

# Electronic Scattering of Graphene Plasmons in the Terahertz Nonlinear Regime

Kelvin J. A. Ooi, Yee Sin Ang, Jin Luo Cheng, Lay Kee Ang, *Senior Member, IEEE*, and Dawn T. H. Tan

**Abstract**—Graphene possesses large Kerr nonlinearities that enabled the realization of energy-efficient nonlinear optical devices. At the same time, electronic scattering of graphene plasmons shows very high energy transfer efficiencies at very low beam energies due to its extremely large plasmon wave vector. We have tapped into these two unique optical properties of graphene to study the potential of an energy-efficient optoelectronic light source and all-optical modulator. We found that  $2\pi$ -phase shift and 94% loss modulation is achievable with just a 25.6-V electron beam passing 10 nm above a graphene waveguide with a Fermi level of 0.1 eV. This also enables nonlinear optical devices to utilize *in situ* generated plasmons without the need of external optics, which could herald the return of vacuum nanoelectronics.

**Index Terms**—nonlinear optics, graphene, Electron beams, plasmons.

## I. INTRODUCTION

THERE is a growing interest in graphene plasmonics of late due to graphene's unique optical properties and thin film structure that makes it a potential candidate for a nanoscale photonic integrated circuit platform [1]. Among these optical properties are graphene's two-dimensional linear optical conductivity and configurable Fermi-level, which enabled very large optical confinement and increased light-matter interaction that can be tailored to any wavelength from the visible to the mid-infrared. At the same time, there is also an increasing number of research activities invested in studying the Kerr nonlinearity of graphene plasmonics. Theoretical and experimental studies consistently found that graphene's Kerr coefficients are at least 1–2 orders higher than the conventional nonlinear dielectrics, and at least on par with metals such as gold [2]–[16]. In addition, a few studies also found that when graphene is used on a plasmonic platform, there is a surface-induced nonlinear enhancement effect which scales with the fourth-power of the optical confinement, and thus further augments the nonlinearity by a thousand to million fold [12]–[16]. In one of our recent studies, we calculated that these enhancements could poten-

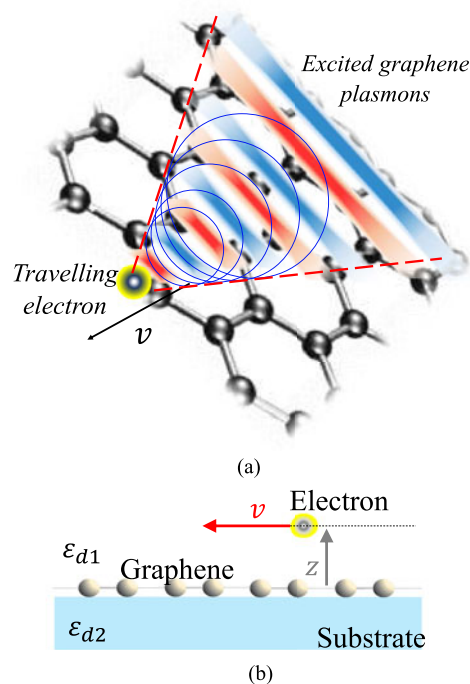


Fig. 1 (a) Schematic impression of a travelling electron generating plasmonic waves on graphene. (b) Side view of the model.

tially drive optical pump intensities and powers down to the sub-MW/cm<sup>2</sup> and pW range respectively [16].

Graphene plasmonics is also studied as a potential candidate for miniaturized light sources. Two categories of plasmon-type light sources exist, from the low-powered tunnelling-junction sources [16]–[20], to the high-powered electron-beam sources [21]–[27]. Electron-beam excited propagating plasmons in metals is a well-studied phenomenon which shows potential for development of compact light sources on nanoscale platforms [21], [22]. With the recent popularity of graphene plasmonics, there has been theoretical studies in electron-beam excitation of graphene plasmons to generate coherent terahertz radiation [23]–[26]. In these studies, it is found that the swift electrons interact strongly with graphene (Fig. 1) to produce high radiation intensities at low powers. The main reasons for this high radiative efficiency are due to the low electron velocities and two-dimensional structure of graphene that enabled large electron-plasmon coupling, as has been studied in detail in our previous paper [26].

The ease of generating high intensity graphene plasmons with electron beams necessitates the introduction of the nonlinear optical conductivity into the physical picture for better modelling accuracy. Besides that, it could also reveal the

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potential of harnessing the electron-plasmon interaction for active manipulation of plasmonic waves. Recently, Gong *et al.* is the first to investigate the direct transformation of graphene plasmons to terahertz radiation through the nonlinear effect of graphene [27]. In this paper, we set forth to quantitatively predict the strength of the total electric field that can be coupled to the propagating graphene plasmonic mode that induces graphene's Kerr nonlinearity, and also showcase its use for active nonlinear switching for electron beams.

## II. FORMALISM

The general expression of the total electromagnetic energy which can be coupled from a moving electron to graphene plasmons is written as [21]

$$U = N \int \hbar\omega\Gamma(\omega)d\omega \quad (1)$$

where  $U$  is the coupled energy,  $N$  is the number of electrons in a bunch,  $\hbar\omega$  is the photon energy, and  $\Gamma(\omega)$  is the energy loss probability of an electron. Specifically looking only at the coupling to surface plasmon modes, we can write the loss probability as [21]

$$\Gamma_{\text{SP}}(\omega) = \frac{2e^2L}{\pi\hbar\omega^2} \cdot K_0\left(\frac{2\omega z}{v\gamma_{\text{LF}}}\right) \cdot \text{Im}\left(\frac{r_p}{\varepsilon_0\varepsilon_d}\right) \quad (2)$$

where  $L$  is the length of energy transfer,  $v$  is the electron velocity,  $z$  is the gap between the electron bunch and graphene,  $\gamma_{\text{LF}}$  is the Lorentz contraction factor,  $K_0(x)$  is the zeroth-order modified Bessel function of the second kind, and  $\varepsilon_d$  is the background permittivity.  $r_p$  is the p-polarized Fresnel reflection coefficient of graphene, which is written as [23], [26]

$$r_p = 1 - \frac{2\varepsilon_{d1}}{\varepsilon_{d1} + \varepsilon_{d2} + \frac{i\sigma_g k_{\text{sp}}}{\varepsilon_0\omega}} \quad (3)$$

where  $\varepsilon_{d1}$  is the incident background permittivity,  $\varepsilon_{d2}$  is the substrate permittivity,  $\sigma_g$  is the Kubo conductivity for graphene [28], and  $k_{\text{sp}}$  is the wave vector of the graphene plasmons.

Next, we also write the relation between the coupled electromagnetic energy and the induced plasmon electric-field strength

$$U = \frac{1}{2}\varepsilon \int |E|^2 dl \quad (4)$$

where  $dl$  is differential length along the electron trajectory.

In a tightly confined graphene plasmonic waveguide, the induced plasmon electric-fields have roughly equal distribution between the in-plane ( $E_L$ ) and out-plane ( $E_T$ ) components according to  $E_T \approx iE_L$  [16]. Taking into account only the in-plane longitudinal electric-fields which can directly induce the nonlinearity of graphene, we finally express the electric-field strength as

$$|E_L|^2 = \frac{1}{\varepsilon} \frac{dU}{dl} = \frac{\hbar}{\varepsilon L} \int_{\omega_{\text{min}}}^{\omega_{\text{max}}} \omega\Gamma_{\text{SP}}(\omega)d\omega \quad (5)$$

From Eq. (5) we can easily see that the electric-field strength can be evaluated by simply taking the integral of  $\omega\Gamma_{\text{SP}}$  with respect to the angular frequency. There are two hard limits to the integral, the lower limit,  $\omega_{\text{min}}$  being the crossing point of  $\omega = kv$  with the plasmon dispersion [21], [26], which can be

obtained from the relation

$$\sigma_{\text{imag}} = v\varepsilon_0\sqrt{2} \quad (6a)$$

where  $\sigma_{\text{imag}}$  is the imaginary conductivity component; and the upper limit,  $\omega_{\text{max}}$  being the surface plasmon resonance (SPR) frequency, given by [26]

$$\omega_{\text{SPR}} \approx \frac{5E_F}{3\hbar} \quad (6b)$$

Finally, we obtain the conductivity modulation  $\Delta\sigma$  through the expression

$$\Delta\sigma_{\text{Re(Im)}} = \frac{3}{4}g\sigma_{\text{Re(Im)}}^{(3)} \frac{|E_L|^2}{1 + |E_L|^2/|E_{\text{th,Re(Im)}}|^2} \quad (7)$$

where  $\sigma^{(3)}$  is the graphene's Kerr nonlinear conductivity which was theoretically derived from our previous papers [9], [10], [16],  $g \approx 4n_{\text{waveguide}}^4$  is the surface-induced nonlinear enhancement factor [12], [16], and  $E_{\text{th}}$  is the threshold saturation field to reduce the optical conductivity by half.

## III. RESULTS

Let us examine the case of a suspended graphene waveguide with a Fermi-level of 0.2 eV. A single electron with energy of 25.6 eV is travelling at a constant speed of  $v = 0.01c$ , where  $c$  is the speed of light in vacuum, at an elevation  $z = 10$  nm above the graphene sheet. In Fig. 2(a) we plot the graphene plasmon (GP) dispersion curve (dash-dotted red line), as well as the 0.01c velocity curve (green dashed line). The moving electron excites the GP waves with a frequency-dependent loss probability calculated from Eq. (2) and is plotted on Fig. 2(b). From the figure, we integrate the energy losses according to Eq. (5) to get the total electric-field strength, and then finally calculate the modulated GP dispersion curve (blue solid line) shown in Fig. 2(a). It can be seen that the modulated GP dispersion curve is bent further below the light line, indicating that the waveguide index and confinement have increased substantially.

Simple Figure-of-Merits (FoM) can be used to describe the phase and loss modulation [16]. The FoM for the phase modulation is defined as

$$\Delta\phi = \frac{\omega}{c} \cdot \Delta n_{\text{waveguide}} \cdot L'_{\text{waveguide}} \quad (8a)$$

where  $\Delta n_{\text{waveguide}}$  is the change of the effective waveguide index, and  $L'_{\text{waveguide}}$  is the effective waveguide length after modulation. While the FoM for the loss modulation is defined as

$$\frac{\Delta\alpha}{\alpha} = \frac{\text{Im}(n'_{\text{waveguide}}) - \text{Im}(n_{\text{waveguide}})}{\text{Im}(n_{\text{waveguide}})} \quad (8a)$$

Both FoMs are plotted in Fig. 2(c) and (d) for the 10–30 THz spectrum. Phase modulation reaches extreme high values near to the SPR frequency (64 THz) as shown in the inset of Fig. 2(c) due to the large change in GP dispersion, but has lower values at the low THz spectrum. Meanwhile, loss modulation has an overall better performance, where a modulation depth of up to 90% is achieved for 30 THz, as shown by the black solid line of Fig. 2(d). These results are entirely consistent with previously reported ones [16].

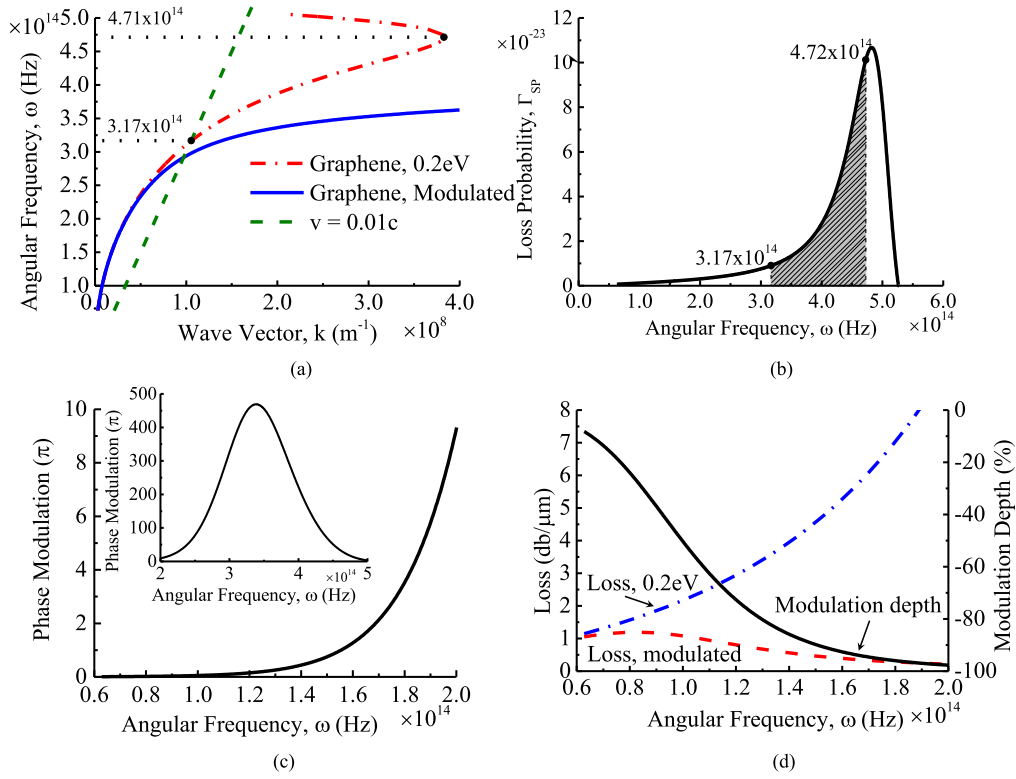


Fig. 2. (a) Graphene plasmon dispersion curves for suspended graphene at  $E_F = 0.2$  eV and modulated by an electron travelling at  $v = 0.01c$  and at  $z = 10$  nm. (b) Integration of the loss probability over frequencies from  $\omega_{\min}$  to  $\omega_{\text{SPR}}$ , represented by the shaded area under the curve. (c) Phase modulation with respect to frequency. (d) Loss modulation and modulation depth with respect to frequency.

We discuss a few ways to modify the modulation strength of the electron beam, which include the Fermi-level of graphene ( $E_F$ ), velocity ( $v$ ) of the moving electron, number of electrons ( $N$ ) in the bunch, and elevation ( $z$ ) of the electron beam. The Fermi-level of graphene bestows the biggest effect, where low Fermi-levels can greatly enhance the modulation strength. In Fig. 3(a) we see that both the phase and loss modulation decrease exponentially with the Fermi-level. Two main contributions come from the high nonlinearities at low Fermi-levels (see Appendix) [9], [10], accompanied by a large waveguide index (Fig. 3(b) right) which augments the surface-induced nonlinear enhancement, as discussed in Eq. (7) [12], [16]. A third, lesser effect comes from the slight increase in the coupling of plasmon electric-fields (Fig. 3(b) left).

Next, we look at the effect of the number of electrons in the bunch and elevation of the electron beam, under the conditions shown in Fig. 4. The initial conditions for number of electrons and elevation are  $N = 2$  and  $z = 10$  nm respectively. Both phase and loss modulation increase for increasing number of electrons and decreasing elevation height respectively. For phase modulation, the effect is exponential as shown in Fig. 4(a), though the decrease in elevation height effects a larger super-exponential increase, while the increase in number of electrons only produces a sub-exponential increase. This discrepancy is less pronounced in the loss modulation shown in Fig. 4(b), where the modulation depth saturates early for  $N < 8$  electrons and  $z > 6$  nm elevation respectively.

Finally, we study the effect of the electron velocity on the modulation strength. Notwithstanding the large difference in

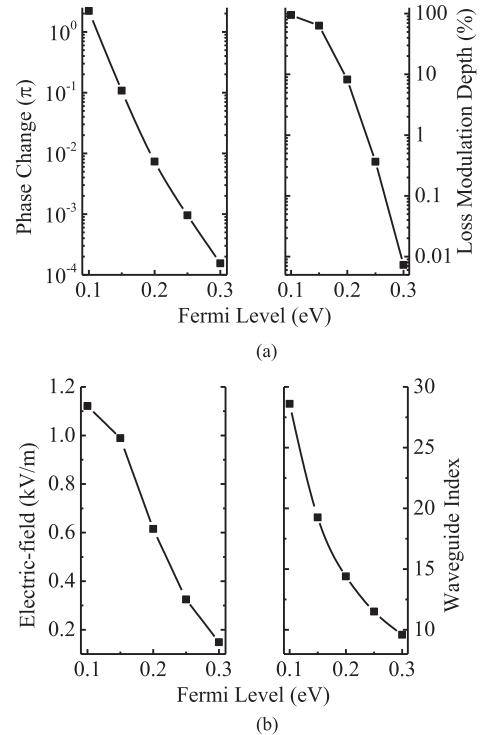


Fig. 3. (a) Phase change and loss modulation depth for an electron travelling at  $v = 0.01c$  and at  $z = 10$  nm, with respect to the Fermi-level of graphene from 0.1–0.3 eV at 10 THz frequency. (b) Corresponding total  $z$ -direction plasmon electric-fields induced on the graphene sheet, and effective index of the graphene plasmonic waveguide.

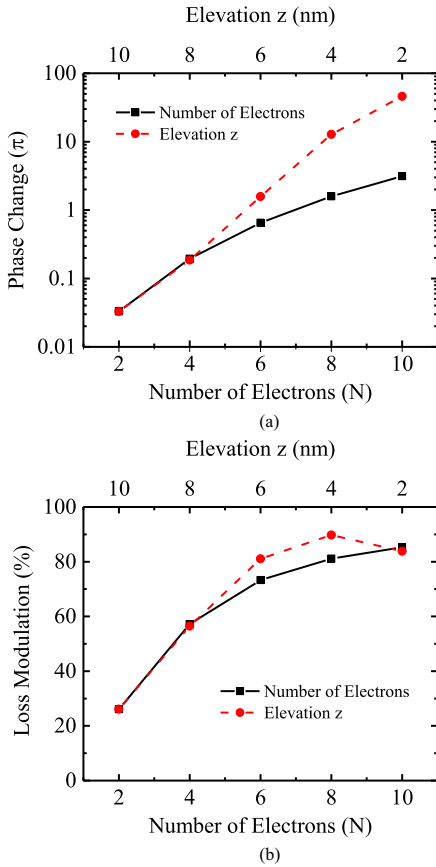


Fig. 4. (a) Phase change with respect to number of electrons and elevation, for electrons travelling at  $v = 0.01c$  above graphene with  $E_F = 0.1$  eV at 10 THz frequency. (b) Corresponding loss modulation depth under the same conditions.

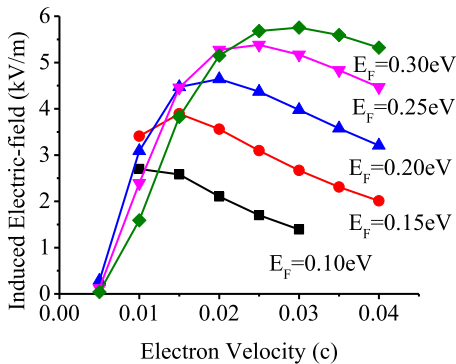


Fig. 5. Induced graphene plasmon electric-field strength as a function of electron velocity for graphene  $E_F = 0.1 - 0.3$  eV,  $N = 1$  and  $z = 10$  nm at 10 THz frequency.

nonlinear coefficients between the various graphene  $E_F$ , here we only look at the total induced plasmon electric-fields as shown in Fig. 5. It is observed that the total generated electric-field strength is generally higher for higher  $E_F$ . This is due to the fact that the SPR frequency scales with  $E_F$ , and hence a wider frequency spectrum is available for the energy transfer between the moving electron and graphene plasmons. Additionally, since the energy transfer also scales inversely with the electron veloc-

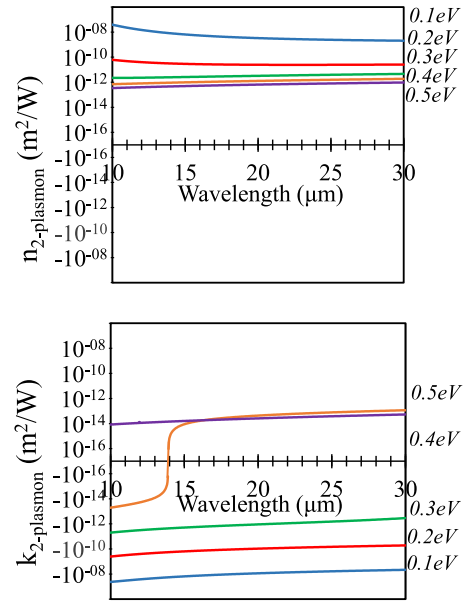


Fig. A1. Nonlinear plasmon refractive indices for different  $E_F$  [16]. © CC-BY 4.0

ity, there is an optimal electron velocity for each graphene  $E_F$  in which the induced plasmon electric-field strength is the highest.

#### IV. CONCLUSION

Putting all observations together, we conclude in general that strong nonlinear modulation through electronic scattering can be achieved when the  $E_F$  is low, electron velocity is two-orders below the speed of light, bunching of just a few electrons, and the elevation of the travelling electrons is within tens of nanometers. To give a ballpark figure, we can obtain a  $2\pi$ -phase change and 94% loss modulation with an electron travelling at  $v = 0.01c$  (25.6 eV) and  $z = 10$  nm above a suspended graphene waveguide with  $E_F = 0.1$  eV. The device performance is extremely efficient considering the fact that current reported all-optical plasmonic modulators require laser fluences in terms of  $\text{mJ cm}^{-2}$  [29], [30], in comparison to the non-relativistic electron beam fluence of only  $4 \mu\text{J cm}^{-2}$  (considering a beam cross-section of  $100 \text{ nm}^2$ ). The nonlinear electronic scattering of graphene plasmons has potential applications in utilizing in-situ optoelectronic generation and control of plasmonic waves without the need of external optics, perhaps heralding and enabling the technology for solid-state vacuum nanoelectronic devices [31].

#### APPENDIX

The steps to get the nonlinear plasmon refractive index of a graphene waveguide has been discussed in detail in Ref. [16], and will be briefly elaborated here.

To get the Kerr coefficients for graphene, the graphene electronic structure is approximated by a two-band tight binding model utilizing only the carbon  $2p_z$  orbital, and then  $\sigma^{(3)}$  (Kerr coefficients) are calculated employing a semiconductor Bloch equation approach [9], [10], taking into account phenomeno-



logical relaxation parameters for both the intraband and the interband transitions.

Now we consider the plasmon refractive index of a graphene plasmonic waveguide

$$n_{\text{plasmon}} = \sqrt{\varepsilon_d - \left( \frac{2\varepsilon_d \varepsilon_0 c}{\sigma(1)} \right)^2} \quad (\text{A.1})$$

Then, we define the modulated plasmon refractive index,

$$n'_{\text{plasmon}} = \sqrt{\varepsilon_d - \left( \frac{2\varepsilon_d \varepsilon_0 c}{\sigma(1) + \Delta\sigma} \right)^2} \quad (\text{A.2})$$

where  $\Delta\sigma = 3\sigma^{(3)}|E|^2/4$ . After that, the nonlinear plasmon refractive indices could be easily defined through

$$n_{2\text{-plasmon}} = \tilde{n}_{\text{plasmon}}/I \quad (\text{A.3})$$

$$k_{2\text{-plasmon}} = \tilde{k}_{\text{plasmon}}/I \quad (\text{A.4})$$

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