of optical conductivity of graphene derived under the Random Phase Approximation (RPA) in the local limit as [26–28]:

\[
\begin{align*}
\sigma(\omega) & = \frac{2e^2\omega_T}{\pi\hbar} \frac{i}{\omega + i\tau} \log \left[ 2 \cosh \left( \frac{\omega_F}{2\omega_T} \right) \right] \\
& + \frac{e^2}{4\hbar} \left[ H \left( \frac{\omega}{2} \right) + \frac{2\omega}{\pi} \int_0^\infty \frac{H \left( \frac{\omega'}{2} \right)}{\omega^2 - \omega'^2} d\omega' \right]
\end{align*}
\]

(4)

where, \(H(x) = \sinh(x)/\cosh(x/2) + \cosh(x/2)\cos(\omega_T x)\), \(\omega_F = E_F/\hbar, \omega_T = k_B T/\hbar, \omega\) is the frequency of incident light, \(e\) is the charge of an electron, \(\tau\) is the Drude relaxation rate, \(T\) is the temperature, and \(k_B\) is the Boltzmann constant. The first term represents the free carrier response of graphene arising due to the intraband transitions. The intraband response is dominant above MIR wavelengths as can be seen in Fig. 2(a–b). The second term describes the contribution of interband transitions, which are the dominant contribution at visible and NIR wavelengths, where the conductance has a constant value of \(G_0 = (e^2/4\hbar)\). The optical conductivity calculated using RPA is consistent with numerous experimental reports in literature [23, 29, 30].

2.3 Numerical Modeling of Graphene

To develop useful devices with graphene it is essential to develop accurate models for numerical simulations. Graphene being a vanishingly thin monolayer poses unique challenges for numerical treatment. There are two main approaches for the numerical implementation of graphene (i) treat graphene as an effective medium with finite thickness \(t_g\) [31] and permittivity given by \(\varepsilon(\omega) = 1 + i\sigma(\omega)/\omega\varepsilon_0 t_g\), (ii) describe graphene with a surface current \(J_s = \sigma(\omega)E\). In the first approach there can be stability and convergence problems if the choice of \(t_g\) is not sufficiently small. Further, an extremely small thickness will add unnecessary computational complexity due to the requirement of a fine mesh size [32]. Hence, the second approach is more suited for grid-based, full-wave finite-element frequency domain simulation methods using for example commercial tools such as COMSOL Multiphysics or CST Studio. The validity of the numerical modeling can be seen in Fig. 3 where the experimentally measured IR reflectivity of single layer graphene (SLG) normalized to reflectivity at charge neutral point (CNP) is shown along with the numerical simulations. When the carrier density is increased SLG becomes more “metallic” and hence reflectivity is higher. The dip seen in the 9–10 \(\mu m\) range is due to the optical phonon in the SiO\(_2\) layer which leads a large reflection peak and an effectively small change as a function of carrier density. The features at 8 \(\mu m\) are related to the epsilon near zero enhanced absorption which appear when reflection from a range of angle of incidence \((\phi)\) is measured. We can see from Fig. 3(b) that simulation at normal incidence alone does not capture the experimental data. To better match the experimental features we averaged simulations over a range of \(\phi\)—the full details of which are discussed in an upcoming paper. The final result of the averaging procedure is shown in Fig. 3(c) which indicates the RPA model is able to capture the behavior of graphene remarkably well considering that the experiments were performed using Chemical Vapor Deposition (CVD) grown with up to 20% inhomogeneity. An important point to note is that the scattering time extracted from our experimental and numerical analysis is about five times smaller than estimate based on DC Drude model [33].
**Figure 2:** Relative contributions of interband and intraband transitions to real part (a) and imaginary part (b) of conductivity. Interband conductivity is dominant at visible frequencies while intraband contribution dominates above mid-infrared frequencies ($n_{g,2D} = 7 \times 10^{11} \text{ cm}^{-2}$, $T = 300 \text{ K}$, $\tau = 3 \times 10^{-14} \text{ s}$); (c) Figure of merit calculations indicating that graphene is a good plasmonic material between 2 µm–20 µm wavelengths. The plasmonic response can be tuned to near-infrared wavelengths by increasing carrier density in graphene.

**Figure 3:** (a) Modulation of IR reflectivity of CVD graphene on Si/300 nm SiO$_2$ substrate as a function of sheet carrier density. Measurements were performed using a Fourier Transform Infrared (FTIR) Spectrometer with microscope accessory (Objective: 15X, N.A. 0.58 Reflectochromat); measurements are normalized to $R_{\text{CNP}}$. (b-c) FEM simulations using a surface current model for graphene (COMSOL Multiphysics) at normal incidence and averaged over a range of angle of incidence ($\phi$); the experimental data was fitted with carrier scattering time ($\tau$) as a fitting parameter in the RPA model, which yields scattering time of 10 fs.

The RPA model cannot be used directly in time domain simulations because the interband conductivity does not have a simple time domain form. Several groups report time domain simulations by either ignoring interband contribution or assuming asymptotic conditions [34–36]. These approximations are not valid at optical wavelengths or room temperature. We recently developed an accurate, multivariate time domain model for graphene using two Padé approximant terms, which accurately captures the time domain response of graphene from visible to IR wavelengths [37, 38]. This model has been already tested by combining critical-point approach [37, 38] with a surface-current enabled FDTD scheme [32, 39].

**2.4 Gating techniques for control of optical properties**

Electrical backgating is the most popular approach to control the charge density in the graphene sheet. In this method, a DC voltage is applied to the silicon substrate separated from the graphene sheet by a thin insulating layer. The induced carrier density can be estimated using a parallel plate capacitor model as $n_{g,2D} = C_{\text{gate}}(V_G - V_{\text{CNP}})/q$, where $C_{\text{gate}} = \varepsilon_{\text{insulator}}/t_{\text{insulator}}$ is the gate capacitance, $V_G$ is the applied gate voltage and $V_{\text{CNP}}$ is the charge neutral point voltage and $q$ is the charge of the electron. Thermally grown 300 nm SiO$_2$, which is typically used as an insulator, gives a gate capacitance of 11.5 nF cm$^{-2}$. A thin insulating layer is usually preferred since it would give better capacitive control over the in-
duced charge. However, when the insulating films are thinner than 100 nm the gate leakage current increases causing the device to breakdown. The Si/SiO$_2$ backgating technique typically yields a highest carrier density of ~7 \times 10^{12} \text{ cm}^{-2} taking into account device area and leakage considerations. The gate capacitive control can be slightly improved by using a high-$k$ material like HfO$_2$ or Al$_2$O$_3$ instead of traditional SiO$_2$ as an insulator [40].

A simple yet effective method of achieving large carrier densities is to use an electrochemical top gate [41], wherein the gate voltage is applied to an electrolyte deposited on top of a graphene sheet (see Fig. 5a). The electrochemical gating scheme exhibits a much higher gate capacitance (~μF cm$^{-2}$) due to the formation of an extremely thin electrical double layer at the interface [42, 43]. Graphene can also be doped by exposure to chemicals such as Nitric Acid (HNO$_3$), Iron (III) Chloride (FeCl$_3$) and Gold (III) Chloride (AuCl$_3$), which leads to large sheets carrier density due to charge transfer reactions [44]. However, chemically doped devices are also known to degrade over time making them less robust compared to back gated devices.

### 3 Graphene as a platform for tunable plasmonic devices

#### 3.1 Fundamentals

Surface plasmons enable the confinement of the incident electromagnetic energy into a subwavelength volume at the resonant wavelength. The characteristic resonant wavelength is extremely sensitive to the local dielectric environment, which can be effectively used to modulate the optical response of the resonance. The changes in optical properties of graphene described in the previous section, will perturb the plasmonic resonance in metallic nanostructures if they are fabricated close to the graphene sheet. The changes in resonance frequency due to this small dielectric perturbation can be estimated using the perturbation theory using the equation [45, 46]

$$\frac{\Delta \omega}{\omega_0} = -\frac{\iiint V dV \left[ \left( \Delta \vec{\mu} \cdot \vec{H}_0 \right) \cdot \vec{H}_0^* + \left( \Delta \vec{e} \cdot \vec{E}_0 \right) \cdot \vec{E}_0^* \right]}{\iiint V dV \left[ \mu \cdot \vec{H}_0 \cdot \vec{H}_0^* + e \cdot \vec{E}_0 \cdot \vec{E}_0^* \right]}$$

$$= -\frac{\Delta W_m + \Delta W_e}{W_m + W_e}$$  \hspace{1cm} (5)

where, $\omega_0$ is the resonance frequency, $E_0$ and $H_0$ are unperturbed electric and magnetic fields, $\Delta \vec{e}$ is the change in dielectric permittivity, $\Delta \vec{\mu}$ is the change magnetic permeability, $W_e$ and $W_m$ are the total electric and magnetic energies, respectively. The change in resonance frequency $\Delta \omega$ is large when a strong perturbation of material parameters overlaps with the electric and magnetic fields of the resonant mode leading to a large numerator contribution in Eq. 5. The integral in the numerator captures the change in the stored electromagnetic energy, whereas the denominator represents the total electromagnetic energy. Therefore, the fractional change in resonance frequency is simply proportional to fractional changes in the total electromagnetic energy. This approach provides a quantitative measure to analyze graphene-plasmonic hybrid geometries, and has been verified for dipole antennas [47] and Fano resonant metasurfaces [48] in MIR wavelengths.

#### 3.2 Tunable MIR plasmonic devices

In recent years, many groups have attempted to combine the dynamic tunability of graphene with plasmonic resonances for IR modulator applications. The extraordinary sensitivity of plasmonic modes to the presence of graphene was first reported in split-ring resonators [49]. However, in that work the influence of carrier density and the exact nature of the interaction was not discussed [50]. Subsequent studies using bow-tie antennas addressed this issue and concluded that graphene modulates the plasmonic resonance strongly in MIR frequencies [51, 52]. The width of the plasmon resonance was found to decrease as the Fermi energy is increased. This is easily explained by considering the allowed interband transitions. At a Dirac point all interband transitions are allowed leading to higher loss and therefore wider resonance. However, as the Fermi energy is increased some of the interband transitions are blocked and hence the width of the resonance is lowered. It should be noted that despite the early promise—up to 25% peak change in transmission at a 5 μm wavelength (see Fig. 4a)—the bow-tie antennas did not show any spectral tunability. Further, the incident electromagnetic energy interacts strongly with graphene only in the narrow (~80 nm × 80 nm) neck region of the bow-tie antenna. In fact, for any dipole antenna at resonance the electromagnetic energy is confined only to the small regions around the edges of the antenna. Therefore, these geometries will not be very sensitive to changes in graphene and are not strongly tunable. An improved design for tunable resonance in MIR frequencies was proposed by Yao et al. where they incorporated ideas from Metal-Insulator-Metal (MIM) waveguides into the antenna geometry [53, 54]. In a clever design they stagger...
two adjacent nanorod antennas so that the electric field is confined to regions between the nanorods—similar to a bonding mode in a MIM waveguide. They report improved tuning range of ~100 nm at 6.5 µm wavelength (see Fig. 4b). The tunability of plasmon resonance could also be improved by using Fano resonant metallic structures, which possess overlapping dipolar and quadrupolar resonances [55, 56]. These resonant modes mutually interfere and produce much sharper spectral features, and hence show greater sensitivity to graphene. A large blue shift (~30 cm$^{-1}$) was shown using Fano resonant metasurface [48, 56] in MIR wavelengths (see Fig. 4c). The progress made by a number of research groups toward graphene-based modulation schemes is tabulated in Table 1. At mid-IR wavelengths MIM mode in plasmonic antennas and Fano resonant antennas have shown promising amplitude and spectral modulation results [56, 57]. Dynamic modulation has not received sufficiently wide attention—with only one report from Harvard University demonstrating 3 dB bandwidth of up to 20 GHz [57]. There is still significant room for further improvement of modulation speed since the intrinsic carrier transport occurs on a picosecond timescale in graphene [58].

### 3.3 Tunable NIR plasmonic devices

There is also tremendous interest in applying graphene to realize modulators and sensors at NIR and visible wavelengths, since it can potentially lead to devices with small footprint and high modulation speeds [59, 60]. It can be seen from Fig. 2c that large changes in conductivity in NIR frequencies occur when the carrier density in graphene is > 1 × 10$^{13}$ cm$^{-2}$. The conventional backgating technique is not adequate to realize such high carrier densities. Therefore, ionic liquid gating discussed earlier is typically used for experiments at NIR and visible wavelengths. Modulation of plasmonic resonance in NIR frequencies was demonstrated in nanorods [61] and dolmen structures [62]. The influence of graphene is much stronger in dolmen structures because of the higher sensitivity of Fano-type resonance. High-$k$ dielectrics like Al$_2$O$_3$, which provide stronger capacitive control on the charge carriers at the same gate voltage, provide yet another direction improving the maximum carrier density in graphene and thereby achieve stronger modulation [57]. In another intriguing report electrically tunable surface plasmon polaritons at visible wavelengths have been realized by exploiting the high localized electric fields at the interface of Ag nanowire and graphene to achieve the huge carrier density needed [63].

### 3.4 Other graphene-plasmonic hybrid devices

In the last few years a number of research groups used plasmonic structures to improve the response of graphene photodetectors. The primary advantage of graphene over traditional photodetectors is its broad spectral sensitivity (due to its unique electronic band structure), fast transport properties [64] and short intrinsic lifetimes [58]. Photodetectors with speed exceeding 40 GHz [65] and intrinsic bandwidth of 260 GHz [58] have been demonstrated. One of the primary challenges for graphene based photodetectors is the small responsivity which is the result of finite optical absorption in single layer graphene. Further, the extraction of photocurrent is efficient only in the regions where there is a field gradient (typically p-n junctions). Echtermeyer et al. showed that by using plasmonic nanostructures it is possible to efficiently localize electromagnetic energy near the p-n junctions, and thereby enhance the efficiency of photodetectors by up to 20 times [66]. The plasmonic nanostructures help in improving the spectral selectivity of photodetectors and could potentially enable multicolor photodetection [67]. In addition to the direct excitation of e-h pairs in graphene, the hot electrons in plasmonic structures have also been shown to contribute to

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<th>Table 1: Comparison of different approaches reported in literature demonstrating modulation of plasmonic resonance with graphene.</th>
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<td>Graphene Ag-nanowire waveguide (63)</td>
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Some of the data is approximated from the experimental results in the references mentioned.
(a) The change reported is obtained by comparing antennas with graphene to antennas without graphene.
(b) Gate capacitance not reported. $n_\text{max}$ extracted from optical reflectivity measurements.
(c) Estimated by assuming a dielectric constant of 8 for AlO$_x$. Alumina provides greater gate control and hence higher carrier density.
(d) Estimate also includes local electric field enhancement due to the nanowire in addition to gate.

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photocurrent—resulting in an 800% increase in photocurrent compared to pristine graphene [68]. However, most of these experiments report improved responsivity at the cost of speed, spectral sensitivity, or device dimensions and are performed at visible wavelengths. Novel device architectures which can blend the tunability and improved responsivity of graphene-plasmonic antenna hybrid photodetector will hold significant potential for practical applications especially at IR wavelengths.

4 Conclusions and future directions

Graphene-antenna hybrid platform has shown considerable potential for the development of compact and ultrafast MIR modulators [56, 57]. There is significant scope for improvement in the NIR wavelength range where there have only been a few demonstrations [61, 62]. An important shortcoming of most of the graphene-plasmonic hybrid devices so far is the work function mismatch at the metal–graphene interface, which leads to Fermi level pinning at the interface [69]. This adversely impacts the charge transport behavior and lowers tunability of the plasmonic resonance. Mueller et al. report that the metal contact induced doping of graphene not only occurs un-
under the contact, but also extends 200–300 nm into the graphene sheet [70]. While it is known that metallic nanostructures cause electron doping of graphene [51], the diffusion length of carriers is presently unclear. This needs to be investigated carefully through scanning photocurrent measurements to optimize the tunability of plasmonic resonance. Another direction for improving the tunability is to increase the number of graphene layers, and thereby improve the optical conductivity [71] and strength of interaction with plasmonic resonance. Study of screening and interlayer coupling in exfoliated multilayer graphene shows that linear band structure model for graphene is valid up to four layers [72], which suggests that some of the unique properties of graphene will be preserved up to a few layers. Lastly, the experimental carrier scattering time obtained by fitting experimental IR reflectivity measurements with RPA model is only about 10 fs. Improving the uniformity of CVD-grown graphene will be very important in improving the scattering time and thereby the electro-optical response. Looking into the future, theoretical studies have shown that graphene patterned down to sizes below 10 nm can also support plasmons [73], and can potentially enable nonlinear phenomena at the single photon level [74]. Advances in the aforementioned challenges can lead to the development of exciting new technologies in addition to the NIR and MIR graphene-plasmonic antenna hybrid devices discussed in this review.

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