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Backscattering in fractal aggregates: theoretical and numerical studies

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Abstract. We have performed a numerical study of multiple scattering in fractal aggregates of different fractal dimensions D. We have found a behaviour which does not correspond to previous work on random media. Our theoretical approach allows us to understand the structure of the backscattered intensity and shows that the coherent backscattering peak due to time-reversal invariance is drowned in other types of interference.

1. Introduction

Over the past few years, fractal structures have been the subject of much attention [1, 2]. Fractal geometry has provided us with many well defined structures which may be used either to represent real objects or to investigate the properties of well known methods in physics [3].

The physical mechanism of aggregation often leads to sparse fractal clusters. One model of aggregation which has been well studied is the diffusion-limited cluster–cluster aggregation model [4, 5]. The fractal character means that each of the clusters is statistically invariant under space dilation, or equivalently, that the average correlations in particle positions inside the same cluster follow the power law. This fractality yields typical features in the physical properties of such objects, but, owing to these long-range correlations, the influence of the fractal structure on physical properties is generally non-trivial and difficult to study.

There have been several studies [6, 7] on multiple scattering on fractal clusters, but presently, none of them have dealt with the coherent backscattering cone. Jullien and Botet have studied geometrical optics in fractals [8], but in this case, the radius of the particles was much larger than the wavelength of the incident beam. An interesting study of coherent backscattering of light in fractals has been done by Akkermans *et al* [9]. In their case, light could only propagate from one particle to its nearest neighbours. In fractal aggregates which are included in a transparent medium, light may exhibit a Levy flight behaviour [15].

On the other hand, multiple scattering in random media has been widely investigated since the 1980s [10]. Most of the theory on coherent backscattering of light in random media and its comparison to electronic wave multiple-scattering effects has been reported recently by Lagendjiik and Van Tiggelen [11].

The physical phenomena leading to a coherent backscattering cone in a random medium are now well known. But as a matter of fact, random media differ from fractal media as the latter have long-range correlation. So it seems logical to study the intensity of light in the backward direction to see whether it remains such a peak for fractal systems.

In section 2, we present the numerical procedure for building our fractal aggregates and for representing the interaction of light with these structures. In section 3, a theoretical approach of the problem is made in order to explain the results (section 4). Finally, section 5 contains a discussion of the comparison between theory and numerical results. Section 6 is the conclusion of the whole article.

2. Numerical procedure

2.1. Variable-D model

We have used a program written by Thouy and Jullien [12] to build aggregates of tunable fractal dimension. It is a generalized cluster–cluster aggregation model which is hierarchical in the sense that only clusters with the same number of particles can stick together. This model has the great advantage of having the fractal dimension D as an input parameter of the program. The idea is to build a new cluster of 2^m by sticking two clusters (built separately) containing 2^{m-1} particles in order that the rate between the quadratic mean gyration radius R and the distance between their centre of mass Γ agrees with the relation

$$\Gamma^2 = 4(4^{1/D} - 1)R^2 + \delta^2 \tag{1}$$

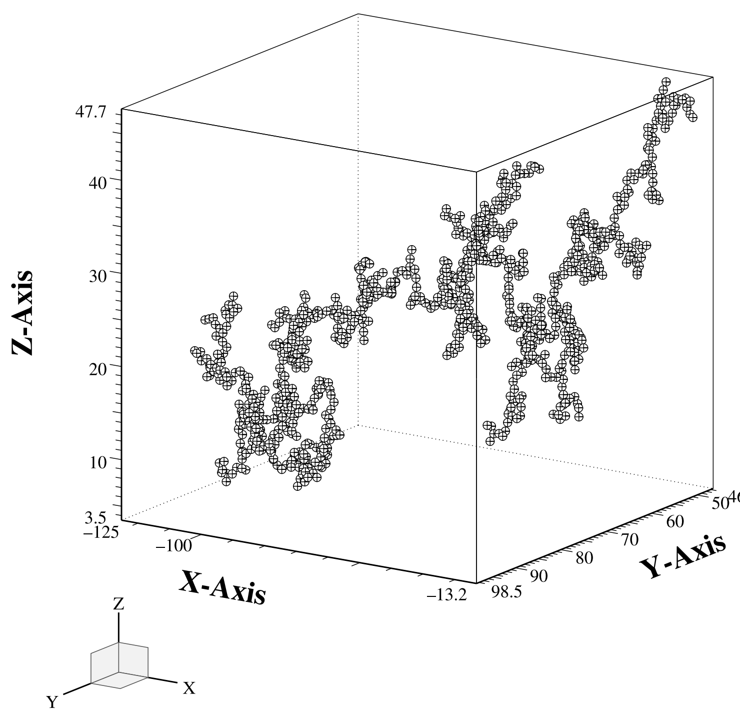

Figure 1. Example of an aggregate composed of 1024 particles and of fractal dimension $D = 1.6$.

where δ is the length of the lattice step. This relation ensures that the mean penetration is adequate for the clusters to stick. Figure 1 represents an example of an aggregate with fractal dimension $D = 1.6$.

2.2. Multiple scattering in the aggregates

To model the interaction of the fractal cluster composed of particles of size $\ll 1$ with an incident monochromatic plane-wave field $\boldsymbol{E}_{\mathrm{inc}}$, we have used the dipolar approximation, which allows us to calculate the far and near electromagnetic field for a given local field on the particle. If the wavelength of the incident radiation is much larger than the radius r of the monomers, the dipolar theory can be applied to each of the N monomers of an aggregate, whatever its size [13]. Let us suppose that this condition is fulfilled. The field radiated $\boldsymbol{E}_{\mathrm{rad}}$ by such a dipole (Rayleigh scattering) simulated by the local incident field $\boldsymbol{E}_{\mathrm{loc}}$ is

$$\boldsymbol{E}_{\mathrm{rad}} = \frac{k^3\,\mathrm{e}^{\mathrm{i}kr}}{4\pi\epsilon_0 kr}\left\{\boldsymbol{p}\left(1+\frac{\mathrm{i}}{kr}-\frac{1}{k^2r^2}\right)+\boldsymbol{n}(\boldsymbol{n}\cdot\boldsymbol{p})\left(-1-\frac{3\mathrm{i}}{kr}+\frac{3}{k^2r^2}\right)\right\} \tag{2}$$

where the dipolar moment $\boldsymbol{p}$ is connected to the local field by

$$\boldsymbol{p} = \epsilon_0\alpha\boldsymbol{E}_{\mathrm{loc}} \tag{3}$$

where α is the polarizability. The unit vector $\boldsymbol{n}$ is defined as $\boldsymbol{r}/r$. In the multiple-scattering regime, the local field is determined by the incident field and by the fields radiated by the other dipoles. One can write a self-consistent equation for the fields:

$$\boldsymbol{E}_{\mathrm{loc}}(\boldsymbol{r}_j) = \boldsymbol{E}_{\mathrm{inc}}(\boldsymbol{r}_j) + \sum_{l\neq j}\boldsymbol{E}_{\mathrm{rad}}(\boldsymbol{r}_j - \boldsymbol{r}_i) \tag{4}$$

where $\boldsymbol{r}_j$ is the position of the jth monomer. The set of equations (2)–(4) can be solved by recursion. But, numerically, one has to be careful about the convergence of the expansion. So, one may compute the field radiated at large distances from the sample.

In our simulation, we took the wavelength of light λ to be equal to 1, so that the size scale of λ allows us to see the fractal correlations. A possible experimental set-up which would correspond to our numerical simulation would be particles of size $\ll 1$, which would be embedded in a transparent matrix. The distances between neighbouring particles would be ~ 1 and their correlation function would keep the fractal properties.

3. Theory

In random media, it is possible to estimate the half-width of the coherent backscattering peak by calculating the mean-square displacement of light, if taken in the diffusion approximation [14]. In fractal media, however, this calculation is more subtle since light exhibits anomalous diffusive behaviour [15].

Generally, for multiple scattering, the phase difference between two optical paths, one being the reciprocal of the other, is given by

$$\phi = (\boldsymbol{k}_i + \boldsymbol{k}_f)\cdot(\boldsymbol{R}_N - \boldsymbol{R}_1). \tag{5}$$

This supposes two-wave interference. If we note θ, the angle to the backward direction and if we use the diffusion approximation, i.e. $|\boldsymbol{R}_N - \boldsymbol{R}_1|^2 = \langle \boldsymbol{R}^2 \rangle \propto f(t_N)$, where $f(t_N)$ is the behaviour of the mean-square displacement as a function of time t_N and velocity of light $c = 1$. So we obtain

$$\phi_N \propto k\theta f(t_N) \tag{6}$$

where ϕ_N is the phase difference for a given time t_N. So,

$$\delta\theta \propto \frac{1}{k} \sum_N \frac{1}{f(t_N)} \tag{7}$$

is the half-width of the coherent backscattering peak. If the mean-square displacement diverges, the half-width of the coherent backscattering peak tends to zero.

For fractal dimensions running between 2 and 3, it is possible to compute the long-time behaviour of the mean-square displacement, using the so-called pore chord distribution introduced by Mering and Tchoubar [16, 17]. The pore chord distribution may be defined here as the number of segments linking two particle centres as a function of the segment length r. Following their article, the pore chord distribution runs as r^{-D+1}.

For instance, if the fractal dimension of the aggregate is $2 < D < 3$, the mean-square displacement behaves as [15]:

$$\langle r^2(t) \rangle = t^2 \big\{1 - (D-1)^{-1} 2 \big(t/t_{\max}\big)^{(2-D)} + \big[(2-D)/(D-1)\big]\big(t/t_{\max}\big)\big\}. \tag{8}$$

If the fractal dimension of the aggregate runs between 1 and 2, the mean-square displacement diverges as [15] t^{-2D+6}.

If we use the diffusion approximation, we see that in fractal media the mean-square displacement is no longer a constant and diverges with time. This time may be approximated by a multiple of the aggregate size and thus has a limit in finite size clusters. But, theory for random media predicts that the coherent backscattering peak has a width which runs as the inverse of the mean-square displacement. So, the faster the mean-square displacement diverges with time, the smaller is the peak half-width. This behaviour is the more obvious for fractal dimensions $D < 2$ where the mean-square displacement diverges as t^{-2D+6}.

Another effect of this pore-chord distribution is related to the fractal dimension D. Indeed, the probability for two optical paths to have the same optical length without being the reverse of each another may be computed by

$$r^D r'^{-D+1} r''^D \tag{9}$$

in the double-scattering case. Here r^D and r''^D are the probability to find two particles at a distance r (respectively r'') from a given point inside the aggregate and r' is the distance between the two particles inside the aggregate. For a random medium, this probability is a constant but for a fractal aggregate it is easy to see that it increases with the aggregate size. This effect decreases for multiple (i.e. more than double) scattering, as the probability for two optical paths to have the same geometrical length is

$$r^{(1)D} r^{(2)-D+1} \ldots r^{(n-1)-D+1} r^{(n)D} \tag{10}$$

in the case of nth-order scattering. So the fractal correlations may induce scattered intensity peaks which are not in the exact backward direction and which may not be washed out by averaging. Moreover, fractal correlations may have a greater effect for fractal dimension D close to 3.

4. Results

In figure 2 the backscattered intensity for a random media is plotted. There is a distinct peak for angle $\pi/2$ in the backward direction. This is characteristic of the time-reversal invariance in random media.

In figure 3(a), we plotted the backscattered intensity for fractal dimension $D = 1.6$ and for an aggregate of 1024 particles. The result is an average over 80 different samples. As one may see, it remains a coherent backscattering peak in this case. This peak is even more obvious in figure 3(b), where we plotted the backscattered intensity for $D = 2.4$ and for the same number of particles.

As predicted in the previous paragraph, there is a size effect on the backscattered intensity, so we have plotted this intensity in figures 4(a) and 4(b) for $D = 1.6$ and $D = 2.4$, respectively with 16 384 particles. Obviously, the coherent backscattering peak no longer remains for $D = 1.6$ and for $D = 2.4$, this peak is higher than twice the background intensity. This behaviour is very different from random media. For these two figures, the results are an average over 12 samples.

Finally, in figures 5(a) and (b), we show the evolution of the backscattered intensity for two different wavelengths ($\lambda = 10, 100$) on 16 384 particles. Results are obtained after averaging over 12 samples.

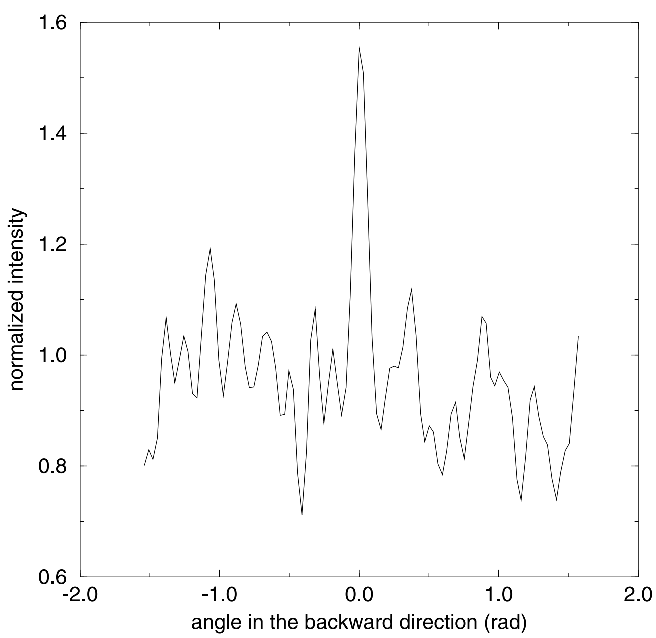

Figure 2. Backscattered intensity (in normalized units) versus the backward angle (in radians) averaged over 80 samples with 1024 particles located randomly. The wavelength here is $\lambda = 1$.

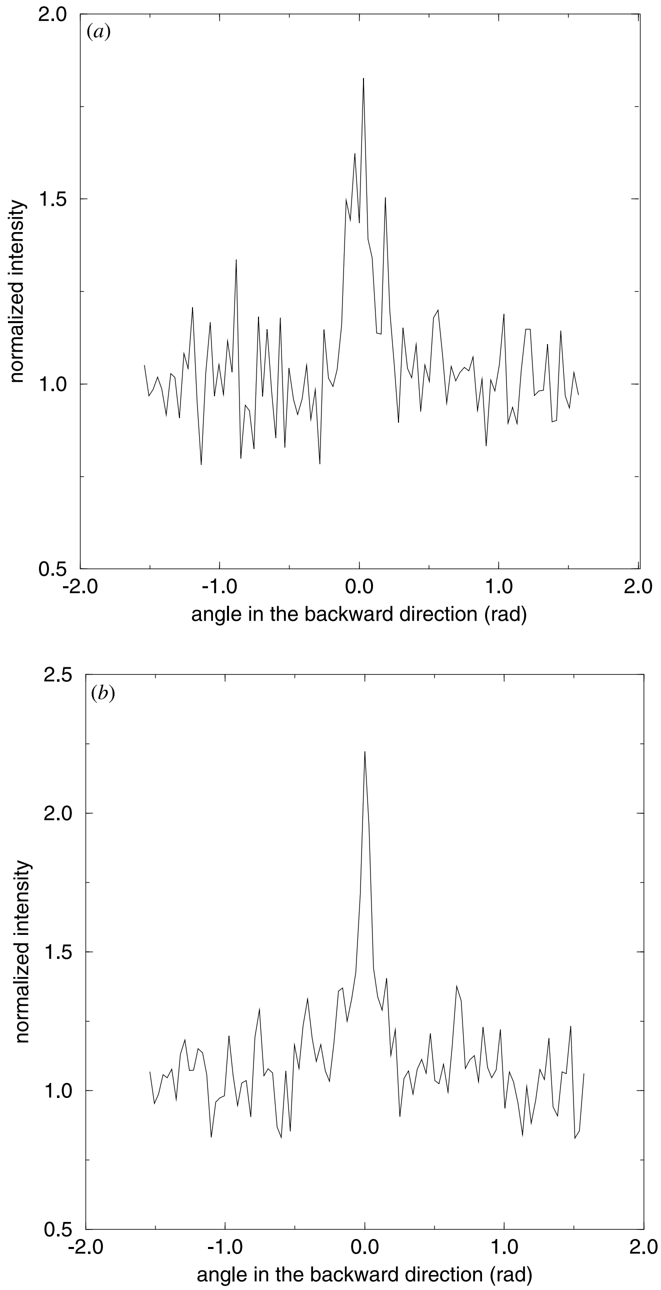

Figure 3. (*a*) Backscattered intensity (in normalized units) versus the backward angle (in radians) averaged over 80 samples with 1024 particles and $D = 1.6$ and $\lambda = 1$. (*b*) Backscattered intensity (in normalized units) versus the backward angle (in radians) averaged over 80 samples with 1024 particles and $D = 2.4$ and $\lambda = 1$.

5. Discussion

As one may see in figures 3(*a*) and (*b*), the coherent backscattering peak exists for light scattering on small fractal aggregates (in this case, there are 1024 particles in the aggregates).

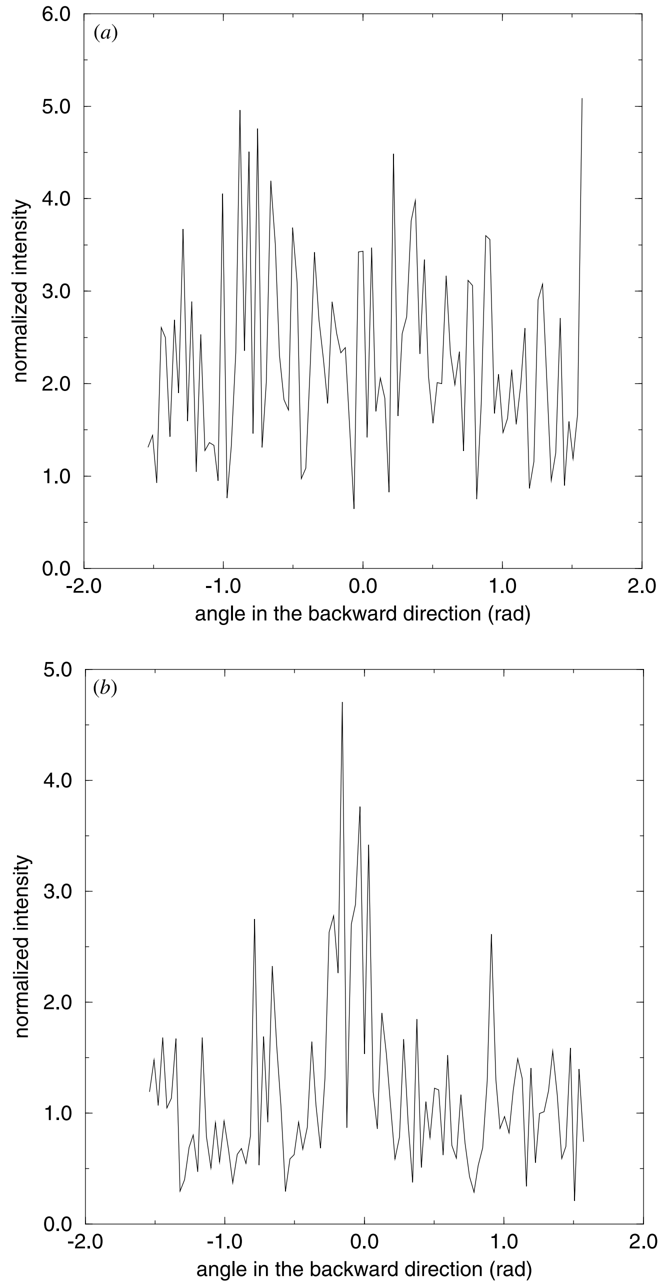

Figure 4. (*a*) Backscattered intensity (in normalized units) versus the backward angle (in radians) averaged over 12 samples with 16 384 particles and $D = 1.6$ and $\lambda = 1$. (*b*) Backscattered intensity (in normalized units) versus the backward angle (in radians) averaged over 12 samples with 16 384 particles and $D = 2.4$ and $\lambda = 1$.

For $D = 2.4$ (figure 3(*b*)), the backscattered peak has an intensity which is smaller than 2 in normalized units. It corresponds to theory for random media which predicts that the coherent backscattering peak has at most an intensity twice as large as the background

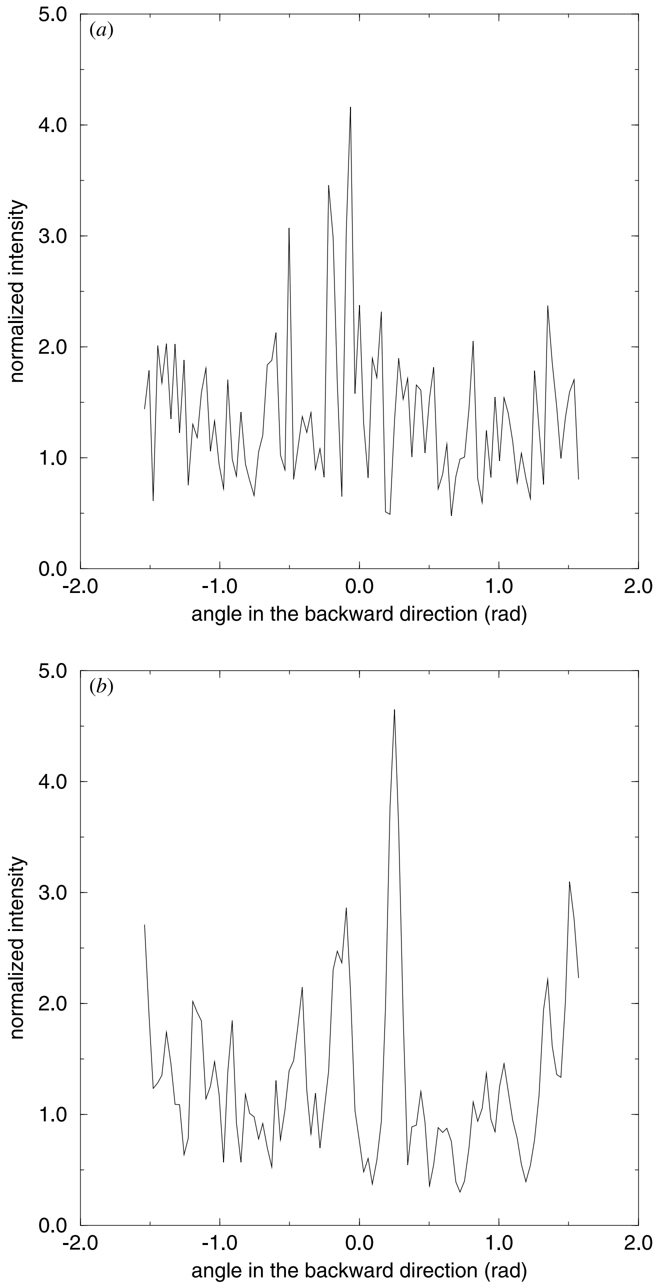

Figure 5. (*a*) Backscattered intensity (in normalized units) versus the backward angle (in radians) averaged over 12 samples with 16 384 particles and $D = 2.4$ and $\lambda = 10$. (*b*) Backscattered intensity (in normalized units) versus the backward angle (in radians) averaged over 12 samples with 16 384 particles and $D = 2.4$ and $\lambda = 100$.

one, as one may see in figure 2. In this case, i.e. for figures 3(*a*) and (*b*), due to size effects, the divergence of the mean-square displacement is not very strong, so the half-width of the coherent backscattering peak is non-zero. Indeed, the half-width of the coherent backscattering peak is inversely proportional to the mean-square displacement. The theory for the mean-square displacement behaviour in fractal media predicts that it diverges, but as the fractal aggregate on which the computation has been made has a finite size which is relatively small, the mean-square displacement cannot overcome a threshold due to the cluster size itself.

For fractal dimension $D = 2.4$ (figure 3(*b*)) and for the random medium (figure 2), the difference between the two coherent backscattering peaks is small. When one approaches the fractal dimension $D = 3$, i.e. the fractal dimension of a random medium, the mean-square displacement has an upper limit (even if it diverges) which is very close to the constant mean-square displacement of a random medium with the same size (1024 particles). This explains the similarity between the backscattered intensities of figures 2 and 3(*b*).

For $D = 1.6$, the divergence of the mean-square displacement is larger than for $D = 2.4$. So the differences between the results given by figures 3(*a*) and 2 are not neglectable. Furthermore, in figure 3(*a*), one may see in the splitting of the backscattering peak, the effects of fractal correlations.

Let us examine figures 4(*a*) and (*b*). The fractal aggregates used for these figures are anisotropic if they are constructed in a three-dimensional space. One may easily understand that the closer the fractal dimension is to 2, the more the aggregates become geometrically linear. In order to avoid the effects of this anisotropy; during computation we have rotated these aggregates in all space directions and we have then computed the backscattered intensity, this is analogous to constructing more aggregates. If we look at figure 4(*a*) (scattered intensity on aggregates of 16 384 particles with $D = 1.6$), we can see that there is no longer a visible coherent backscattering peak. In fact, the coherent backscattered intensity due to time-reversal invariance still exists, but its half-width tends to zero due to divergence of the mean-square displacement, so the coherent backscattering peak disappears. Indeed, for this size of aggregates, the mean-square displacement has anomalous behaviour which has a limit which is very large compared to the mean-square displacement for a random medium of the same size. In this case (a large number of particles in the cluster and small fractal dimension), the light has a Levy flight behaviour: either it is scattered several times between close particles or it propagates between two particles which are very far away from each other. So the mean-square displacement diverges and the coherent backscattering peak narrows.

The backscattered intensity maximum in figure 4(*b*) has a magnitude which is larger than three times the background intensity. This may be explained by the fact that the interference due to fractal correlations is added to the interference due to time-reversal invariance, i.e. several optical paths may have the same optical length without having the same geometrical path. This phenomenon is not washed out by averaging and is more obvious for an aggregate with fractal dimension $D = 2.4$ than for $D = 1.6$ because (see equation (9)) as aggregates with fractal dimension $D = 1.6$ get close to a line, the number of optical paths with the same length and different geometrical paths are less numerous. The peaks near the angle $\theta = 0$ are even larger than the one in the exact backward direction. This indicates that fractal correlations have a greater effect than that due to time-reversal invariance.

Finally, the effect of the wavelength is shown in figures 5(*a*) and (*b*). Figure 5(*a*) is a computational result performed for the same aggregates as for figure 4(*b*) (i.e. with $D = 2.4$). In fact, taking a larger wavelength is equivalent to taking a smaller aggregate with the same wavelength $\lambda = 1$. The final figure (figure 5(*b*)) which represents the scattered intensity with $\lambda = 100$ on isolated aggregates, shows one large peak (which is due to fractal correlations) but also has a shape which gets closer to the dipolar scattering one.

6. Conclusion

We have shown here that due to the divergence of the mean-square displacement of light in fractal aggregates, the half-width of the coherent backscattering peak tends to zero. This effect is mostly visible for large fractal aggregates, as the mean-square displacement has a threshold which is proportional to the aggregate size. Moreover, another effect which differs from random media is the action of fractal correlations that leads to the presence of other peaks, not only in the exact backward direction.

Two studies may now be carried out: the relationship between the backward intensity and the correlations in the aggregate and an experimental study. The experimental procedure would be to analyse backscattering on silica aerogels, which are known to be fractal objects in a given domain.

Acknowledgments

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