Efficiencies of Aloof-Scattered Electron Beam Excitation of Metal and Graphene Plasmons

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Abstract—We assessed the efficiencies of surface plasmon excitation by an aloof-scattered electron beam on metals and graphene. Graphene is shown to exhibit high energy transfer efficiencies at very low electron kinetic energy requirements. We show that the exceptional performance of graphene is due to its unique plasmon dispersion, low electronic density and thin-film structure. The potential applications of these aloof-scattered graphene plasmons are discussed in aspects of coherent radiation.

Index Terms—electron beams, graphene, surface plasmons, slow light

I. INTRODUCTION

The interaction of swift electrons with matter provides pathways for evanescent sources of light [1]. One of these pathways involves the excitation of surface plasmons (SPs), which is usually generated by bombardment of swift electrons onto a thin metal film, as has shown by Ritchie in 1957 [2]. Another method to excite SPs is the aloof-scattering of electrons which was demonstrated by Lecante et al. in 1977 [3], whereby the electron trajectory does not intersect the metal, as schematically shown in Fig. 1. This method involves low-energy transfer and thus minimizes sample damage, in addition to opening up other interesting radiation pathways, such as the plasmon-controlled Cherenkov radiation as has been recently shown by Liu et al. [4].

The aloof-scattering of electrons to excite plasmons on metals is well-studied in the field of electron-loss spectroscopy [1], both in theoretical formulation and experimental observation. In this paper, however, we want to study the aloof-scattering of electrons on graphene, which is still a relatively new area of research with potentially exciting new physics [5].

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II. CONDITIONS FOR ALOOF-SCATTERING EXCITATION OF SURFACE PLASMONS

An aloof-scattered electron can favourably-couple to surface plasmon polaritons (SPPs) of propagation constant given by $k_{\text{sp}}=\omega/v$, where $\omega$ is the radian frequency and $v$ is the velocity of the electron. As such, the velocity and hence kinetic energy of the electron required to excite SPPs are greatly dictated by the plasmon dispersion of the metal. The plasmon dispersion relation for odd SPP modes on a metal of finite thickness is given by the following equation [6]

$$\coth\left(\frac{k_{\text{sp}}^2 - \omega^2 \mu_0 \varepsilon_0 \varepsilon_m}{2\varepsilon_d}\right) = -\frac{\varepsilon_m}{\varepsilon_d} \sqrt{\frac{k_{sp}^2 - \omega^2 \mu_0 \varepsilon_0 \varepsilon_d}{k_{sp}^2 - \omega^2 \mu_0 \varepsilon_0 \varepsilon_m}}$$

(1)

where $\varepsilon_m = \varepsilon_{\infty} - \omega_p^2 / \left(\omega (\omega + i\gamma)\right)$ is the metal permittivity, $\omega_p$ is the plasma frequency, $\varepsilon_{\infty}$ is the high-frequency dielectric constant of the metal, and $\gamma$ is the relaxation frequency. $\varepsilon_d$ is the dielectric background permittivity, $\varepsilon_0$ is the free space permittivity, $\mu_0$ is the free space permeability, and $\Delta$ is the metal thickness.

A. SPP Excitation on Bulk Metals

For bulk metals, we can approximate the system to be an infinite half-space by taking $\lim(\Delta \to \infty)$ in (1). This brings us to the result of the commonly seen dispersion relation

$$k_{sp} = \frac{\omega}{c} \sqrt{\frac{\varepsilon_d \varepsilon_m}{\varepsilon_d + \varepsilon_m}}$$

(2)

where the real part of $k_{sp}$ can be written as
The plasmon dispersion curve saturates for large $k_{sp}$ at the SP resonance (SPR) frequency when the denominator $\left[1 - \chi^2 \left(\varepsilon_d + \varepsilon_x\right)^2\right]$ goes to zero, giving the relation

$$\omega_p = \sqrt{\frac{\omega_p^0}{\varepsilon_d + \varepsilon_x}}$$

At $\varepsilon_d$ and $\varepsilon_x = 1$, we have $\omega_p = \omega_p^0 \sqrt{2}$, which was the result first observed experimentally by Ritchie [2]. The SPR frequency hence scales with the square-root of the 3-D electronic density, $\omega_p \propto \sqrt{N_{3D}}$.

The excitation of SPPs by aloof-scattering of electrons happens favourably at the frequency where the velocity line intersects the plasmon dispersion line, as shown in Fig. 2. Faster electrons can excite SPPs of lower $k_{sp}$ at the expense of higher electron kinetic energy. While it is possible to excite SPPs of higher $k_{sp}$ using slower electrons, these SPPs would gradually lose their polaritonic nature and become electrostatic SPs.

B. SPP Excitation on Thin Metal Films and Graphene

Here we want to make the case for doped graphene as an efficient material to excite SPPs by aloof-scattering of electrons. Graphene is an efficient material for two important reasons: firstly, it is a 2-D thin film of vanishing thickness; secondly, graphene’s SPR frequency scales with the square-root of the 2-D electronic density as $\omega_p \propto \sqrt{N_{2D}}$, where the N$_{2D}$ is usually in the order of $10^{16}$ m$^{-2}$, which is significantly lower than the N$_{3D}$ for metals [7].

A 2-D thin film of vanishing thickness will have an increased $k_{sp}$. Hypothetically, if the metal films can be made extremely thin (e.g. 1nm thickness), we can take lim(Δ→0) in (1) and thus the dispersion relation becomes

$$k_{sp} = \sqrt{\frac{2\varepsilon_d}{\omega_m\Delta}} + \varepsilon_d \left(\frac{\omega}{c}\right)^2$$

and

$$\text{Re}(k_{sp}) \approx \frac{2\omega^2 \varepsilon_d}{\omega_p^0 \left[1 - \chi^2 \varepsilon_x^2\right]}$$

As $k_{sp}$ scales inversely with $\Delta$, we can achieve very high $k_{sp}$ for low $\omega$ and $\nu$, as illustrated in Fig. 3.

We further examine the case for graphene, where the plasmon dispersion is given in terms of the 2-D conductivity

$$k_{sp} = \frac{\omega}{c} \sqrt{\varepsilon_d - \left(\frac{2\varepsilon_d \varepsilon_0 c}{\sigma}\right)^2}$$

The optical conductivity, $\sigma$ is obtained from both the semi-classical model and random-phase approximation, given by

$$\sigma(\omega) = \frac{ie^2 E_F}{\pi \hbar^2 (\omega + i\gamma)} + \frac{ie^2}{4\pi \hbar} \ln \left(\frac{2E_F}{\omega + (\omega + i\gamma)\hbar}\right)$$

where the first term on the right represents the intraband conductivity and the second term represents the interband conductivity [8]. The constant $e$ is the electronic charge, $\hbar$ is the reduced Planck’s constant,
where $L$ is the length of energy transfer, $z$ is the impact parameter, $\gamma_{LF}$ is the Lorentz contraction factor, and $K_0(x)$ is the zeroth-order modified Bessel function of the second kind. It is immediately observed that $\Gamma_{SP}$ scales inversely with $v^2$, thus the propensity for slower electrons to transfer energy is greater than that for faster electrons. Meanwhile, $K_0(x) \propto - \ln(x)$ for small $x$ and $K_0(x) \propto \exp(-x)$ for large $x$ [12], and thus energy transfer improves either when electrons travel closer to the surface, travel with a non-relativistic velocity, or generate lower $k_{sp}$ SPPs.

Finally, the last term in (12) is the energy loss function, which represents the propensity of the material to accept the energy transfer, and is given in terms of the $p$-polarized Fresnel reflection coefficient $r_p$. In general, the energy loss function for thin metal films and graphene would be very high when compared to bulk metals, because the optical fields could be excited on both sides of the film surface.

A. Energy Loss Function of Bulk Metals

The energy loss function of bulk metals could be derived as

$$\text{Im} \left( \frac{r_p}{\varepsilon_0 \varepsilon^d} \right) = \text{Im} \left( \frac{2}{\varepsilon_0 \varepsilon^d + \varepsilon^m} \right) \approx \frac{\omega \gamma}{\varepsilon_0 \varepsilon^d} \left[ 1 - x^2 (\varepsilon^d + \varepsilon^m) \right]$$

(13)

The energy loss function scales proportionally to $\omega$ and $\gamma$ and inversely proportional to $\omega_p$.

B. Energy Loss Function of Thin Metal Films

For thin metal films, $r_p$ is modified to the expression

$$r_p = \frac{\varepsilon_m - \varepsilon_d}{\varepsilon_d + \varepsilon_m} \left[ 1 - \exp(-2k_{sp} \Delta) \right]$$

$$\approx \frac{2k_{sp} \Delta}{\exp(2k_{sp} \Delta) - 2k_{sp} \Delta - 1}$$

(14)

For $k_{sp} \cdot \Delta << 1$, $r_p$ is a very large number. Thus, the energy loss function for thin film metals will be very large below the SPR frequency. Near the SPR frequency, however, (14) will be reduced to $r_p$ for bulk metals.

C. Energy Loss Function of Graphene

We can write $r_p$ for graphene as [5]
where $\varepsilon_{d1}$ is the incident background permittivity and $\varepsilon_{d2}$ is the substrate permittivity. Below the SPR limit, the energy loss-function for graphene is derived to be

$$F_d = \frac{n \hbar c}{E_F} \left( \frac{1}{\sqrt{1 - (\gamma/c)^2}} - 1 \right)$$

which scales proportionally to $\omega$ and $\gamma$ and inversely to $E_F^2$ like the case for bulk metals.

### D. Energy Transfer Performance Comparison

In Fig. 5 we show some relevant parameters for the performance comparison of the energy transfer efficiency. The materials used for the comparison are aluminium and silver (material parameters taken from ref. [13,14]), and graphene ($\gamma$ assumed to be $2 \times 10^{12}$Hz). In Fig. 5(a) and 5(b), the energy loss-function and loss probability are plotted using the expression given in (12). The energy transfer length in the calculations are normalized to 1m. In Fig. 5(c), the electron kinetic energy at the condition for favorable coupling is obtained using the usual relativistic kinetic energy equation

$$KE = m_e c^2 \left( \frac{1}{\sqrt{1 - (\gamma/c)^2}} - 1 \right)$$

Finally, in Fig. 5(d), the energy transfer efficiency is obtained from the ratio of the product of loss probability with unit plasmon energy, $\hbar \omega$, over the electron kinetic energy.

Several features stood out in the figures. Graphene’s energy loss-function is exceptionally high owing to its low $N_{2D}$ values, and grows even larger near the SPR frequency. However, the graphene’s loss probability near the SPR frequency is limited by the the $K_0(x)$ term in (12) due to large $k_{sp}$ values. Meanwhile, thin metal films have overall better energy transfer performance compared to their bulk counterparts, as has been predicted from (14), except that the performance deteriorates near the SPR frequency when the large $k_{sp}$ values becomes the limiting factor, similar to graphene. Overall, from Fig. 5(d), the energy transfer efficiency for graphene is on average 4-orders higher than thin metal films, and 10-orders higher than bulk metals.

In Fig. 6 we looked at some electric-field maps of the PIC-simulated electron-excited plasmons using VORPAL [15]. To facilitate comparison, we selected 3 material systems with corresponding electron speeds that excite a characteristic $k_{sp}$ of $\sim 40 \mu$m$^{-1}$: (a) bulk silver and electron beam speed of 0.45c, (b) 1nm silver film and electron beam speed of 0.155c, and (c) 0.2eV doped graphene and electron beam speed of 0.0178c. The electric-field maps capture the moment when the electron has traversed 400–500nm along the material surface. It is
observed that even with the vastly-varied electron beam speeds (and hence the electron kinetic energies), the excited plasmon electric-fields are in the same order of magnitude, which clearly corroborates with the high excitation energy efficiency of graphene plasmons compared to silver plasmons.

IV. POTENTIAL APPLICATIONS OF ALOOF-SCATTERED GRAPHENE PLASMONS

Graphene’s high energy transfer efficiency could be used for applications in generating coherent radiation. For example, one of the possible applications is the plasmon-controlled Cherenkov radiation as mentioned previously. Liu et al. have shown that the radiation spectrum of Cherenkov radiation, which is usually broadband, could be made frequency-selective by first exciting the metal plasmon modes, and the radiation frequency can be tuned over a narrow spectral range in the visible and UV regime by tuning the electron beam velocity [4]. We took the configuration of Liu et al. and replaced the metal layer with graphene. Fig. 7 shows that for a 100keV electron beam, we could potentially excite Cherenkov radiation modes in the 1–2 THz frequency spectrum. Thus, graphene could potentially be used for efficient THz radiation sources. Moreover, the graphene Cherenkov radiation spectrum has increased tunability: we can tune both the electron beam velocity and also the Fermi level of graphene through an electrostatic-gating.

In cases where low energy electron beams are preferred and Cherenkov-type of radiation is not possible, it is still possible to induce coherent radiation from gratings [16]. Liu et al. has recently shown that the coherent radiation generated from graphene-on-gratings are 400 times stronger than conventional dielectric materials.

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