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# Interplay of nonlocal response, damping, and low group velocity in surface-plasmon polaritons

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## ABSTRACT

The miniaturization of metal structures down to the nanoscale has been accompanied with several recent studies demonstrating plasmonic effects not explainable by classical electromagnetic theory. Describing the optical properties of materials solely through the bulk dielectric function has been augmented with quantum mechanical corrections, such as the electron spill-out effect and nonlocal response. Here, we discuss the latter and its implications on the waveguiding characteristics, such as dispersion and group velocity, of the surface-plasmon polariton mode supported at a metal-air interface.

**Keywords:** Plasmonics, nonlocal response, waveguides.

## 1. INTRODUCTION

Recent advances in nanofabrication<sup>1,2</sup> have opened up new avenues for the experimental exploration of plasmonic effects, which cannot be explained by classical electromagnetic theory. In particular, very small metal particles on the order of few nanometers support localized surface plasmons, which show size-dependent resonance shifts and linewidth broadening that are not reproduced using classical simulations.<sup>3-9</sup> In addition, the bonding plasmon modes of closely-spaced dimers are significantly altered from classical predictions when the gap between the particles decreases down to the sub-nanometer range.<sup>10-17</sup>

Simultaneous with the experimental efforts, significant theoretical progress has taken place to provide an understanding of the observed effects by accounting for nonlocal response<sup>18-24</sup> and the electron spill-out effect<sup>25-28</sup> in metals. In this regard, the simple and semi-classical generalized nonlocal optical response (GNOR)<sup>22,23</sup> model has been successful in unifying the description of both small individual nanoparticles and dimers with gaps down to approximately 0.5 nm. In this paper, we study the implications of nonlocal response on waveguiding.<sup>29,30</sup> Specifically, we employ the GNOR model to study the dispersion relation and group velocity of the surface-plasmon polariton (SPP) supported at a metal-air interface. We compare the GNOR results with the nonlocal hydrodynamic model (HDM)<sup>18</sup> and the common local-response approximation (LRA).

## 2. NONLOCAL RESPONSE

Before we discuss the details of the SPP supported at a metal-air interface, we first introduce the governing electromagnetic equations that describe the optical properties of metals. Our starting point is Maxwell's wave equation for non-magnetic metals in the frequency domain

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}, \omega) = \omega^2 \mu_0 \mathbf{D}(\mathbf{r}, \omega) + i\omega \mu_0 \mathbf{J}(\mathbf{r}, \omega), \quad (1)$$

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where  $\mathbf{E}$ ,  $\mathbf{D}$ , and  $\mathbf{J}$  denote the electric field, the displacement field, and the current density, respectively. The displacement field describes the response of the bound charges to external electric fields, while the current density describes the response of the free charges. As we will see, this distinction is not necessary in the LRA as one can combine both responses into a frequency-dependent permittivity  $\varepsilon(\omega)$  of the metal. However, for the cases of nonlocal response theories, the distinction is necessary as only the freely-moving electrons are responsible for nonlocality. Considering first the LRA, we connect the displacement field and current density field to the electric field in the usual manner

$$\mathbf{D}(\mathbf{r}, \omega) = \varepsilon_0 \varepsilon_{\text{core}}(\omega) \mathbf{E}(\mathbf{r}, \omega), \quad (2)$$

$$\mathbf{J}(\mathbf{r}, \omega) = \sigma(\omega) \mathbf{E}(\mathbf{r}, \omega), \quad (3)$$

where  $\varepsilon_{\text{core}}(\omega)$  is the frequency-dependent permittivity due to the bound charges and  $\sigma(\omega)$  is the conductivity of the free electrons. Inserting the constitutive relations in Eqs. (2) and (3) into the wave equation in Eq. (1) allows to rewrite the right-hand side as

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}, \omega) = \left(\frac{\omega}{c}\right)^2 \varepsilon(\omega) \mathbf{E}(\mathbf{r}, \omega), \quad (4)$$

where we have defined

$$\varepsilon(\omega) = \varepsilon_{\text{core}}(\omega) + i \frac{\sigma(\omega)}{\omega \varepsilon_0}, \quad (5)$$

which describes the response of both bound and free charges. We stress that this approach is only applicable for the LRA. Assuming a Drude model for the conductivity, we then arrive at the Drude dielectric function

$$\varepsilon(\omega) = \varepsilon_{\text{core}}(\omega) - \frac{\omega_p^2}{\omega^2 + i\gamma\omega}, \quad (6)$$

which we will use to describe the response of the metal in the LRA.

For the nonlocal response models based on the hydrodynamic approach,<sup>18,31,32</sup> the constitutive relation for the free electrons [i.e., Eq. (3)] turns out to be a differential equation which can be written as<sup>20,21,23</sup>

$$\frac{\eta^2}{\omega^2 + i\gamma\omega} \nabla [\nabla \cdot \mathbf{J}(\mathbf{r}, \omega)] + \mathbf{J}(\mathbf{r}, \omega) = \sigma \mathbf{E}(\mathbf{r}, \omega). \quad (7)$$

Here, we emphasize the presence of the nonlocal velocity parameter  $\eta$ , which depends on the choice of nonlocal model. We also see that for  $\eta \rightarrow 0$ , we retrieve the LRA relation in Eq. (3). For the HDM, where nonlocality arises due to the inclusion of the Thomas–Fermi energy in the description of the free electrons, the nonlocal velocity is given as

$$\eta_{\text{HDM}} = \sqrt{3/5} v_F, \quad (8)$$

where  $v_F$  is the Fermi velocity. In the GNOR model, where the diffusive currents of the free electrons are also accounted for,<sup>22</sup> the nonlocal velocity is

$$\eta_{\text{GNOR}} = \sqrt{(3/5)v_F^2 + D(\gamma - i\omega)}, \quad (9)$$

with  $D$  denoting the diffusion constant. Hence, accounting for the nonlocal response of the free electrons relies on solving the coupled differential equations given by Eqs. (1) and (7), regardless of choice of specific model. Additionally, we point out that  $\eta$  is complex-valued in the GNOR model, while completely real-valued in HDM. This distinction will become important when discussing the SPP of the metal-air interface. In the following, we present the SPP solutions to the coupled equations for the simple case of a metal-air interface.

### 3. METAL-AIR SPP

The SPP supported at a metal-air interface constitutes an advantageous framework for studying the implications of nonlocal response in plasmonics, since only material parameters, without any size parameters, come into play. The dispersion relation governing the SPP at a metal-air interface accounting for both nonlocal response and retardation has been solved in detail previously,<sup>33,34</sup> so we simply state the result here

$$1 = -\frac{\varepsilon\kappa_d}{\kappa_m} - \frac{k^2}{\kappa_{nl}\kappa_m} \frac{\varepsilon - \varepsilon_{core}}{\varepsilon_{core}}, \quad (10)$$

where  $k$  is the SPP propagation constant,  $\kappa_d = \sqrt{k^2 - (\omega/c)^2}$ ,  $\kappa_m = \sqrt{k^2 - (\omega/c)^2\varepsilon}$ ,  $\kappa_{nl} = \sqrt{k^2 - k_{nl}^2}$ , and  $k_{nl} = \sqrt{\omega^2 + i\gamma\omega - \omega_p^2/\varepsilon_{core}/\eta}$ . For large values of  $k$  the simpler nonretarded approximation ( $c \rightarrow \infty$ ) becomes accurate, where the dispersion relation is given by

$$1 = -\varepsilon - \frac{k}{\kappa_{nl}} \frac{\varepsilon - \varepsilon_{core}}{\varepsilon_{core}}. \quad (11)$$

In the following, we numerically solve the SPP dispersion relations for three different cases: (i) a lossless Drude metal, (ii) a lossy Drude metal, and (iii) for gold with experimental bulk parameters taken from literature.<sup>35</sup> We compare the results of the GNOR model with the HDM and LRA. In all cases we use the following free-electron parameters appropriate for gold:<sup>36</sup>  $\hbar\omega_p = 9.02$  eV,  $\hbar\gamma = 0.071$  eV,  $v_F = 1.39 \times 10^6$  m/s, and  $D = 1.90 \times 10^{-4}$  m<sup>2</sup>/s.

#### 3.1 Lossless Drude

The first case is for a lossless Drude metal, where the dielectric function is simply  $\varepsilon = 1 - (\omega_p/\omega)^2$ . Hence, from a classical perspective, there is no dissipation of energy in the material. The SPP dispersion relations for the GNOR, HDM, and LRA approaches are displayed in Fig. 1(a-b). In the LRA, we notice the well-known large- $k$  limit at frequency  $\omega_{sp} = \omega_p/\sqrt{2}$  in the real part of the effective index  $n_{eff} = k/(\omega/c)$ , while the imaginary part vanishes due to the absence of material loss. For the HDM, we see that the real part of the effective index does not saturate at the classical limit  $\omega_{sp}$  but continues to increase for larger frequencies.<sup>37</sup> The rate of increase in the real part of the effective index is determined by the magnitude of the real part of the nonlocal velocity  $\text{Re}(\eta_{HDM})$ . Again no propagation loss is present due to the lack of loss in the material. For the GNOR model, the real part of the effective index behaves as in the HDM, albeit with a faster increase due to  $\text{Re}(\eta_{GNOR}) > \text{Re}(\eta_{HDM})$ . However, a profound change is seen in the imaginary part of the effective index, which is now finite and increases with frequency. Thus, even in the absence of material loss, the SPP experiences propagation losses due to nonlocal response. Physically, this interesting effect occurs since the GNOR model mimics Landau damping, which is present even in a collisionless plasma. Mathematically, the finite propagation loss is a direct consequence of the nonlocal velocity  $\eta$  being complex-valued [Eq. (9)]. Finally, the nonretarded approximations are displayed in dashed lines, and we see, as anticipated, that the agreement is excellent in the large- $k$  limit.

Besides the dispersion relation, another important physical parameter describing the SPP is the group velocity given as

$$v_g = \frac{1}{\frac{\omega}{c} \frac{\partial \text{Re}(n_{eff})}{\partial \omega}} \quad (12)$$

which is displayed in Fig. 1(c). Here, we notice the striking difference between the nonlocal models and the LRA. In the LRA, the group velocity vanishes when the frequency approaches  $\omega_{sp}$ , while the nonlocal models show a finite group velocity at all frequencies. The HDM displays a lower limit to the group velocity, which is captured by the nonretarded approximation (dashed lines). Hence, by utilizing the simpler nonretarded dispersion relation, we can analytically calculate the group velocity in the HDM. For a lossless Drude metal, we find

$$v_g^{\text{HDM}} = \frac{\omega^2}{\omega^2 + \omega_p^2/2} \sqrt{3/5} v_F. \quad (13)$$

We note that for frequencies significantly above the surface plasmon frequency  $\omega_p/\sqrt{2}$ , the group velocity of the SPP approaches the nonlocal velocity in the HDM  $\sqrt{3/5}v_F$ .

### 3.2 Lossy Drude

We now weakly increase the material losses by introducing a small but finite Drude damping rate  $\gamma$  into the dielectric function. The SPP dispersion relation for this case is shown in Fig. 2(a-b). Considering first the result in the LRA, we observe a dramatic change compared to the lossless case. In particular, the SPP propagation constant stays finite at all frequencies and significant propagation loss occurs close to the surface plasmon frequency  $\omega_{\text{sp}}$ . The large change from lossless to slightly lossy is a direct consequence of the need to regularize the nonphysical situation of a vanishing SPP group velocity from the lossless case. Indeed, a vanishing group velocity in the presence of even minute material losses would lead to infinite propagation loss,<sup>38</sup> which would not be physically meaningful. Instead, the SPP dispersion relation in the LRA dramatically bends back to finite values of effective index. In both the HDM and GNOR models, the change in the real part of the effective index is less dramatic due to the finite group velocity. We do however observe an increase in the imaginary part of the effective index for the HDM as a consequence of the weak material losses. In contrast, the GNOR model is not altered by the presence of weak material losses, since the nonlocal loss channel (i.e., Landau damping) dominates.

### 3.3 Gold

As the final case, we study a realistic plasmonic system consisting of a gold-air interface. The bulk dielectric function for gold  $\epsilon_{\text{exp}}(\omega)$  is taken from literature.<sup>35</sup> To calculate the contribution from the bound charges, we use the recipe  $\epsilon_{\text{core}}(\omega) = \epsilon_{\text{exp}}(\omega) + \omega_{\text{p}}^2/(\omega^2 + i\gamma\omega)$ .<sup>19</sup> The SPP dispersion relation is shown in Fig. 3(a-b). Here, we notice no observable difference between LRA and the nonlocal response models as a consequence of the significant material losses in gold, which dominate the nonlocal effects. Some possibilities to increase the effects of nonlocal response are to consider thin ( $< 10$  nm) metal films or small gaps between metal layers, but these systems are beyond the scope of this work.

## 4. CONCLUSIONS

We have numerically investigated the surface-plasmon polariton mode supported at a metal-air interface within the frameworks of the local-response approximation and two nonlocal response models; the hydrodynamic model and the generalized nonlocal optical response model. For the cases of none-to-weak material losses, we find significant differences between the nonlocal theories and the local-response approximation. In particular, we notice in the lossless case that the generalized nonlocal optical response model gives rise to a finite propagation length as a consequence of Landau damping; a damping mechanism which is present even in a collisionless plasma.

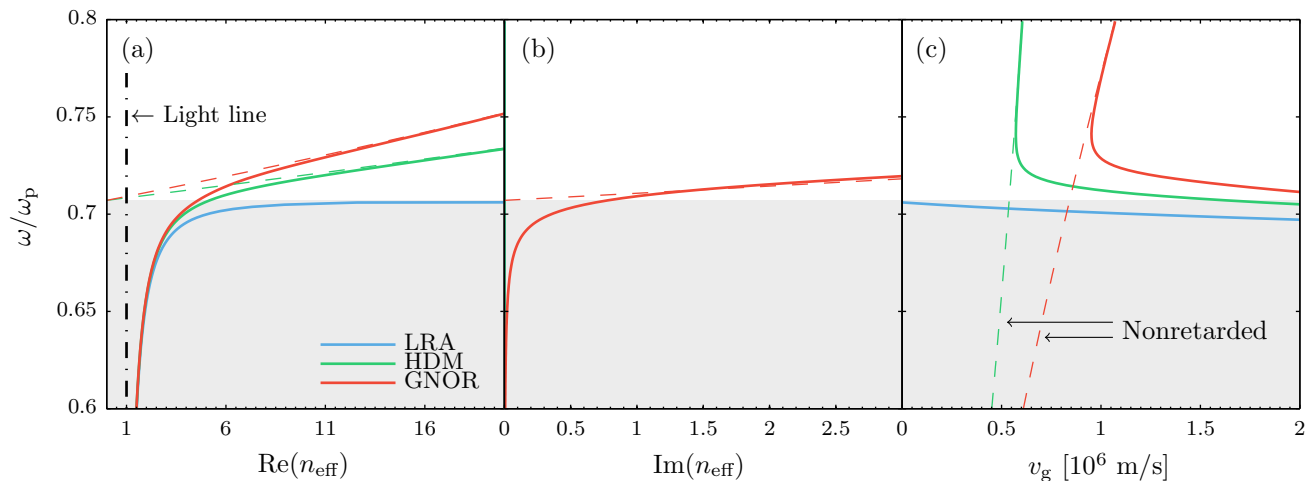


Figure 1. (a-b) Effective index  $n_{\text{eff}}$  of the surface plasmon polariton supported at a metal-air interface for a lossless Drude metal. The dashed lines show the nonretarded solutions. (c) Group velocity  $v_g$  of the SPP.

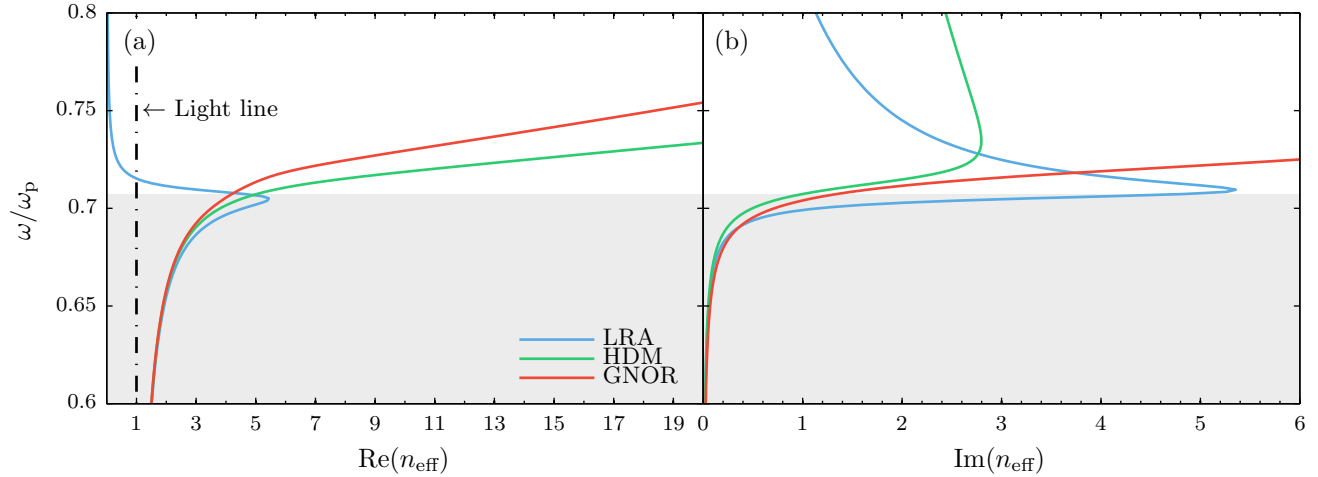


Figure 2. (a-b) Effective index  $n_{\text{eff}}$  of the surface plasmon polariton supported at a metal-air interface for a lossy Drude metal, i.e., with a finite Drude damping rate  $\gamma$ .

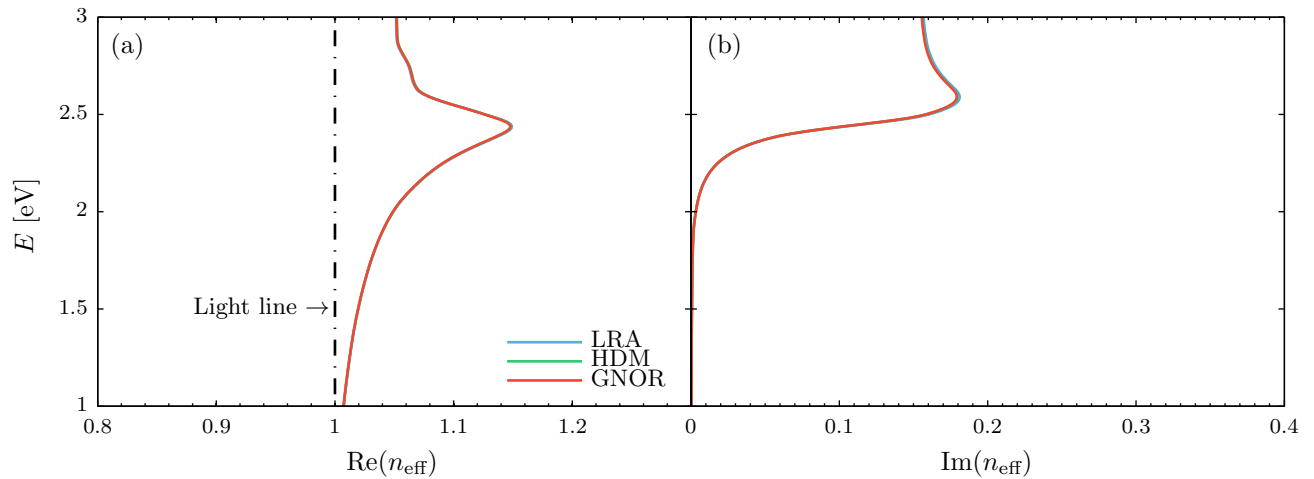


Figure 3. (a-b) Effective index  $n_{\text{eff}}$  of the surface plasmon polariton supported at a gold-air interface. Experimental bulk parameters for gold taken from literature.<sup>35</sup>

However, when taking into account the realistic material parameters of gold, we find that the effects of nonlocal response are masked due to too large material losses.

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