

Homogenization of Plasmonic Nanocluster Metamaterials

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Metamaterials and plasmonics based on micro- and nanostructured metallic-dielectric composites are bringing an important revolution to the microwave and optics fields due to their potential for enabling the realization of novel physical properties unattainable from natural materials, such as isotropic negative refraction, slow light, near-field enhancement, as well as EM focusing and energy transfer beyond the diffraction limit. Such artificial composite structures owe their peculiar properties both to the constituent materials which comprise their elementary building blocks and to their specific spatial arrangement. The use of homogenization methods can provide a convenient characterization of EM-wave-matter interaction by describing metamaterials as bulk homogeneous materials with effective parameters that take into account their inherent qualities and complex nature. While the concept of homogenization theory is easily applied to the long-wavelength limit, e.g. the microwave regime where true sub-wavelength structures can be fabricated, 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. As a consequence, metamaterials can be characterized by nonnegligible spatial dispersion effects.

A general approach to homogenize nonmagnetic periodic metamaterials capable of providing a comprehensive description of both spatial and frequency dispersion phenomena has been recently introduced (M. G. Silveirinha, *Phys. Rev. B*, 75, 115104, 2007). This homogenization formalism has been subsequently applied to derive generalized Lorentz-Lorenz (GLL) formulas for the effective parameters of a dielectric crystal with a single inclusion per unit cell, provided that particle interaction can be described by the dipolar terms only (M. G. Silveirinha, *Phys. Rev. B*, 76, 245117, 2007).

The objective of this work is to extend the aforementioned GLL method 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). 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 and have been proposed as building blocks for new magnetic and negative index materials at optical frequencies (A. Vallecchi *et al.*, *Opt. Express* 19, 2754-2772, 2011). While the electric and magnetic responses of NC metamaterials can be computed by considering each NC as a single inclusion with given polarizabilities and using the GLL formulas valid when each unit cell contains a single inclusion per unit cell, it is expected that a more accurate description can be accomplished by 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.