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EM Characterization of Raspberry-Like Nanocluster Metamaterials

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Introduction

Modern nanochemistry has developed efficient techniques to manipulate nanoscale objects with a highly advanced degree of control. Chemically-engineered nanoparticles can be synthesized with a large choice of sizes, shapes, constituent materials and surface coatings, and further assembled spatially into self-assembled structures, either spontaneously or in a directed manner [1]. Advances in particle self-assembly and the quasi unlimited range of nanostructures with controlled architectures and functions available suggest that such assemblies may also provide a simple route to metamaterials at infrared and visible length scales. Indeed, nanochemistry and self-assembly strategies are able to inexpensively produce materials whose inner structure is natively in the right range of sizes for optical and infrared applications and can provide fully three-dimensional (3D) structures, thus opening the way to the fabrication of 3D-metamaterial samples of finite volume of the highest importance to many applications. Such metamaterials may be used, for example, to create 3D homogeneous, isotropic negative index materials (NIMs), with simultaneously negative permittivity and magnetic permeability, cloaking devices or light-based circuits manipulating local optical electric fields rather than the flow of electrons.

In this work we investigate certain EM properties of metamaterials formed by densely arrayed clusters of plasmonic nanoparticles, which will be referred to as nanoclusters. Nanoclusters are formed by a number of metal nanocolloids (possibly enclosed within a dielectric shell) attached to a dielectric core, as in the examples shown in Fig. 1, and can be easily realized and assembled by current state-of-the-art nanochemistry techniques. Such a kind of structure generalizes the concept of nanorings originally proposed in [2] to realize a magnetic media at visible frequencies and has been recently shown in [3] to have the potential of providing resonant isotropic optical magnetism. An approximate model based on the single dipole approach (SDA) [4], [5] in conjunction with the multipole expansion [6] of the scattered field is used here to evaluate the electric and magnetic polarizabilities of the nanocluster. Then, the permittivity and permeability of the composite medium are estimated by the Maxwell Garnett homogenization model. Results obtained by this approximate method will be compared with data from full-wave simulations, focusing on the characterization of the nanocluster resonant isotropic electric and magnetic responses to an

incident wave field, and the possibility to realize an isotropic NIM at optical frequencies.

Nanocluster Modelling

The colloidal silver particles are evenly distributed around the dielectric core to obtain a compact ensemble. Namely, the positions of the colloids is derived by maximizing the minimum distance between them, that can be reformulated as a minimization problem of the potential energy of N particles on the surface of a sphere of unit radius [7]. It is noted that certain minimum configurations achieved by this approach coincide with those of the Platonic solids consisting of equilateral triangles or some Archimedean semiregular polyhedra and their duals. Two examples of nanoclusters are illustrated in Fig. 1. The configuration shown in Fig. 1(a) consists of 12 silver nanocolloids of radius 17 nm covering a silica particle of radius 15.3 nm, whereas the nanocluster in Fig. 1(b) comprises 96 silver nanoparticles of radius 8 nm on a 34.8-nm-radius silica particle. The overall dimension of the nanoclusters D_t (i.e., the overall diameter of the clusters) are very similar: $D_t = 98.7$ nm and $D_t = 101.7$ nm for the 12 and 96 particle nanoclusters, respectively.

The geometrical symmetry and the small optical size of clusters sketched in Fig. 1 ensure isotropic electric and magnetic-dipole responses. At one frequency of the incident light, the induced overall (equivalent) electric dipole of the nanocluster dominates when the polarization of the colloidal particles is mainly parallel to the electric field of the linearly polarized incident light. At a different frequency the induced magnetic dipole dominates, and the polarization of colloidal particles is mainly azimuthal (rotationally symmetric) with respect to the direction of the incident magnetic field. In other words, the applied magnetic field forms, like in [2], effective polarization nanorings around the silica core. Both *electric* and *magnetic* resonant frequencies originate from the combination of plasmonic resonances of the individual colloidal nanospheres.

As customary, in the context of the SDA model [4], [5], the metallic nanospheres are simply characterized by their induced electric dipoles. For the polarizability of each single nanosphere we use the following quasistatic expression:

$$\alpha = 4\pi\epsilon_0\epsilon_h \frac{\alpha_{LL}}{1 - i(2k^3/3)\alpha_{LL}}, \quad \alpha_{LL} = a^3 \frac{\epsilon_m - \epsilon_h}{\epsilon_m + 2\epsilon_h} \quad (1)$$

where a is the radius of the nanosphere and ϵ_m and ϵ_h are the relative permittivity of the metal and of the host medium, respectively. The relative permittivity of metal is predicted by the Drude model

$$\epsilon_m = \epsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\gamma)} \quad (2)$$

where ω_p is the plasma radian frequency and γ is the damping frequency. The time harmonic convention $\exp(-i\omega t)$ is assumed here. In accordance with [2], for silver we assume $\epsilon_\infty = 5$, $\omega_p = 1.37 \times 10^{16}$ rad/s, and $\gamma = 27.3 \times 10^{12}$ s⁻¹.

Fig. 1. Two examples of metal nanoclusters with different number and size of nanocolloids evenly distributed over a dielectric core: (a) cluster made of 12 nanocolloids of radius 17 nm each; (b) cluster of 96 nanocolloids of radius 8 nm each. Coordinate units are meters

Homogenization and Effective Material Parameters

The equivalent electric and magnetic dipoles associated to the nanocluster are determined by the multipole expansion of the scattered field [6] and used to derive the electric and magnetic polarizabilities. Then, the effective permittivity and permeability of an array of nanoclusters are obtained by the Maxwell Garnett homogenization model. These parameters are plotted in Figs. 2(a-b) and 2(c-d) for a cubic lattice of the nanoclusters shown in Fig. 1(a) and 1(b), respectively, placed in a host matrix with $\epsilon_h = 2.2$. The number density of nanoclusters per unit volume is $N_d = (D_t)^{-3}$. The effective parameters are estimated at three different field polarizations and propagation directions of the illuminating wave, i.e., $\epsilon_{xx}^{eff} = \epsilon_{xx}^{eff}(\hat{\mathbf{k}}_i = -\hat{\mathbf{z}})$, $\epsilon_{yy}^{eff} = \epsilon_{yy}^{eff}(\hat{\mathbf{k}}_i = -\hat{\mathbf{x}})$, $\epsilon_{zz}^{eff} = \epsilon_{zz}^{eff}(\hat{\mathbf{k}}_i = -\hat{\mathbf{y}})$, and correspondingly $\mu_{xx}^{eff} = \mu_{xx}^{eff}(\hat{\mathbf{k}}_i = -\hat{\mathbf{y}})$, $\mu_{yy}^{eff} = \mu_{yy}^{eff}(\hat{\mathbf{k}}_i = -\hat{\mathbf{z}})$, $\mu_{zz}^{eff} = \mu_{zz}^{eff}(\hat{\mathbf{k}}_i = -\hat{\mathbf{x}})$, where the unit vector $\hat{\mathbf{k}}_i$ denotes the incident plane wave propagation direction. Not surprisingly, the electric and magnetic responses of the material barely depend on the incidence wave direction and polarization. Indeed, the electromagnetic isotropy of the clusters comes out from their symmetry (see Fig. 1), whereas their compact sizes ($< \lambda/4$) results in negligible spatial dispersion. A wide band permittivity resonance and a narrower magnetic activity are observed in Fig. 2. The regions where permeability is negative are accompanied by high losses. However, there are frequency regions where permeability significantly differs from unity with limited losses. This suggests a clear and economical way to envisage artificial materials that exhibit magnetism at optical frequency.

Fig. 2. Effective permittivity and permeability for a cubic lattice with number density $N_d = (D_t)^{-3}$ of the nanoclusters from Fig. 1 in a host matrix with $\epsilon_h = 2.2$. (a), (b) refer to the cluster in Fig. 1(a); (c), (d) refer to the cluster in Fig. 1(b).

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