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ELECTROSTATICS OF A NANOWIRE INCLUDING NONLOCAL EFFECTS

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Abstract—We develop a method for calculating transverse static polarizability (per unit length) of a bulk nanowire by taking in to account the temporal and spatial dispersion. To describe these phenomena, we developed analytical theory based on local random-phase approximation and plasmon pole approximation. Our theory is very general in the sense that it can be applied to any material which can be characterized by a bulk dielectric function of the form $\varepsilon(\omega, k)$. The theory is applied to calculate the transverse static polarizability of dielectric nanowire.

1. INTRODUCTION

It is well known that the electronic and optical properties of very small structures, such as nanosphers [1–4], nanowires and nanocavities [5–7], nanocylinders and wires [7–9], are very different from those of the corresponding bulk materials because of surface effects. Nanosystems possess unique properties different from those of macroscopic materials when characteristic lengths govern their properties. Therefore, the spatial dispersion becomes much more important when the characteristic size of the particle or distances between them becomes comparable to the characteristic scale of the system [1, 2, 7].

One of the consequences of such a small size of the system is that the electric field **E** and the displacement vector **D** are related by a nonlocal relationship instead of the usual local relation. Assuming that fields are weak enough that displacement vector **D** can be obtained by perturbation theory, one can obtain integral linear-response relation in terms of the corresponding fields (i.e., relations between **D** and electric

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field **E**

$$\mathbf{D}(\mathbf{r}) = \int_V \varepsilon\left(\mathbf{r}, \mathbf{r}', \omega\right) \mathbf{E}\left(\mathbf{r}'\right) d^3 r' \tag{1}$$

here $\varepsilon(\mathbf{r}, \mathbf{r}', \omega)$ is the dielectric response function.

The displacement at point $\mathbf{r}$ depends on the values of the electric field at neighboring points $\mathbf{r}'$ (spatial dispersion). A spatially dispersive medium is therefore also called a *non-local* medium. This effect can be observed at interface between different media or in metallic objects with sizes comparable with the mean-free path of electrons. In most cases of interest, the effect of the spatial dispersion is very week; therefore we can assume that the materials of the system are isotropic. Otherwise, ε would have been a tensor, which would make no principal difficulty but make formulas somewhat more complicate. However, on the other hand temporal dispersion, is a widely encountered phenomenon and it is important to take it accurately into account.

As we discussed above the relation (1) is non-local. However, one can use mathematically equivalent description in Fourier domain which is local,

$$\mathbf{D}(\mathbf{k}) = \varepsilon(\mathbf{k})\mathbf{E}(\mathbf{k}), \tag{2}$$

Here we have introduced the corresponding arguments in Fourier domain for the electric field

$$\mathbf{E}(\mathbf{k}) = \int \mathbf{E}(\mathbf{r}) e^{(i\mathbf{k}\cdot\mathbf{r} - i\omega t)} d^3 r \tag{3}$$

and one can get similar expressions for other quantities.

1.1. Nonlocal Dielectric Function

Energy $\hbar\omega$ and momentum k dependence of the nonlocal dielectric function $\varepsilon(k, \omega)$ is due to the spatial correlation between the induced charge density and lattice ions, an effect which prevents the pulling up of electrons at short distances. There are some situations where the momentum transfer is important, and a local dielectric response, $\varepsilon(\omega)$, is not enough to accurately describe the interaction between electrons and sample. One of them is electron microscopy. Also, when two interfaces interact in very close proximity, or when large scattering angles of the incident beam are used, the higher values of transferred momentum are essential. In such a situation, one needs to consider a nonlocal dielectric function to describe the medium.

To describe the k dependance of the dielectric response, one can consider well-known Lindhard formula [10–12], which is one of the closed solutions in the theory of Fermi systems that explicitly gives

the nonlocal dielectric response function (longitudinal) $\varepsilon(k,\omega)$.

$$\varepsilon(k,\omega) = 1 + \frac{3\omega_p^2}{k^2 v_F^2}\left[1 - \frac{\omega}{2kv_F}\ln\left(\frac{\omega + kv_F}{\omega - kv_F}\right)\right] \tag{4}$$

where ω_p is plasma frequency and v_F the electron speed at the Fermi surface. The complex function in Eq. (4), $\ln(\frac{\omega+kv_F}{\omega-kv_F})$ is defined as $\ln(\frac{\omega+kv_F}{\omega-kv_F}) = \ln|\frac{\omega+kv_F}{\omega-kv_F}| - i\pi$ for $(\frac{\omega+kv_F}{\omega-kv_F}) < 0$. Note that $\varepsilon(k,\omega)$ has a non-zero imaginary part, which describes optical losses, only when $\omega < kv_F$. These optical losses can be connected to the excitation by a field of incoherent electron-hole pairs. This phenomenon is called Landau damping, which is described by Eq. (4). The Landau damping actually is dephasing, where coherent field oscillations are transformed into incoherent electron hole pairs, but the total energy of the system is not changed. Landau damping is fulfilled when the size of the system is comparable to or less than the correlation length, l_c, then condition, $\omega < kv_F$, satisfies this limit.

This situation can be also highly important for nanooptics: which is a modern branch of optical science that explores how optical frequency radiation can be confined on the nanoscale, i.e., 1–100 nm (much smaller than the optical wavelength). Such nanolocalized fields are due to the interaction processes (oscillation of polarization charges) between electromagnetic radiation and conduction electrons at metallic interfaces or in small metallic nanostructures, leading to an enhanced optical near field of sub-wavelength dimension. Such oscillations on the nanoscale are called surface plasmons.

Another example of such a behavior of k dependence of dielectric response is given by plasmon pole approximation [13] which accounts for the free electron oscillations and partially for the electron-hole pair generation in the material.

$$\varepsilon(k,\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma) - \frac{3}{5}k^2 v_F^2} \tag{5}$$

Actually, this is a useful approximation (random-phase approximation) of Eq. (4). One can show that in the large k limit Eq. (5) tends to the energy-momentum relationship of a single electron, $\omega = k^2/2$ while in the small k limit it reproduces the bulk-plasmon dispersion relation. This approximation has been used in three- and two-dimensional systems, with good agreement with more sophisticated treatment and experimental results.

Nonlocal corrections of the response should produce a difference between the surface mode positions and excitation strengths. Nonlocal effects have been largely studied in the planar geometry. In particular,

different features of spatial dispersion have been considered to obtain the image potential outside a metallic semi-infinite planar interface [14–17]. On the other hand, for nanoparticles or nanowires, where the induced charge density is always limited into very small regions, and therefore strongly interacting, this correlation is expected to be stronger. Therefore, nonlocal effects are likely to be more relevant.

2. MODEL AND EQUATIONS

Let us assume that an external electric field $\mathbf{E} = E_0(\omega)\hat{x}$ is applied to a nanowire whose radius is a, and the material is described by nonlocal dielectric function $\varepsilon(k,\omega)$ [see Fig. 1]. Here retardation is neglected: size of the nanowire is much smaller than the wavelength of the excitation field.

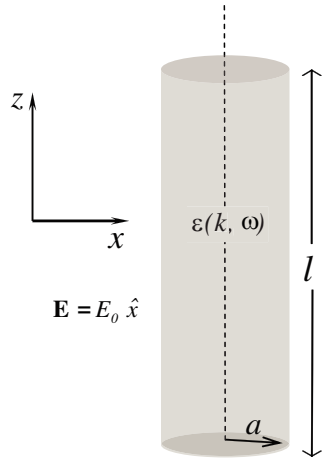

Figure 1. Schematics of a cylindrical nanowire of radius a with the axis oriented along the z axis characterized by a nonlocal dielectric function $\varepsilon(k,\omega)$. The external electric field oriented in x axis.

The potential $V(\mathbf{r})$ and radial component of the displacement vector $\mathbf{D}_r(\mathbf{r})$ for $r > a$ are given by

$$V(\mathbf{r}) = -E_0 r\cos\varphi + \frac{\wp}{r}\cos\varphi \tag{6}$$

$$\mathbf{D}_r(\mathbf{r}) = \mathbf{E}_r(\mathbf{r}) = E_0\cos\varphi + \frac{\wp}{r^2}\cos\varphi \tag{7}$$

where $\wp$ is the induced line dipole with dipole moment per unit length $\wp/\ell$ and orientation along x. Here φ is the angle between the vectors $\mathbf{r}$ and $\mathbf{E}$.

Inside the wire ($r < a$), we can write

$$\nabla \cdot \mathbf{D} = 0 \quad \text{and} \quad \nabla \times \mathbf{E} = 0. \tag{8}$$

Following Ritchie and Marusak [14], we will assume an infinite fictitious medium in order to solve for the fields inside the wire. This medium satisfies the following conditions: (i) Maxwell's equations are continued to the $r > a$ region of the infinite medium, (ii) The fields inside the real wire are the same as the ones of the infinite medium with the same response function, (iii) The normal component of the displacement $\mathbf{D}_r$ is discontinuous in $r = a$ in this infinite system, but the tangential components are continuous. Therefore, we introduce a uniform dielectric medium with a fictitious cylindrical surface of charge at $r = a$ which acts as a source for $\mathbf{D}$.

Because of this, $\nabla \cdot \mathbf{D} = 0$, which is normally valid in an infinite continuous medium, does not hold on the surface of the wire. Furthermore, $\mathbf{D}(\mathbf{r}) = \int \varepsilon(|\mathbf{r} - \mathbf{r}'|, \omega)\mathbf{E}(\mathbf{r}')d^3r'$ by our assumption and $\nabla \times \mathbf{E} = 0$ everywhere. Therefore, now we can introduce new potential function $V_D(\mathbf{r})$

$$\mathbf{D}(\mathbf{r}) = -\nabla \mathbf{V}_D(\mathbf{r}) \tag{9}$$

Next we define

$$\mathbf{V}_D(\mathbf{k}) = \int V_D(\mathbf{r}) \exp(-i\mathbf{k} \cdot \mathbf{r}) d^3r \tag{10}$$

and its inverse

$$\mathbf{V}_D(\mathbf{r}) = \frac{1}{(2\pi)^3} \int V_D(\mathbf{k}) \exp(-i\mathbf{k} \cdot \mathbf{r}) d^3k \tag{11}$$

in this infinite system. Similar relations for the total potential $V(\mathbf{r})$ and its Fourier transform $V(\mathbf{k})$ can be defined.

We note that outside the cylindrical surface of the fictitious medium, $V(\mathbf{r})$ and $V_D(\mathbf{r})$ must be of the form

$$V(\mathbf{r}) = \widetilde{\mathcal{V}}(r)\cos\varphi, \quad \text{and} \quad V_D(\mathbf{r}) = \widetilde{\mathcal{V}}_D(r)\cos\varphi \tag{12}$$

Expressions in Eq. (8) together with Eq. (9) reduce to $\nabla^2 V_D = 0$. Multiplying this equation by $e^{-i\mathbf{k}\cdot\mathbf{r}}$ and integrating over $\mathbf{r}$, we get

$$-k^2 V_D(\mathbf{k}) + a\zeta \left[\int e^{-i\mathbf{k}\cdot\mathbf{r}} \cos\varphi d\varphi dz \right]_{r=a} = 0. \tag{13}$$

Although $d\widetilde{\mathcal{V}}_D/dr$ is continuous at the actual cylinder-vacuum interface, it is discontinuous at $r = a$ in our fictitious medium. Therefore, the ζ in the Eq. (13) can be written as

$$\zeta = -\left[\frac{d\widetilde{\mathcal{V}}_D}{dr} \right]_{r=a^+} + \left[\frac{d\widetilde{\mathcal{V}}_D}{dr} \right]_{r=a^-} \tag{14}$$

We expand $e^{i\mathbf{k}\cdot\mathbf{r}}$ in cylindrical coordinates by using the relation [18]

$$e^{(i\mathbf{k}\cdot\mathbf{r})} = e^{i\lambda z}\sum_{m=-\infty}^{m=\infty} i^m J_m(pr)e^{im(\varphi-\varphi_p)} \tag{15}$$

where $\mathbf{k} = k\hat{k} = p\hat{\rho} + \lambda\hat{z}$ is Fourier transform variable in cylindrical coordinates (p, φ_p, λ), and J_m is the Bessel function of order m. After integrating Eq. (13) over z and φ we get

$$k^2 V_D(\mathbf{k}) + \frac{2\pi a}{\lambda}\zeta J_1(pa)\cos\varphi_p = 0. \tag{16}$$

Then one can show that

$$V_D(k) = -\frac{2\pi a}{\lambda k^2}\zeta J_1(pa)\cos\varphi_p \tag{17}$$

and,

$$V(k) = -\frac{2\pi a}{\lambda k^2\varepsilon(k,\omega)}\zeta J_1(pa)\cos\varphi_p. \tag{18}$$

Now we come back to the space coordinate using equations like Eq. (11). For $r < a$, this gives

$$V_D(\mathbf{r}) = \frac{\zeta r}{2}\cos\varphi \quad \text{and} \quad V(r) = \frac{a\zeta}{2}\mathcal{F}(r)\cos\varphi \tag{19}$$

where

$$\mathcal{F}(r) = -\frac{i}{\pi}\int \frac{e^{i\lambda z}J_1(pa)J_1(pr)}{\lambda\left(p^2+\lambda^2\right)\varepsilon\left(\sqrt{p^2+\lambda^2},\omega\right)}pdpd\lambda. \tag{20}$$

We will take this potential as the one corresponding to our real system inside the wire ($r < a$) and apply the boundary conditions at $r = a$. We can match $V(r,\omega)$ and $\mathbf{D}(\mathbf{r})$ obtained from Eqs. (9) and (19) with the corresponding quantities outside the wire, given by Eqs. (6) and (7) at $r = a$. Then one can get the following system of equations

$$-E_0a^2 + \wp = a^2\zeta\mathcal{F}(r) \quad \text{and} \quad E_0a^2 + \wp = -\frac{1}{2}\zeta a^2. \tag{21}$$

Our final result for $\wp$ can be written as

$$\wp = E_0a^2\left(\frac{1-2\mathcal{F}(a)}{1+2\mathcal{F}(a)}\right) \tag{22}$$

where $\mathcal{F}(a) = \mathcal{F}(r)|_{r=a}$. When $\varepsilon(k,\omega) \to \varepsilon(\omega)$, i.e., in the local limit $\mathcal{F}(a) \to [2\varepsilon(\omega)]^{-1}$ and $\wp$ reduces to the familiar expression found in textbooks [19], $\wp = E_0a^2(\frac{\varepsilon(\omega)-1}{\varepsilon(\omega)+1})$ and finally we define $\delta(\omega) \equiv \wp/E_oa^2$. The $\delta(\omega)$ can now be calculated from Eq. (22). Of particular interest is the Im$[\delta(\omega)]$, because it is proportional to the absorption coefficient of the nanowire and its variation with frequency can be demonstrated.

3. APPLICATIONS

Plasmon pole approximation: In this approximation, using Eq. (5) for the dielectric function, one can determine the function $\mathcal{F}(a)$ given by Eq. (20) as,

$$\mathcal{F}(a) = -\frac{i}{\pi}\int \frac{e^{i\lambda z}\left[\xi_2^2 - \left(\lambda^2+p^2\right)\right][J_1(pa)]^2}{\lambda\left(\lambda^2+p^2\right)\left[\xi_1^2-(\lambda^2+p^2)\right]}pdpd\lambda. \tag{23}$$

where $\xi_1 = \sqrt{[\omega(\omega+i\gamma)-\omega_p^2]}/\beta$, $\xi_2 = \sqrt{\omega(\omega+i\gamma)}/\beta$ and $\beta = \sqrt{3/5}v_F$. One can easily determine the function $\mathcal{F}(a)$ given by Eq. (23) [20]

$$\mathcal{F}(a) = -\xi_1^{-2}\left(\xi_1^2-\xi_2^2\right)[I_1(ia\xi_1)K_1(ia\xi_1)] \tag{24}$$

Therefore, our final result for $\delta(\omega)$ takes the form

$$\delta(\omega) = \frac{\xi_1^2 + 2\left(\xi_1^2-\xi_2^2\right)[I_1(ia\xi_1)K_1(ia\xi_1)]}{\xi_1^2 - 2\left(\xi_1^2-\xi_2^2\right)[I_1(ia\xi_1)K_1(ia\xi_1)]} \tag{25}$$

To illustrate nonlocal effects, we present in Fig. 2 the Log$[\alpha(\omega)]$ (where $\alpha(\omega) \equiv \mathrm{Im}[\delta(\omega)]$) for the nanowire as a function of the ω/ω_p. Here we take $\omega_p = 8.16\,\mathrm{eV}$, $\gamma = 0.001\omega_p$ and $v_F = 10^6\,\mathrm{ms}^{-1}$.

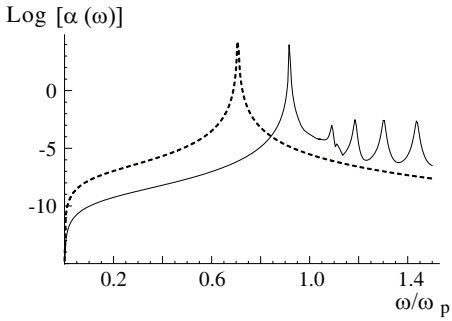

Figure 2. Variation of the Log$[\alpha(\omega)]$ with ω/ω_p for 1 nm radius nanowire. The dashed curve shows the result of the local theory.

The calculated spectrum of a nanowire of radius 1 nm is shown in Fig. 2. The results for the local effects are shown by the dashed curve, and have only one peak, at the frequency $\omega_p/\sqrt{2}$. With the inclusion of nonlocal effects, the main peak blueshifted from its classical value of $\simeq 0.7\omega_p$ to $\simeq 0.9\omega_p$ and more important series of small peaks appear above the bulk plasma frequency. These subsidiary peaks are due to the excitation of bulk plasmons. These peaks are in good agrement

with the peaks that have been predicted in metal nanowires [7]. This figure may be compared with Ruppin's result given in [7] and with good agreement. In particular, our result predicts (just like [7]) a shift in the main peak and series of small peaks above plasma frequency. Our result also agrees with that of [7] as far as the relative heights of the main peak and these secondary peaks are concerned. The slight discrepancy in the positions of the main and secondary peaks is presumably due to the use of retardation effects in his work. Therefore, in [7] both the longitudinal and transverse dielectric functions were considered. In [7], all expressions and the final results depend on only one wave vector. However, in the present approach the final results depend on all wave vectors. In other words, in [7] the authors assumed that $1/\varepsilon(k,\omega)$ had sharp maximum at a particular wave vector. If the function $1/\varepsilon(k,\omega)$ has broad maximum then Eq. (20) is more reliable.

We now investigate the influence of the nanowire radius on the position and the relative heights of the main peak and secondary peaks. When we increase nanowire radius from 1 nm to 2 nm, qualitatively, we have a similar situation as in Fig. 2. As shown in Fig. 3(a), even though the number of secondary peaks increases, their relative amplitudes decrease. As expected, the position of the main peak redshifted from its classical position. As shown in Fig. 3(b), this behavior continue when the radius of the nanowire increases further.

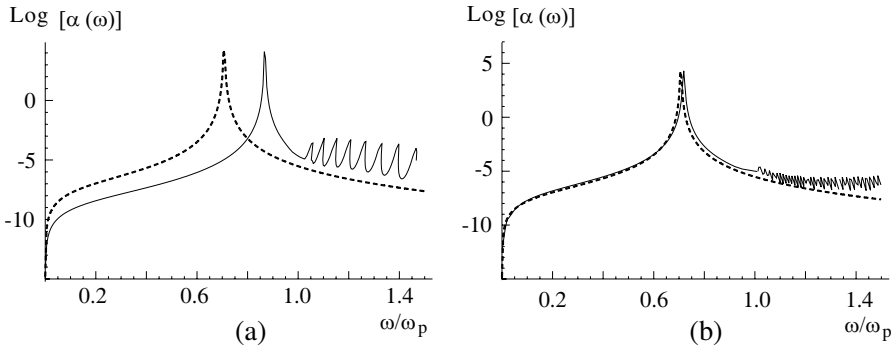

Figure 3. Same figure as Fig. 2. (a) For 2 nm and (b) for 5 nm radius nanowire.

Due to their small amplitudes, detecting any of the secondary peaks above the plasma frequency would be difficult. In a thin nanowire, the electron collisions with the surface suppress the electron mean free path. This will contribute to enhancement of the γ in the dielectric function. As a result, it will further suppress the amplitudes of the secondary peaks. Since the position of the main peak dose not

depend on the γ, the main peaks might be observable in very small nanowire.

4. CONCLUSION

We have developed a simple theoretical framework for the transverse static polarizability of a nanowire that allows the inclusion of nonlocal effects. Compared to a local approach, a blueshift in the energy of the plasmons is obtained. In particular, our results are significant for very thin wires, where the nonlocal effects are much more relevant. In a sense, our theory is more general and can even be applied to a nonlocal response function given in Eq. (4) (not shown here). We hope that our work will be useful in studying the optical properties of nanowires. Nonlocal effects in materials commonly used in plasmonics such as gold and silver can be addressed within this model with the use of an appropriate nonlocal response function. Finally, our expression for $\delta(\omega)$ can provide a simple but accurate way for taking into account both the size and the frequency dependence of the polarizability.

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