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Non-local optical response of two-dimensional arrays of metallic nanoparticles

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Abstract

The optical absorption spectra of two-dimensional (2D) arrays of spatially dispersive metallic nanoparticles is examined. The corresponding non-local dielectric function of the spheres is provided by (a) the hydrodynamic approximation and (b) the Lindhard theory. More specifically, it is shown that the adoption of either types of non-local dielectric functions (hydrodynamic or Lindhard-type) for dilute 2D arrays of spheres does not alter the structure of the optical absorption spectrum but leads to a blue-shift of all its distinct features. This effect becomes more prominent as the radius of the nanoparticles decreases. However, for a close-packed arrangement of spheres, a non-local dielectric function provides a significantly different optical spectrum than the local one, due to the dominant role of near-field effects, which strongly depend on the choice of the dielectric function of the metal.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Plasmonics constitutes a new, emerging field of condensed matter which deals with phenomena associated with the interaction of light with surface modes in metallic nanostructures. Of particular interest is the interaction of light with arrays of metallic nanoparticles and nanohole arrays (for a recent review see [1] and references therein). Recent advances in electron-beam lithography and self-assembly nanofabrication techniques enable the preparation of well-defined systems of nanoparticles with a tailored shape, size and arrangement, and allow for the observation of new, interesting and potentially useful physical phenomena [2–6].

The theoretical treatment of light interaction with arrays of metallic nanoparticles is principally studied by means of the multiple-scattering theory either in its bulk [7, 8] or layer formulation [9–14]. Usually, this treatment is based on the assumption of a local electromagnetic (EM) response, which means that the dielectric function describing the optical response of metals depends only on the angular frequency ω and is independent of the wavevector \mathbf{k} . In a recent work [15], a non-local dielectric function for the metal has been assumed for dilute three-dimensional crystals of metal-coated dielectric

spheres without, however, introducing dramatic effects in the absorption spectra compared to a typical local Drude-type dielectric function. In the past, the adoption of a non-local optical response for metals has been employed for single metal particles [16–19] and small aggregates of such [20, 21] as well as for single metal nanoshells [22]. The common observation of the above studies is a blue-shift of the dipole surface plasmon resonance and a small secondary structure due to the excitation of bulk-plasmon resonances.

The present work presents rigorous electrodynamics calculations of the optical response of two-dimensional (2D) ordered arrays of metal nanoparticles by means of the layer-multiple-scattering (LMS) method [23–25]. The metal nanoparticles are assumed to be spatially dispersive and are described by a wavevector-dependent dielectric function according to the hydrodynamic [31] and Lindhard [32] models. The optical absorption spectra from arrays of spatially dispersive particles are compared against those obtained for particles described by a local, Drude-type dielectric function. The prominent features of the absorption spectrum obtained for a non-local dielectric function are blue-shifted relative to those of the local case. The shift increases with decreasing the sphere radius for constant surface coverage. For an array

of touching spheres, the optical absorption spectra in the local and non-local treatments differ markedly. For small values of the metal loss factor, the assumption of a non-local response induces an evident fine structure of bulk-plasmon resonances. When the electron scattering at the particle boundary is taken into account, the loss factor is greatly enhanced, leading to a suppression of fine details of the absorption spectrum. However, even in this case, the absorption spectra in the local and non-local pictures differ significantly. Namely, when a local Drude-type dielectric function is employed, the absorption spectrum shows an unusually strong absorption tail for very low frequencies, which is neither predicted in the non-local treatment nor has ever been measured experimentally in such systems. The paper is organized as follows. Section 2 presents a brief outline of the theory used in this paper. Section 3 presents results for 2D periodic arrays of spheres and section 4 concludes the paper.

2. Theory

We consider a single nonmagnetic spherical scatterer of radius S described by a transverse dielectric function, ϵ_T , and a longitudinal one, ϵ_L . The scatterer is assumed to be embedded within a nonmagnetic host homogeneous material whose dielectric function is denoted by ϵ_h . For this case, the E -components of the corresponding scattering T -matrix have been provided by Ruppin [16]

$$T_{El}(\omega) = \left\{ \left\{ j_1'(q_L r) \left[j_1(q_s r) \frac{\partial}{\partial r} (r j_1(q_h r)) \epsilon_s - j_1(q_h r) \times \frac{\partial}{\partial r} (r j_1(q_s r)) \epsilon_h \right] - c_1 j_1(q_h r) \right\} \left\{ j_1'(q_L r) \left[h_1^+(q_h r) \times \frac{\partial}{\partial r} (r j_1(q_s r)) \epsilon_h - j_1(q_s r) \frac{\partial}{\partial r} (r h_1^+(q_h r)) \epsilon_s \right] \right\}^{-1} \right\}_{r=S} \quad (1)$$

where

$$c_1 = l(l+1) \frac{j_1(k_L S)}{q_L S} j_1(k_T S) (\epsilon_T - \epsilon_h). \quad (2)$$

For $c_1 = 0$, equation (1) takes the usual form, valid for a local dielectric function. j_l is the spherical Bessel function and h_l^+ the spherical Hankel function. $q_h = \sqrt{\epsilon_h} \omega / c$ whilst q_T and q_L are the transverse and longitudinal wavenumbers within the sphere, respectively. They are given by

$$q_T^2 = \epsilon(q_T, \omega) \omega^2 / c^2 \quad (3)$$

and

$$\epsilon(q_L, \omega) = 0. \quad (4)$$

The H -component of the T -matrix remains the same as in the case of a scatterer described by a single (transverse) local dielectric function. In the derivation of equation (1) the electric-field spherical-wave expansion within the sphere, apart from the transverse components, also contains a longitudinal component [16].

Within the hydrodynamic model [31], the transverse ϵ_T and longitudinal dielectric functions are given by

$$\epsilon_T(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)} \quad (5)$$

$$\epsilon_L(q, \omega) = 1 - \frac{\omega_p^2}{\omega^2 - \beta q^2 + i\omega\gamma}, \quad (6)$$

where ω_p is the bulk plasma frequency of the metal and γ is the loss factor. $\beta = \frac{3}{5} v_F^2$ where v_F is the Fermi velocity of the metal. Equation (5) is the ordinary Drude dielectric function which is widely used to describe metals within the local response approximation. Substituting equation (6) into equation (4) we obtain the longitudinal wavenumber within a metal sphere

$$q_L^2 = (\omega^2 + i\omega\gamma - \omega_p^2) / \beta. \quad (7)$$

Within the Lindhard [32] theory, ϵ_T and ϵ_L are provided by [16]

$$\epsilon_T(q, \omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)} \frac{3}{2\alpha^2} \left(\frac{1 + \alpha^2}{\alpha} \arctan \alpha - 1 \right) \quad (8)$$

$$\epsilon_L(q, \omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)} \frac{3}{\alpha^2} \left(1 - \frac{\arctan \alpha}{\alpha} \right) \times \left[1 + \frac{i}{\omega\gamma} \left(1 - \frac{\arctan \alpha}{\alpha} \right) \right]^{-1} \quad (9)$$

where

$$\alpha^2 = - \frac{q^2 v_F^2}{(\omega + i\gamma)^2}. \quad (10)$$

The corresponding wavenumbers q_T and q_L of the Lindhard-type dielectric functions are found by substituting equations (8) and (9) into equations (3) and (4), respectively. The latter are solved numerically using the corresponding q_T, q_L of the hydrodynamic model as starting values. Having calculated q_T and q_L in either models (hydrodynamic or Lindhard), one can calculate the corresponding T -matrix from equation (1) for a single metal sphere and from that solve the scattering problem for a 2D array of spheres by means of the LMS method [23–25]. The LMS method is ideally suited for the calculation of the transmission, reflection and absorption coefficients of an EM wave incident on a composite slab consisting of a number of layers which can be either planes of non-overlapping particles with the same 2D periodicity or homogeneous plates. For each plane of particles, the method calculates the full multipole expansion of the total multiply scattered wave field and deduces the corresponding transmission and reflection matrices in the plane-wave basis. The transmission and reflection matrices of the composite slab are evaluated from those of the constituent layers.

3. Results

The theory presented in the previous section is applied to the case of a 2D square array of metallic (silver) spheres of radius S occupying the sites of a square lattice of period a (see figure 1). In what follows, we will use as frequency unit the bulk plasma frequency of the metal ω_p , and as length unit the quantity c/ω_p . For silver, $\hbar\omega_p = 9.2$ eV [26, 27], and hence the length unit is $c/\omega_p = 21.46$ nm. The bulk loss factor is $\hbar\gamma_b = 0.02$ eV. We note that the local Drude dielectric function (equation (5)) is not sufficiently accurate for a wide range of frequencies [28, 29] as it does not take into account the interband transitions taking place in the metal. However,

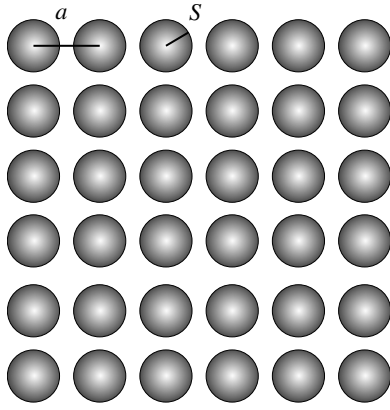


Figure 1. A square lattice of spheres of radius S and period a , where $S/a = 0.4$.

to the best of our knowledge, an analytic non-local dielectric function which goes beyond the Drude model is not at hand and, therefore, interband phenomena are not included in our calculations.

Figure 2 shows absorption spectra for light incident normally on 2D square arrays of silver spheres of different radii and lattice constants for a fixed surface coverage of spheres, $f = \pi(S/a)^2 = 0.503$. In order to achieve converged results, we have taken the cutoff l_{\max} in the angular-momentum expansion of the EM field to be equal to 5. We have also taken into account 29 reciprocal-lattice vectors in the plane-wave expansion of the EM field. For each case, figure 2 depicts the absorption curves for spheres described by local and non-local dielectric functions. It is worth noting that both the hydrodynamic and Lindhard models for the non-local dielectric function of the metal yielded the same results. So, the calculations were performed for the hydrodynamic model (equations (5) and (6)) as it is simpler than the Lindhard one. The parameter β in equation (6) is taken to be $\beta/c^2 = 1.29 \times 10^{-5}$ corresponding to the Fermi velocity $v_F = 1.4 \times 10^6 \text{ m s}^{-1}$ of silver. In figure 2, one observes that the main consequence stemming from the adoption of a non-local dielectric function is an overall shift towards higher frequencies (blue-shift) of the structure of the absorption spectrum. The shift is caused by the introduction of longitudinal waves within the silver spheres which modify the internal electric field and correspondingly the resonance condition [16]. However, it is evident that the blue-shift is much more apparent as the radius of the spheres becomes smaller. This is due to the fact that the longitudinal waves which are taken into account in a non-local treatment of the scattering from the arrays of spheres, are of evanescent nature. Therefore, for large enough spheres their contribution to the total electric field is appreciable only close to the surface of the sphere. As the spheres become smaller, the area where the longitudinal waves are important increases, giving rise to more distinguishable effects when compared to local treatment of scattering where longitudinal waves are not allowed. The above are in accordance with previous non-local effective-medium treatments [30].

The structure which is obvious in both local and non-local treatments of the EM response of the metallic spheres stems from the surface plasmon resonances taking place at

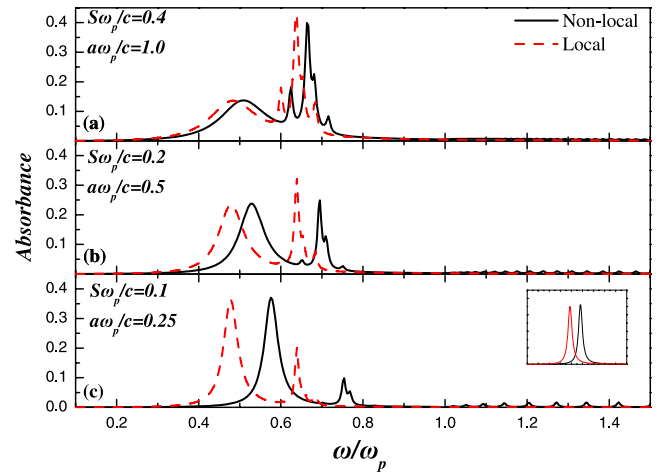


Figure 2. Absorption spectra of normally incident light for a square lattice of silver ($\hbar\omega_p = 9.2 \text{ eV}$, $\hbar\gamma_b = 0.02 \text{ eV}$, $v_F = 1.4 \times 10^6 \text{ m s}^{-1}$) spheres for different sphere radii and lattice constants. For all curves, the surface coverage is the same, $f = \pi(S/a)^2 = 0.503$. The inset shows the absorption spectrum for $S\omega_p/c = 0.1$, in the dipole approximation. The solid lines refer to metallic spheres described by a non-local dielectric function whilst the broken lines to those described by a local one.

each sphere. Of course, there is strong interaction between the surface plasmon resonances of neighbouring spheres, giving rise to broader absorption lines. The lowest frequency minimum corresponds to a dipole excitation of the metallic spheres which is around the single-sphere surface resonance $\omega_{sp} = \omega_p/\sqrt{3}$. This is also evident from the inset of figure 2(c) which shows that the main absorption peaks are practically reproduced from the dipole approximation ($l_{\max} = 1$). The second lowest absorption peak corresponds to a quadrupole surface plasmon, etc. It is evident that the higher-multipole absorption peaks become more pronounced as the spheres become larger, a phenomenon which is evident already from the absorption cross-section of a single metallic sphere [33].

In figure 2(c) and for frequencies above the bulk plasma frequency, i.e. $\omega/\omega_p > 1$, we observe a fine structure of small absorption peaks which is apparent only for spatially dispersive spheres (non-local dielectric function). This structure stems from the excitation of bulk plasmons at the spheres of the array [16]. The bulk plasma absorption peaks are also slightly evident for larger spheres (see figure 2(b) for $\omega/\omega_p > 1$). We note, however, that in an experimentally obtained absorption spectrum it is very hard to observe bulk-plasmon excitations since the actual losses of a metallic nanoparticle are much larger than those of bulk metal due to the additional scattering of the conduction electrons at the boundary of the particle. The latter effect obscures the observation of fine details in the absorption spectrum (see the discussion below). The bulk plasma excitations in metallic nanoparticles can possibly be confirmed by electron energy-loss spectroscopy as is the case for bulk metals.

It is well known that the effect and the significance of a non-local dielectric function is much more dramatic when near-field phenomena come into play. For example, non-local dielectric functions must be used for accurate calculations of

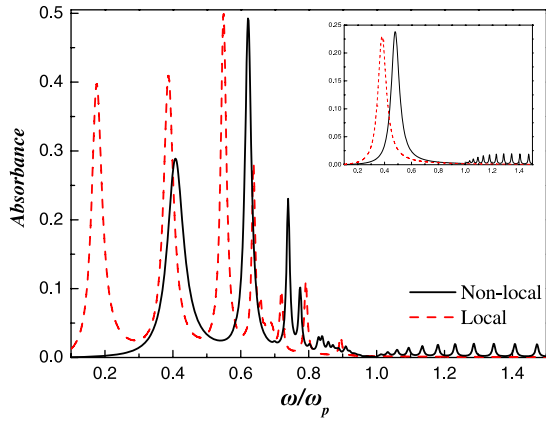


Figure 3. Absorption spectra of normally incident light for a square lattice of close-packed metallic spheres with $S\omega_p/c = 0.125$ and $a\omega_p/c = 0.25$. The inset shows the same absorption spectra in the dipole approximation. The solid line refers to metallic spheres described by a non-local dielectric function whilst the broken line to those described by a local one.

the van der Waals forces between nanoparticles, especially when the particles are in very close proximity [34]. Also, when calculating the thermal near field close to a metallic surface a non-local optical response should be employed [35]. Therefore, we have also studied the case where the spheres of the array are touching, i.e. $S/a = 0.5$ in figure 1. The corresponding absorption spectrum is shown in figure 3 and it is obtained for 129 reciprocal-lattice vectors and $l_{\max} = 15$. It is worth noting, however, that the corresponding curve for the non-local dielectric function converged for fewer angular-momentum terms, i.e. $l_{\max} = 12$. This may be due to the fact that when near-field effects are dominant (such as the case of touching spheres) a non-local dielectric function is a more natural choice than the local one [20].

From figure 3 it is obvious that the non-local and local treatments provide drastically different absorption spectra. For example, the local dielectric function predicts an absorption maximum around $\omega/\omega_p \sim 0.15$ which is absent in the case of a non-local dielectric function. As can be also seen from the inset of figure 3, this low-frequency peak is not evident in the dipole approximation ($l_{\max} = 1$) but is formed progressively as l_{\max} increases. This may seem to be a counterintuitive result since the higher-multipole surface plasmon resonances of a single metallic sphere, and accordingly the corresponding resonance bands generated by the interaction of neighbouring spheres, always lie above the dipole contribution (in frequency). However, since metal covers about 78.5% of the 2D space in a close-packed arrangement of spheres, the optical response of such an array can be alternatively described as that of a homogeneous metal containing air holes (cavities) of nanometre scale. Each air hole is defined as the space between four close-packed spheres. In this picture, the transmission/absorption resonances stem from the interaction of the surface-cavity modes of neighbouring holes rather than from the interaction of modes of neighbouring spheres. In the first case (interaction of neighbouring air holes) the cavity modes generated from higher-multipole contributions lie below

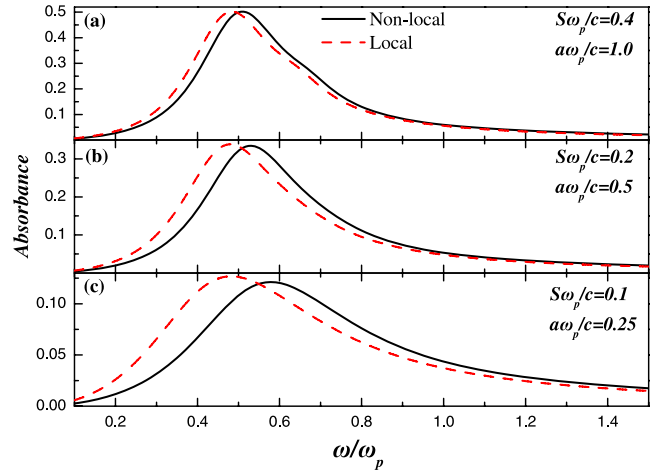


Figure 4. The same as figure 2 but with scattering at the particle boundary taken into account in the loss factor γ . For (a) $\hbar\gamma = 0.13$ eV, (b) $\hbar\gamma = 0.23$ eV, and (c) $\hbar\gamma = 0.45$ eV.

the dipole ones in frequency as has been shown for the case of spherical air holes in a homogeneous metal [36–38]. A 2D array of touching spheres is in the cross-over regime between a network topology (air cavities in a metal) and a cermet one (metal bodies in air) where these two regimes exhibit entirely different characteristics. For example, a system with network topology possesses a cutoff frequency below which no propagation is allowed [36] whilst in the cermet topology such a frequency is absent. Therefore, there is no unambiguous physical picture illuminating the mechanism of optical absorption in the case of touching spheres, although a rigorous numerical calculation is at hand. The only exception is the peak structure above ω_p which is due to the bulk plasmon excitations of the spheres.

In figures 2 and 3 we have neglected the additional light absorption which takes place within the silver particles due to the scattering of the electrons at the particle boundary. In this case, the corrected loss factor γ is provided by [27]

$$\gamma = \gamma_b + v_F S^{-1}. \quad (11)$$

As is evident from the above, the correction becomes significant as the particle size decreases. In order to take into account the effect of the electron scattering at the particle boundary, we have recalculated the absorption spectra of figure 2 for the suitable loss factor in each case: for $S\omega_p/c = 0.4$ ($S = 8.58$ nm) the loss factor is $\hbar\gamma = 0.13$ eV, for $S\omega_p/c = 0.2$ ($S = 4.29$ nm) $\hbar\gamma = 0.23$ eV, and for $S\omega_p/c = 0.1$ ($S = 2.15$ nm) $\hbar\gamma = 0.45$ eV. It is evident that the fine structure of figure 2 is lost. Namely, all the higher-multipole peaks below ω_p are submerged within the dipole one [27] whilst the weak bulk-plasmon excitations above ω_p are evidently widened and lost. However, the blue-shift of the absorption peak introduced by the adoption of a non-local dielectric function is still present and becomes more obvious as the particle size decreases. We note that for very small spheres such as those of figures 2(c) and 4(c), the blue-shift introduced by the non-locality of the dielectric function is compensated by a red-shift when a more rigorous, microscopic treatment is employed [39, 40].

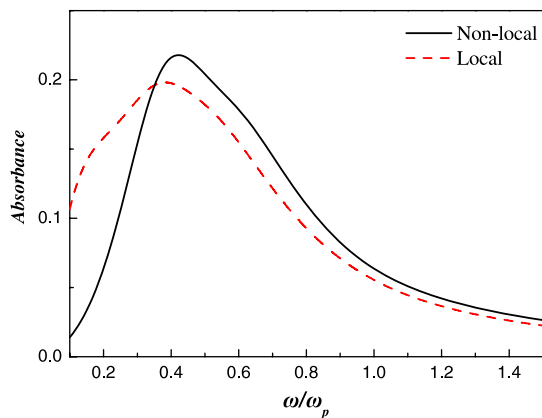


Figure 5. The same as figure 3 but with scattering at the particle boundary taken into account in the loss factor, i.e. $\hbar\gamma = 0.36$ eV.

Finally, in figure 5, we have recalculated the absorption spectra of figure 3 with the electron scattering at the particle boundary taken into account; the latter enhances dramatically the loss factor γ . Again, the dominant effect is the merging of all distinct peaks of figure 3 into a single one, in both local and non-local treatments. Also, the blue-shift of the non-local absorption peak is evident. However, the surprising result of figure 5 is the wide absorption tail in the long-wavelength limit (low frequencies) obtained for the local Drude-type dielectric function only, implying an enhanced absorption in the infrared (IR) region. We note enhanced IR absorption has been predicted theoretically and observed experimentally only for fractal aggregates of metal nanoparticles excluding other topologies of close-packed structures either periodic or random [41]. On the contrary, the non-local treatment does not predict significant absorption in the long-wavelength limit in accordance with previous work. The latter justifies the adoption of a non-local treatment in close-packed topologies such as that of figure 5.

4. Conclusions

The effect of a non-local dielectric function in the absorption spectra of 2D arrays of metallic nanoparticles has been studied by means of rigorous electrodynamic calculations. Both hydrodynamic and Lindhard types of non-local dielectric functions have generated the same absorption spectra. When these spectra are compared with those obtained from the assumption of a local, Drude-type, dielectric function, the overall effect is a blue-shift of the absorption peaks stemming from the surface plasmon excitations at the spheres of the array. In the case of a square array of touching metallic spheres the assumption of non-locality in the optical response of the metallic spheres leads to a significantly modified absorption spectrum compared to the local case. Namely, the use of a local Drude-type dielectric function introduces a strong absorption tail for very low frequencies in contrast to the non-local treatment. Since this effect has not been reported experimentally for periodic close-packed systems (only fractal geometries demonstrate this effect) we may conclude that the assumption of a non-local Drude-type function for close-packed metallic systems constitutes a far more natural choice.

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