Are there novel resonances in nanoplasmonic structures due to nonlocal response?

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ABSTRACT

In tiny metallic nanostructures, quantum confinement and nonlocal response change the collective plasmonic behaviour with resulting important consequences for e.g. field-enhancement and extinction cross sections. Here we report on nonlocal resonances in the hydrodynamical Drude model for plasmonic nanostructures that have no counterpart in the local-response Drude model. Even though there are no additional resonances in the visible due to nonlocal response, plasmonic field enhancements are affected by nonlocal response. We present both analytical results for simple geometries and our numerical implementation for arbitrary geometries, and address computational issues related to the several length scales involved.

Keywords: nanoplasmonics, nonlocal response, hydrodynamical Drude model, field enhancement, COMSOL

1. INTRODUCTION

Sub-wavelength metal nanostructures are studied for many reasons, for example for their abilities to confine and enhance optical fields,[1] to sense optical properties of surrounding media, to work as antennae or waveguides and to interact with individual quantum light sources.[2,3] Miniaturization is an ongoing process in nanoplasmonics, enabled by a scale invariance of solutions to Maxwell's equations.[1] It is challenging to fabricate ever smaller metal structures and to efficiently excite surface plasmons in them.[4,5] Another type of challenge is to ascertain that nanoplasmonic structures indeed have the designed properties such as sub-wavelength spatial field distributions. Increasingly, electron beams are employed as probes to this end,[6] in particular electron energy loss spectroscopy (EELS)[7–9] of which the energy resolution has greatly improved in recent years, and cathodoluminescence.[10]

More compact structures also have the advantage of less ohmic loss during transport, although the smaller structures have a relatively larger surface whereby loss at metal surfaces becomes increasingly important. Besides loss, another fundamental challenge for nanoplasmonics is nonlocal optical response, and the one we will address here. It is also known as spatial dispersion. In general, the response of a medium to a certain wave is nonlocal, if the medium has more ways to transport energy than via that wave, as will be explained in Sec. 2. Examples are superconductors, doped semiconductors, and metals. Here we focus on the latter case, in particular on light interacting with noble metals such as gold and silver. Strictly speaking, in metals the optical response is always nonlocal, but it is usually safe to neglect this, so not all books on plasmonics need to be rewritten. However, as we will explain and illustrate below, its effects become noticeable both for nanometer-sized metal structures and for larger metal nanostructures with nanometer-sized spatial variations.

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With non-negligible nonlocal response, the concept of the refractive index, even of a spatially inhomogeneous refractive index $n(\mathbf{r},\omega)=\sqrt{\varepsilon(\mathbf{r},\omega)}$, becomes inadequate. In a medium with nonlocal response, the linear optical polarization field $\mathbf{P}(\mathbf{r},\omega)$ at a position $\mathbf{r}$ is not a function of the electric field $\mathbf{E}(\mathbf{r},\omega)$ only at that position, but rather by a weighted spatial integral over $\mathbf{E}(\mathbf{r}',\omega)$, with weights peaking at $\mathbf{r}'=\mathbf{r}$:

$$\mathbf{P}(\mathbf{r},\omega)=\varepsilon_0\int \mathrm{d}^3\mathbf{r}'\,\chi(\mathbf{r},\mathbf{r}',\omega)\mathbf{E}(\mathbf{r}',\omega). \tag{1}$$

The response is smeared out or filtered in the sense that an infinitely localized electric field $\mathbf{E}(\mathbf{r}',\omega)=\mathbf{E}_0\delta^3(\mathbf{r}'-\mathbf{r}_0)$ gives rise to a smeared-out polarization field $\mathbf{P}(\mathbf{r},\omega)=\varepsilon_0\chi(\mathbf{r},\mathbf{r}_0,\omega)\mathbf{E}_0$. In the hydrodynamical Drude model that we will use to describe nonlocal response in metals, the smearing out is caused by charge density waves, a collective movement of the free electrons induced by the light. In the usual local-response limit, $\chi(\mathbf{r},\mathbf{r}',\omega)$ reduces to $\chi(\mathbf{r},\omega)\delta^3(\mathbf{r}-\mathbf{r}')$. Then there is no smearing out, as Eq. (1) shows: in the local-response limit, infinitely localized electric fields give rise to infinitely localized polarization fields.

Finding solutions to Maxwell's equations becomes harder with nonlocal media, but several exact solutions exist for simple geometries such as wires[11,12] and spheres[13] and plane surfaces.[14] These exact solutions are important, both because they give insight in the new physics emerging in the nonlocal-response regime, and because wire and sphere geometries happen to be important in nanoplasmonics, since they can be fabricated and have many applications. We recently studied analytically how optical properties of plasmonic nanowires are influenced by nonlocal response,[12] thereby also identifying the cause of conflicting results in the recent literature. In Sec. 3 we present some further analytical results on nanowires.

For all but the simplest geometries, numerical methods are needed to solve Maxwell's equations. That is the situation for local response and even more so for nonlocal response. While for local response problems standard software packages exist, such Maxwell solvers do not exist if the response becomes nonlocal. We developed a nonlocal-response add-on to COMSOL that can tackle arbitrary geometries, tested it to be quite accurate, and studied plasmonic dimers with it, which are known to give strong locally enhanced field strengths.[15] We present some further numerical results for field enhancement near plasmonic dimers in Sec. 4, and conclude in Sec. 5. But first we introduce the theory in Sec. 2.

2. THEORY: HYDRODYNAMICAL DRUDE MODEL

In the usual local-response Drude model, the free-electron density in a bulk metal is a constant n_0 that is known to influence the plasma frequency $\omega_\mathrm{p}^2=n_0e^2/(\varepsilon_0 m_0)$. By contrast, in tiny metal structures described by the hydrodynamical Drude model, the density varies both in space and time. The theory allows for longitudinal pressure waves in the electron density, analogous to sound waves. The free electrons are described collectively by their density $n(\mathbf{r},t)$ and velocity vector field $\mathbf{v}(\mathbf{r},t)$. In the linearized hydrodynamical Drude model, the density and velocity are expanded around the equilibrium values in the absence of light, and equations of motion are derived for the external-field induced first-order corrections that are assumed to be small. In that linearized regime, the current density becomes $\mathbf{J}(\mathbf{r},t)=-en_0(\mathbf{r})\mathbf{v}(\mathbf{r},t)$. Coupled wave equations can be derived for the electric field and for the current density.[12,16] The Maxwell wave equation for the electric field

$$\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\boldsymbol{E}=\frac{\omega^2}{c^2}\boldsymbol{E}+i\omega\mu_0\boldsymbol{J} \tag{2}$$

is driven by the current density, while in turn the wave equation for the current density

$$\beta^2\boldsymbol{\nabla}\left[\boldsymbol{\nabla}\cdot\boldsymbol{J}\right]+\omega\left(\omega+i\gamma\right)\boldsymbol{J}=i\omega\omega_\mathrm{p}^2\varepsilon_0\boldsymbol{E} \tag{3}$$

is driven by the electric field. The term $\beta^2\boldsymbol{\nabla}\left[\boldsymbol{\nabla}\cdot\boldsymbol{J}\right]$ with β proportional to the Fermi velocity is the novelty here; without it, the equation would reduce to Ohm's law $\mathbf{J}=\sigma(\omega)\mathbf{E}$ where $\sigma(\omega)$ is the Drude conductivity, and the response would be local. The usual Maxwell boundary conditions are not enough to find a unique solution to these coupled equations (2) and (3). Additional boundary conditions are needed. Before choosing the level of description for the equilibrium charge density profile $n_0(\mathbf{r})$ across a metal-dielectric interface, there is not a unique answer to the question how many additional boundary conditions are needed.[17] For simplicity we neglect

quantum tunneling of electrons across the interface, thereby approximating $n_0(\mathbf{r})$ across the interfaces by a step function that vanishes outside the metal and assumes the constant bulk value n_0 inside the metal. After that choice, it immediately follows from charge conservation that the component of $\mathbf{J}$ normal to the interface vanishes at the interface. This already counts as an additional boundary condition, and this one is enough to find unique solutions to the coupled wave equations.[17] The logic behind our neglect of quantum tunneling is that nonlocal response will become important at length scales of 10 nm or less, as we will see, whereas quantum tunneling is important at distances of 1 nm or smaller.[18,19]

In a series of recent papers, for example in Refs. 20, 21 (the latter being an SPIE proceedings by ourselves) and based on older work on excitons,[22] a modification of the hydrodynamic model was used, leading to quite spectacular predictions of novel resonances in the visible due to nonlocal response, where the only modification consists of replacing the gradient-of-the-divergence term $\boldsymbol{\nabla}\left[\boldsymbol{\nabla}\cdot\boldsymbol{J}\right]$ in the wave equation (3) by the Laplacian $\nabla^2\boldsymbol{J}$. We recently found out and showed that this modified model is of limited use, since its predictions in all but the local-response limit are quite different from the hydrodynamical model itself.[12] We only summarize our findings here. The justification to use the modified model could be that the solutions are easier to find and do not differ much from the ones for the hydrodynamical model, or at least to derive that such is the case in some parameter regime. There seems to be a justification in the quasi-static regime, where the curl of the electric field is assumed to vanish. For by taking the curl of Eq. (3), one finds that the curl of $\mathbf{J}$ is proportional to the curl of $\mathbf{E}$. By elementary vector analysis it then follows that in the quasistatic regime we can replace $\boldsymbol{\nabla}\left[\boldsymbol{\nabla}\cdot\boldsymbol{J}\right]$ in the wave equation (3) by the Laplacian term $\nabla^2\boldsymbol{J}$. Why does this only *seem* to be a justification for the modified model, which in Ref. 12 we called the 'curl-free model'? Because Eq. (3) with the Laplacian contains no longer the information that $\mathbf{E}$ and $\mathbf{J}$ were assumed to have vanishing curl, and because for consistency with the quasistatic approximation the double-curl term in the other wave equation (2) should then also be left out. However, this double-curl term is kept in the curl-free model, so that one can say that the curl-free model amounts to taking the quasi-static limit in only one of the two coupled equations of the linearized hydrodynamical model. We have shown that the solutions of the coupled wave equations in the curl-free model consequently are not curl free.[12] In summary, there is no justification for the curl-free model in the quasi-static limit. This qualitative analysis does not tell how much the solutions of the hydrodynamic and the curl-free model differ quantitatively, but we address this issue for a specific geometry in Sec. 3.

However, before doing calculations for a specific geometry, it is useful to eliminate the electric field from the coupled wave equations (2) and (3), thereby obtaining separate equations for the curl and for the divergence of the current density, respectively:

$$\left(\beta^2\nabla^2+\omega^2+i\omega\gamma-\omega_{\mathrm{p}}^2\right)\boldsymbol{\nabla}\cdot\boldsymbol{J}=0, \tag{4}$$

$$\left[c^2\nabla^2+\omega^2\varepsilon(\omega)\right]\boldsymbol{\nabla}\times\boldsymbol{J}=0. \tag{5}$$

Without damping, they become the Boardman equations.[23] Their importance lies in the fact that for arbitrary geometry, Eq. (4) has exponentially decaying solutions of $\boldsymbol{\nabla}\cdot\boldsymbol{J}$ for $\omega<\omega_{\mathrm{p}}$ and propagating solutions with some damping for $\omega>\omega_{\mathrm{p}}$. This shows quite generally that new resonances can occur due to nonlocal response, but only above the plasma frequency, for those frequencies for which the propagating longitudinal waves become resonant standing waves in nanometer-sized confined geometries.

3. ANALYTICAL RESULTS FOR NANOWIRES

We study the extinction cross section of infinitely long nanowires with nonlocal response to normally incident TM-polarized light.[12] We expand the fields inside and outside the metal in terms of Bessel functions, which the reader may not find surprising, but for the hydrodynamic model the important novelty is that inside the metal there is a combination of divergence-free ('transverse') and curl-free ('longitudinal') waves, see also Ref. 11. The fields inside and outside the metal are then glued together so as to satisfy Maxwell's boundary conditions plus additional boundary conditions.

Our main findings are that in local response, there is a well-known extinction maximum at $\omega_{\mathrm{sp}}=\omega_{\mathrm{p}}/\sqrt{2}$, while the hyrodynamical Drude model predicts a blueshift of that resonance. This blueshift is larger for smaller wire radius. Furthermore, for nanowires of radius smaller than 10 nm, new nonlocal resonances become visible, but indeed only above the plasma frequency, in agreement with the general observation in Sec. 2 based on the

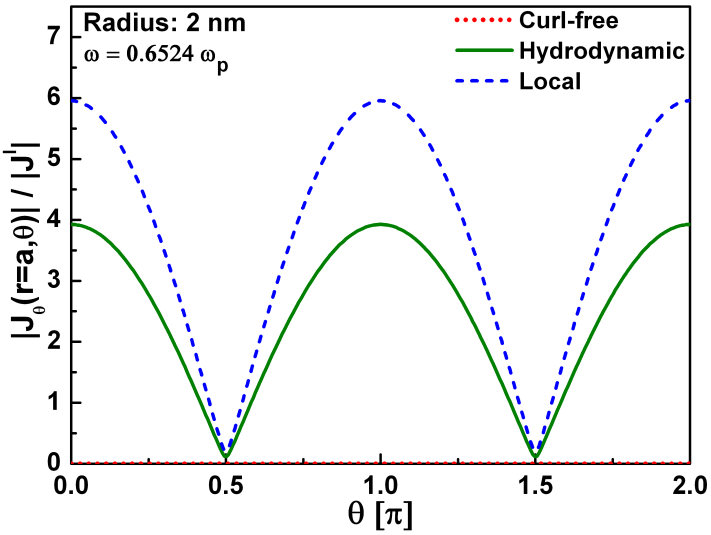

Figure 1. Tangential component of the current density around the $r = a$ boundary of an infinite cylinder with pure plasma response as a function of the cylindrical angle θ. The values are scaled to the incident current density $\mathbf{J}^{\mathrm{I}}$. The wire radius is $a = 2\,\mathrm{nm}$ and frequency of the incident light is $\omega = 0.6524\omega_{\mathrm{p}}$.

generalized Boardman equations (4) and (5). The spacing between these resonances grows as the wire radius decreases. These resonances are interpreted as confined standing-longitudinal-wave resonances. The longitudinal waves are evanescent below the plasma frequency, but propagating above ω_{p}. Whereas in local-response theories external light does not interact with bulk plasmons, the hydrodynamic model does predict resonances of confined longitudinal modes excited by light. On top of those, the 'curl-free model'[20,21] also predicts new resonances due to nonlocal response at visible frequencies, but those are spurious resonances that we showed have no counterpart in the hydrodynamic model itself.[12]

The local-response model, the hydrodynamical Drude model, and the 'curl-free' model all have different associated boundary conditions at metal-dielectric interfaces: respectively, two Maxwell boundary conditions, those two plus a vanishing of the normal component of $\mathbf{J}$, and those three plus a vanishing tangential components of $\mathbf{J}$. Thus in the curl-free model, a unique solution is not found by only specifying the normal component of the current density when assuming stepwise equilibrium free-electron density profiles. Let us now consider the current density on the boundary of the metal surface. Figure 1 shows the tangential component of the current density when going around the illuminated nanowire, where $\theta = 0$ corresponds to the side where the incident plane wave originates from.[24] All current densities are scaled to the 'incident current density', which is computed in terms of the incident electric field $\mathbf{E}^{\mathrm{I}}$ as $\mathbf{J}^{\mathrm{I}} \equiv \sigma(\omega)\mathbf{E}^{\mathrm{I}}$. The figure shows that the tangential current density at the interface for the hydrodynamical Drude model essentially follows the sinusoidal behavior of the local model, but with smaller amplitude, whereas the curl-free model has a forced vanishing tangential component of $\mathbf{J}$.

We also compare the fields inside the nanowires for the three models. In Figure 2 we depict the current components along the cut through the middle of the nanowire in the direction of the incident light.[24] The radial component of $\mathbf{J}$ vanishes identically for all three models: the incident light does not generate currents perpendicular to the direction of its electric field. On the other hand, the tangential components of $\mathbf{J}$ show remarkably different behavior. The local-response value is (almost) constant across the $a = 2\,\mathrm{nm}$ nanowire, as expected for such a sub-wavelength structure. The hydrodynamic tangential component of $\mathbf{J}$ is also quite flat inside the metal, with a value smaller than in the local theory, as we found in Fig. 1. The curve does bend somewhat down near the interfaces, which is a hint of the Fermi wavelength as a new length scale besides the wire radius and the optical wavelength. Quite different is the tangential current in the curl-free model. It vanishes at the interface, dictated by an additional boundary condition, but inside the metal we see a resonant standing wave pattern at the particular frequency chosen. There is no corresponding resonance in the hydrodynamical model. Thus the resonance obtained in the curl-free model is unphysical, and Fig. 2 illustrates its limited use.

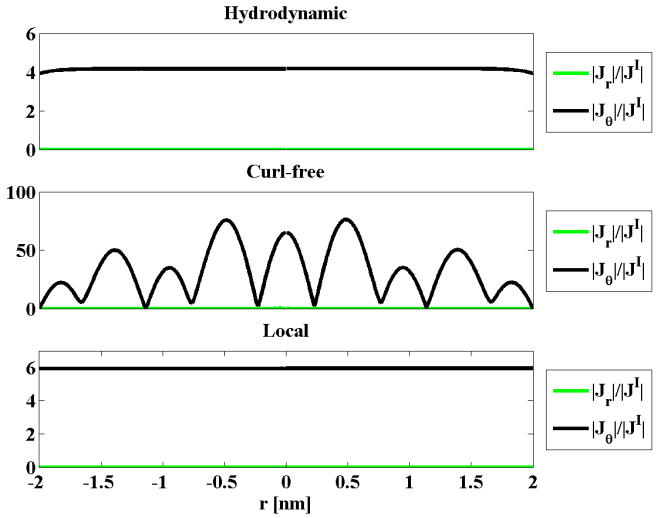

Figure 2. Radial component $\mathbf{J}_\mathrm{r}$ and tangential component $\mathbf{J}_\theta$ of the current density $\mathbf{J}$ in the nanowire along the horizontal midline ($\theta = 0$) as a function of the radial position r inside the cylinder for the local Drude, the hydrodynamic and the curl-free models. Interband transitions are neglected. The radius is $a = 2\,\mathrm{nm}$ and the frequency $\omega = 0.6503\omega_\mathrm{p}$.

4. NUMERICAL RESULTS FOR NANOWIRE DIMERS

We also implemented the hydrodynamic Drude model numerically, extending the capabilities of COMSOL as a Maxwell solver for nonlocal response.[15] This allows us to calculate solutions to the hydrodynamic model for arbitrarily shaped nanoplasmonic nanowires. For details about the implementation we also refer to our Ref. 21, although it discusses implementing the problematic curl-free model rather than the linearized hydrodynamic model. Information about numerical convergence can be found in Ref. 15. Important difference between simulating local-response models and the nonlocal hydrodynamic model is that the size of the numerical grid should be small compared to the smallest length scale in the problem. For local response this is typically the size of the plasmonic nanostructure, while for nonlocal response problems the smallest length scale is typically the Fermi wavelength, of order 1 nm. Numerical calculations for the nonlocal model are therefore usually computationally heavier. We can write the coupled wave equations as a single partial integro-differential equation in terms of the electric field alone. This would show that the response is nonlocal indeed, but as a numerical method it is not efficient. So we rather solve the coupled differential equations for $\mathbf{E}$ and $\mathbf{J}$.

Figure 3 shows the spatial profile of the field enhancement of a cylindrical dimer, both for local and for nonlocal response. Although there are no large differences, the nonlocal response gives a somewhat broader field distribution with lower amplitude. Figure 4 shows the frequency dependence of the field enhancement in between the dimers, again for local and for nonlocal response. Clearly, the field enhancement peaks are generally lower for nonlocal response, and shifted to the blue. At a fixed frequency, the nonlocal field enhancement may exceed or be equal to the value for local response. In agreement with our theoretical analysis, we observe no new resonances below the plasma frequency (at 8.9 eV for Au and Ag) in our numerical investigations, but nevertheless the differences between local and nonlocal response can be considerable for closely spaced dimers, even when the differences would be small for the individual wires when isolated.

5. CONCLUSIONS

Nonlocal response becomes noticeable in metals with nanometer-sized features. Our analytical work on the hydrodynamical model agrees quantitatively with our versatile numerical implementation in COMSOL. New resonances due to nonlocal response are predicted to appear above the plasma frequency only. In the visible spectrum, hybridization resonances of plasmonic dimers are blueshifted and appear weaker in the hydrodynamic Drude model.

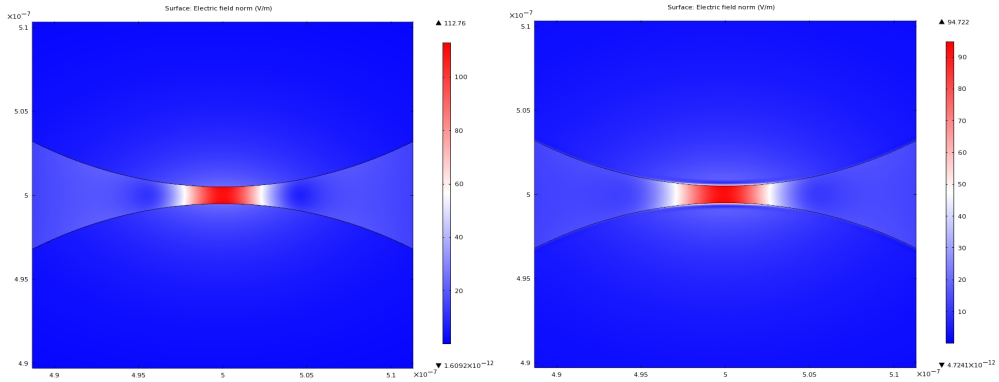

Figure 3. Norm of the electric field in [V/m] for a cylindrical dimer with radii of 25 nm and distance 1 nm, at 3.684 eV. The left panel shows the field distribution for local response, the right panel for the hydrodynamic Drude model.

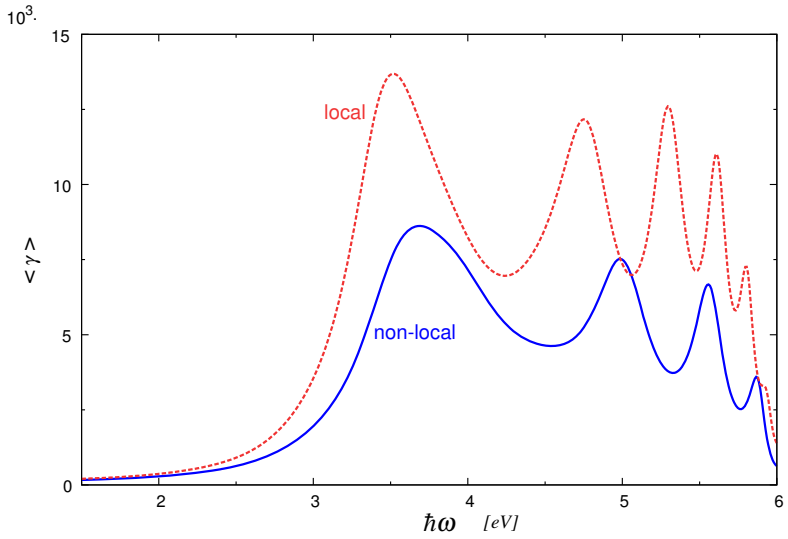

Figure 4. Averaged field enhancement $\langle\gamma\rangle$ as a function of frequency, in between two cylindrical dimers of radii 25 nm at a distance of 1 nm, for local response and for the hydrodynamical Drude model. The average is taken on the interval connecting the nearest points on the cylinders.

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