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A Spectral Behavior of Fractal Aggregates in the Quasi-static Approximation

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Abstract

In this paper we present a method to calculate optical properties of small fractal clusters of spheres constructed in a recursive manner in the quasistatic approximation. To calculate optical properties of octahedral generator of six spheres we used the dipole-dipole approximation developed in Shalaev theory. After S iterations we received a fractal cluster of N particles with determined optical properties.

1. Introduction

Electromagnetic phenomena in random metal-insulator composites, such as rough thin films, cermets, colloidal aggregates and other, have been intensively studied for the last two decades [1]. These media typically include small nanometer – scale particles or roughness features. Often nanocomposites, within a certain interval of length-scale, are characterized by a random fractal, i.e. scale – invariant structure. Fractals look similar at different scales; in other words, a part of the object resembles the whole [2]. In this paper we study optical absorption by deterministic, recursively constructed three – dimensional fractal aggregates consisting of spherical metallic particles. Usually, it is quite difficult to calculate optical properties of fractal clusters containing a large number of particles. The most convenient way is to use the scale – invariant properties of the fractal structure. To illustrate the geometrical construction of the fractal, consider a cluster of $N = 6$ spheres as shown in Fig.1. Here we depict how the first two stages of such a construction can be built from individual spherical particles. First we calculate the optical properties of a small cluster – generator of six spheres. Further we shall use these aggregates as generators for an iterative procedure to obtain the fractal system after a number of recursive iterations. The optical parameters of these generated clusters are assigned to a new “effective particle”, which instead will participate in the iteration process. Finally, after S iterations, we receive a fractal cluster of N particles with determined optical parameters. For generators containing six spheres we have applied a method of taking into account pair dipole-dipole interactions between particles within the cluster, developed in the works of Shalaev [3-4].

2. Polarizability of a Cluster and Recursive Approach

We assume that the fractal cluster is located in a continuous dielectric matrix with permittivity $\varepsilon_0 = 1$. We also assume that the size of generator and of the all cluster after S iterations is small compared with the wavelength of incident radiation. This fact will allow us to neglect retardation effects and describe whole system in quasi-static limit. Following the results of previous works [3-4], we can write induced dipole moments of the generator in the form:

$$d_{i,\alpha} = \alpha_0 \left(E_\alpha^{(0)} + \sum_{i \neq j} W_{ij,\alpha\beta} d_{j,\beta} \right) \tag{1}$$

$$W_{ij,\alpha\beta} = \langle i\alpha | W | j\beta \rangle = \left[\delta_{\alpha\beta} - 3\frac{(r_{ij})_\alpha (r_{ij})_\beta}{r_{ij}^2} \right] \frac{1}{r_{ij}^3} \tag{2}$$

where $W_{ij,\alpha\beta}$ is a quasi-static interaction operator between each pair of particles; $\alpha, \beta = x, y, z$ are the coordinates of particles in three dimensional space; $r_{ij} = |r_i - r_j|$ is a distance between particles; r_i and r_j are the origins of spheres i and j, respectively.

$$\alpha_0 = Ba^3 = \frac{\varepsilon - \varepsilon_0}{\varepsilon + 2\varepsilon_0} a^3 \tag{3}$$

is the usual dipole polarizability of the spherical particle with radius a. The polarizability tensor of the i-th particle interacting with all neighboring $j \neq i$ particles can be found from Eq. (1-2) [3-4]:

$$\alpha_{i,\alpha\beta}(\omega) = \sum_{j,n} \frac{\langle i\alpha | n \rangle \langle n | j\beta \rangle}{\alpha_0^{-1}(\omega) + w_n} \tag{4}$$

where w_n and $\langle i\alpha | n \rangle \langle n | j\beta \rangle$ are eigenvalues and eigenvectors of the interaction matrix W respectively, i.e. $W|n\rangle = w_n |n\rangle$. The average polarizability of the generator is given by:

$$\alpha(\omega) = \frac{1}{3N} \sum_i Tr\left[\alpha_{\alpha\beta}^{(i)}\right] \tag{5}$$

where N is a number of particles in generator.

Consider now the octahedral cluster with N = 6 spheres shown in Fig. 1 with sphere radius a and distance between nearest neighbors $R = 2a\sigma$. According to the iteration scheme [5], the radius of the equivalent sphere after the first step of iteration (S = 1) is $a(1) = R/2 + a = a(1+\sigma)$. The same procedure can be applied, for example, to the octahedral generator with N = 7 spheres, where after the first iteration the equivalent radius becomes $a(1) = a(1+\sqrt{2}\sigma)$. Taking into account self-similar properties of fractal cluster, we can assume that after S iterations the radius of the single "effective sphere" in the final cluster is [6]

$$a(s) = a(1+\sigma)^s \tag{6}$$

for the N = 6 generator. After S steps of the iteration process for N = 6 generator, the recursive relation for $B^{(s)}$, which should be used for calculation of "effective sphere" polarization, has a form:

$$B^{(s)} = (1+\sigma)^{-3} \sum_n \frac{\langle i\alpha | n \rangle \langle n | j\beta \rangle}{\left(B^{(s-1)}\right)^{-1} + w_n a^3 (1+\sigma)^{3(s-1)}} \tag{7}$$

Therefore, the extinction cross-section of the fractal cluster with generator N = 6 after S iterations should be:

$$\sigma_e^{(s)} = 4\pi k N^s \operatorname{Im}\left[a^3 (1+\sigma)^{3(s-1)} \sum_n \frac{\langle i\alpha | n \rangle \langle n | j\beta \rangle}{\left(B^{(s-1)}\right)^{-1} + w_n a^3 (1+\sigma)^{3(s-1)}} \right] \tag{8}$$

where $k = {2\pi}/{\lambda}$ and λ denotes the wavelength of incident radiation in the system.

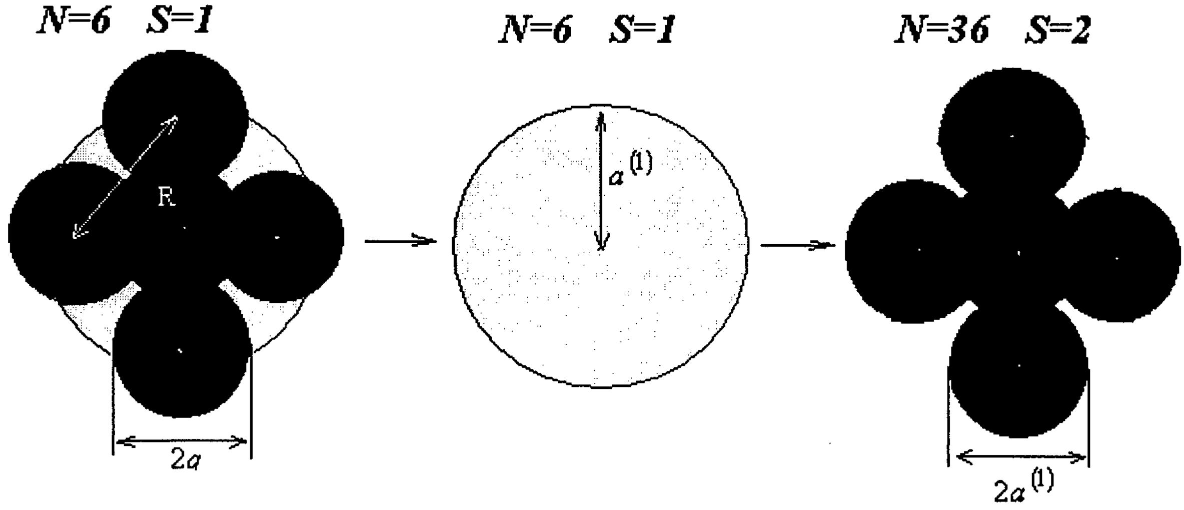

Fig. 1 .The iterative procedure of cluster construction when a cluster of spheres N=6 is replaced by a single equivalent sphere, which is used to construct a larger cluster in a self-similar manner. The fractal dimension of the resulting fractal cluster is $D = {\ln(6)}/{\ln(1+\sigma)}$

3. Results and Discussion

We have applied this theory to the deterministic fractal of metallic particles $N = 6$ depicted in Fig. 1 for the $S = 1$, 2 iteration steps. In Fig. 2 we present the results of our calculations by plotting the extinction efficiency of the fractal cluster N = 6 as a function of $\omega = {\omega}/{\omega_p}$, where ω_p is a plasma frequency of metallic particles. We have specified the properties of the particle material by the values $\omega_p = 1.37 \cdot 10^{16} s^{-1}$, $\gamma = 7.14 \cdot 10^{13} s^{-1}$. The average size of particles $a = 30\,\mathring{A}$ and diameter $d = 61\,\mathring{A}$, gives us the parameter $\sigma = 1.017$. We observe that the main features of the spectra are present already in the dipolar approximation of the Shalaev theory [3-4]. Our results show that the response of the system is strongly dependent on the volume fraction of particles in cluster, which decreases rapidly with arising number S of iterations. This is very interesting, because in the normal non-fractal structures the volume fraction of particles should be approximately constant with increasing number of iterations (i.e. number of particles in the cluster $N \to \infty$). In the same time the magnitude of spectra also decreases with arising number S. Indeed, the self-similar properties of fractal clusters, which save its geometrical structure, after S iterations make them more "transparent". We have also observed that, in particular, the low frequency peak shift to lower frequencies as parameter σ decreases. It should be noted that the two peaks in Fig.2. are in qualitative agreement with theoretical results obtained by application of Ausloos theory [7]. In experiments on random fractal metal aggregates, a two-peak absorption, qualitatively similar to Fig.2 has been observed [8-11]. We are not aware of any data on the behavior of these peaks as a function of aggregate size or particle separation. For linear chains, the low frequency peak shift to low frequency with increasing number of particles [12]. It would be interesting to investigate whether this is the case for anisotropic fractals also. Our method could, in principle, be used for two-scale anisotropic fractal structures. The present method has large similarities with Discrete Dipole Approximation [13] largely used by astrophysicists. This may point a way to extend the method to large particle clusters beyond the quasistatic limit.

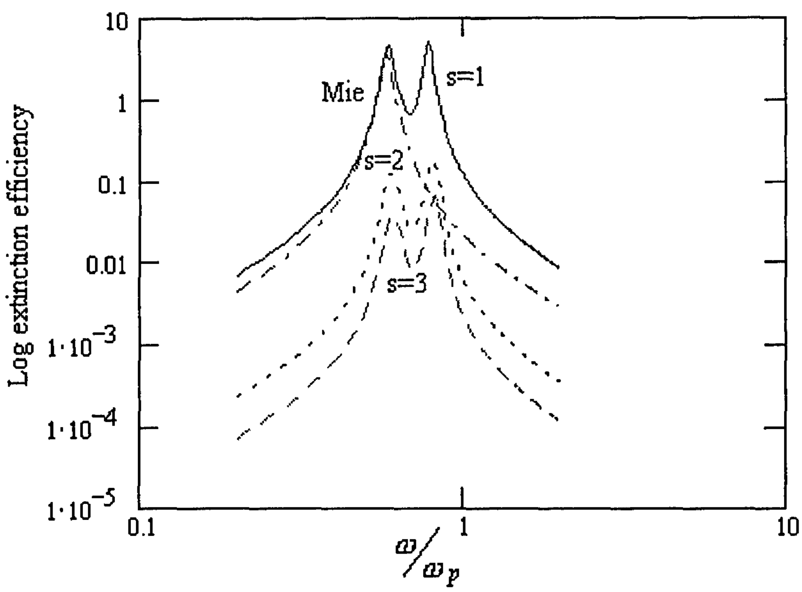

Fig.2. The logarithm of the extinction efficiency as a function of normalized frequency for the generator of 6 spheres (Fig.1.) at σ =1.017. For comparison the results at the first (s=1), second (s=2) and third (s=3) stages of iteration process are shown. The dot-dashed curve gives results of direct application of Mie theory to given cluster of 6 spheres considered as a one spherical particle.

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