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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 properties. 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:
\begin{equation}
\alpha_0 = \frac{\varepsilon - \varepsilon_0}{\varepsilon + 2\varepsilon_0} a^3
\end{equation}

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]:

\begin{equation}
\alpha_{i, \alpha \beta}(\omega) = \sum_{j, n} \frac{\langle \omega | n \rangle \langle n | j \beta \rangle}{\alpha_{0, i \alpha \beta}}
\end{equation}

where \( w_n \) and \( \langle \omega | n \rangle \langle n | j \beta \rangle \) 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:

\begin{equation}
\alpha(\omega) = \frac{1}{3N} \sum_i Tr \left[ \alpha_{i, \alpha \beta} \right]
\end{equation}

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]

\begin{equation}
a(s) = a(1 + \sigma)^s
\end{equation}

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:

\begin{equation}
B(1 + a)^{-3} \sum_n \frac{\langle \omega | n \rangle \langle n | j \beta \rangle}{(B(1))^{-1} + w_n a^{3(1 + \sigma)^{3(s-1)}}}
\end{equation}

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

\begin{equation}
\sigma_x^{(s)} = 4\pi N a^3 \text{Im} \left[ a^{3(1 + \sigma)^{3(s-1)}} \sum_n \frac{\langle \omega | n \rangle \langle n | j \beta \rangle}{(B(1))^{-1} + w_n a^{3(1 + \sigma)^{3(s-1)}}} \right]
\end{equation}
where \( k = \frac{2\pi}{\lambda} \) and \( \lambda \) denotes the wavelength of incident radiation in the system.

\[
N=6 \quad S=1 \quad N=6 \quad S=1 \quad N=36 \quad S=2
\]

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 

\[
D = \frac{\ln(6)}{\ln(1 + \sigma)}
\]

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 \( \omega_p \) is a plasma frequency of metallic particles. We have specified the properties of the particle material by the values \( \omega_p = 1.37 \cdot 10^{16} \text{s}^{-1}, \gamma = 7.14 \cdot 10^{13} \text{s}^{-1} \). The average size of particles \( a = 30 \text{Å} \) and diameter \( d = 61 \text{Å} \), 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 \( \sigma \) 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 \( \alpha = 1.017 \). 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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References


