Small-angle multiple neutron scattering in fractal media

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Multiple small-angle neutron scattering from fractals is considered. It is shown that the mean free path ℓ in many cases is small compared to the commonly used sample thickness L. For multiple scattering when $\ell \ll L$ the width of the intensity distribution q_L is proportional to L^{μ}, where $\mu = 1/(4-D_s)$ and $\mu = 1/(D_v - 2)$ for the surface and volume fractals, respectively, and $D_{s,v}$ is the corresponding dimensionality of the fractals. In both cases $\mu > \frac{1}{2}$ and the multiple scattering is a superdiffusion. The large-q tail of the intensity is the same as observed for single scattering. However, parts of the total intensity for $q < q_L$ and $q > q_L$ have the same order of magnitude and transition to the quasi-Guinier regime occurs at $q \ll q_L^{(\Delta)} < \xi^{-1}$ where ξ is cutoff for behaviour. It is shown that the failure of the diffusive random-walk description of the small-angle multiple scattering takes place in all cases when the mean square of the scattering angle $\overline{\vartheta^2}$ for the single scattering is an ill-defined quantity.

I. INTRODUCTION

The small-angle x-ray and neutron-scattering techniques are widely used as tools for studying large-scale inhomogeneities in condensed matter. The scattering experiments reveal that in many cases the scattering intensity $I(q)$ at large q deviates from the Porod law $I(q) \sim q^{-4}$ and falls off as $q^{-\Delta}$ where $\Delta < 4$. (See for example Ref. 1 and references therein.)

In the case of volume or mass fractals one has $\Delta = D_v$ where $D_v < 3$ is the fractal dimension.[1–4] For surface fractals $\Delta = 6 - D_s$ where $2 < D_s < 3$ is the corresponding dimension.[4,5] In the first case $\Delta < 3$ and in the second one $3 < \Delta < 4$. It means that the volume and surface fractals may be readily distinguished experimentally. The Porod law is restored if $D_v = 3$ and $D_s = 2$.[4–6] As a rule the fractal structure is limited by an upper length cutoff or the fractal correlation length ξ and at $q \lesssim \xi^{-1}$ we have a transition to the Guinier region.[7,3]

However, all these results are applicable in the single-scattering regime only, i.e., if the sample thickness L is small compared to the mean free path ℓ. As we will show below, the opposite situation $\ell \ll L$ may occur in real experimental systems. In this paper we present results of the theoretical study of the small-angle multiple scattering from the fractal systems. Similar consideration has been done in Refs. 8 and 9 for the cases when the single scattering on finite-size inhomogeneities was described by the Born and quasiclassical approximations, respectively. The multiple scattering from critical fluctuations has been considered in Ref. 8. It should be noted that the double critical scattering in iron has been studied experimentally using polarized neutrons.[10] Below we will restrict ourselves to multiple neutron scattering only because for x rays the absorption has to be taken into account.

We will estimate the mean free path ℓ for the fractal scattering and show that in some cases it may be of order of 10^{-3}–10^{-1} cm. Then we will evaluate the intensity of the multiple small-angle scattering. It will be shown that the width of the intensity distribution is characterized by the momentum $q_L^{(\Delta)}$ which is given by

$$q_L^{(\Delta)} = \frac{1}{2\xi}\left(\frac{L}{g_\Delta \ell}\right)^{\mu}, \tag{1}$$

where $\Delta = D_v$ and $6 - D_s$ for the volume and surface fractals, respectively, and $g_\Delta \sim 1$. We see that $q_L^{(\Delta)} \sim L^{\mu_{s,v}}$, where $\mu_v = (D_v - 2)^{-1} > 1$ and $1 > \mu_s = (4 - D_s)^{-1} > 1/2$. In both cases characteristic momentum $q_L^{(\Delta)}$ increases with L faster than in the case of random walks when $q_L^{\rm RW} \sim L^{1/2}$. This behavior of $q_L^{(\Delta)}$ is an example of the so-called anomalous diffusion or superdiffusion.[11] It is related to the fact that we have not well defined mean scattering angle $(\overline{\vartheta^2})^{1/2} \ll 1$ for a single scattering event.

Indeed, we have by the definition

$$\overline{\vartheta^2} = \left(\int d\Omega\, \vartheta^2 d\sigma/d\Omega\right) \Big/ \int d\Omega\, d\sigma/d\Omega, \tag{2}$$

where $d\sigma/d\Omega$ is the cross section for the single scattering event. Obviously $\overline{\vartheta^2}$ is well defined in the small-angle region if $d\sigma/d\Omega$ decreases faster than q^{-4}. However, in our case $\Delta < 4$. The exponent $\Delta = 4$ may be considered as a higher critical dimension for the anomalous small-angle multiple scattering. This value takes place for $D_v = 3$ (see below) and $D_s = 2$, when the fractals turn into the finite-size inhomogeneities and for the Coulomb scattering.[12] At the same time $\Delta = 2$ is a lower critical dimension. Strictly speaking if $\Delta \le 2$ the scattering cannot be considered as a small-angle one because the total cross section is not saturated by the small q region, and below we restrict ourselves by the condition $D_v > 2$. The multiple scattering for $\Delta = 2$ has been studied in Ref. 8.

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We see that the random-walks description of the small-angle multiple scattering is not valid in all physically important cases. Even in the Porod case when $\Delta = 4$ the width of the intensity distribution is proportional to $[(L/\ell)\ln(L/\ell)]^{1/2}$ instead of $(L/\ell)^{1/2}$.[8]

This paper is organized as follows. In Sec. II we estimate the mean free path ℓ for the fractal systems. Section III is devoted to the multiple scattering. The conclusions are presented in Sec. IV.

II. NEUTRON MEAN FREE PATH IN FRACTAL MEDIA

We estimate now the mean free path for neutron propagation in the fractal media. For this purpose we need to know the order of magnitude of the cross section. We consider the volume and surface fractals separately and the limiting case of finite-size inhomogeneities.

a. Volume fractals. For definiteness we consider a medium where fractals are pores which sizes r are distributed in the range $R_0 < r < \xi$ and $R_0 \ll \xi$. In this case the density-density correlation function has the form[3,4]

$$\langle\rho(r)\rho(0)\rangle = \left(\frac{N_0^2 D_v}{4\pi}\right)\left(\frac{R_0}{r}\right)^{3-D_v}\exp\left(-\frac{r}{\xi}\right), \quad (3)$$

where N_0 is the atomic density between the pores and the factor N_0^2 ensures the correct physical dimension. As a result for the single fractal cross section we have[3,4]

$$\frac{d\sigma}{d\Omega} = K_0^4 V_{D_v} R_0^{3-D_v} \times\frac{D_v\Gamma(D_v-1)\sin[(D_v-1)\tan^{-1}(q\xi)]}{(4\pi)^2 q(q^2+\xi^{-2})^{(D_v-1)/2}}, \quad (4)$$

where $K_0^2 = 4\pi|b|N_0$, b is the corresponding scattering length, $V_{D_v} = V_3(R_0/\xi)^{3-D_v}$, $V_3 = \xi^3$, and $\Gamma(x)$ is the gamma function. We define here the fractal volume V_{D_v} in such a way that it has the proper physical dimension cm^3 and the factor $(R_0/\xi)^{3-D_v}$ describes reduction of the total mass of the matter compared to the dense case in the region of the range of ξ. It should be noted that Eq. (4) is the simplest expression which gives the proper large-q dependence of the cross section q^{-D_v} for $D_v < 3$ and the Porod law q^{-4} for $D_v = 3$. Some other expressions were discussed in Ref. 3. We restrict ourselves to Eq. (4) because our results are not sensitive to the precise form of the cross section at $q \lesssim \xi^{-1}$.

If $D_v = 3$ we get from Eq. (4)

$$\frac{d\sigma}{d\Omega} = \frac{3K_0^4 V_3}{8\pi^2(q^2+\xi^{-2})^2\xi}. \quad (5)$$

For $q\xi \gg 1$ this expression coincides by the order of magnitude with the Born cross section for the scattering from the sphere of the radius ξ calculated in the Born approximation, which may be represented as[13]

$$\left(\frac{d\sigma}{d\Omega}\right)_{sh} = 3K_0^4 V_{sp}(\xi)/(8\pi\xi q^4). \quad (6)$$

if one replaces $\cos^2 q\xi$ by its mean value 1/2 and uses the well-known expression for the neutron potential energy in the medium[13]

$$U = 2\pi b N_0 \hbar^2/m = \hbar^2 K_0^2 b/(|\, b\,|). \quad (7)$$

Comparing Eqs. (5) and (6) we conclude that for $D_v = 3$ fractals turn into the system of pores with the sizes of the order of ξ.

At $D_v = 2$ from (4) we have

$$\frac{d\sigma}{d\Omega} = \frac{K_0^4 V_2 R_0}{8\pi^2(q^2+\xi^{-2})}, \quad (8)$$

where $V_2 = \xi^2 R_0$.

From (4) we obtain for the total cross section

$$\sigma_{D_v} = I_{D_v} K_0^4 R_0^3 V_{D_v}\,(\xi/R_0)^{D_v}(k\xi)^{-2}, \quad (9)$$

where k is the neutron wave number and

$$I_{D_v} = \frac{D_v\Gamma(D_v-1)}{8\pi}\int_0^\infty \frac{dx\sin[(D_v-1)\tan^{-1}x]}{(x^2+1)^{(D_v-1)/2}}. \quad (10)$$

The mean free path ℓ is determined in the usual way as $\ell = (\sigma n_F)^{-1}$ where n_F is the fractal density. If the medium porosity is related only with fractals we have for their concentration[14]

$$n_F = \frac{\Delta N}{N_0 V_{D_v}}, \quad (11)$$

where $\Delta N/N_0 = 1 - c$ is the fraction of the sample occupied by pores. As a result the mean free path is given by

$$\frac{1}{\ell_{D_v}} = I_{D_v} K_0^4 \frac{\Delta N}{N_0}\frac{\xi}{k^2}\left(\frac{R_0}{\xi}\right)^{3-D_v} \quad (12)$$

and for $D_v = 3$ and 2 we obtain, respectively,

$$\frac{1}{\ell_3} = 3K_0^4\xi\frac{\Delta N/N_0}{16\pi k^2} \quad (13)$$

and

$$\frac{1}{\ell_2} = \frac{K_0^4 R_0}{4\pi k^2}\frac{\Delta N}{N_0}\ln(2k\xi). \quad (14)$$

In the case of the spherical pores using the corresponding expression for the total cross section[13] we get $1/\ell_{sp} = 2\pi/\ell_3$, i.e., both quantities have the same order of magnitude as they should have. The numerical factor 2π is related to the particular definition of the correlation function given by Eq. (3).

For $D_v = 2$ the q dependence of the cross section (8) is the same as in the case of the critical fluctuations if one neglects the small Fisher parameter η[15]:

$$d\sigma/d\Omega = Zb^2\left[a^2(q^2+\xi^{-2})\right]^{-1}, \quad (15)$$

where $Z \sim 1, a$ is of the order of the interatomic spacing and $\xi = a(T-T_c)^{-\nu}$ where ν is the critical range exponent. For ferromagnets above the Curie temperature values of Z and a are listed, for example, in Ref. 16. The corresponding mean free path has the form[17]

$$\frac{1}{\ell_{\rm cr}} = \frac{2\pi b^2 N_0 Z}{(ka)^2}\ln(2k\xi). \tag{16}$$

Expressions (14) and (16) coincide if $2R_0N_0 = Za^{-2}$ and $\Delta N/N_0 = 1$. Physically it means that pores disappear and all atoms of the system are involved in the fluctuations.

Equations (9) and (10) are valid for $D_v > 2$ only when the total cross section σ_{D_v} is saturated at small scattering vectors $q \sim \xi^{-1}$, i.e., when the scattering may be considered as a real small-angle one. In this case a broad region of the sample thickness L may exist where we have the small-angle multiple scattering (SAMS). However, if $D_v < 2$ the large $q \sim k$ give the main contribution to the total cross section and the scattering intensity becomes almost isotropic after few scattering events and as a result we have not the SAMS at all. The case $D_v = 2$ is a boundary case where all q between ξ^{-1} and $2k$ contribute to the cross section. The multiple scattering for $D_v = 2$ has been studied theoretically in Ref. 8. The double critical scattering in iron was observed using polarized neutrons in Ref. 10 and good agreement was found with theory. Below we will restrict ourselves to $D_v > 2$ only.

Let us estimate now a possible order of magnitude of ℓ_{D_v}. For most solids we have $K_0 \sim 10^{-2}$ Å^{-1}, the coefficient I_{D_v} given by Eq. (10) is of the order of 0.1, and we get

$$\ell_{D_v} \sim 10^9 \text{ Å}^4 \frac{k^2}{\xi}\frac{N_0}{\Delta N}\left(\frac{\xi}{R_0}\right)^{3-D_v}. \tag{17}$$

If we put $k = 1.0$ Å^{-1}, $\xi = 10^4$ Å, $R_0 = 10$ Å, and $D_v = 2.5$ we obtain $\ell_{D_v} = 3\times 10^{-2}$ cm$(N_0/\Delta N)$. The similar estimation for $D_v = 3$ gives a much smaller value for $\ell_3 = 10^{-3}$ cm$(N_0/\Delta N)$. We see that if the ratio $\Delta N/N_0$ is not very small, the mean free path may be much less than the sample thickness L in the real experimental conditions. We note also that ℓ_{D_v} decreases strongly with k^2 and ξ^{-1}.

b. Surface fractals. We consider now a system of pores with the size of the order of ξ and the fractal surface characterized by the dimension D_s restricted by the condition $2 \le D_s < 3$. In Refs. 18, 5, and 19 it was shown that the small-angle scattering may be expressed as

$$\frac{d\sigma}{d\Omega} = 4\pi(bN_0)^2 V\, c(1-c)\frac{1}{q}\int_0^\infty dr\, r\,\gamma(r)\sin qr, \tag{18}$$

where V is the sample volume, $1-c = \Delta N/N_0$, and if $r \ll \xi$ one gets

$$\gamma(r) = 1 - \frac{V_b(r)}{4c(1-c)V}, \tag{19}$$

where $V_b(r)$ is the volume of the boundary layer of the thickness r at the surface of the pores. For the surface fractal $V_b(r)$ can be expressed as[5]

$$V_b \sim \frac{\delta V}{\xi^3} S_\xi \left(\frac{\xi}{r}\right)^{D_s-2} r, \tag{20}$$

where δV is the part of the volume V occupied by pores, the ratio $\delta V/\xi^3$ is of the order of the total number of pores in the volume V, $S = \xi^2$ is of the order of the pore surface measured at a scale of the order of ξ and the factor $(\xi/r)^{D_s-2}$ describes the enhancement of the pore surface if it is measured at a scale $r < \xi$.

It is convenient to rewrite Eq. (20) in the following form:

$$V_b = C_b V_3(\xi) N_p\, (\xi/r)^{D_s-3}; \tag{21}$$

here $N_p = \delta V/\xi^3$ is of the order of the total pore number in the sample and $C_b \sim 1$ is a nonuniversal constant. As a result of Eqs. (18), (19), and (21) we get for the scattering cross section at $q \gg \xi^{-1}$[5]

$$\frac{d\sigma}{d\Omega} = \frac{K_0^4 V_3(\xi) C_b \Gamma(5-D_s)\sin[(D_s-1)\pi/2]}{16\pi q^3 (q\xi)^{3-D_s}}. \tag{22}$$

If $D_s \to 3$ the cross section vanishes. This result has been discussed in Refs. 6 and 19. At $D_s = 2$ Eqs. (6) and (22) have the same order as they should have, because in this case the fractal surface turns into a smooth one.

We did not get an interpolation expression for all q, similar to Eq. (4). We can only estimate the total cross section using Eq. (22) and ξ^{-1} as a low q cutoff. As a result we obtain

$$\frac{1}{\ell_{D_s}} = I_{D_s} K_0^4 \frac{\Delta N}{N_0}\frac{\xi}{k^2}, \tag{23}$$

where

$$I_{D_s} \sim \frac{1}{8} C_b \Gamma(5-D_s)\sin\left[\frac{\pi(D_s-1)}{2}\right]. \tag{24}$$

Comparing Eqs. (23) and (13) we see that the mean free path for pores with fractal and smooth boundaries have the same order of magnitude. Correspondingly the estimation of ℓ_{D_s} coincides with that given above for ℓ_3.

III. MULTIPLE SCATTERING

The general theory of small-angle multiple scattering (the Molière theory) is presented in Ref. 12. It was shown in Ref. 8 that it is actually based on two assumptions: (1) the total cross section is saturated in the small-angle region; (2) the mean free path is large compared to the size of inhomogeneities ξ. In our case both conditions hold if $D_v > 2$ and for all values of D_s.

According to Refs. 12 and 8 for the intensity of the multiple scattering we have

$$I(q) = \frac{1}{2\pi}\int_0^\infty dn\, n J_0\left(\frac{qn}{k}\right) \times\left[\exp\left(-\frac{LF_n}{\ell}\right) - \exp\left(-\frac{L}{\ell}\right)\right], \tag{25}$$

where $F_n = (\sigma_0 - \sigma_n)/\sigma_0$

$$\sigma_n = \frac{2\pi}{k^2}\int_0^\infty dq\,q\,\frac{d\sigma}{d\Omega}\,J_0\left(\frac{nq}{k}\right) \tag{26}$$

is the coefficient in the expansion of $d\sigma/d\Omega$ in Legendre polynomials, and the approximate expression $P_n(\cos\vartheta) = J_0(\vartheta n)$ is used, which holds if $\vartheta \ll 1$.[13] Obviously σ_0 coincides with the total cross section. For $L \ll \ell$ Eq. (25) gives the intensity of the single scattering which is proportional to L.

From Eqs. (4) and (22) we see that in both cases $d\sigma/d\Omega$ may be represented as $Aq^{-\Delta} f_\Delta(q\xi)$, where A is a constant, $\Delta = D_v$ or $6 - D_s$, $f_\Delta(x) = 1$ if $x \gg 1$ and $q^{-\Delta} f_\Delta(q\xi)|_{q\to 0} < \infty$. As a result we obtain

$$F_n = \int_0^\infty dx\,x^{-\Delta+1}[1 - J_0(\vartheta_0 n x)]f_\Delta(x) \times \left[\int_0^\infty dx\,x^{-\Delta+1} f_\Delta(x)\right]^{-1}, \tag{27}$$

where $\vartheta_0 = 1/(k\xi)$.

For $L \gg \ell$ the main contribution to $I(q)$ is in the region where $F_n \ll 1$. In this case we see from Eq. (27) that $n\vartheta_0 \ll 1$ and in the numerator we may replace $f_\Delta(x)$ by unity. As a result we get

$$F_n = (\vartheta_0\, n/2)^{\Delta-2}/g_\Delta, \tag{28}$$

where g_Δ is given by

$$g_\Delta = \frac{(\Delta-2)\Gamma(\Delta/2)}{\Gamma(2-\Delta/2)}\int_0^\infty dx\,x^{1-\Delta} f_\Delta(x). \tag{29}$$

From Eqs. (25) and (28) we obtain for $L \gg \ell$

$$I(q) = \frac{k^2}{2\pi q^2}\int_0^\infty dx\,x J_0(x) \times \exp\left[-\left(\frac{L}{\ell g_\Delta}\right)\left(\frac{x}{2q\xi}\right)^{\Delta-2}\right], \tag{30}$$

where $\Delta = D_v$ and $\Delta = 6 - D_s$ for the volume and surface fractals, respectively. Equation (1) is an immediate consequence of this expression.

It is convenient to rewrite this equation as

$$I(q) = \frac{\mu k^2}{2\pi[q_L^{(\Delta)}]^2}\int_0^\infty dy\,y^{2\mu-1} J_0\left(\frac{q}{q_L^{(\Delta)}}y^\mu\right)e^{-y}, \tag{31}$$

where $\mu = (\Delta - 2)^{-1}$.

Using the well-known expressions[13] $\int_0^\infty dq\,q\,J_0(qy) = 4\delta(y)/y$ and $\int_0^\infty dy\,\delta(y) = 1/2$ one can easily check that the total scattering intensity is equal to unity as it should be in the case of multiple scattering when all particles are declined by the sample.

If we put $\Delta = 4$ and $g_\Delta = 1$ in Eq. (30) we get the random-walks result

$$I(q) = \frac{k^2}{4\pi(q_L^{\mathrm{RW}})^2}\exp\left[-\left(\frac{q}{2q_L^{\mathrm{RW}}}\right)^2\right], \tag{32}$$

where $q_L^{\mathrm{RW}} = (L/\ell)^{1/2}/(2\xi)$.

However, for $\Delta = 4$ we obtain from Eq. (27)

$$F_n = (\vartheta_0 n/2)^2 \ln\frac{1}{\vartheta_0 n} \tag{33}$$

and the intensity is described by Eq. (32) if we replace q_L^{RW} by

$$q^{(4)} = [(L/\ell)\ln(L/\ell)]^{1/2}/(2\xi). \tag{34}$$

It is the result for the finite-size inhomogeneities considered in Ref. 8. We see that now we have random walks with steps which increase logarithmically with the sample thickness L. Such a behavior is related to the fact that according to Eq. (2) the mean square of the scattering angle is ill defined also in the Porod case.

For the case $\Delta = 2$ we have

$$F_n = [\ln(1/n\vartheta_0)]/\ln(k\xi) \tag{35}$$

and the scattering remains the small-angle one in rather narrow region of L. Corresponding expressions are presented in Ref. 8. For $\Delta = 3$ from Eq. (30) we get

$$I(q) = \frac{k^2 q_L^{(3)}}{2\pi(q^2 + q_L^{(3)\,2})^{3/2}}, \tag{36}$$

where $q_L^{(3)} = L/(2g_3\ell\xi)$. As was shown in Ref. 9 this expression describes the multiple scattering from spheres with radii $R = g_3\xi$ if the following conditions are fulfilled. (1) The single scattering is a diffraction on impenetrable sphere and is given by

$$\frac{d\sigma}{d\Omega} = k^2 R^4 J_1^2(qR)/(qR)^2. \tag{37}$$

(2) The concentration of spheres is low and the mean free path

$$\ell = 2RV/3\Delta V \gg R, \tag{38}$$

where $\Delta V/V$ is the relative volume of the sample occupied by the spheres. The diffractional scattering is accompanied by refraction. In Ref. 9 it was shown that Eq. (36) describes the multiple scattering at the condition

$$L \gg \ell[kR(U/E)]^2 \ln[kR(U/E)], \tag{39}$$

where E is the neutron energy, U is given by Eq. (7), and $kR(U/E) \gg 1$.

Let us consider now the form of the intensity distribution as function of q. First of all for the forward scattering intensity calculated from Eq. (31) we get

$$I(0) = \frac{\mu k^2\Gamma(2\mu)}{2\pi[q_L^{(\Delta)}]^2} = \frac{2\mu(k\xi)^2}{\pi}\left(\frac{g_\Delta\ell}{L}\right)^{2\mu}\Gamma(2\mu), \tag{40}$$

where $2\mu = 2/(D_v - 2) > 2$ and $2 > 2\mu = 2/(4 - D_s) > 1$ for the volume and surface fractals, respectively. In both cases $I(0)$ decays with L faster than L^{-1} as in the

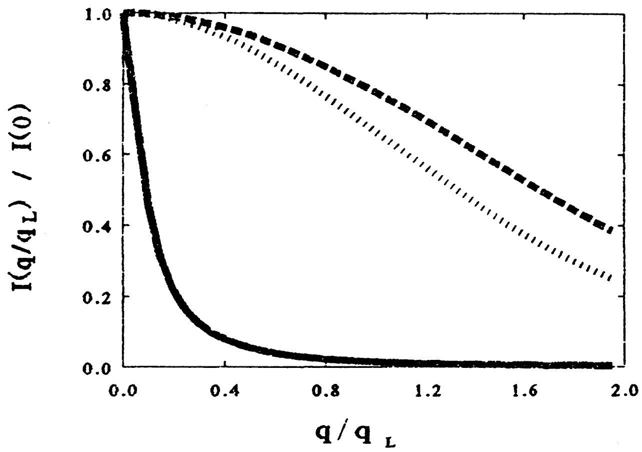

FIG. 1. The central part of the normalized multiple-scattering intensity as a function of q/q_L for the volume fractals with $D_v = 2.5$ (full line), surface fractals with $D_s = 2.5$ (dotted line), and random walks (dashed line).

random-walks case [see Eq. (32)] which corresponds to two-dimensional diffusion. Such a behavior is a characteristic feature of the superdiffusion.[11]

In the small-q region where $q < q_L^{(\Delta)}$ Eq. (31) for $I(q)$ may be expanded in a power series in q^2 and for $I(q)$ we have the quasi-Guinier expression

$$I(q) = I(0)\left[1 - \frac{1}{3}q^2 R_g^2(L)\right], \tag{41}$$

where we for the effective radius of the gyration obtain

$$R_g^2(L) = \frac{3\Gamma(4\mu)}{4\Gamma(2\mu)[q_L^{(\Delta)}]^2}. \tag{42}$$

Here the numerical coefficient is minimal for $\mu = 1/2$, i.e., for the random walks and increases with μ. This increase is rather moderate for the surface fractals ($1/2 < \mu < 1$) and becomes very strong for the volume fractals. For example, $[q_L^{(\Delta)} R_g(L)]^2 = 0.75$, 1.26, and 630 for $\mu = 1/2$ (RW), $\mu = 2/3$ ($D_s = 2.5$), and $\mu = 2$ ($D_v = 2.5$), respectively. We see that in the range $(q/q_L^{(\Delta)}) \lesssim 1$ the intensity distribution contracts with increasing value of μ as it is shown in Fig. 1. For the volume fractals this contraction is very strong and the quasi-Guinies region almost disappears as it is seen in Fig. 1. One can show that in this case the q^2 expansion of $I(q)$ is an asymptotic one.

If $q \gg q_L^{(\Delta)}$ we can expand the exponent in Eq. (30) and obtain the same expression as for the single scattering. However, now it holds for $q \gg q_L^{(\Delta)}$ only. Moreover, the total intensity in the single-scattering case is proportional to $(L/\ell) \ll 1$. For the multiple scattering it is equal to unity because all particles are declined by the sample and contributions from two regions: $q < q_L^{(\Delta)}$ and $q > q_L^{(\Delta)}$ have the same order of magnitude. The single-scattering behavior of the intensity at $q \gg q_L^{(\Delta)}$ is related to the rare scattering events and the corresponding mean free path is larger than L. The low-q multiple scattering before and after the rare large-q event declines the particle on $q \sim q_L^{(\Delta)}$ and cannot change appreciably the intensity distribution for $q \gg q_L^{(\Delta)}$.

IV. CONCLUSIONS

The main result of our study is the following. The random-walk diffusional description of the small-angle multiple scattering fails in many physically significant cases because the mean square of the scattering angle $\overline{\vartheta^2}$ for the single scattering event is an ill-defined quantity. Even in the case of finite-size inhomogeneities the step of the random walks logarithmically increases with the sample thickness L.

We estimate the mean free path ℓ for the neutron propagation in the presence of the large-scale inhomogeneities using the value for $K_0^2 = 4\pi N_0 b$ typical for porous or two component solids and show that ℓ may be much less than commonly used sample thickness L and in this case the multiple scattering is important. In this case the intensity distribution divides into two parts: the tail where $q \gg q_L^{(\Delta)}$ and the central part where $q \lesssim q_L^{(\Delta)}$, where $q_L^{(\Delta)}$ is given by Eq. (1). The tail has the same form as for the single scattering and its intensity is proportional to the sample thickness L. For $q \ll q_L^{(\Delta)}$ the scattering intensity has the quasi-Guinier form given by Eq. (41). However, this approximation is hardly applicable for the volume fractals where the effective radius of the gyration is very large and the q^2 expansion of the intensity $I(q)$ is an asymptotic one. In any case the effective radius of the gyration $R_g(L) \sim 1/q_L^{(\Delta)}$ is less than the fractal size ξ. Correspondingly for the reliability of the determination of ξ from the crossover from the large-q scattering to the Guinier regime one should check if the width of the central part of the intensity distribution is L independent.

Above we estimated the mean free path using the value $K_0^2 = 4\pi N_0 |b|$ for the porous solids. This estimation is applicable, for example, for the coal porosity discussed in Ref. 5. However, this estimation fails for a lot of other systems where the variation of bN_0 originating from disorder is much less. As an example one can mention volcanic rocks,[20] aggregated colloid particles,[1,21,22] and similar systems. In all these cases the mean free path is much larger than estimated above. However, from Eqs. (12)–(14) and (23) we see that ℓ is proportional to the neutron energy and the multiple scattering should be important for the sufficiently cold neutrons. We note also the multiple scattering remains the small-angle one if the sample thickness L is much less than the critical value L_0 determined by the condition $q_L^{(\Delta)} \sim k$. From Eq. (1) we obtain

$$L_0 \sim (k\xi)^{\Delta-2}\ell. \tag{43}$$

It may be shown that this expression has the same order of magnitude as the transport mean free path determined by

$$1/\ell_{\mathrm{tr}} = n_F \int d\Omega\,(d\sigma/d\Omega)(1-\cos\vartheta). \tag{44}$$

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