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Nonlocal formalism for nanoplasmonics: Phenomenological and semi-classical considerations

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Abstract

The plasmon response of metallic nanostructures is anticipated to exhibit nonlocal dynamics of the electron gas when exploring the true nanoscale. We extend the local-response approximation (based on Ohm's law) to account for a general short-range nonlocal response of the homogeneous electron gas. Without specifying further details of the underlying physical mechanism we show how this leads to a Laplacian correction term in the electromagnetic wave equation. Within the hydrodynamic model we demonstrate this explicitly and we identify the characteristic nonlocal range to be $\xi_{NL} \sim v_F/\omega$ where v_F is the Fermi velocity and ω is the optical angular frequency. For noble metals this gives significant corrections when characteristic device dimensions approach ~1–10 nm, whereas at more macroscopic length scales plasmonic phenomena are well accounted for by the local Drude response. © 2013 Elsevier B.V. All rights reserved.

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1. Introduction

The interaction of light with the free electrons in noble metals has led to a range of novel plasmonic phenomena and a versatile platform for a variety of new applications [1–5]. In particular, nanofabrication technologies and chemical synthesis are now allowing the plasmonics community to explore and manipulate lightmatter interactions at sub-wavelength length scales, taking advantage of the spatially rapid oscillations of surface-plasmon polaritons and their ability to localize their energy in very small metallic volumes and structures [6].

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The understanding of the optical response of plasmonic structures has been successfully developed within the common framework of the local-response approximation (LRA) with Ohm's law $J(\mathbf{r}) = \sigma(\mathbf{r})\mathbf{E}(\mathbf{r})$ as the constitutive equation [7,8]. However, the ability to fabricate and experimentally explore yet smaller metallic nanostructures has recently stimulated new theoretical developments aiming at quantum phenomena in nanoplasmonic systems [9–11] and the most recent experimental developments [12–16] have clearly made a call for theory developments going beyond the LRA. Spatial dispersion due to nonlocal response is one of the extensions of the LRA formalism which have been studied extensively in more recent years [17–27]. While the commonly employed LRA is inherently a description without any intrinsic length scales [7,8], the new developments naturally introduce fundamental length scales associated with the quantum wave

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dynamics of the electron gas. As a consequence, plasmon polaritons cannot sustain spatial oscillations beyond a cut-off wave number ω/v_F [28,29], where v_F is the Fermi velocity of the electron gas. However, even before reaching this cutoff we anticipate important nonlocal corrections in noble-metal nanostructures with characteristic dimension approaching the 1–10 nanometer regime (see Fig. 1).

For light interaction with arbitrarily shaped plasmonic structures, we begin from general considerations of nonlocal response and treat the case of short-range corrections to the LRA. The main result of this phenomenological analysis is that, irrespectively of the detailed underlying physical mechanism, nonlocal corrections appear in Maxwell's wave equation for the electrical field through an additional Laplacian operator term. Next, we turn to a specific hydrodynamic model and derive this result explicitly. Finally, we briefly address the importance of nonlocal response by dimensional analysis.

2. Nonlocal response formalism

We consider the interaction of light with metallic nanostructures in the linear regime, but with a general and spatially nonlocal response, i.e.

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}) = \left(\frac{\omega}{c}\right)^2 \int d\mathbf{r}' \boldsymbol{\varepsilon}(\mathbf{r}, \mathbf{r}') \mathbf{E}(\mathbf{r}').$$
 (1)

In the following we will focus on the electron plasma itself, for simplicity leaving out any interband effects. For comparison to the commonly employed framework, we note that within the LRA the two-point dielectric function simplifies to $\varepsilon(\mathbf{r}, \mathbf{r}') \approx \varepsilon_D \delta(\mathbf{r} - \mathbf{r}')$ with the usual Drude dielectric function

$$\varepsilon_D = 1 + \frac{i\sigma}{\varepsilon_0 \omega} = 1 - \frac{\omega_p^2}{\omega(\omega + i/\tau)}$$
(2)

where ω_p is the plasma frequency, σ is the Ohmic conductivity, and $1/\tau$ is the damping rate. In this case, the integral in the integro-differential equation (Eq. (1)) is readily performed and we arrive at the ordinary partial-differential equation (PDE) for the local-response dynamics of plasmonic systems.

In the following we will use different approaches to get more insight into the nonlocal response function $\varepsilon(\mathbf{r}, \mathbf{r}')$ associated with the plasmon response of the electron gas in metals.

3. Phenomenological considerations

First, we note that the local approximation with a delta-function response is an overall very good

approximation and modeling based on this is indeed offering very good accounts of the majority of plasmonic phenomena observed in experiments. Thus, in our attempt to account for nonlocal response it seems adequate to only slightly relax the delta-function response. Consequently, we turn to a general nonlocal response function $\varepsilon(\mathbf{r}, \mathbf{r}')$ which is only short-range and with a characteristic nonlocal length $\xi_{\rm NL}$, such as in a Gaussian representation of a delta-function response (see Fig. 2). For convenience, we write the response function as

$$\varepsilon(\mathbf{r},\mathbf{r}') = \varepsilon_D \delta(\mathbf{r} - \mathbf{r}') + f(|\mathbf{r} - \mathbf{r}'|), \qquad (3)$$

i.e. with a local-response Drude contribution and with a small nonlocal correction associated with a homogeneous and isotropic plasma. In accordance with the above discussion we assume that f satisfies

$$\int d\mathbf{r} f(r) \ll |\varepsilon_D|,\tag{4a}$$

$$\int d\mathbf{r} \, rf(r) = 0,\tag{4b}$$

$$\int d\mathbf{r} \, r^2 f(r) = \xi_{\rm NL}^2. \tag{4c}$$

The approach in Eq. (3) is strongly inspired by a recent phenomenological approach by Ginzburg and Zayats [30]. Rather than using a particular f as a smearing function in numerical simulations we here proceed analytically. Due to the short-range behavior of f we may conveniently Taylor expand the slowly varying electrical field in the integrand of Eq. (1) around the point **r**. To second order in $(\mathbf{r'} - \mathbf{r})$ this gives

$$E_{j}(\mathbf{r}') \simeq E_{j}(\mathbf{r}) + \left[\nabla E_{j}(\mathbf{r})\right] \cdot \left[\mathbf{r}' - \mathbf{r}\right] + \left[\mathbf{r}' - \mathbf{r}\right]^{T} \left[\hat{\mathbf{H}} E_{j}(\mathbf{r})\right] \left[\mathbf{r}' - \mathbf{r}\right],$$
(5)

where the Hessian matrix $\hat{\mathbf{H}}$ has elements $\hat{H}_{ji} = \frac{\partial^2}{(\partial_j \partial_i)}$ and j = x, y, z. Next, substituting into Eq. (1) and performing the integral it is clear that by assumption the zero-order term in the expansion contributes negligibly compared to the Drude contribution (Eq. (4a)). For symmetry reasons, the first-order terms and the second-order cross terms vanish identically as they involve the first moment of f (Eq. (4b)). Consequently, the leading correction comes from the diagonal terms of the Hessian which we conveniently write as $(1/2)[\nabla^2 E_j(\mathbf{r})][\mathbf{r}' - \mathbf{r}]^2$, i.e. involving the second moment of f (Eq. (4c)). With these steps we now get

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}) = \left(\frac{\omega}{c}\right)^2 \left[\varepsilon_D + \mathcal{C}_{\mathrm{NL}} \nabla^2\right] \mathbf{E}(\mathbf{r}),$$
 (6)

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