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#### 3D Isotropic Magnetic and Negative Index Nanocluster Metamaterials

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#### Abstract

We investigate the optical properties of metamaterials formed by collections of three-dimensional clusters of plasmonic nanoparticles. This type of nanoclusters (NCs) consist of spherical constellations of metallic nanospheres arranged around a central dielectric sphere and support collective plasmon modes among which the most relevant are those associated with the induced electric and magnetic dipole resonances. We discuss the effective electric and magnetic polarizabilities of NCs, emphasizing their isotropic properties, and we show that dense packed arrays of NCs can be used to realize metamaterials with negative effective permittivity and permeability, and even negative effective refractive index at visible frequencies.

### 1. Introduction

Modern nanochemistry and self-assembly techniques are able to inexpensively produce fully three-dimensional (3D) metamaterials for optical and infrared applications [1]. Among several types of chemically synthesized nanostructures, 2D and 3D clusters of self-assembled plasmonic particles are increasingly attracting interest for their use as building blocks for new 3D homogeneous magnetic materials, negative index materials (NIMs) [2-6], cloaking devices or light-based circuits manipulating local optical electric fields rather than the flow of electrons. In fact, in these kinds of structures plasmonic particles, that do not directly interact with an incident magnetic field, can be arranged to force the electric field to circulate in the plane orthogonal to an incident magnetic field, inducing an overall magnetic resonance [2]. Besides, such a magnetic-based plasmonic resonance coexists with the individual electric resonance supported by each of the nanoparticles composing a cluster.

In this work we analyze the optical properties of metamaterials formed by close-packed arrangements of plasmonic nanoclusters (NCs). Similarly to the structures considered in [4], such NCs are formed by a number of metal nanocolloids (possibly enclosed within a dielectric shell) attached to a dielectric core of variable size, as in the examples shown in Fig. 1, and can be easily realized and assembled by current state-of-the-art nanochemistry techniques. An approximate model based on the single dipole approximation (SDA) [7], in conjunction with the multipole expansion of the scattered field is used here to evaluate the electric and magnetic polarizabilities of a few sample NC configurations. In order to obtain a highly isotropic behaviour of the NCs, a most-regular-disposition criterion is applied to locate the particles around the central cores, leading to completely regular or pseudo-regular NCs, depending on the chosen particle number. The resonances (in particular the electric and magnetic dipolar ones) of this kind of structures can be tuned by varying the number of particles, their separation, and the permittivity of the host material. We show that cluster geometries can be engineered to exhibit electric and magnetic resonances in the same frequency range, which opens the possibility to realize an isotropic negative index material at optical frequencies.

### 2. Nanocluster Modeling

NCs are made by spherical constellations of metallic nanospheres evenly distributed around a dielectric or (dielectric coated) metallic core to obtain a compact and regular ensemble. Compactness of NCs is instrumental in minimizing spatial dispersion effects, and therefore particles are closely packed around the central core. As a consequence, the dimension of the silver nanoparticles determines the maximum number of nanoparticles that can be clustered around the core. A sample NC comprising 48 silver nanoparticles is illustrated in Fig. 1. Interparticle spacing (d) can be controlled by coating the particles with polymer shells, as shown in Fig. 1.

There are two main resonant modes of the NCs we are interested in, namely the electric and magnetic dipole resonances [2, 4]. At the electric resonance, the NC overall induced electric dipole dominates, as a result of the polarization of the silver nanoparticles being mainly parallel to the incident electric field. At the magnetic resonance, the induced magnetic field. In other words, the applied magnetic field forms, like in [2], effective polarization nanorings around the silica core, that are equivalently seen as effective magnetic dipoles. Both electric and magnetic resonances of the NC originate from the collective plasmonic resonances.



Figure 1. (a) Sample NC geometry comprising 48 metallic nanoparticles, and (b) sketch of a NC metamaterial with simple cubic lattice.

## 3. Effective Permittivity and Permeability of Nanocluster Metamaterials

In Fig. 2 we show the effective permittivity and permeability of a composite formed by a periodical arrangement of the 48-element NC shown in Fig. 1. The material effective parameters, estimated by the Maxwell Garnett homogenization model, are plotted for two filling fraction values f = 0.52 and f = 0.74, corresponding to close-packed simple cubic and face-centered-cubic (fcc) lattice, respectively. The magnetic permeability exhibits a resonance at a frequency slightly below the resonance of the effective permittivity. As apparent, the strong electric resonance provides very large positive and negative values of the permittivity also for the lattice with lower cluster concentration (simple cubic lattice). Though absorption losses are significant at the electric resonance, as the large imaginary part of the effective permittivity can be exploited with negligible losses. The magnetic resonance is weaker than the electric one, but strong enough to make the permeability reach negative values, even for the lower filling fraction lattice. For the considered NC the magnetic resonance does not fall within the negative region of the effective permittivity, so that a negative index behavior is not achieved. However, tuning of resonances can be achieved by changing the separation between the particles and the dielectric environment of the cluster, or using different cluster geometries and more complex individual particles.



Figure 2. (a) Permittivity and (b) permeability of a close-packed 3D periodic array of the 48-element NC in Fig. 1 for simple cubic (f = 0.52, black lines) and fcc (f = 0.74, red lines) lattices. Real and imaginary parts of the parameters are shown in solid and dashed lines, respectively.

#### 5. Conclusion

We have examined the optical properties of 3D metamaterials formed by close-packed arrangements of plasmonic NCs formed by a number of core-shell metal-dielectric nanocolloids attached to a central dielectric core, that can be easily realized and assembled by current state-of-the-art nanochemistry techniques. A most-regular-disposition criterion is applied to locate the particles around the central cores in order to obtain highly isotropic behavior of the NCs. By combining a SDA with the multipole expansion of the scattered field we have shown that 3D nanoclusters are useful for creating isotropic electric and magnetic activity which can be tuned and even superimposed by proper design of the NC. Such electric and magnetic activities are inherited by the composite material created by packing NCs.

### 6. Acknowledgments

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