Supplementary material to "Scattering from surface fractals in terms of composing mass fractals"

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In Sec. I some important issues concerning small-angle scattering (SAS) are discussed. In Sec. II general remarks about SAS from mass fractals are presented. In Sec. III the influence of polydispersity on SAS curves from surface fractals, is shown, and in Sec. IV is presented a figure with a power-law distribution of balls.

I. THEORETICAL BACKGROUND FOR SMALL-ANGLE SCATTERING

In a very good approximation, the differential cross section of a sample exposed to a beam of neutrons, X-rays or light is given by [1, 2] $\mathrm{d}\sigma/\mathrm{d}\Omega = |A(\boldsymbol{q})|^2$, where $A(\boldsymbol{q}) \equiv \int_{V'} \rho_{\mathrm{s}}(\boldsymbol{r})e^{i\boldsymbol{q}\cdot\boldsymbol{r}}\mathrm{d}^3r$ is the total scattering amplitude, V' is the total volume irradiated by the incident beam, and the scattering length density $\rho_{\mathrm{s}}(\boldsymbol{r})$ is defined with the help of Dirac's δ function: $\rho_{\mathrm{s}}(\boldsymbol{r}) = \sum_j b_j\delta(\boldsymbol{r}-\boldsymbol{r}_j)$. Here, $\boldsymbol{r}_j$ are the positions of microscopic objects like atoms or nuclei with the scattering lengths b_j.

Let us consider a sample consisting of rigid homogeneous objects of the density ρ_{m}, which are immersed into a solid matrix of density ρ_{p}, and suppose that their spatial positions and orientations are uncorrelated (this assumes that the concentration of the objects in the solid matrix is low enough). Then the scattering intensity (differential cross section per unit volume of the sample) can be written as

$$I(q) \equiv \frac{1}{V'}\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = n|\Delta\rho|^2V^2\left\langle|F(\boldsymbol{q})|^2\right\rangle, \qquad (1)$$

where n is the concentration of the objects in the irradiated volume, $\Delta\rho = \rho_{\mathrm{m}} - \rho_{\mathrm{p}}$ is the scattering contrast, V is the volume of each object and $F(\boldsymbol{q})$ is the normalized scattering amplitude (form factor) of the object

$$F(\boldsymbol{q}) = \frac{1}{V}\int_V e^{-i\boldsymbol{q}\cdot\boldsymbol{r}}\mathrm{d}\boldsymbol{r}, \qquad (2)$$

obeying the condition $F(0) = 1$. Here, the symbol $\langle\cdots\rangle$ stands for the ensemble averaging over all orientations of the objects. If the probability of any orientation is the same, then it can be calculated by integrating over all directions of the scattering vector $\boldsymbol{q}$ [3].

It is easy to derive a few useful properties of the form factor (2), which are valid for a particle of arbitrary shape.
i) Scaling: if we scale all the lengths of the particle as $l \to \beta l$ then $F(\boldsymbol{q}) \to F(\beta\boldsymbol{q})$.
ii) Translation: if the particle is translated $\boldsymbol{r} \to \boldsymbol{r}+\boldsymbol{a}$ then $F(\boldsymbol{q}) \to F(\boldsymbol{q})\exp(-i\boldsymbol{q}\cdot\boldsymbol{a})$.
iii) Rotation: if the particle is rotated with an orthogonal matrix $\boldsymbol{r} \to \hat{O}\boldsymbol{r}$ then $F(\boldsymbol{q}) \to F(\hat{O}^{\mathrm{T}}\boldsymbol{q})$. Recall that the inverse of an orthogonal matrix is equal to the transpose of it $\hat{O}^{-1} = \hat{O}^{\mathrm{T}}$, where $(\hat{O}^{\mathrm{T}})_{ij} = \hat{O}_{ji}$.
iv) Additivity of the nonnormalized scattering amplitude: if a particle consists of two not overlapping subsets I and II, then $F(\boldsymbol{q}) = \big(V_IF_I(\boldsymbol{q}) + V_{\mathrm{II}}F_{\mathrm{II}}(\boldsymbol{q})\big)/(V_{\mathrm{I}}+V_{\mathrm{II}})$.

The average over all directions of the scattering vector $\boldsymbol{q}$ in Eq. (1) is analogous to diffraction with an uncollimated beam in optics [4]: the interference patterns of plane waves, coming from different directions, superimpose upon each other. This results in strong *spatial incoherence* [5]: for the subsets *I* and *II*, the correlator $\langle F_I(\boldsymbol{q})F_{II}(\boldsymbol{q})\rangle$ decays when $q \gg 2\pi/r$, where r is of order of the distance between their centers [4]. This indicates the border between the coherent regime (where the scattering *amplitudes* V_IF_I and $V_{II}F_{II}$ should be added) and incoherent regime (where the scattering *intensities* $\langle|V_IF_I|^2\rangle$ and $\langle|V_{II}F_{II}|^2\rangle$ should be added). This can be illustrated by a simple example of the SAS intensity from two point-like objects, placed rigidly the distance l apart. If each of them has the unit amplitude, the intensity is written as $I(q) = \langle|e^{i\boldsymbol{q}\cdot\boldsymbol{r}_1} + e^{i\boldsymbol{q}\cdot\boldsymbol{r}_2}|^2\rangle$, which yields after averaging over the solid angle

$$I(q) = 2\left(1+\frac{\sin ql}{ql}\right). \qquad (3)$$

A fast decay of the coherence can be seen from Fig. 1 when $ql \gg 2\pi$.

For a "primary" object like a ball or cube of total size l, the intensity $\langle|F(\boldsymbol{q})|^2\rangle$ is of order one in the Guinier range $q \lesssim 2\pi/l$ and decays as $1/q^4$ in the Porod range $q \gtrsim 2\pi/l$ [1].

Almost all scattering properties of a complex object can be understood by means of the above simple properties of composing "primary" objects and transitions from coherent to incoherent scattering regimes. In the next section, we outline and explain some basic properties of mass and surface fractals.

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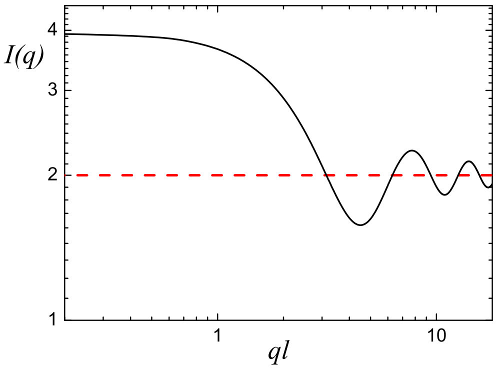

FIG. 1. (Color online) The SAS intensity (3) from an ensemble of two point-like objects with unit amplitude, placed rigidly the distance l apart but randomly oriented. One can see the transition from the coherent regime [$I(q) = 4$] to the incoherent regime [$I(q) = 2$] when $q \gg 2\pi/l$: only a very few minima and maxima with decaying amplitudes are quite pronounced. The fast decay of the correlator $\langle e^{i\boldsymbol{q}\cdot(\boldsymbol{r}_2-\boldsymbol{r}_1)}\rangle$ is due to the average over all directions of the scattering vector $\boldsymbol{q}$, which is analogous to diffraction with an uncollimated beam in optics [4]: the interference patterns of plane waves, coming from different directions, superimpose upon each other. This results is the same as if the strong spatial incoherence of the incident beam is realized.

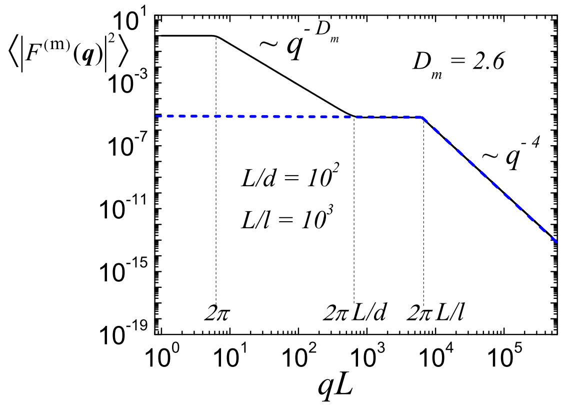

FIG. 2. (Color online) Generic normalized SAS intensity from mass fractals with a single scale (solid black line). The intensity shows the presence of the four main regimes: Guinier (at small q), fractal (at intermediate q), plateau (at larger q), and Porod (at high q). The characteristic lengths L, d, and l are explained in the text. The blue dashed line shows the approximation of completely uncorrelated primary objects, composing the mass fractal. The scattering intensity of the object (like cube or ball) consists of the Guinier and Porod regions only. Note that a typical experimental SAS tool has the dynamic Q-range $q_{max}/q_{\min}$ about two or three orders, so only a part of the shown curve can be observed in practice.

II. A MASS FRACTAL WITH A SINGLE SCALE

The scattering properties of mass fractals with a single scale were studied in detail in the previous publications [4, 6].

For a mass fractal of the total length L, composed of p small "primary" structural units of size l separated by distances d ($l \lesssim d \ll L$), the normalized form factor can be estimated qualitatively by the formula

$$\langle|F^{(\mathrm{m})}(\boldsymbol{q})|^2\rangle \simeq \begin{cases} 1, & q \lesssim 2\pi/L, \\ (qL/2\pi)^{-D_\mathrm{m}}, & 2\pi/L \lesssim q \lesssim 2\pi/d, \\ (d/L)^{D_\mathrm{m}}, & 2\pi/d \lesssim q \lesssim 2\pi/l, \\ (d/L)^{D_\mathrm{m}}(ql/2\pi)^{-4}, & 2\pi/l \lesssim q, \end{cases} \tag{4}$$

(see Fig. 2). Here p is of the order of $(L/d)^{D_\mathrm{m}}$ in accordance with the definition of the fractal dimension.

Such a fractal can be constructed with a simple iteration rule (an example is the Cantor dust): a "primary" object like a ball or cube or another simple shape generates k objects of the same shape but of the size scaled by the factor β_s, which is smaller than one in general. The initial single object (zero iteration) has the size of order r_0. Then after n iterations, the total number of the objects is equal to $p = k^n$, and they all are put somehow inside a form of the total size L. The distances between the objects and their sizes are of order $d = \beta_\mathrm{s}^n L$ and $l = \beta_\mathrm{s}^n r_0$, respectively. The mass fractal has the Hausdorff dimension D_m obeying the relation [7] $k\beta_\mathrm{s}^{D_\mathrm{m}} = 1$.

Equation (4) explicitly shows that the SAS intensity of mass fractal is characterized by the four main regions: Guinier at $q \lesssim 2\pi/L$, fractal at $2\pi/L \lesssim q \lesssim 2\pi/d$, a plateau at $2\pi/d \lesssim q \lesssim 2\pi/l$, and Porod regime at $q \gtrsim 2\pi/l$.

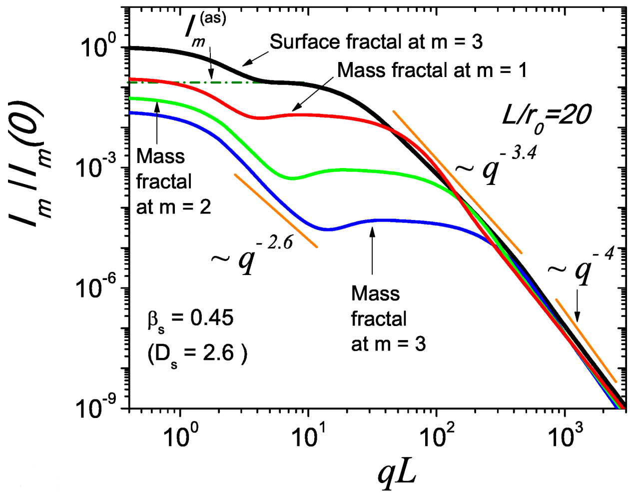

FIG. 3. (Color online) Polydisperse scattering with relative variance $\sigma_\mathrm{r} = 0.4$.

We make a few remarks here. First, the intensity in the Guinier range is actually parabolic: $I(q) \simeq I(0)(1-R_\mathrm{g}^2 q^2/3)$, where R_g is the radius of gyration. This parabolic behavior of the intensity is ignored in the above estimations for the sake of simplicity. Second, the *mass* fractal region appears due to *spatial correlations* between the composing "primary" units [4, 6]. For this reason, the fractal region of the mass fractal is determined by the maximal and minimal distances between the centers of the structural units. Third, the plateau at $2\pi/d \lesssim q \lesssim 2\pi/l$ in the scattering intensity can be consid-

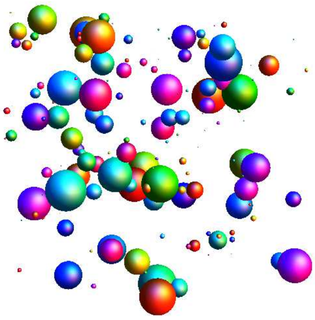

FIG. 4. (Color online) A set of balls distributed over sizes.

ered as a Guinier region for the primary unit (which is of the same size l), because the spatial correlations between different units are not important in this region, and thus the total intensity is equal to p times the intensity of the primary unit (see the discussion in Sec. I). For the normalized intensity of primary globular unit of size l, one can adopt the Porod-law relation

$$\left\langle |F_0(\boldsymbol{q})|^2 \right\rangle \simeq \begin{cases} 1, & q \lesssim 2\pi/l, \\ (ql/2\pi)^{-4}, & 2\pi/l \lesssim q. \end{cases} \tag{5}$$

As discussed above, it coincides with the last two rows in Eq. (4) up to the factor $(d/L)^{D_{\mathrm{m}}} = 1/p$, which appears due to the chosen normalization of the total intensity of mass fractal at zero momentum. The latter is equal to p^2 times the intensity of the primary unit (the coherent regime). Then neglecting all the spatial correlations *between* the primary objects (units), composing the fractal, yields the scattering intensity shown by the dashed (blue) line in Fig. 2. Fourth, the "pure" power-law functions with different exponents, given by Eq. (4) and shown in Fig. 2, is a simplification of an actual behaviour of the intensity. Actually, there is a complex pattern of maxima and minima superimposed on the power-law decays. However, this pattern is smeared and can disappear completely when the polydispersity is developed [4, 6].

III. POLYDISPERSE FRACTAL FORM FACTOR

In most cases, a real system consists of fractals of various sizes and forms (polydispersity). We can model polydispersity by considering an ensemble of the fractals with different lengths l of the initial cube taken at random (that is, l is here the length of the initial cube and the ratio l/r_0 is held constant over the ensemble. Note that in the previous sections, we denote the length of the initial cube L, while in the presence of polydispersity, L is the mean value of the cube length over the ensemble.

The distribution function $D_N(l)$ of the fractal sizes is defined in such a way that $D_N(l)\mathrm{d}l$ gives the probability of finding a fractal whose size falls within the range $(l, l+\mathrm{d}l)$. We consider here quite common log-normal distribution

$$D_N(l) = \frac{1}{\sigma l (2\pi)^{1/2}} \exp\left(-\frac{[\log(l/L) + \sigma^2/2]^2}{2\sigma^2} \right), \tag{6}$$

where $\sigma = [\log(1+\sigma_{\mathrm{r}}^2)]^{1/2}$. The quantities L and σ_{r} are the mean length and its coefficient of variation (that is, the ratio of the standard deviation of the length to the mean length), called also relative variance

$$L \equiv \langle l \rangle_D\,, \quad \sigma_{\mathrm{r}} \equiv (\langle l^2 \rangle_D - L^2)^{1/2}/L, \tag{7}$$

where $\langle \cdots \rangle_D \equiv \int_0^\infty \cdots D_N(l)\mathrm{d}l$. Therefore, by using Eqs. (1) and (6) the polydisperse intensity becomes (see Fig. 3)

$$I_m^{(\mathrm{s})}(q) = n\,|\Delta\rho|^2 \int_0^\infty \left\langle \left| F_m^{(\mathrm{s})}(\boldsymbol{q}) \right|^2 \right\rangle V_m^2(l) D_N(l)\mathrm{d}l, \tag{8}$$

where $F_m^{(\mathrm{s})}(\boldsymbol{q})$ is the scattering amplitude of the Cantor-like surface fractal, given by Eq. (18) of the main paper.

IV. RANDOM DISTRIBUTION OF BALLS

A typical set of balls whose radii follow some distribution is displayed in Fig 4.

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