Generalized Lorentz-Lorenz Method for the Retrieval of Plasmonic Nanocluster Metamaterial Effective Parameters

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***Abstract*— Generalized Lorentz-Lorenz formulas are developed for the effective parameters of metamaterials composed of a periodic arrangement of plasmonic nanoclusters (NCs). Each NC exhibits both electric and magnetic polarizabilities but its dimension is in general not very subwavelength. The obtained formulas for the metamaterial effective electric and magnetic characteristics duly consider both electric and magnetic polarizabilities and naturally describe the effects of frequency and spatial dispersion. Homogenization results for a material considered here show the effects of spatial dispersion near the fundamental electric resonance.**

Index Terms—Metamaterials; constitutive relations; scattering; electromagnetic multipoles.

I. INTRODUCTION

Metamaterials and plasmonics based on micro- and nano-structured metallic-dielectric composites are bringing an important revolution to the microwave and optics fields. In fact, metamaterials enable the realization of novel physical properties that are unattainable from natural materials. Examples of unconventional EM behaviors in such advanced materials include isotropic negative refraction, slow light, near-field enhancement with controlled polarization, as well as EM focusing and energy transfer that beats the diffraction limit. Such exotic artificial composite structures owe their peculiar properties both to the constituent materials which comprise their elementary building blocks and to their specific spatial arrangement.

Microwave and optical metamaterials rely on our understanding of EM-radiation-matter interaction, and the use of homogenization methods can provide a convenient characterization of such an interaction by describing metamaterials as bulk homogeneous materials with effective parameters that take into account their inherent qualities and complex nature, similarly to what is done for natural materials. While the concept of homogenization theory is easily being applied to the asymptotic long-wavelength limit, e.g. the microwave regime where true sub-wavelength structures may be fabricated with ease, the optical regime challenges the underlying hypothesis of a true subwavelength unit cell. Indeed, for artificial materials the size of the lattice constant is typically only moderately smaller than the wavelength of light, in contrast to natural materials where the wavelength-to-lattice ratio is several orders of magnitude larger, even at optical frequencies. As a consequence, metamaterials can be characterized by nonnegligible spatial dispersion effects [1].

A general approach to homogenize nonmagnetic periodic metamaterials has been recently introduced [2] that is capable of providing a comprehensive description of both spatial and frequency dispersion phenomena. The above homogenization formalism has been subsequently applied to derive a generalization of the classical Lorentz-Lorenz formulas [3] for a dielectric crystal comprising one particle per unit cell, in the hypothesis that particle interaction can be described by the dipolar terms only [4].

The objective of this work is to extend the generalized Lorentz-Lorenz (GLL) method proposed in [4] to the case when the unit cell contains more than one inclusion, for application to the homogenization of a class of metamaterials formed by periodic arrangements of spherical plasmonic nanoclusters (NCs). Clusters of self-assembled plasmonic nanoparticles have been proposed as building blocks for new magnetic and negative index materials (NIMs) at optical frequencies [5]-[7]. Indeed, in this type of structures, plasmonic particles are arranged to force the electric field to circulate in the plane orthogonal to the incident magnetic field, inducing an overall magnetic resonance that coexists with the individual electric resonance supported by each constituent nanoparticle. In particular, spherical NCs formed by a number of silver nanocolloids enclosed within a thin dielectric shell and attached to a dielectric core of variable size can provide isotropic electric and magnetic resonances in three-dimensions (3D) [7]. At first, we compute the electric and magnetic responses of NC metamaterials by considering each NC as a single inclusion, whose polarizbilities are numerically retrieved from the multipolar expansion of its scattered field, and using the GLL formulas developed in [4] valid when each unit cell contains a single inclusion per unit cell. GLL formulas valid for multiple inclusions in the unit cell of a 3D periodic array, rigorously taking into account interaction among the constituent particles within the unit cell and across the array, will be presented in the extended abstract along with relevant homogenization results for sample NC metamaterials.

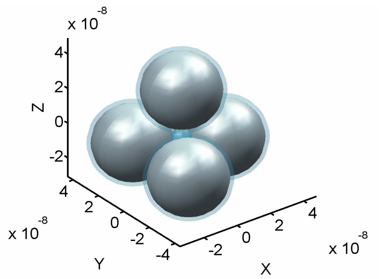
II. Homogenization of Nanocluster Metamaterials

The approach of determining the effective parameters of NC metamaterials by considering each NC as a single inclusion and using the GLL formulas in [4] is illustrated in the following by application to the two sample tetrahedral and icosahedral NC configurations shown in Fig. 1(a) and Fig. 3(a), respectively.

The permittivity of the constituent nanoparticles is estimated by the Drude model $\varepsilon_m = \varepsilon_\infty - \omega_p^2 / [\omega(\omega + i\gamma)]$, where the parameters for silver are assumed as: $\varepsilon_m = 5$ $\omega_p = 1.37 \times 10^{16}$ rad/s, $\gamma = 27.3 \times 10^{12}$ rad/s. Both considered NCs are immersed in a medium with relative permittivity $\varepsilon_h =$ 2.2, and their polarizabilities are determined in terms of the scattering coefficients of the multipolar expansion of the scattered field through the procedure outlined in [7]. As a consequence of substantial symmetry and subwavelength dimension of the NCs, the magneto-electric terms are several orders of magnitude smaller than the electric and magnetic polarizabilities and can be neglected. It is therefore assumed that the NC response is nonbianisotropic, and that the relations between the local fields and the NC induced dipole moments are

$$\boldsymbol{p}_e = \varepsilon_0 \varepsilon_h \overline{\overline{\boldsymbol{\alpha}}}_{ee} \boldsymbol{E}_{loc}, \quad \boldsymbol{p}_m = \overline{\overline{\boldsymbol{\alpha}}}_{mm} \boldsymbol{H}_{loc} \tag{1}$$

Under these assumptions, the dielectric and magnetic functions of the metamaterial can be obtained by the GLL formulas [4]



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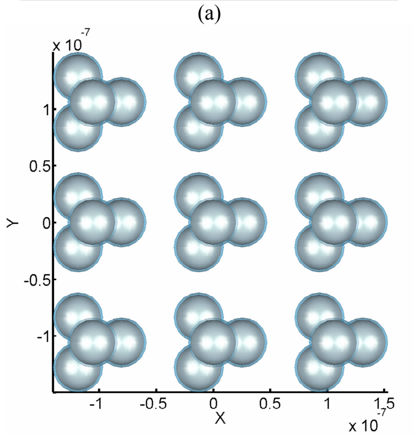
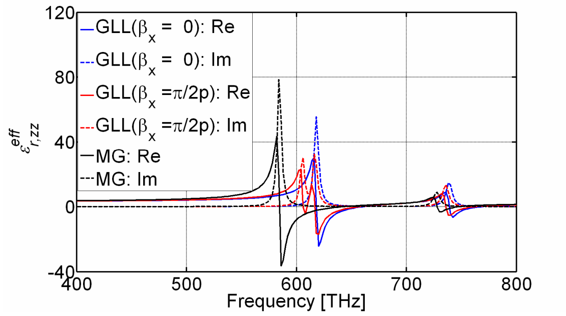


(b)

Figure 1. (a) Tetrahedral NC formed by 4 solid silver nanospheres of radius a = 20 nm arranged around a central dielectric particle with radius of 4.9 nm with a surface-to-surface separation between particles of 4 nm. The overall size of the NC is $D = 98$ nm. (b) Simple cubic lattice of tetraahedral NCs. The lattice period is p = 106 nm.



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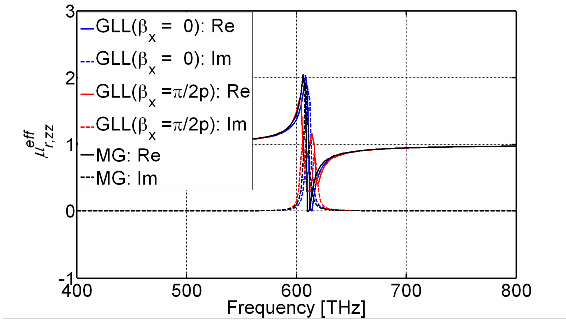


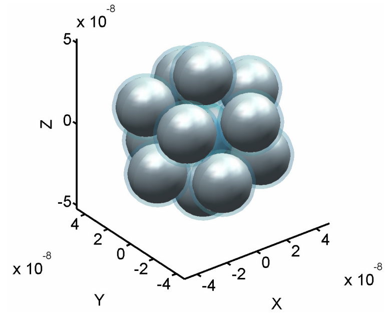
(b)

Figure 2. (a) Effective permittivity and (b) permeability of the tetrahedral NC composite material from Fig. 1 for wave vector $\boldsymbol{\beta} = \beta_x \hat{\boldsymbol{x}}$, with variable $\beta_x = 0, \pi/2$.

$$\begin{aligned} \overline{\overline{\boldsymbol{\varepsilon}}}_r^{eff} &= \varepsilon_h \overline{\overline{\boldsymbol{I}}} + \varepsilon_h \left(V_{cell} \overline{\overline{\boldsymbol{M}}}_{mm} \right)^{-1} \overline{\overline{\boldsymbol{\alpha}}}_{ee} \\ \overline{\overline{\boldsymbol{\mu}}}_r^{eff} &= \overline{\overline{\boldsymbol{I}}} + \left(V_{cell} \overline{\overline{\boldsymbol{M}}}_{ee} \right)^{-1} \overline{\overline{\boldsymbol{\alpha}}}_{mm} \end{aligned} \tag{2}$$

where $\overline{\overline{\boldsymbol{M}}}_{ee,mm} = \overline{\overline{\boldsymbol{A}}}_{mm,ee} - \overline{\overline{\boldsymbol{A}}}_{me,em} \left[\overline{\overline{\boldsymbol{A}}}_{ee,mm} \right]^{-1} \overline{\overline{\boldsymbol{A}}}_{em,me}$, $\overline{\overline{\boldsymbol{A}}}_{ee,mm} = \overline{\overline{\boldsymbol{I}}} - \overline{\overline{\boldsymbol{\alpha}}}_{ee,mm} \overline{\overline{\boldsymbol{C}}}_{\text{int}}$, $\overline{\overline{\boldsymbol{A}}}_{em} = -\zeta_h \overline{\overline{\boldsymbol{\alpha}}}_{ee} \overline{\overline{\boldsymbol{C}}}_{e,m}$, $\overline{\overline{\boldsymbol{A}}}_{me} = (\zeta_h)^{-1} \overline{\overline{\boldsymbol{\alpha}}}_{mm} \overline{\overline{\boldsymbol{C}}}_{e,m}$, ζ_h denotes the host medium intrinsic impedance, and $\overline{\overline{\boldsymbol{C}}}_{\text{int}}$ and $\overline{\overline{\boldsymbol{C}}}_{e,m}$ are the interaction dyadics introduced in [4].

In Fig. 2 are plotted the zz components of the effective permittivity and permeability of the composite material formed by arranging the tetrahedral NCs from Fig. 1(a) on a simple cubic lattice with period $p = 106$ nm (Fig. 1(b)). The propagation is along the x axis, i.e. the wave vector is $\boldsymbol{\beta} = \beta_x \hat{x}$, and the effective parameters are computed by the GLL formulas in (2) for $\beta_x = 0,\ \pi/2$. The corresponding results obtained by Maxwell Garnett (MG) homogenization formulas are also shown in Fig. 2 for the purpose of comparison. It is observed that the effective permittivity predicted by the GLL method exhibits a noticeable dependence on β_x near the electric resonance, and that the behaviour of both the electric and magnetic responses markedly changes at variable β_x values. Such spatial dispersion effects are not taken into account by the MG theory.



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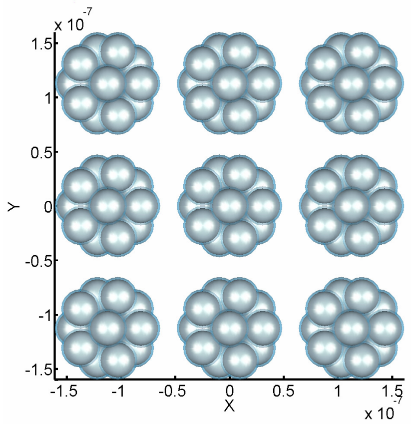
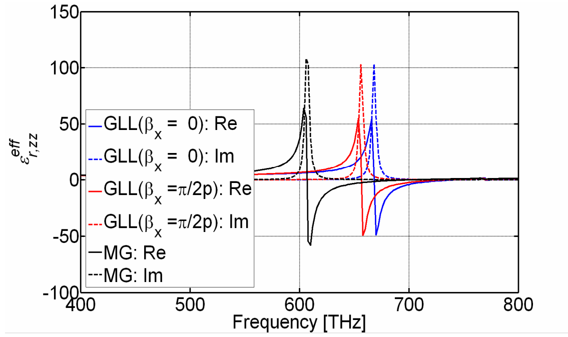


(b)

Figure 3. (a) Icosahedral NC formed by 12 solid silver nanospheres of radius a = 16 nm arranged around a central dielectric particle with radius of 16.2 nm with a surface-to-surface separation between particles of 4 nm. The overall size of the NC is $D = 104.5$ nm. (b) Simple cubic lattice of icosahedral NCs. The lattice period is p = 112.5 nm.

Results for the effective permittivity and permeability of the composite material formed by the periodical arrangement of the icosahedral NCs depicted in Fig. 3(b) are shown in Fig. 4. The period of the simple cubic metamaterial lattice is $p = 112.5$ nm. The wave vector is assumed to be $\boldsymbol{\beta} = \beta_x \hat{x}$, and the effective parameters computed by the GLL formulas in (2) for $\beta_x = 0,\ \pi/2$ are compared with MG results. Also this metamaterial composite exhibits spatial dispersion effects near the electric resonance the position of which varies for variable β_x values.



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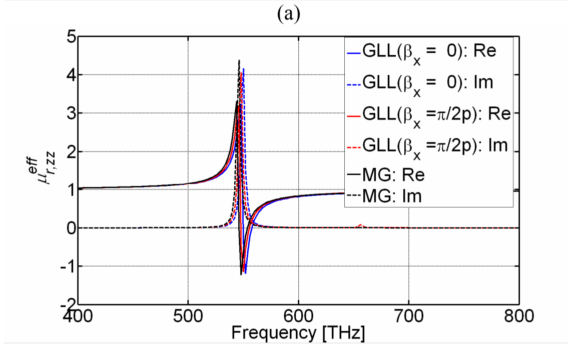

(b)

Figure 4. (a) Effective permittivity and (b) permeability of the icosahedral NC composite material from Fig. 3 for wave vector $\boldsymbol{\beta} = \beta_x \hat{\boldsymbol{x}}$, with variable $\beta_x = 0, \pi/2$.

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