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Effective analysis of arrays of nanospheres for near-field enhancement and subwavelength imaging in the optical region

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ABSTRACT

Recently there has been a number of studies of single-layer and double-layer arrays of small resonant particles made of a noble metal. The intense interest to these structures is caused by their promising properties for near-field enhancement and subwavelength imaging applications, especially in the optical range. They have substantial advantages over the structures containing DNG (double negative) materials as they are easier in fabrication and may mitigate the problem of losses. So far the super-resolution properties were theoretically investigated only for the arrays of a finite extent. In this work we consider single-layer and multilayer infinite arrays. This formulation allows to build a highly effective algorithm and to consider both the problem of excitation of a periodic structure by a single dipole and the modal properties of the structure. The field produced by a single dipole source is effectively described by using the array scanning method, accelerated by the Ewald method. Each subwavelength sphere is represented as an electric dipole scatterer. Special attention is given to the investigation of the number of layers influence on local field enhancement and to the study of the field distribution between the layers.

Keywords: nanostructures, periodic structures, nanoparticles, plasmons, local field enhancement

1. INTRODUCTION

It is well known that there is the diffractive limit for conventional lenses based on curved surfaces. No conventional lens can focus light onto an area smaller than a square wavelength. This is because they can transport only the propagating (visible) part of the spatial spectrum produced by a source. An alternative was suggested by Pendry [1]. He has shown that a slab of DNG (double negative) material can cancel the decay of the evanescent waves [1] that can thus be all “transported” on the other side of the material. Before, Veselago [2] theoretically predicted lensing properties of the DNG material which were the evident consequence of the Snell’s law for the visible spectrum of a source. A DNG material is able to focus all propagating and evanescent components of the radiated spectrum of a source, and only losses may limit the perfect reconstruction of the source image. Pendry has also shown the possibility to use a silver film which possesses only negative permittivity to transport the evanescent part of spectrum [1], although in this case the visible spectrum cannot propagate through such a “lens”. This possibility was confirmed in the experiment where subwavelength details were reproduced on the photoresist by illumination through a thin silver film [3]. This phenomenon can be explained by the excitation of the surface plasmon polaritons at the interfaces, which support certain values of evanescent spectrum.

It is natural to assume that structures that support an “interface” mode may be able to transport a part of the image evanescent spectrum. One of possible realizations was suggested in [4,5]. It contains two planar arrays of nanoscatterers that support slow modes. The authors considered arrays of silver and gold nanospheres and have shown the possibility to achieve local field enhancement and super resolution using these structures. The main advantage of this approach is that it does not require a bulk DNG medium.

Note that in [5] the array had a finite extent. In this work we consider a different formulation to analyze single-, double- and multilayer periodic array of metal nanospheres with infinite extent along two directions, excited by a single electric dipole source on one side. The presented effective formulation is based on the array scanning method (ASM) [6-11], that expresses the field produced by a single source in a periodic environment via a spectral integral over the Brillouin zone.

We treat metal spheres as electric dipolar scatterers. This simplification is generally used when the spheres are much smaller than the wavelength [4,5,12-19]. We used the Drude model for the representation of permittivity of a particle material. Since the problem is periodic in two directions, the required periodic dyadic Green function (GF) is accelerated with the Ewald method [20-22]. The interactions between spheres take into account all the radiative and reactive parts of the dipole fields, with the self-polarization term extracted from the Ewald acceleration scheme. This approach is very efficient for the case of dipole excitation and for mode analysis.

When one or more layers of arrayed nanospheres are excited by a dipole, they are able to provide local field enhancement and super resolution effects on the other side of the stacked layers. Field enhancement has been related to the modal dispersion characteristics. We analyze how the field is transported by the resonant mode of the periodic structure across the layers, and we show subwavelength field localization on the other side of the layers.

2. MODEL OF EXCITATION OF AN INFINITE ARRAY OF METALLIC NANOSPHERES BY A SINGLE ELECTRIC DIPOLE

The structure under investigation consists of an infinite array (made of one or more layers) of metallic nanospheres, periodic along the x and y directions (Fig. 1), with periods a and b , respectively. The radius of spheres r_p is considerably smaller than the wavelength λ in the host material. The positions of the spheres are $\mathbf{p}_{lmn} = ma\hat{x} + nb\hat{y} + z_l\hat{z}$, $m, n = 0, \pm 1, \pm 2, \dots$, $l = 1, \dots, N$. Here the bold symbols denote vectors and the unit vectors are denoted by a hat ($\hat{}$). We focus on multilayer configurations with layers at longitudinal positions $z_1, z_2, \dots, z_N$, where N is the number of layers.

Fig. 1. Geometry of the problem. (a) 3D view of a single-layer structure.. (b) Multi-layer structure. Only the y - z cross-section cut is shown.

The array is excited by a dipole source with dipole moment $\mathbf{p}_S$ placed at $\mathbf{r}_S = x_S\hat{x} + y_S\hat{y} + z_S\hat{z}$. The field excited by a single electric dipole in proximity of an infinite periodic structure can be efficiently calculated using the array scanning method (ASM) [6-11]. Accordingly, the solution of the problem in Fig. 1 is retrieved from the solutions of the associated auxiliary problem, where the periodic array of spheres is excited by a periodic system of linearly-phased dipoles

$$\mathbf{p}_{S,mn} = \mathbf{p}_S e^{-j\mathbf{k}_t \cdot \mathbf{p}_{mn}} \quad (1)$$

placed at positions $\mathbf{r}_{S,mn} = \mathbf{r}_S + ma\hat{x} + nb\hat{y}$, $m, n = 0, \pm 1, \pm 2, \dots$. Here $\mathbf{p}_{mn} = ma\hat{x} + nb\hat{y}$ and $\mathbf{k}_t = k_x\hat{x} + k_y\hat{y}$ is the transverse excitation wavevector. The field $\mathbf{E}(\mathbf{r}, \mathbf{r}_S)$ at an arbitrary point $\mathbf{r}$ produced by a single dipole source at $\mathbf{r}_S$ is given by [11]

$$\mathbf{E}(\mathbf{r}, \mathbf{r}_S) = \frac{ab}{(2\pi)^2} \int_{-\pi/a}^{\pi/a} \int_{-\pi/b}^{\pi/b} \mathbf{E}^\infty(\mathbf{r}, \mathbf{r}_S, \mathbf{k}_t) dk_x dk_y, \quad (2)$$

where $\mathbf{E}^\infty(\mathbf{r}, \mathbf{r}_S, \mathbf{k}_t)$ is the field at $\mathbf{r}$, in the presence of the periodic array of spheres, produced by the periodic dipoles at $\mathbf{r}_{S,mn} = \mathbf{r}_S + \mathbf{p}_{mn}$, with the transverse wavevector $\mathbf{k}_t$, integrated over the Brillouin zone. We assume the time harmonic dependency $\exp(j\omega t)$, which is suppressed here and below.

To solve the problem of periodic excitation we use the formula which relates the dipole moment $\mathbf{p}^\infty$, representing a sphere at the position $\mathbf{r}$, with the *local* electric field $\mathbf{E}^{\text{loc}}(\mathbf{r}, \mathbf{r}_S, \mathbf{k}_t)$ produced by the array of sources $\mathbf{p}_{S,mn}$ plus that of all the metal spherical nanoscatterers except for the reference one [4,5,12,19]

$$\mathbf{p}^\infty = \alpha \mathbf{E}^{\text{loc}}(\mathbf{r}, \mathbf{r}_S, \mathbf{k}_t). \quad (3)$$

Here α is the polarizability of a metal sphere,

$$\alpha = \pi \varepsilon_0 \varepsilon_h \left(\frac{\varepsilon_m + 2\varepsilon_h}{4r_p^3 (\varepsilon_m - \varepsilon_h)} + j \frac{k^3}{6} \right)^{-1}, \quad (4)$$

ε_h is the relative permittivity of the host medium, ε_m is the relative permittivity of metal represented here using the Drude model

$$\varepsilon_m = 1 - \frac{\omega_p^2}{\omega(\omega - j\omega_D)}, \quad (5)$$

where ω_p and ω_D are the plasma and damping radian frequency for the adopted metal, and $k = \sqrt{\varepsilon_h} k_0$, where k_0 is the vacuum wavenumber.

Considering equation (3) for the spheres of each layer at positions $\mathbf{p}_{l00}$, $l=1, \dots, N$, represented by electric dipole moments $\mathbf{p}_l^\infty$, and using the GF representation for $\mathbf{E}^{\text{loc}}(\mathbf{r}, \mathbf{r}_S, \mathbf{k}_t)$ we obtain the following matrix equation:

$$\left(\frac{1}{\alpha} \mathbf{I} - \underline{\mathbf{G}}^\infty(z_l \hat{\mathbf{z}}, z_l \hat{\mathbf{z}}, \mathbf{k}_t) \right) \mathbf{p}_l^\infty - \sum_{\substack{r=1 \\ r \neq l}}^N \underline{\mathbf{G}}^\infty(z_l \hat{\mathbf{z}}, z_r \hat{\mathbf{z}}, \mathbf{k}_t) \mathbf{p}_r^\infty = \underline{\mathbf{G}}^\infty(z_l \hat{\mathbf{z}}, \mathbf{r}_S, \mathbf{k}_t) \mathbf{p}_S, \quad l=1, \dots, N. \quad (6)$$

Here $\underline{\mathbf{G}}^\infty(\mathbf{r}, \mathbf{r}', \mathbf{k}_t)$ is the electric-field dyadic GF for the periodically phased array of dipoles

$$\underline{\mathbf{G}}^\infty(\mathbf{r}, \mathbf{r}', \mathbf{k}_t) = \sum_{m,n=-\infty}^{\infty} \underline{\mathbf{G}}(\mathbf{r}, \mathbf{r}' + \mathbf{p}_{mn}) e^{-j\mathbf{k}_t \cdot \mathbf{p}_{mn}} = \frac{1}{\varepsilon_0 \varepsilon_h} \left[k^2 G^\infty(\mathbf{r}, \mathbf{r}', \mathbf{k}_t) \mathbf{I} + \frac{\partial^2 G^\infty}{\partial \mathbf{r}^2}(\mathbf{r}, \mathbf{r}', \mathbf{k}_t) \right], \quad (7)$$

$\mathbf{I}$ is the identity dyadic, $G^\infty(\mathbf{r}, \mathbf{r}', \mathbf{k}_t)$ is the corresponding 2D periodic scalar GF

$$G^\infty(\mathbf{r}, \mathbf{r}', \mathbf{k}_t) = \sum_{m,n=-\infty}^{\infty} e^{-j\mathbf{k}_t \cdot \mathbf{p}_{mn}} G(\mathbf{r}, \mathbf{r}' + \mathbf{p}_{mn}), \quad (8)$$

and $G(\mathbf{r}, \mathbf{r}')$ is the scalar GF

$$G(\mathbf{r}, \mathbf{r}') = \frac{e^{-jk|\mathbf{r}-\mathbf{r}'|}}{4\pi|\mathbf{r}-\mathbf{r}'|}. \quad (9)$$

The symbol $\partial^2 G^\infty / \partial \mathbf{r}^2$ in (7) is defined as

$$\frac{\partial^2 G^\infty}{\partial \mathbf{r}^2}(\mathbf{r}, \mathbf{r}_S) \cdot \mathbf{p}_S \equiv \nabla \left(\nabla G^\infty(\mathbf{r}, \mathbf{r}_S) \cdot \mathbf{p}_S \right). \quad (10)$$

The function $\tilde{\mathbf{G}}^\infty(\mathbf{r}, \mathbf{r}', \mathbf{k}_t)$ in (6) is the same dyadic GF as in (7) without the $(m, n) = (0, 0)$ term, and thus it is not singular at $\mathbf{r} = \mathbf{r}'$. Therefore, the Mexican hat $(\hat{})$ denotes here the regularized GF defined as $\hat{\mathbf{G}}^\infty(\mathbf{r}, \mathbf{r}', \mathbf{k}_t) \equiv \mathbf{G}^\infty(\mathbf{r}, \mathbf{r}', \mathbf{k}_t) - \mathbf{G}(\mathbf{r}, \mathbf{r}')$.

Once the solutions $\mathbf{p}_l^\infty$, $l = 1, \dots, N$ of equation (6) have been determined, the electric field at an arbitrary position $\mathbf{r}$, produced by the array of exciting dipoles, phased with the transverse wavevector $\mathbf{k}_t$, are then evaluated by

$$\mathbf{E}^\infty(\mathbf{r}, \mathbf{r}_S, \mathbf{k}_t) = \sum_{l=1}^N \hat{\mathbf{G}}^\infty(\mathbf{r}, z_l \hat{\mathbf{z}}, \mathbf{k}_t) \cdot \mathbf{p}_l^\infty + \mathbf{G}^\infty(\mathbf{r}, \mathbf{r}_S, \mathbf{k}_t) \cdot \mathbf{p}_S, \quad (11)$$

The periodic GFs (as well as the regularized periodic ones) were calculated using the Ewald method [20-22], which has exponential convergence and allows calculating the GF for complex wavevectors and frequencies.

3. “INTERFACE” MODES IN PERIODIC ARRAYS OF SILVER NANOSPHERES

Note that the same equation (6) with zero in the right part describes the modes in the array of nanospheres. In Fig.2 the dispersion curves for a single-layer of silver nanospheres are shown ($0 \leq k_x \leq \pi/a$ and $k_y = 0$). The radius of the nanospheres is $r_p = 28$ nm, the periods are $a = b = 65$ nm. The Drude model for silver permittivity in (5) assumes $\omega_p = 2\pi f_p$ with $f_p = 914$ THz. The spheres were considered lossless to find only real modes ($\omega_D = 0$). The relative permittivity of the media hosting the nanospheres is $\varepsilon_h = 1$. The three supported modes correspond to the dipole moments $\mathbf{p}^\infty$ parallel to the x , y and z -axis. The small boxes in Fig.2 represent the regions where the mode polarized along z has a flat dispersion curve. This peculiarity is important to create the sought super resolution, as was already discussed in [4].

Fig. 2. Dispersion curves for the single-layer array of silver nanospheres.

4. LOCAL FIELD ENHANCEMENT

We show here the possibility to obtain the local field enhancement by arrays of silver nanospheres. In all cases the source is situated at the coordinate origin, its dipole moment is directed along the z -axis and its magnitude is chosen as $p_{S,z} = 10^{-30} \text{ C}\cdot\text{m}$. The first layer is placed at $z_1 = a = b = 65 \text{ nm}$. In the case of a multi-layer array of nanospheres, the inter-layer distance is $h = 2a$. The relative permittivity of the media hosting the nanospheres is $\epsilon_h = 1$. The field is calculated in the observation plane $z_{\text{obs}} = 2Na$, at the distance a above the last layer.

The radius of nanospheres is $r_p = 65 \text{ nm}$. The values of $\omega_p = 2\pi f_p$ and $\omega_D = 2\pi f_D$ for silver, necessary for the calculation of ϵ_m in (5), are assumed as in [5] where $f_p = 914 \text{ THz}$ and $f_D = 5.13 \text{ THz}$.

In Fig. 3 the local field (or near-field) enhancement

$$\left| E_z(\mathbf{r}_{\text{obs}}) \right|^2 / \left| E_z^{\text{inc}}(\mathbf{r}_{\text{obs}}) \right|^2, \quad (12)$$

defined as the ratio between the field intensity with and without the array at the position $\mathbf{r}_{\text{obs}} = 2Na\hat{\mathbf{z}}$, is shown versus frequency for different numbers of layers $N = 1, 2, 3, 4$. The field without the array corresponds to the field E_z^{inc} of an isolated dipole. The near-field enhancement is observed in a wide frequency range for any number of layers, and increases when increasing the number of layers.

Fig. 3. Local-field enhancement for different numbers of array layers, defined as the ratio of the field intensity with and without the arrays.

The most significant maxima of field enhancement for the double-layer structure occurs at frequencies $f = 509.5\text{THz}$, $f = 537.5\text{THz}$, $f = 569\text{THz}$. The frequencies relative to the strongest local field enhancement in Fig. 3 correspond to the flat parts of the dispersion curves in Fig. 2. They alternate with the minima at frequencies $f = 516.5\text{THz}$, $f = 553.25\text{THz}$. The flatter the part of the dispersion curve of the excited modes (Figs.2) the wider the part of the evanescent spatial spectrum, which is reconstructed at the observation point and at a specified frequency, due to the resonant array. The presence of the array results in the excitation of a strong field in the vicinity of the layers since several spectral components (waves with different wavenumbers) are excited by the source and are able to transmit their evanescent field to the observation point. The field enhancement is particularly strong for a large number of layers. Therefore the device made by layers of nanospheres can be used to amplify certain evanescent field components, with respect to what happens in a homogeneous medium.

5. FIELD ACROSS LAYERS

To gain deeper understanding of the physical mechanism we calculated the field across layers (Figs. 4-6). We considered the frequencies of maximum local field enhancement for double-layer array. Nevertheless Fig. 3 shows that these frequencies are close to the maximum of the local field enhancement for any number of layers. The plots (a) and (b) represent the x - and z - components, respectively, of the E field along the z -axis with $x = a/2$ and $y = 0$ (in all cases considered the field component E_y is vanishing for symmetry reasons). The plot (c) represents the z -component of the field along the z -line with $x = 0$, $y = 0$ (there, both E_x and E_y vanish, and the field has singularities at the positions of the source and spheres). The vertical dash-dot lines indicate the z -positions of the layers.

Figures 4-6 show that the field decays away from each layer but it is able to strongly excite the subsequent resonant layer. The maintenance of the evanescent field across the layers can be interpreted as an “amplification” when compared to the field without layers (the incident field).

Fig. 4. Field distributions across layers for $f = 509.5$ THz .

Fig. 5. Field distributions across layers for $f = 537.5$ THz .

Fig. 6. Field distributions across layers for $f = 569$ THz .

6. SUBWAVELENGTH FIELD CONCENTRATION

Referring to the geometry in Fig. 1b, Fig. 7 shows the field distributions along the line $x = 0$, in the image plane $z_{obs} = 2Na$, for the case of double-layer array ($N = 2$) and at the indicated frequencies. The half-power width of the “image” is marked by arrows. These results clearly show that subwavelength field concentration (“focusing”) is achieved for the frequencies corresponding to the maximum local field enhancement. Since the field across the layers is made of the evanescent part of the excited spectrum, it has large transverse wavenumber components and it is thus able to reconstruct the subwavelength field localization far from the source. For all three field enhancement maxima in Fig. 7, the half-power width of the “image” is much less than a quarter of wavelength.

Fig.7. Field distributions along the line $x = 0$ in the image plane $z_{obs} = 2Na$ for the double-layered structure ($N=2$) at the frequencies of maximum and minimum field enhancement (see Fig. 3).

7. CONCLUSION

The local field enhancement of infinite arrays of metallic nanospheres are investigated at optical frequencies, for a dipole source oriented orthogonally to the array plane. Earlier, similar investigations were performed for arrays of finite extent and only a few features were stressed. The main advantages of the present formulation are that certain physical phenomena are not hindered by truncation effects, and the possibility to use more efficient algorithms to numerically facilitate the investigation of multilayer arrays, based on the array scanning method (ASM) combined with the Ewald method.

When the stacked layers are excited by a vertical dipole, we have shown the mechanism of the evanescent field “transportation” on the other side of the layers (resonance excitation). It has been shown that the local field enhancement is a general property of the arrays of nanoparticles supporting a mode. It is observed in a wide frequency range, but the

extreme values correspond to the flattest parts of the dispersion curve. We have also investigated the occurrence of the subwavelength field concentration on the other side of the layers.

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