Influence of randomness on the effective dielectric function of metal-dielectric composites for metamaterial purposes

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Abstract

The effective dielectric function of metal-dielectric composites consisting of spherical particles randomly distributed in a matrix is numerically investigated. The study, based on the spectral density theory, shows that local field fluctuations may critically affect the composite performance for metamaterial applications. It appears that randomness significantly modifies the effective dielectric function of composites consisting of metal inclusions embedded in dielectric matrix. The effect is less remarkable for dielectric inclusions in a metal matrix.

1. Introduction

Metal-dielectric composites consisting of ordered ensembles of isolated spherical particles embedded in a matrix have been focus of renewed interest due to their potential for metamaterial applications [1-4]. Less attention has been paid to random systems although they may be more suitable for mass production and low cost metamaterial fabrication [5]. Actually, the influence of randomness has been shown to critically affect metamaterial performance [2, 5]. In this work we analyze the effective dielectric function of composites consisting of randomly distributed and equally sized spheres embedded in a matrix. The analysis is performed in the general framework of the spectral density theory and is applied to the case of i) metal particles in dielectric matrix and ii) dielectric particles in metal matrix.

2. Spectral representation of effective dielectric function of composites

A general description of the effective dielectric function (ε_{eff}) of composites consisting of a matrix with dielectric function ε_1 and embedded inclusions with dielectric function ε_2 is given by the spectral density theory [6]. This theory states that ε_{eff} has the following integral representation:

$$\varepsilon_{eff} = \varepsilon_1 \left(1 - p \int_0^1 \frac{g \ u, p}{t - u} \, du \right) \tag{1}$$

where *u* is an integral variable, $t = \varepsilon_1 / (\varepsilon_1 - \varepsilon_2)$, *p* is the filing fraction of inclusions in the composite and g(u, p) is the so-called spectral density function. The above expression, known as Bergman representation, presents ε_{eff} as a sum of poles that can be identified as resonances related to the topology of the system. These resonances appear for values of the parameter *t* between 0 and 1 and have a weight given by g(u, p). The spectral density function depends uniquely on the geometrical arrangement of the composite. Therefore the Bergman representation explicitly separates the effects of the composite topology through g(u, p) and the effects of the specific dielectric functions of the components through the parameter *t*.

For a given topological arrangement of the composite, it is possible to calculate g(u, p). For the case of a composite consisting of a monodisperse distribution of polarizable spheres randomly located in a matrix, a numerical procedure was proposed in reference [7]. The approach basically consists on aver-

aging g(u, p) of a large number of random systems generated by Monte-Carlo simulations. For each system, g(u, p) is found by imposing periodic boundary condition on the Monte-Carlo-simulated unit random cell and accounting for the electromagnetic coupling among particles. Fig 1 (left) shows the spectral density of random systems at different filling fractions of inclusions, computed as described above and taking into account the dipolar coupling among particles. The spectral density function for an ordered system of particles arranged in single cubic cell is given by a delta function located at u = (1-p)/3. The finite width of the spectral density function for the case of random system is due to the local field fluctuations resulting that resonances of the system occur for a range of *t* values rather than for a single *t* value as in the case of an ordered system.



Fig. 1: Spectral density of disordered system of particles at different filling fraction p (left). Real and imaginary part of the parameter t for SiO₂-Ag composites (right).

According to Bergman representation (equation 1), it appears that the closer the values of t to the real interval [0, 1] the stronger the influence of the composite topology on the effective dielectric function. This is actually the case of many metal-dielectric composites as shown in Fig. 1 (right) for SiO₂-Ag composites in the optical range

3. Application to metal-dielectric composites

The effective dielectric function for a system consisting of Ag particles with p = 1/3 and embedded in a SiO₂ matrix is shown in Fig.2 for both, ordered and disordered systems. The ordered system is characterized by a narrow surface plasmon resonance while for the disordered system the resonance is much broader and red-shifted, due to the finite width of g(u, p) (Fig.1). The broadening results in lower quality factors (Re(ε_{eff})/Im(ε_{eff})) for many plasmonic applications and prevents the composite to achieve very large large negative values of ε_{eff} . However, ordered systems can achieve very large negative values of ε_{eff} , enabling those systems to be part of negative refractive index structures [3].





The situation for a metal matrix with dielectric inclusions is shown in Fig.3. In this case, the discrepancies between the ordered and disordered systems are limited to a much narrower spectral range. The reason is that the range of values of t where the spectral density function is non-zero affects a shorter spectral range than for the metal-in-dielectric case, as evidenced in Fig.1 (right). Thus, incorporation of dielectric inclusions in metal matrix allows tuning of the metal-like effective dielectric function, what can be useful for tunable superlens and negative -index structures [8]. The present simulations suggest that randomness on the spatial distribution of inclusions virtually does not affect the tuning potential.



Fig. 3: Real (left) and imaginary (right) part of the effective dielectric function for systems consisting of ordered and disordered SiO₂ particles embedded in a Ag matrix with a filling fraction of particles p = 1/3.

4. Conclusion

The influence of randomness on the effective dielectric behaviour of systems of particles embedded in a matrix has been studied through numerical simulations of the spectral density function. It was found that randomness affects the composite properties in a much wider range for metal-in-dielectric systems that dielectric-in-metal systems.

References

- [1] I. Romero and F. J. García de Abajo, Anisotropy and particle size-effects in nanostructured plasmonic metamaterials, *Optics Express*, vol 17, 22012, 2009
- R. Sainidou and F. J. García de Abajo, Plasmon guided modes in nanoparticle metamaterials, *Optics Express*, vol 16, 4499, 2008
- [3] C. Rockstuhl, F. Lederer, C. Etrich, T. Pertsch and T. Scharf, Design of an artificial three-dimensional composite metamaterial with magnetic resonances in the visible range of the electromagnetic spectrum, *Physical Review Letters* vol. 99, 017401, 2007
- [4] C. Rockstuhl and T. Scharf, A metamaterial based on coupled metallic nanoparticles and its band-gap property, *Journal of Microscopy*, vol. 229, 281, 2008
- [5] V. A. Tamma, J. H. Lee, Q. Wu, and W. Park, Visible frequency magnetic activity in silver nanocluster metamaterial, *Applied Optics*, 49, A11, 2010
- [6] D. J. Bergman, The dielectric constant of a composite material–A problem in classical physics, *Phys. Rep.*, vol. 43, 377, 1978
- [7] B. Cichocki and B. U. Felderhof, Electrostatic spectrum and dielectric constant of nonpolar hard sphere fluids, *J. Chem. Phys.*, 90, 4960, 1989
- [8] J. Sancho-Parramon, V. Janicki, and H. Zorc, On the dielectric function tuning of random metal-dielectric nanocomposites for metamaterial applications, *Optics Express*, 18, 26915, 2010