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Robustness of the fractal regime for the multiple-scattering structure factor

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ABSTRACT

In the single-scattering theory of electromagnetic radiation, the *fractal regime* is a definite range in the photon momentum-transfer q, which is characterized by the scaling-law behavior of the structure factor: $S(q) \propto 1/q^{d_f}$. This allows a straightforward estimation of the fractal dimension d_f of aggregates in *Small-Angle X-ray Scattering* (SAXS) experiments. However, this behavior is not commonly studied in optical scattering experiments because of the lack of information on its domain of validity. In the present work, we propose a definition of the multiple-scattering structure factor, which naturally generalizes the single-scattering function $S(q)$. We show that the mean-field theory of electromagnetic scattering provides an explicit condition to interpret the significance of multiple scattering. In this paper, we investigate and discuss electromagnetic scattering by three classes of fractal aggregates. The results obtained from the TMatrix method show that the fractal scaling range is divided into two domains: (1) a genuine fractal regime, which is robust; (2) a possible anomalous scaling regime, $S(q) \propto 1/q^{\delta}$, with exponent δ independent of d_f, and related to the way the scattering mechanism uses the local morphology of the scatterer. The recognition, and an analysis, of the latter domain is of importance because it may result in significant reduction of the fractal regime, and brings into question the proper mechanism in the build-up of multiple-scattering.

1. Introduction

The investigation of the optical response of disordered matter is an intricate problem. The most common tools to extract information from the scattering data are the trial-and-error methods. However, due to a large number of relevant parameters, the results are tedious to obtain and may prove to be ambiguous. For this reason, direct methods allowing computation of system parameters from the scattering data are highly desirable.

In the single-scattering case, a few direct methods are known. For example, the fractal dimension of a finite aggregate of particles is given by a special scaling law of the *structure factor*, $S(q)$, in the fractal regime [1]:

$$S(q) \propto (qa)^{-d_f}. \quad (1)$$

Here, $q = |\mathbf{q}|$ is the magnitude of the momentum transfer, a is the typical microscopic particle size, and d_f is the fractal dimension of the aggregate. This fractal scaling law is valid in the range $1/R_g < q < 1/a$, where R_g is the mean radius of gyration. The above relation in q-space reflects the corresponding scaling law of the pair-correlation function, $g(r)$, in real space [1]:

$$g(r) \propto r^{d_f - 3}, \quad (2)$$

valid in the range $a < r < R_g$.

From the introduction of X-ray generators a century ago [2] to the present synchrotron facilities [3], the structure

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factor has turned out to be a major tool to characterize correlations between the particle positions over many length scales [4]. The relation (1) is widely used in small-angle X-ray scattering (SAXS) [2] to study the morphology of fractal aggregates [5] or fractal surfaces [6]. We can also obtain the specific surface of the system from the *Porod regime* [7], and the mean gyration radius from the *Guinier regime* [8].

On the other hand, most light-scattering experiments involve multiple-scattering processes, which are intractable using simple mathematical tools. However, previous studies have shown that Eq. (1) may remain valid for scattering measurements even though multiple-scattering events are suspected to be present [9,10]. Experimentalists widely use the above single-scattering scaling law for the characterization of various fractal aggregates [11], though they also claim that minor differences exist between the experimental data and the transmission electron microscopy (TEM) analysis of the fractal dimension. Other results suggest that Eq. (1) might not be valid when multiple-scattering is present. A theoretical argument due to Berry and Percival [12] claims that the scaling relation in Eq. (1) should fail for aggregates with mass-fractal dimension $d_f > 2$. However, it is not clear what relation should replace Eq. (1) in that case. Results of numerical computations [13,14] suggest that Eq. (1) is valid for the case of $d_f > 2$, but with an effective exponent different from the fractal dimension.

In this paper, we present detailed theoretical and numerical results to clarify this important issue. The paper is organized as follows. In Section 2, we introduce the static structure factor with multiple-scattering condition. Section 3 describes the mean-field multiple-scattering theory for the dipolar and multipolar regimes. In Section 4, we compare theoretical and numerical results for three different types of cluster aggregates. In Section 5, we discuss the scaling behavior of the structure factor. Finally, we conclude with a summary in Section 6.

2. General definition of the static structure factor

Consider a monochromatic electromagnetic wave of amplitude E_0 and wavelength λ, illuminating an aggregate of N spherical particles of radius a. We assume elastic light-scattering, with the wave-vector $\mathbf{k}_{\rm inc}$ (of modulus $k = 2\pi/\lambda$) defining the propagation direction of the incident beam. The scattered wave-vector, $\mathbf{k}_{\rm sca}$ (with the same modulus k), defines the observation direction. For the general case of a randomly-oriented aggregate, the intensity $I_N(\mathbf{q})$ of the scattered wave depends on qa, where q is the magnitude of the scattering vector $\mathbf{q} = \mathbf{k}_{\rm inc} - \mathbf{k}_{\rm sca}$.

Assuming separation of the optical properties and the spatial distribution of the particles, $I_N(\mathbf{q})$ is related to the structure factor $S_N(\mathbf{q})$ as

$$I_N(\mathbf{q}) = |E_0|^2 f(N) F(\mathbf{q}) S_N(\mathbf{q}), \tag{3}$$

the form factor $F(\mathbf{q})$ being the intensity scattered by a single particle.

We make the following observations:

(1) The structure factor $S_N(\mathbf{q})$ is a positive function which can be conveniently normalized such that $S_N(\mathbf{0}) = 1$. This function contains information about the spatial distribution of the particles. For the case of a single particle (i.e., $N=1$), $S_1(\mathbf{q}) = 1$.

(2) The scaling factor $f(N)$ is related to the forward-scattering scaling as

$$f(N) = \frac{I_N(\mathbf{0})}{|E_0|^2 F(\mathbf{0})}. \tag{4}$$

In most cases, this function behaves as a power-law: $f(N) \propto N^{\alpha}$, with an exponent $0 < \alpha \le 2$ [5].

The simplest way to define the structure factor is then via the normalized ratio of the scattered intensity from the N particles (I_N) and the scattered intensity from a single particle (I_1):

$$S_N(\mathbf{q}) = \frac{I_N(\mathbf{q})}{I_N(\mathbf{0})} \cdot \frac{I_1(\mathbf{0})}{I_1(\mathbf{q})}. \tag{5}$$

Now, I_N being written as the product of a function of the optical parameters and a function involving the spatial distribution of the particles, the definition (5) results in a quantity which depends essentially on the mass distribution of the aggregate.

The Rayleigh–Debye–Gans (RDG) theory provides a framework in which the separation between the optical and geometrical properties is realized [15]. Indeed, the RDG theory tells us that the single-scattering of the incoming wave by a collection of N electromagnetic dipoles is the dominant process due to the weak electric polarizability of the particles inside the aggregate. The structure factor in Eq. (5) is then written as the square modulus of the Fourier transform of the density distribution of the scattering system [4]:

$$S_N(\mathbf{q}) = \left| \frac{1}{N} \sum_{j=1}^{N} e^{i\mathbf{q}\cdot\mathbf{r}_j} \right|^2 \tag{6}$$

$$S_N(\mathbf{q}) = \frac{1}{N}\left(1 + \rho \int (g(\mathbf{r}) - 1) e^{i\mathbf{q}\cdot\mathbf{r}}\, d\mathbf{r}\right). \tag{7}$$

In Eq. (6), $\mathbf{r_j}$ denotes the position of the jth particle in the aggregate. In Eq. (7), $g(\mathbf{r})$ is the pair-correlation function [1], and $\rho = N/V$ is the particle number density in a given volume V.

For a fractal scatterer of radius of gyration R_g, which is an aggregate of spherical particles of radius a, the main features of the RDG structure factor are as follows:

(1) $qa < a/R_g$ is the *Guinier regime* [8], which depends only on the parameter qR_g.

(2) $a/R_g < qa < 1$ is the *fractal regime*, which is characterized by Eq. (1). We focus on this regime in the next section.

(3) $1 \ll qa$ is the *Porod regime* [7], which is not relevant for the present paper.