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SMALL-ANGLE NEUTRON SCATTERING FROM 3D MONODISPERSE VICSEK FRACTALS

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Малоугловое рассеяние нейтронов
на монодисперсных 3D-фракталах

Вычислена интенсивность малоуглового рассеяния нейтронов на систему монодисперсных, хаотично ориентированных 3D Vicsek фракталов. Фрактальная система получена путем создания последовательности приближений, начинающихся с куба с размером сторон l как инициатора. Учитывая взаимодействия между частицами, мы применяем смешанное интегральное уравнение Роджерса–Юнга, чтобы получить парную функцию распределения $g(r)$ и структурный фактор $S(q)$ для системы. Парный потенциал, используемый в уравнении Роджерса–Юнга, выбран так, чтобы учитывать число рук рассеивателей.

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Small-Angle Neutron Scattering from 3D Monodisperse
Vicsek Fractals

The small-angle neutron scattering intensity for a system of monodisperse, randomly oriented 3D Vicsek fractals has been computed. The fractal system is obtained by generating a sequence of approximations starting with a cube of edge l as an initiator. When the interactions among particles are taken into account, we apply the mixed Rogers–Young integral equation to obtain the pair distribution function $g(r)$ and structure factor $S(q)$ for the system. The pair potential used in Rogers–Young (RY) closure is chosen so as to take into account the arm number of the scatterers.

The investigation has been performed at the Frank Laboratory of Neutron Physics, JINR.

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1. INTRODUCTION

The theoretical study of the structural properties of matter such as numerous polymeric structures, aqueous solutions, magnetic liquids or magnetic elastomers is often achieved by employing scattering methods [1–4]. For those materials which show a self-similarity (fractal systems) on a finite range at microscopic scale, the scattering techniques are an essential tool of investigation and different models to reveal their structure have been proposed [5–7]. In this paper the small-angle neutron scattering (SANS) [4, 8, 9] is considered as a scattering method for evaluating the intensity from a system of monodisperse nonrandom 3D Vicsek fractals [10, 11]. The approach for the scattered intensity presented here was successfully employed for other similar systems of nonrandom fractals (see [12]). Methods for obtaining the scattered intensity from nonrandom and random fractals can be found also in [13–16]. Vicsek fractals, constructed by a deterministic set of iterative steps, can serve as a model for hyperbranched macromolecular structures [17, 18], and small-angle neutron scattering techniques provide us with more information on the microstructure of such objects, not obtainable by other methods.

Spatial correlations in the system are described in terms of the pair-distribution function $g(r)$, related to the probability of finding the center of any particle at a distance r from the center of a given particle, by solving the Ornstein–Zernike (OZ) [19] nonlinear integral equation. The derivation of the pair-distribution function as described above and based on the formalism developed in the theory of simple classical fluids [21] is followed by the determination of the structure factor $S(q)$, related to the pair-distribution function by

$$S(q) = 1 + 4\pi \frac{N}{V} \int_0^\infty [g(r) - 1] r^2 \frac{\sin(qr)}{qr} dr, \tag{1}$$

where N is the number of particles in a volume V and q is the module of the scattering vector $\mathbf{q}$.

Construction of the Vicsek fractals in three dimensions is shown in Fig. 1 and is closely related to the construction of the Menger sponge [5].

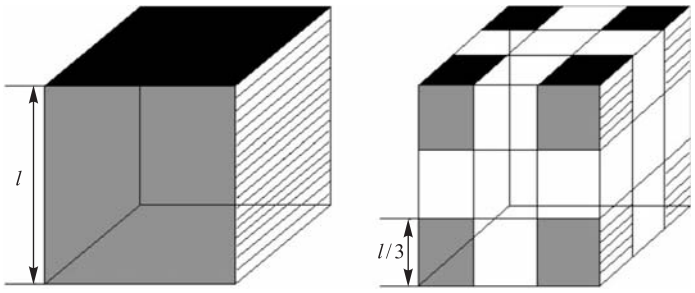

Fig. 1. Construction of the 3D Vicsek fractals

The initiator in the construction is a cube. The first iteration towards the final object, the generator is formed by dividing the initial cube of length l into 27 smaller cubes of length $l/3$. From each face, the cubes of side length $l/3$ from the corners are kept and the others are removed (Fig. 1). This leaves a prefractal composed of 9 smaller cubes (eight in the corners of the initial cube and one in the center) each scaled down by one-third. Every edge of the new cubes is divided again into three parts leaving 81 cubes scaled down by one-ninth from the original cube. Repetead iteration of this construction leads to the 3D Vicsek fractals.

The purpose of this paper is to obtain a mathematical model which describes the small-angle neutron scattering from 3D Vicsek fractals. The next section starts with a theoretical background, followed by the determination of the form factor and structure factor respectively.

2. THEORETICAL BACKGROUND

The link between fractal systems and scattered intensity is the fractal dimension [22], a fundamental characteristic for fractal systems, and it is obtained as the slope of scattered intensity against the module of the scattering vector on a double logarithmic scale. In a system of Vicsek fractals, which belongs to the category of mass fractals [23–25] objects (aggregates of primary particles or subunits), the distribution of mass has the property that the mass inside a spherical surface of radius r is given by [4]

$$M(r) \approx r^{d_f}, \tag{2}$$

where d_f is the mass-fractal dimension, a number between 1 and 3 that can be fractional. The value of d_f is determined from

$$d_f = \frac{\log(N)}{\log(1/\epsilon)}, \tag{3}$$

where N is the number of self-similar objects and ϵ the scaling ratio, in our case $\epsilon = 1/3$. For Vicsek fractals, the mass-fractal dimension is

$$d_f = \frac{\log(9)}{\log(1/(1/3))} = 2. \tag{4}$$

The smaller the value of d_f, the more open the structure is, and as d_f is reduced to 1, the object becomes a line if it remains singly connected.

The scattered intensity on an absolute scale for any interacting particulate system of scatterers can be expressed as

$$I(q) = (\Delta\rho)^2 V_p^2 N_p \langle P_n(q) \rangle S(q) \tag{5}$$

with $\Delta\rho = \rho_p - \rho$ the scattering contrast; and ρ_p and ρ are the scattering length densities of the particles and the surrounding medium, respectively; $\langle \ldots \rangle$ denotes an ensemble average, V_p is the mean particle volume, N_p is the number of particles per unit volume, P_n is the scattering form factor of particles for nth approximant and has the expression:

$$P_n(q) = \left| \frac{1}{V_{p_n}} \int\limits_{V_{pn}} e^{i\mathbf{qr}} \right|^2. \tag{6}$$

$S(q)$ is the structure factor which can be obtained by Fourier transform of the pair-distribution function $g(r)$. Recall that the vector $\mathbf{q}$ is defined by

$$\mathbf{q} = \mathbf{k}_0 - \mathbf{k_1}, \tag{7}$$

where $\mathbf{k}_0$ and $\mathbf{k}_1$ are, respectively, the incident and scattered wave vectors of the neutrons. During a small-angle neutron experiment, one measures the elastic scattering, for which $|\mathbf{k}_0| = |\mathbf{k}_1| = 2\pi/\lambda$, where λ is the wavelength of the neutrons. As a consequence, the vector $\mathbf{q}$ has the magnitude

$$|\mathbf{q}| = \frac{4\pi}{\lambda} \sin\left(\frac{\theta}{2}\right), \tag{8}$$

where θ is the scattering angle.

2.1. Form Factor of 3D Vicsek Fractals. When the volume fraction of the particles inside the sample is less than approximately 2–3%, it is reasonable to analyze the scattering in terms of randomly oriented, noninteracting particles (i.e., we neglect the structure factor and set $S(q) = 1$). In this so-called dilute limit, the particles scatter independently, the total scattering is the sum of the scattering from each particle and has the expression

$$P_n(q) = (\Delta\rho)^2 V_{p_n}^2 N_p \left\langle \left| \frac{1}{V_{p_n}} \int\limits_{V_{p_n}} e^{i\mathbf{qr}} d\mathbf{r} \right|^2 \right\rangle . \tag{9}$$

We will denote

$$F_n(\mathbf{q}) = \frac{1}{V_{p_n}} \int\limits_{V_{p_n}} e^{i\mathbf{qr}} d\mathbf{r} \tag{10}$$

the square root of the scattering form factor of the nth approximation. The origin of the volume integration is taken at the center of the cube and $\mathbf{r}$ is a vector from its origin to the volume element $d\mathbf{r}$. From equation (9) the form factor for the first approximant (Fig. 1) is obtained by dividing the cube with edge l into 27 cubes with edge $l/3$ and keeping 9 cubes following the procedure described above and it has the expression

$$F_1(\mathbf{q}) = G_1(\mathbf{q}) F_0\left(\frac{\mathbf{q}}{3}\right), \tag{11}$$

where

$$G_n(\mathbf{q}) = \frac{1}{9} \sum_{j=-1}^{1} \sum_{k=-1}^{1} \sum_{l=-1}^{1} \omega_{jkl} e^{i\mathbf{q}\mathbf{R}_{jkl}^n}, \tag{12}$$

with

$$\omega_{jkl} = \delta_{j0}\delta_{k0}\delta_{l0} + (1-\delta_{j0})(1-\delta_{k0})(1-\delta_{l0}), \tag{13}$$

a quantity which ensures that the expression (11) includes only the cubes of edge $l/3$ formed from the initiator, δ_{ij} is the Kronecker delta symbol, $\mathbf{R}_{jkl}^1 = (j\mathbf{x} + k\mathbf{y} + l\mathbf{z})l/3$ is the vector from the center of the zero-order approximant (cube of edge l) to the center of the cubes with edge $l/3$, and $\mathbf{x}$, $\mathbf{y}$, $\mathbf{z}$ are the unit vectors in the directions of x, y, and z axes. The origin of the x, y, z axes is chosen to be in the center of the initiator, and the axes are oriented so that each cube face is perpendicular to one of the axes. $F_0(\mathbf{q})$ is the form factor of the initiator.

The form factor for the 2nd approximant is obtained by repeating the procedure on the $F_0(\mathbf{q}/3)$, and by induction, the nth approximation will have the expression

$$F_n(\mathbf{q}) = \prod_{i=1}^{n} G_i(\mathbf{q}) F_0\left(\frac{\mathbf{q}}{3^n}\right). \tag{14}$$

Substituting Eq. (13) into Eq. (12) and further into (10), averaging over orientation and taking into account that for $\mathbf{q}$ small enough we have $F_0(\mathbf{q}/3^n) \approx 1$, the form factor for the nth approximation will have the following expression:

$$P_n(q) = P_n(0) \sum_{\alpha=-T_n}^{T_n} \sum_{\beta=-T_n}^{T_n} \sum_{\gamma=-T_n}^{T_n} A^n_{\alpha\beta\gamma} \frac{\sin(q\rho^n_{\alpha\beta\gamma})}{q\rho^n_{\alpha\beta\gamma}}, \tag{15}$$

where the indices α, β, γ are connected to the indices j, k, l through the relations

$$\begin{aligned} &\alpha = j + j' \text{ with } -1 \leqslant j \leqslant 1 \text{ and } -1 \leqslant j' \leqslant 1, \\ &\beta = k + k' \text{ with } -1 \leqslant k \leqslant 1 \text{ and } -1 \leqslant k' \leqslant 1, \\ &\gamma = l + l' \text{ with } -1 \leqslant l \leqslant 1 \text{ and } -1 \leqslant l' \leqslant 1. \end{aligned} \tag{16}$$

The form factor at zero is $P_n(0) = (\Delta\rho)^2 V^2_{p_n} N_p$; $\rho^n_{\alpha\beta\gamma}$ is given by $\rho^n_{\alpha\beta\gamma} = l/3^n(\alpha^2 + \beta^2 + \gamma^2)^{1/2}$ and

$$A^{n+t}_{\alpha\beta\gamma} = \sum_{p=p_{\min}}^{p_{\max}} \sum_{r=r_{\min}}^{r_{\max}} \sum_{s=s_{\min}}^{s_{\max}} A^n_{prs} A^t_{\alpha-3^t p, \beta-3^t r, \gamma-3^t s} \tag{17}$$

with

$$p_{\max} = \text{Minint}\left(T_n,\ \frac{\alpha + T_t}{3^t}\right) \tag{18}$$

and

$$p_{\min} = \text{Maxint}\left(-T_n,\ \frac{\alpha - T_t}{3^t}\right). \tag{19}$$

For $r_{\max}$ and $r_{\min}$, in Eqs. (17) and (18) α is replaced by β, and for $s_{\max}$ and $s_{\min}$, α is replaced by γ. One defines $\text{Maxint}(x, y) = x^{\max}_{\text{int}}$ when $x > y$ and $\text{Maxint}(x, y) = y^{\max}_{\text{int}}$ when $x < y$, respectively, as the largest integers not greater than x or y. Analogously, $\text{Minint}(x, y) = x^{\min}_{\text{int}}$ when $x < y$, and $\text{Minint}(x, y) = x^{\min}_{\text{int}}$ when $x > y$ are, respectively, the smallest integers not less than x or y. In Eq. (15) $T_n = 3^n - 1$ and A_{jkl} for $n = 1$ is evaluated from Eq. (12). For $n > 1$ the coefficients $A^n_{\alpha\beta\gamma}$ are evaluated from A^1_{jkl} using the relation

$$\prod_{i=1}^{n+t} (G_i(\mathbf{q}))^2 = \left(\prod_{i=1}^{n} (G_i(\mathbf{q}))^2\right) \left(\prod_{j=1}^{t} \left(G_j\left(\frac{\mathbf{q}}{3^n}\right)\right)^2\right). \tag{20}$$

Using Eq. (15), the scattering form factors $P_n(q)/P_n(0)$ for the first four iterations were plotted against ql in Fig. 2. The relative error resulting from relation (14) is

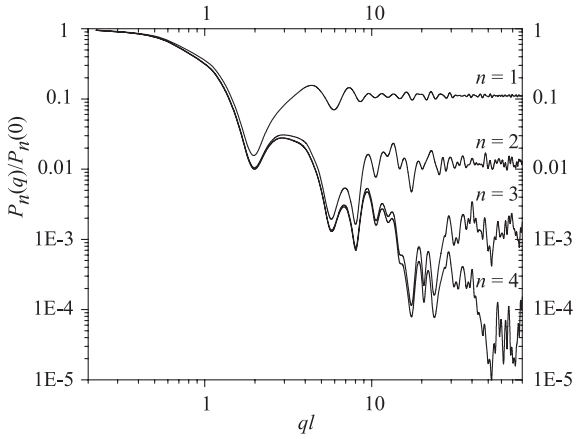

Fig. 2. Form factors for Vicsek fractals for the first four iterations, from Eq. (15)

given by $\frac{1}{12}\left(\frac{ql}{3^n}\right)^2$ and will be less than c if [12]

$$\frac{1}{12}\left(\frac{ql}{3^n}\right)^2 < c, \tag{21}$$

where c is a given accuracy. For $c = 1\%, 2\%, 3\%$ the maximum values for ql for which Eq. (16) is available, are summarized in the table.

Maximum values for ql for which Eq. (15) has the accuracy given by c

	$n = 1$	$n = 2$	$n = 3$	$n = 4$
$c = 1\%$	1.03	3.11	9.35	28.05
$c = 2\%$	1.46	4.40	13.22	39.68
$c = 3\%$	1.8	5.4	16.2	48.6

2.2. Interacting 3D Vicsek Fractals. The description of interactions between scatterers is given by the structure factor $S(q)$, defined as

$$S(q) = 1 + \rho\tilde{h}(q) \tag{22}$$

with $\tilde{h}(q)$ denoting the Fourier transform of the total correlation function $h(r)$. $h(r) = g(r) - 1$ is connected to the direct correlation function through the OZ equation which has the form

$$h(r) = c(r) + \rho \int dr' c(|r - r'|)h(r'), \tag{23}$$

where ρ is the density of the system, defined as the number N of particles enclosed in the macroscopic volume V. The RY closure [20] adopted here for solving Eq. (23) interpolates continuously between HNC [27] and PY approximations [28], namely:

$$g(r) = e^{-\beta u(r)} \left(1 + \frac{e^{\gamma(r) f(r) - 1}}{f(r)}\right), \tag{24}$$

where $\gamma(r) \equiv h(r) - c(r)$, and $f(r)$ is the mixing function:

$$f(r) = 1 - e^{-\alpha r}, \tag{25}$$

where α is an adjustable parameter used to achieve thermodynamic consistency between the virial and compressibility pressures [21]. We choose the interparticle potential [29] between scatterers as

$$\beta u(r) = \begin{cases} \dfrac{5}{18} f^{\frac{3}{2}} \left(-\ln\left(\dfrac{r}{\sigma}\right) + \dfrac{1}{1 + \dfrac{\sqrt{f}}{2}} \right) & \text{if } r \leqslant \sigma \\ \dfrac{5}{18} f^{\frac{3}{2}} \dfrac{1}{1 + \dfrac{\sqrt{f}}{2}} \dfrac{\sigma}{r} e^{-\dfrac{\sqrt{f}(r-\sigma)}{2\sigma}} & \text{if } r > \sigma \end{cases}, \tag{26}$$

where $\beta = 1/k_B T$ is the inverse temperature, k_B is Boltzmann's constant, T is the absolute temperature, f is the functionality of the scatterers, in our case $f = 8$,

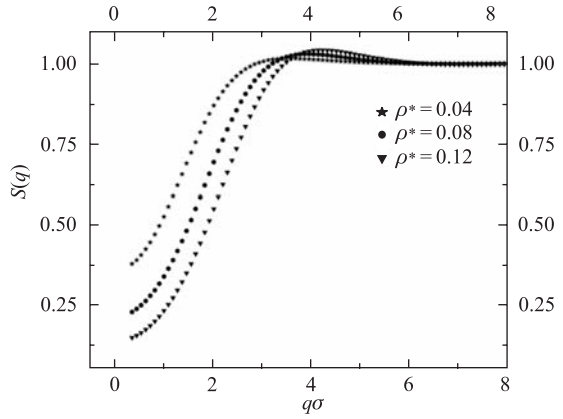

Fig. 3. Theoretical structure factor for 3D Vicsek fractals at different values of densities

and σ is the spatial extent of scatterers, connected to the length of the cube through $\sigma = l\sqrt{3}$. The potential $u(r)$ is a combination between a logarithmic behavior for $r \leqslant \sigma$ and a Yukawa form for $r > \sigma$.

In the development of the theories presented before, σ is chosen to be the unit of length and $\bar{\rho} = \rho\sigma^3$ the dimensionless density (which appear in Eq. (22)). In order to obtain the radial distribution function of the system, we use the potential given by Eq. (26) together with the RY closure Eq. (21) and solve the OZ equation (20) for three dimensionless densities: $\bar{\rho_1} = 0.04$, $\bar{\rho_2} = 0.08$ and $\bar{\rho_3} = 0.12$. The results presented in Fig. 3 show a structure factor similar to a smoothed step function and very small values of the peaks. With increasing density, the values of q for which the peaks appear are shifted to the right.

3. CONCLUSIONS

In this paper we have studied the small-angle neutron scattering intensity from a system of interacting 3D Vicsek fractals. Employing an algorithm which has been shown to be able to furnish the form factor from a system of mass fractals [12], we obtained the form factor from a system of 3D Vicsek fractals, which can be considered as a model for various polymeric structures.

According to the Eq. (4), the mass-fractal dimension of 3D Vicsek fractals is 2. On other hand, it is well known that nonfractal objects with Euclidean dimension 3 give an intensity proportional to q^{-2} [4] (e.g., scattering from a thin disk). A similar situation occurs when for certain surface-fractals systems with surface fractal dimension $D_f = 2$ [26] (e.g., Mandelbrot set) scattering gives an intensity proportional to $q^{-(6-D_f)} = q^{-4}$ a decay obtainable also from scattering from spheres [4]. In either case, an a priori knowledge about the structure of the scatterers is necessary in order to reveal the fractal or nonfractal nature of the system. Small-angle neutron scattering alone does not differentiate them.

To take into account the interactions between scatterers, we have applied the Rogers–Young mixed integral equation. The closure interpolates between the PY and HNC equations. The structure factor for three small valued densities is similar to a smoothed step function. The increase of density is followed by an increase of height of the peaks and displacement towards the right. Applications of this model to real structures similar to those studied in this paper are in our future plan.

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