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A modelling study of scattering behaviour of 2D DBM fractals

Zeng Yanwei†‡, Wei Wang†§ and Sergio Meriani†‖

† International Centre for Science and Technology, ICS-Trieste, Italy

‡ Department of Materials Science & Engineering, Nanjing University of Chemical Technology, Nanjing 210009, People's Republic of China

§ Department of Physics, Nanjing University, Nanjing 210008, People's Republic of China

‖ Department of Materials Engineering, University of Trieste, Trieste 34127, Italy

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Abstract. A scattering equation has been developed by considering power-law correlation and geometrical details of fractals, and then used to describe the scattering behaviour of circular and sharply bounded two-dimensional random fractal patterns generated with the dielectric breakdown model (DBM). It is shown that the scattering equation may, on the whole, reasonably describe the ideal coherent scattering profiles calculated by starting from the coordinates of all scatterers that make up the model fractals, while an excellent fit to the scattering profiles requires taking into account the additional contribution from the correlation of local density fluctuations. A scaling invariance properly between the scattering intensity profiles from fractals of different sizes has also been manifested and discussed.

1. Introduction

Over the past decades, fractal phenomena ubiquitously present in a variety of physical, chemical, and biological systems, which display fractal features such as dentritic growth, diffusion-limited aggregation, dielectric breakdown, viscous fingering, electrochemical deposition, growth of bacterial colonies, etc, have experienced extensive study of both theoretical and experimental aspects [1, 2]. In order to experimentally identify and recognize fractal features of a specific structure or pattern occurring in nature or the laboratory, several useful methods and techniques have been developed. From the box-counting principle, for instance, the imaging analysis of aggregate structures under the electron microscope (usually digitized and computerized) has been used for directly detecting their fractal nature [3]. Nevertheless, the coherent scattering of x-rays, neutrons or laser light by fractal objects seems to be one of the most convenient and effective tools for evaluating the structural parameters or correlationships of fractals. Such a technique is based on the fact that fractal objects usually possess a density correlation characterized by power law $c(r) \sim r^{Df-3} f(r/\xi)$, with $f(r/\xi)$ being called the scaling function. As a consequence, quasi-power-law $I(q) \sim q^{-Df}$ scattering intensity profiles emerge in the 'fractal regime' of scattering space [4–6] and the corresponding fractal dimension D_f can be directly read out from a double-logarithmic plot of the scattering intensity versus the momentum transfer. For a quantitative interpretation of the scattering data from real fractal objects, however, the scaling function $f(r/\xi)$ usually requires particular considerations for the presence of finite-size effects arising from the correlation cut-off at a large scale of real fractal aggregates or clusters. It seems that Sinha *et al* [7] first phenomenologically assumed the function $f(r/\xi)$ as an exponential $\exp(-r/\xi)$ to take into account this finite-size effect with the parameter ξ interpreted as the aggregate size. Also, a function $\exp[-(r/\xi)^2]$ was raised instead of $\exp(-r/\xi)$ in [8]. It is clearly evident that the advantages of such *a priori* setting of scaling functions allow one to readily establish a mathematic expression of structure factor for the fitting analyses of scattering data. Several examples using such a scaling function can be found in the scattering researches on colloidal aggregates of particles of gold, silica and others [9–11], but it should be noted that a scaling function such as $\exp(-r/\xi)$ is not universal since it only gives a rough structural picture of some boundaries of fractals, which in reality, as argued in our research concerning the finite-size effect on the scattering behaviour [12], may show more complications than could be described by a simple exponential $\exp(-r/\xi)$ due to the presence of structural inhomogeneity and diverse geometrical structures of the boundaries. Therefore, a blind use of a simply preset scaling function to data-fitting analyses would probably lead to ill evaluated structural parameters when the correlation function of the fractals does not follow the preset one, even though sometimes the final fitting could be mathematically well accepted.

In the present work, we report on an investigation of the scattering behaviour of some circular and sharply bounded 2D fractals generated via computer with the dielectric breakdown model (DBM). The research consists in, first, working out coherent scattering intensities by starting from point scatterers of the model fractals, which might be well regarded as ideal coherent scatterings; secondly, quantitatively interpreting these scattering data by incorporating circular and sharp boundaries of the model fractals into a scattering equation built up on the basis of point-scattering theory together with fractal power-law correlation. It is shown that the scattering equation, in which a scaling function different from Sinha's was constructed according to the particulars of the model fractals' boundaries and physically well clarified, provides a fairly good fit to those ideal scattering profiles over the entire q range in the studies. In contrast, the use of scaling function $\exp(-r/\xi)$ failed to give a good enough description when the structural parameters were evaluated with the right values. It is more interesting that an excellent fit to the ideal scattering profiles can be achieved when an additional contribution ascribed to the correlation of local fluctuation of density distribution inside the fractals is taken into account. This research, in fact, by exemplifying sharply bounded fractals, has proved the importance and the feasibility of establishing a correct scattering equation to reveal the structural details of real fractals, which may well be of more complicated boundaries than the present one, such as gradual transition boundaries characterized by some 'width', for instance. Moreover, the present work suggested an additional contribution to the scattering intensity from the correlation of local fluctuations of scatterer density distribution. This argument is supposed to deserve further study and more attention when a detailed description of fractal aggregates by the scattering technique is involved.

2. Dielectric breakdown modelling and scattering of 2D fractals

The 2D fractal patterns used in the present modelling study were generated through a standard growth procedure of the DBM [11], in which the Laplace equation $\nabla^2 U = 0$ is numerically simulated on a 2D point lattice with unit space between any two neighbouring points in longitudinal and latitudinal directions. At the beginning, a seed 'particle' is placed at a lattice point chosen as the centre of the lattice, from which a circumference of radius $R = 200$ units is assumed. The boundary conditions are set such that $U_{out} = 1$ on the outer circumference and $U_{in} = 0$ at the inner occupied sites. During the pattern's growth process, the growth probability at the ith perimeter bond is chosen as $p_i = \nabla U_i / \Sigma_j I \nabla U_j I$ for every single-step growth. Successively, such a growth mechanism allows a fractal pattern to grow up to a desired size. Figure 1 illustrates a typical 6000-point fractal pattern generated by the above procedure.

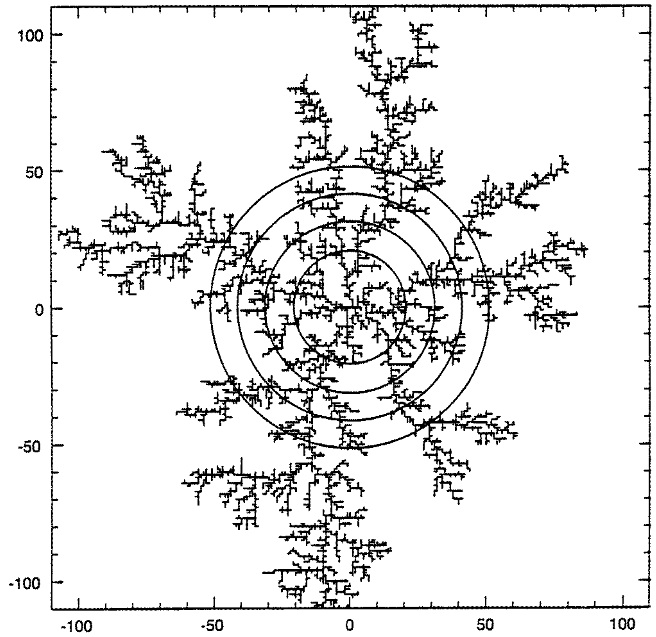

Figure 1. A typical 2D fractal pattern generated via DBM, marked with concentrical circles representative of the off-trimming traces with different radii $R = 20$, 30, 40 and 50 in lattice units.

Over ten such fractals of about 6000 points in each, an average density–density correlation function $c(r) \sim \langle \rho(r')\rho(r'+r) \rangle$ was found to be of a power law: $c(r) \sim r^{-0.3}$, with the mean fractal dimensionality $D_f = 1.7$. The circular and sharply bounded fractal patterns for modelling scatterings were then obtained by peripherally trimming fresh fractals at distances $R = 20$, 30, 40 and 50 (in lattice units) from the centre.

According to the point-scatterer diffraction theory, as in the case of a 3D point assembly, the elastical scattering intensity from a 2D point system hit by a beam of electromagnetic radiation (x-rays, light) or neutrons, normal to the pattern plane, will be given by

$$I_{eu}(\boldsymbol{q}) = I f(\boldsymbol{q}) I^2 N_p [1 + 1/N_p \Sigma\Sigma \exp(\mathrm{i}\boldsymbol{q} \cdot \boldsymbol{r}_{ij})] \tag{1}$$

where N_p is the total number of point scatterers; $I f(\boldsymbol{q}) I^2$ is usually called the form factor corresponding to the intensity scattered by one individual scatterer; $\boldsymbol{q}$ stands for the momentum-transfer's component parallel to the pattern plane. Its modulus $I \boldsymbol{q} I = 2\pi \sin(2\theta)/\lambda$, with θ being the angle between the momentum transfer and the plane, and λ the wavelength of the incident beam. After taking an angular average of (1) due to the random radian orientation of planar point assemblies, which are considered parallel to one another, a structure factor can be obtained:

$$S(q) = I_{eu}(q) / I f(q) I^2 N_p = [1 + 1/N_p \Sigma\Sigma J_0(q r_{ij})] \tag{2}$$

where $J_0(qr_{ij})$ is the zero-order Bessel function of the first kind, and r_{ij} refers to the distance from the ith to the jth scatterer.

The factor $S(q)$ is actually a structure-related function of q, depending on the details of the distribution of scatterers in the plane. By using (2), the simulation of ideal coherent scatterings from circularly trimmed fractals of various radii has been performed. As an example, the averaged scattering profile over ten model fractals of radius $R = 30$ is plotted by squares in figure 2. For the fractals of radii $R = 20$, 40 and 50, the corresponding scattering profiles can be seen in figure 4, below.

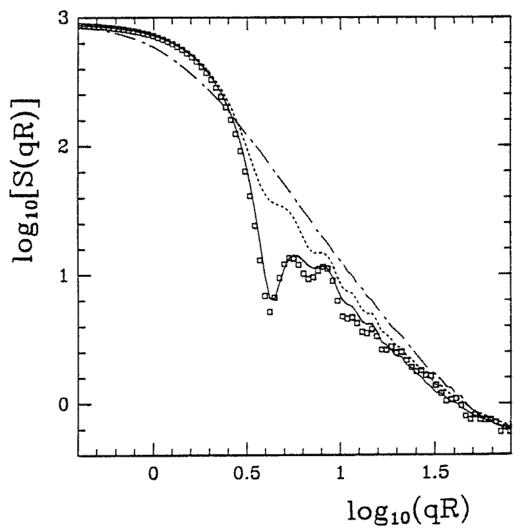

Figure 2. A comparison between the scattering intensity profiles: squares, calculated directly by (2); dotted line, given by (7) together with (6) and (8); solid line, by (7) together with (6), (8), (10) and (11); dot–dashed line, by (7) with $\Phi(r)K(r)$ replaced by scaling function $\exp(-r/\xi)$.

3. The scattering equation for planar fractals

From the point of view of practical structure analysis, it is evidently unfeasible to define a fractal object of random structure as exactly as in the case of crystalline matter. Owing to its dilation symmetry, however, a fractal structure is expected to be statistically characterized and described by a set of structural parameters. In order to achieve this goal by starting from the scatterings by fractals instead of their scatterers' coordinates, one needs to set up a scattering equation in terms of the density correlation and geometrical details of the fractals.

According to the point-scattering theory, as in the case of a 3D system, the scattering intensity for a 2D point-scatterer assembly is given by

$$I_{eu}(\boldsymbol{q}) = If(\boldsymbol{q})I^2 \iint_{-\infty}^{+\infty} \rho(\boldsymbol{u})\rho(\boldsymbol{u}')\sigma(\boldsymbol{u})\sigma(\boldsymbol{u}') \exp(-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{u}) \exp(\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{u}') \,\mathrm{d}\boldsymbol{u}\,\mathrm{d}\boldsymbol{u}' \qquad (3)$$

where $If(\boldsymbol{q})I^2$ has the same meaning as in (1); $\rho(\boldsymbol{u})$ is the scatterer density distribution function consistently defined in the whole plane for an infinite scattering object; and $\sigma(\boldsymbol{u})$ refers to the shape function used to characterize the shape, size, and boundary structure of a finite-sized scatterer assembly.

By setting $\boldsymbol{u}' = \boldsymbol{u} - \boldsymbol{r}$ and noting that the above integration with respect to $\boldsymbol{u}$ may be effectively performed only in the overlapping region of $\sigma(\boldsymbol{u})$ and $\sigma(\boldsymbol{u}+\boldsymbol{r})$, (3) can be rewritten as

$$I_{eu}(\boldsymbol{q}) = If(\boldsymbol{q})I^2 N_p \int_{-\infty}^{+\infty} \Phi(\boldsymbol{r})K(\boldsymbol{r})g(\boldsymbol{r}) \exp(-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r})\, \mathrm{d}\boldsymbol{r} \tag{4}$$

where $\Phi(\boldsymbol{r}) = \Phi_0(\boldsymbol{r})/A$, with $\Phi_0(\boldsymbol{r})$ being the convolution square of the shape function $\sigma(\boldsymbol{u})$, A the total surface area occupied by the fractal pattern; $N_p = A\langle\rho(\boldsymbol{u})\rangle_A$ (hereafter the angular brackets stand for an average: $\langle * \rangle = (\int * \,\mathrm{d}u/A)$; $g(r) = \langle\rho(\boldsymbol{u})\rho(\boldsymbol{u}+\boldsymbol{r})\rangle_A/\langle\rho(\boldsymbol{u})\rangle_A$, the ensemble average correlation function of infinite fractals; and $K(\boldsymbol{r})$ is a newly introduced function we call the inhomogeneity function with the following definition:

$$K(\boldsymbol{r}) = \langle\rho(\boldsymbol{u})\rho(\boldsymbol{u}+\boldsymbol{r})\rangle_{A'(r)}/\langle\rho(\boldsymbol{u})\rho(\boldsymbol{u}+\boldsymbol{r})\rangle_A \tag{5}$$

where $A'(r)$ refers to the overlapping region of the functions $\sigma(\boldsymbol{u})$ and $\sigma(\boldsymbol{u}+\boldsymbol{r})$. The function $K(\boldsymbol{r})$ accounts for the influences of density inhomogeneity present within the fractals on their scattering intensity. It can be deduced that $K(\boldsymbol{r})$ is equivalent to the ratio $\langle\rho(\boldsymbol{u})\rangle_{A'(r)}/\langle\rho(\boldsymbol{u})\rangle_A$ by viewing the fact that every point that belongs to the fractal object has the same type of environment when considered as the origin. So it can be numerically evaluated by using the power-law density function for fractals:

$$K(r) = D_f/[2\pi R^{Df}\Phi(r)] \int_{A'(r)} r^{Df-2}\, \mathrm{d}A. \tag{6}$$

For the same reason as mentioned in section 2, an angular average of (4) gives rise to a general structure factor for random 2D fractals:

$$S(q) = 2\pi \int_0^{\infty} \Phi(r)K(r)g(r)rJ_0(qr)\, \mathrm{d}\boldsymbol{r} \tag{7}$$

where $J_0(qr)$ is also the zero-order Bessel function of the first kind. By substituting the general correlation function for 2D fractals $g(r) = \kappa r^{Df-2}$ into (7) and noticing that $\Phi(r)$, the reduced convolution square of shape function for a disc of radius R, is given by [14]

$$\Phi(r) = \begin{cases} 2/\pi[\cos^{-1}(r/2R) - (r/2R)\sqrt{1-(r/2R)^2}] & r < 2R \\ 0 & r > 2R \end{cases} \tag{8}$$

a scattering equation for circular and sharply bounded fractals can be completely defined from (7).

4. Discussions

4.1. The scaling function and scattering profiles

By comparing (7) with the usual form of structure factor used in the literature [7], it is easy to note that the produce of $\Phi(r)$ and $K(r)$ is exactly in the place of the scaling function $f(r/\xi)$. Such an interesting comparison allows one to derive from (7) a complete definition of the scaling function for finite-sized fractals. Physically, one can see that the scaling function $f(r/\xi)$ comprises two parts: (i) the convolution square of shape function $\Phi(r)$, only determined by the geometrical form of fractals, and (ii) the inhomogeneity function $K(r)$, as a function of fractal dimensionality, reflecting the structural inhomogeneity inside the disordered fractals and assuming unity for any 2D aggregates with $D_f = 2$. At this point, it is clearly evident that for disordered fractal systems, the scaling function $f(r/\xi)$ is not always as simple as $\exp(-r/\xi)$ or something unfathomable but a function entirely dependent on the shape, size, and structural inhomogeneity of the fractals. It can be well mathematically constructed by considering fractal correlations and geometrical details of the fractals concerned. Undoubtedly, this way will lead one to achieving the disclosure of boundary structures of finite-sized fractals beside their fractal correlation properties.

By using (7) together with (6) and (8), a scattering profile for the model fractal patterns of radius $R = 30$ has been calculated, where N_p and D_f are evaluated with the statistical averages over ten model fractals of the same radius R. As shown in figure 2, the dotted line indicates the corresponding scattering profile, accompanied by the one given by (2) in squares. It can be seen that both curves show non-linear scattering profiles with a few rises and falls in the middle part of q, and fairly well coincide with each other, especially in the lower and higher q ranges. In sharp contrast, the scattering intensity calculated by using a simple exponential scaling function $f(r/\xi)$ in place of $\Phi(r)K(r)$ and evaluating ξ with a mean radius of gyration of the model fractals shows an almost straight profile in the intermediate q range, as shown in figure 2 (dot–dashed line), lying much farther from the square line than the dotted one. Obviously, the use of scaling function $\exp(-r/\xi)$ tends to yield a scattering profile quite close to the ideal straight line in a bi-logarithmic plot, which is often expected and stressed in some literature. However, here the case is absolutely not the same as described by scaling function $\exp(-r/\xi)$ because of the large differences between the sharply cut edge and the boundary exponentially diffusing into infinity. Therefore, it can be concluded that whether or not a linear scattering regime could be found for real fractal structures is also strongly related to the particulars of scaling function $\Phi(r)K(r)$, not only to power-law correlation function $g(r)$.

4.2. The additional contribution from the local density fluctuations

Although the scattering profiles derived from (7) and (2) can be in fairly good coincidence, an evident discrepancy between two scattering curves appears in the intermediate q range, especially in the first valley region, where the scattering curve given by (2) shows a deeper sink than the one given by (7). By such a comparison, one may see that the scattering equation developed in terms of power-law correlation, circular shape, and sharp boundary conditions can only fairly well describe the scattering behaviour of the model fractals when the structural parameters involved in it are evaluated with the values from the averaged fractal structures.

With the purpose of clarifying the factors that may be responsible for such a discrepancy, the correlation length spectra $\phi(r)$ (the population of scatterer pairs of different separation r) for the model fractals have been calculated both analytically and directly by using coordinates of the occupied sites in fractals, referred to as $\phi_a(r)$ and $\phi_d(r)$ and plotted in figure 3. For analytical calculation of correlation length spectra, the following formula adopted from (7) was used:

$$\phi_a(r) = 2\pi\Phi(r)K(r)g(r)r. \tag{9}$$

As illustrated in figure 3, $\phi_d(r)$ appears like a fluctuating function despite of an average over ten model fractals, showing a fluctuation of symmetrically enhanced amplitudes about the intermediate length scale, whereas the analytical spectrum $\phi_a(r)$ has a continuous and smooth profile. Moreover, it can be seen that $\phi_a(r)$ shows an evident deviation from the mean value of $\phi_d(r)$, which was obtained by multiple smoothing of the $\phi_d(r)$ curve and might be considered as the correct statistical description of the fluctuating correlation length spectrum. The presence of the difference between $\phi_a(r)$ and the mean value of $\phi_d(r)$ seems to indicate that some other factors should be taken into account in order to find a perfect

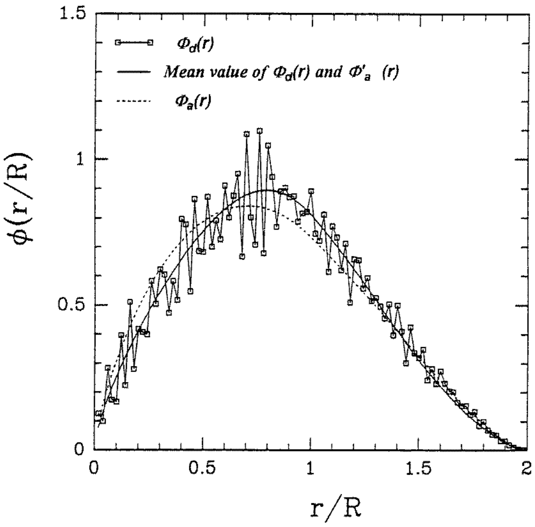

Figure 3. A comparison between the correlation length spectra $\phi(r/R)$: squares, $\phi_d(r/R)$ averaged over ten fractals; dotted line, $\phi_a(r/R)$ obtained from analytical calculation by (9) combined with (6) and (8); solid line, $\phi_a(r/R)$ modified by (9) together with (6), (8), (10) and (11), exactly coinciding with the mean value of $\phi_d(r/R)$, obtained after multiple smoothing of $\phi_d(r/R)$.

fit between two scattering profiles in addition to the power-law correlation and geometrical details such as shape, size, and boundary state.

By carefully examining each term in (3), one may find that a random fractal structure actually possesses a density distribution that cannot be mathematically continuous and analysable everywhere and should always exhibit some degree of local fluctuations. Taking such a factor into account, therefore, the genuine density function is supposed to be written as $\rho(\boldsymbol{r}) = \rho^0(\boldsymbol{r}) + \delta\rho(\boldsymbol{r})$, with $\rho^0(\boldsymbol{r})$ being the mean value of $\rho(\boldsymbol{r})$, and $\delta\rho(\boldsymbol{r})$ the random deviation from $\rho^0(\boldsymbol{r})$. Accordingly, such a consideration will have the function product $K(r)g(r)$ in (7, 9) re-expressed as

$$K(r)g(r) = \frac{\langle\rho^0(u)\rho^0(u+r)\rangle_{A'(r)}\langle\rho^0(u)\rho^0(u+r)\rangle_A}{\langle\rho^0(u)\rho^0(u+r)\rangle_A\langle\rho^0(u)\rangle_A}\left[1 + \frac{\langle\delta\rho(u)\delta\rho(u+r)\rangle_{A'(r)}}{\langle\rho^0(u)\rho^0(u+r)\rangle_A}\right]. \qquad (10)$$

On the right-hand side of (10), the term prior to the square brackets is actually the usual contribution of $K(r)g(r)$ and can be obtained by analytical calculation with the help of (6) and the usual power-law correlation function. As can be seen inside the brackets, however, a non-zero modifying factor is added when the correlation of local density fluctuations is not zero. Obviously, such an additional contribution is expected to be universally present in random fractals because no reason can be found to make sure that the local density fluctuations are not correlated everywhere in random fractals. By comparing $\phi_a(r)$ with the mean value of $\phi_d(r)$, the modifying factor in (10) has been numerically simulated for the model fractals used and can be expressed approximately by

$$\frac{\langle\delta\rho(u)\delta\rho(u+r)\rangle_{A'(r)}}{\langle\rho^0(u)\rho^0(u+r)\rangle_A} = 0.18\sin^2(r\pi/2R). \qquad (11)$$

where an amplitude of 0.18 reflects the degrees of local density fluctuations and their correlation. By using (10,11) for $K(r)g(r)$, a modified correlation length spectrum $\phi'_a(r)$

has been derived from (9). It yields almost the same curve as the mean value of $\phi_d(r)$ does with no visible discrepancy between them, as shown in figure 3. What is more interesting, as shown in figure 2, the scattering profile (solid line) calculated by using (7) with $K(r)g(r)$ evaluated by (10,11) gives an excellent fit to the ideal experimental scattering profile over the whole q range in the studies.

4.3. General scaling invariance

As seen in (6,8), the scaling function $\Phi(r)K(r)$ has its variables that can be well reduced by the fractal size R. This property allows (7) to be transformed into

$$S(q^*) = 2\pi\kappa R^{Df} \int_0^\infty \Phi(r^*)K(r^*)r^{*Df-1}J_0(q^*r^*)\,\mathrm{d}r^* \tag{12}$$

where the dimensionless quantities $q^* = q/R$ and $r^* = r/R$. Now if we let

$$\int_0^\infty \Phi(r^*)K(r^*)x^{*Df-1}J_0(q^*r^*)\,\mathrm{d}x^* = S_0(q^*, D_f)$$

and note the normalization condition, $S(q^* = 0) = N_p$, the total number of the scatterers involved in a given fractal pattern, which varies as a function of the size R and dimensionality D_f of the fractals, $N_p = 2\pi\kappa R^{Df}/D_f$, it follows that

$$S(q^*)/N_p = S_0(q^*, D_f)/S_0(0, D_f). \tag{13}$$

This equation, being of great importance for exploring the scattering behaviour of fractal structures, indicates that the q^* dependence of reduced scattering intensities $S(q^*)/N_p$ is solely determined by dimensionality D_f, disregarding the size of the fractals concerned, if they have the analogous boundary conditions and follow the same correlationship. It is interesting to note that this property has been excellently demonstrated through forming scattering profiles for the model fractals of different radii: $R = 20$, 30, 40 and 50. The ideal coherent scattering profiles for these model fractals are illustrated in figure 4, where the inset box shows their collapse. It is clear that no matter how large the fractals are, their reduced intensity curves are well coincident with one another. Physically, one can conclude that this scaling property results from the self-similarity of the fractals, and also expect that it is helpful to monitor the dynamic growth of fractal aggregates. If a growing aggregate maintains its boundary structure, shape, and interparticle correlationship during its growth process, its scattering profiles measured at different time intervals are supposed to abide by (13). Otherwise, any deviation from (13) may imply some changes in boundary structure, shape or correlationship between the particles.

5. Conclusions

In summary, the investigation of scattering behaviour by 2D model fractals has shown that the ideal scattering profiles calculated by starting from the exact coordinates of each scatterer inside the fractals can be fairly well described by a scattering equation, which is strictly developed in terms of the point-scattering theory and power-law correlation, as well as the boundary structure of the model fractals, when the relevant structural parameters in the scattering equation are evaluated with the values averaged over ten model fractal patterns of the same size. In contrast, the description by using scaling function $\exp(-r/\xi)$ is quite poor even though the correct values of structure parameters are adopted. Moreover, by taking into account the additional contribution to the scattering behaviour from the

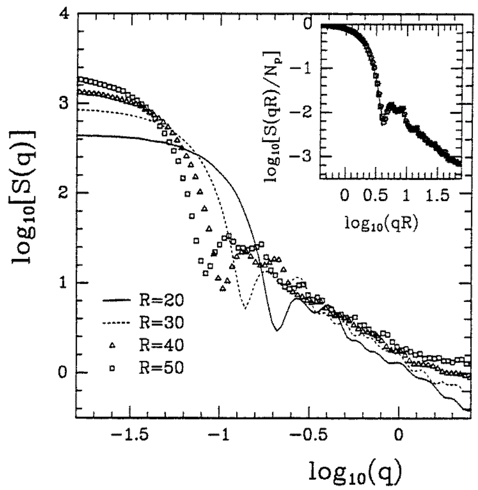

Figure 4. Scattering profiles $S(q)$ for the circularly trimmed fractals with different radii. The inset shows their collapse after scaling.

correlation of local density fluctuations inside the random fractals, the remaining discrepancy between the ideal scattering profile and the scattering equation in the intermediate q range can be almost completely eliminated. This significant result manifests that a particular scattering intensity profile from real fractals is not only governed by the power law, regular inhomogeneity, and their geometrical details, but also impacted to a certain degree by the correlation from local density fluctuations. In addition, a scaling invariance between the scattering intensity profiles for the fractals, which possess the same fractal correlation and analogous boundary structures, but different sizes, has been revealed and demonstrated. This interesting scattering property appears to be useful for monitoring the real growth of fractal aggregates.

Acknowledgments

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