CORRELATION PROPERTIES OF LIGHT BACKSCATTERED MULTIPLY FROM FRACTAL AGGREGATES OF PARTICLES UNDER BROWNIAN MOTION

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ABSTRACT

Studies on the phenomena of light scattered multiply from strongly absorbent media have been of benefit to metrology in biophysics, chemistry, and other areas. In this article, a temporal autocorrelation function of time-varying light backscattered multiply from a fractal aggregate of particles under Brownian motion suspended in an absorbent medium is investigated by means of Monte Carlo simulation. Temporal statistics of the backscattered light in time regions longer and shorter than the relaxation time depend on the fractal dimension and agree with the case of the homogeneously random media, respectively. The angular dependence of light backscattered multiply from fractal media varies dramatically depending on the fractal dimension in comparison with that from homogeneously random media. The influence of absorption generates the time shift in the temporal autocorrelation function in such a way that they vary approximately in accordance with a square root of the absorption coefficient but are independent of the fractal dimension. © 1999 Society of Photo-Optical Instrumentation Engineers. [S1083-3668(99)00502-X]

Keywords multiple scattering; fractal aggregates; Monte Carlo simulation; diffusive transport theory; diffusing wave spectroscopy.

1 INTRODUCTION

Static and dynamic light scattering techniques have played an important role in the physiology and biorheology.[1–6] Since a biological tissue is generally composed of complex structures with dimensions ranging from several tens of nm to several mm, a fractal-like structure has been taken into consideration to investigate the relation between the optical properties and the structural features in the laboratory framework.[7–9]

Dynamic light scattering by a dense medium with the fractal-like structure is of current interest in this investigation. The particle aggregation is assumed to be a model of the physiological and biorheological dense media without dilution, for example, the blood coagulation, the blood plasma aggregation, the gelation and aggregation of the gastric mucin and so on. The diffusive wave spectroscopy (DWS)[10,11] technique is used to analyze the temporal properties of multiply backscattered light from a particle aggregate, in which a temporal autocorrelation function (TAF) is numerically obtained by Monte Carlo simulations. The angular dependence of the TAF on the fractal dimension and the absorption of the medium is discussed in connection with a scattering-order distribution function (SODF) and a total path-length distribution function (TPDF). As a result, it is shown that the fractal property of the scattering medium and the absorption affects the relaxation time and generates a time shift in the TAF, respectively.

2 SIMULATIONS

Monte Carlo simulations are carried out to trace all scattering events and trajectories of the light backscattered multiply from fractal aggregates of particles. Suppose that an aggregate medium exists in the space of $Z>0$ in the absolute coordinate system (X,Y,Z) and that the plane light polarized along the X direction is incident on the medium normally to its boundary plane between air and the medium. The incident light undergoes successive scattering events by many particles, where the scattering angle at the mth scattering event and the distance between the mth and $(m-1)$-th scattering events are denoted by θ_m and r_m, respectively. Finally, the scattering light emerges from the medium in the

direction of the backscattering angle θ which is defined as an angle between the direction of the emerging light and the direction opposite to the incident light. The emerging light produces an intensity distribution in far field which has an intensity peak referred to as enhanced backscattering in the direction of retroreflection. The light varies in time due to the Brownian motion of all scattering particles so that the TAF of the multiply backscattered light monotonically decreases by an accumulative effect of all particle movements.

With regard to multiple scattering in the fractal aggregate of particles, the free path-length distribution $p(r)$ was derived theoretically as[12]

$$p(r_m) = \frac{(D-2)}{l_D} \left(\frac{r_m}{l_D} \right)^{D-3} \exp \left[-\left(\frac{r_m}{l_D} \right)^{D-2} \right] \quad (1)$$

in the range of $2.0 < D \leq 3.0$. In Eq. (1), D and l_D denote a fractal dimension and an effective mean free path-length for the fractal aggregate. The simulation is conducted for a 10% suspension of polystyrene latex beads with diameters of 100 nm and a refractive index of 1.59. It is illuminated by the polarized plane wave from an Ar^+ laser with a wavelength of $k_i = 514.5\,nm$. Under this condition, the scattering event at each particle may be approximated by Rayleigh–Debye scattering.[13]

The TAF of the multiply backscattered light is given by[14]

$$\gamma(\tau, \theta) = \sum_{n=1}^{\infty} p(n, \theta) \left\langle \exp \left(-\frac{4\tau}{\tau_0} \sum_{m=1}^{n} \sin^2 \frac{\theta_m}{2} \right) \right\rangle_{\theta_m} \quad (2)$$

under the assumption that each particle in the medium moves independently of each other due to Brownian motion even after the particles have aggregated. In Eq. (2), τ and $\tau_0 = (D_p k_i^2)^{-1}$ denote a delay time and a single scattering relaxation time in the homodyne correlation function of the time-varying amplitude scattered from the particle with the diffusion constant D_p. The TAF of the multiply backscattered light has an angular dependence which is generated by the interference effect producing the backscattering enhancement. $p(n, \theta)$ denotes a fraction of the intensity emerging in the direction of the backscattering angle θ after a sequence of n scattering events to the total intensity in the same direction.

By taking a polarization effect into consideration, $p_X(n, \theta)$ and $p_Y(n, \theta)$ for co- and cross-polarized components of the backscattered light are defined by

$$p_X(n, \theta) = \langle I_{nX} \rangle_{ex} \bigg/ \sum_{n=1}^{\infty} \langle I_{nX} \rangle_{ex} \quad (3a)$$

and

Fig. 1 Temporal autocorrelation functions (TAF) of (a) the co-polarized and (b) the cross-polarized components of light scattered multiply from three different media with dimensions of $D = 2.5$, 2.8, and 3.0 in the direction of retroreflection. The vertical and horizontal axes denote a logarithm of the TAFs and a square root of the delay time, respectively. The black circles stand for the relaxation times τ_c which were estimated to be $\tau_c = 2/A^2$ by fitting the TAFs to $\exp(-A\sqrt{\tau})$.

$$p_Y(n, \theta) = \langle I_{nY} \rangle_{ex} \bigg/ \sum_{n=2}^{\infty} \langle I_{nY} \rangle_{ex}, \quad (3b)$$

respectively. $\langle I_{nX} \rangle_{ex}$ and $\langle I_{nY} \rangle_{ex}$ denote the nth order scattering intensity distributions for the co- and cross-polarized components and are numerically calculated by using the data obtained by Monte Carlo simulations. To include the effect of absorption with the absorption coefficient μ_a, it is assumed that the scattered light intensity decreases in accordance with $\exp(-\mu_a s)$ as the path length s of the scattered light increases. By substituting Eq. (3) into Eq. (2), the angular dependence of the time-varying light backscattered multiply from the fractal aggregate of particles can be investigated numerically and quantitatively.
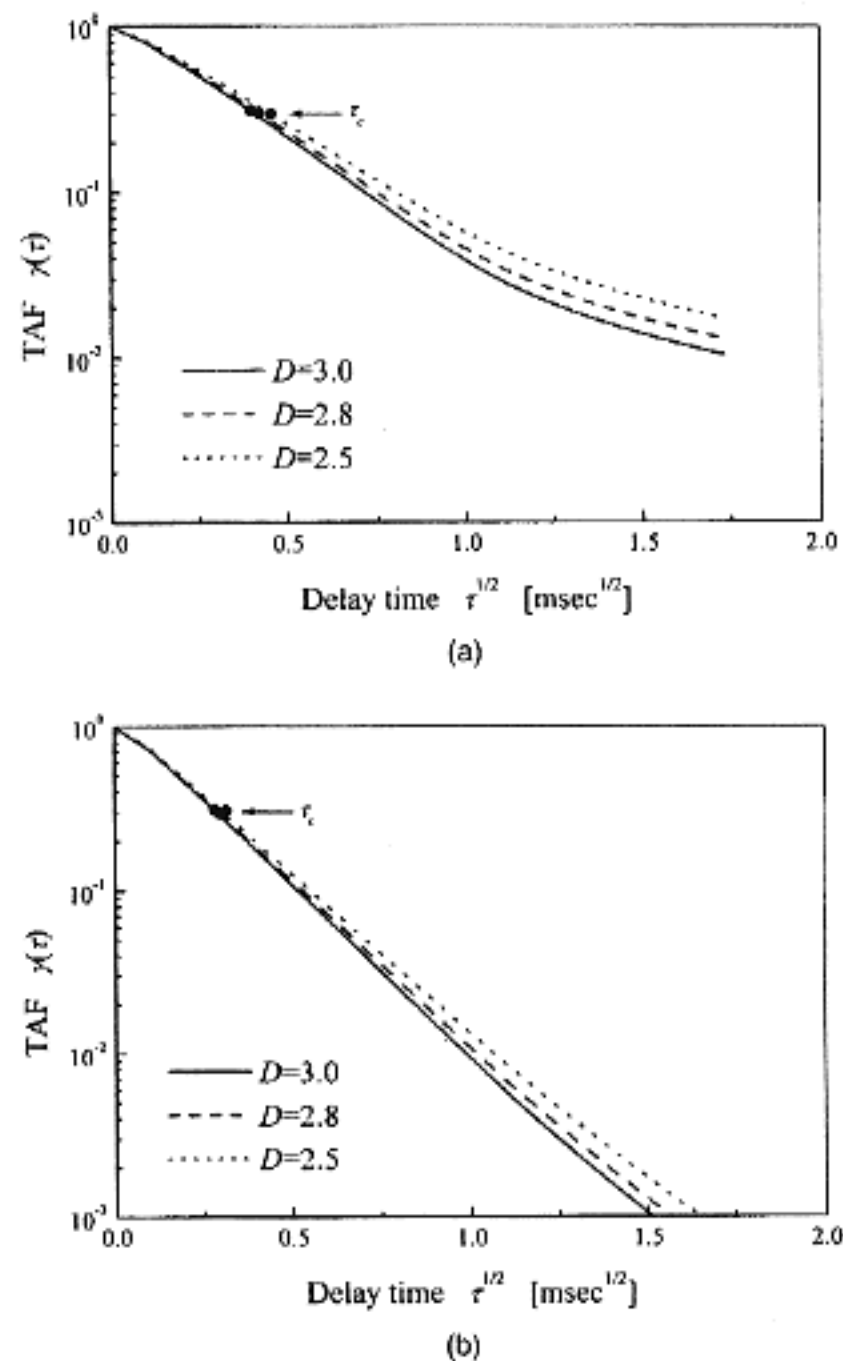
3 RESULTS AND DISCUSSIONS

Figures 1(a) and 1(b) show the TAFs of the co- and cross-polarized light amplitudes backscattered multiply from the fractal aggregates of particles obtained numerically for three different dimensions of $D = 2.5$, 2.8, and 3.0. In Figure 1, the black circles stand for the relaxation times τ_c which are estimated to be $\tau_c = 2/A^2$ by fitting the TAFs to $\exp(-A\sqrt{\tau})$ that was derived theoretically under the diffusion approximation.[10,11] The TAF shows the following features depending on the polarization and the aggregation of particles: the TAF becomes wider with a decrease of the dimension; the variations of the TAF become more outstanding with an increase of the delay time; and the variation of the TAF is larger for the co-polarized component than for the cross-polarized one.

In further discussion of the properties of the TAF, the two types of TAFs expressed by discrete and continuous versions for homogeneously random media are taken into consideration. The former is expressed as a summation of the decay function $\exp(-\tau n/2\tau_0 n^*)$ weighted by the SODF $p(n,\theta)$ ranging from a single to infinity in scattering order and is given by

$$\gamma(\tau,\theta) = \sum_{n=1}^{\infty} p(n,\theta) \exp(-\tau n/\tau_0 n^*), \qquad (4)$$

where n^* denotes the averaged scattering order where the direction of light propagating in the medium is fully randomized. The latter is expressed as an integration of the decay function $\exp(-\tau s/\tau_0 l^*)$ weighted by the TPDF $p(s,\theta)$ ranging from zero to infinity in total path length and is given by

$$\gamma(\tau,\theta) = \int_0^{\infty} p(s,\theta) \exp(-\tau s/\tau_0 l^*) ds, \qquad (5)$$

where the transport mean free path length l^* denotes the averaged path length where the direction of light propagating in the medium is fully randomized, and s is the total path length of light propagating with nth scattering order. For homogeneously random media, Eq. (4) is equal to Eq. (5) because of $p(s,\theta) = p(nl,\theta)$. It should be noted in the derivation of Eq. (5) that the lower limit of the integration was assumed to be reduced from l^* to zero. However, this assumption will break down whenever light with a path length shorter than l^* contributes strongly to the TAF. Nevertheless, the Laplace transform relation of Eq. (5) between the TAF and the TPDF has been fairly well confirmed in experiments.[15–17]

The TPDFs and the SODFs of light backscattered multiply from fractal media with $D = 2.5$, 2.8, and 3.0 are shown in Figures 2 and 3, respectively. As seen from Figure 2, the TPDF apparently depends on the dimension of the medium in the region of $s < l_D$, and becomes independent of the dimension
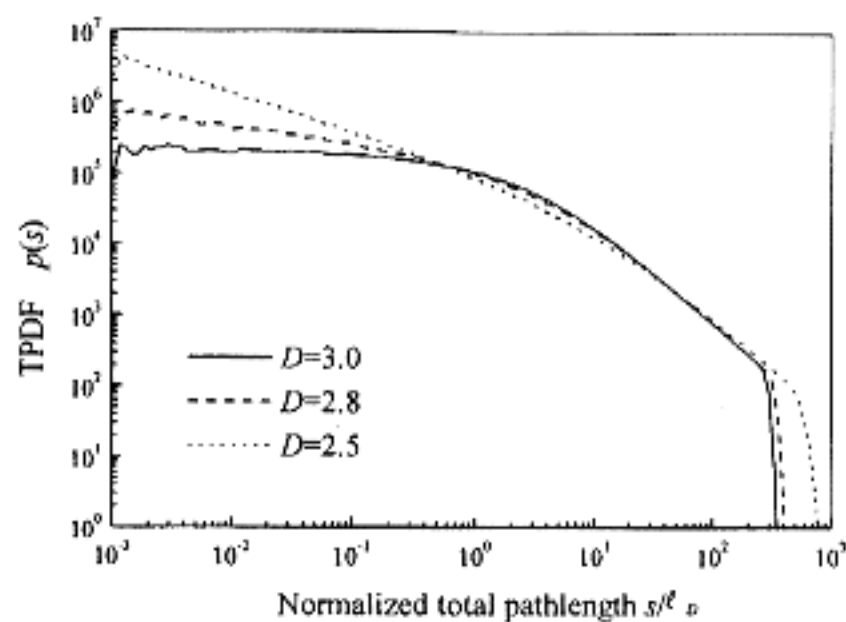
Fig. 2 TPDFs of light backscattered multiply from three different media with dimensions of $D = 2.5$, 2.8, and 3.0, where the horizontal axis is normalized by the effective mean free path length l_D. Both the horizontal and vertical axes are plotted on a logarithmic scale.

in the region of $s > l_D$. In the former region, the dominant contributions with low scattering orders are not yet randomized in direction and, consequently, a decreasing TPDF rate in a logarithmic plot varies asymptotically following the line of $D - 3$. In the latter region, the direction of light scattered with high scattering order is fully randomized so that the scattered light loses information about the spatial properties of aggregated particles. Therefore, the TPDF becomes independent of the fractal dimension. On the other hand, it is seen from Figure 3 that the SODF depends on the dimension only at $n \approx 1$ in such a way that the probability increases slightly with a decrease of the dimension.

Due to the assumption of a medium with a volume fraction of 10% and particle diameter of 100 nm, the approximations of $l_D^* = l_D/\langle 1 - \cos\theta \rangle \approx l_D$ and $n^* \approx 1$ are satisfied in the simulation. Although
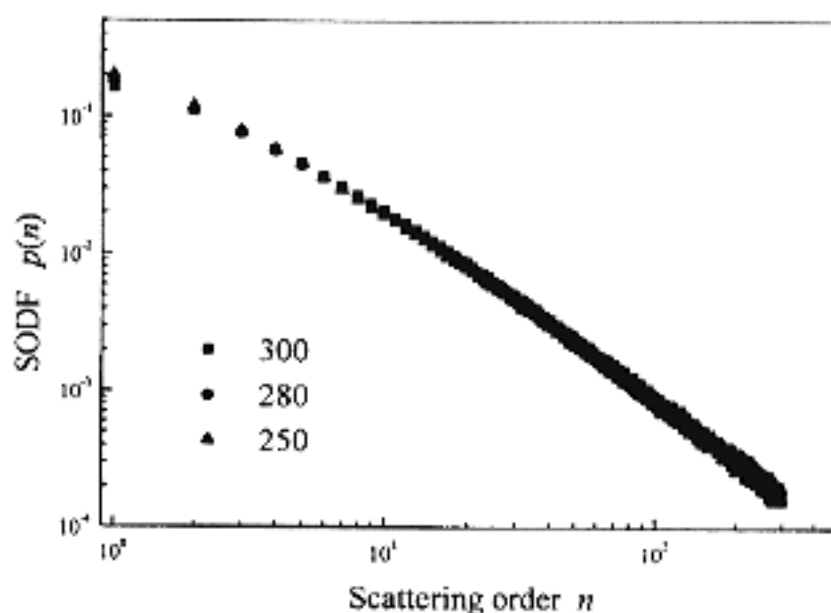

Fig. 3 SODFs of light backscattered multiply from three different media with dimensions of $D = 2.5$, 2.8, and 3.0. The vertical axis is plotted on a logarithmic scale.
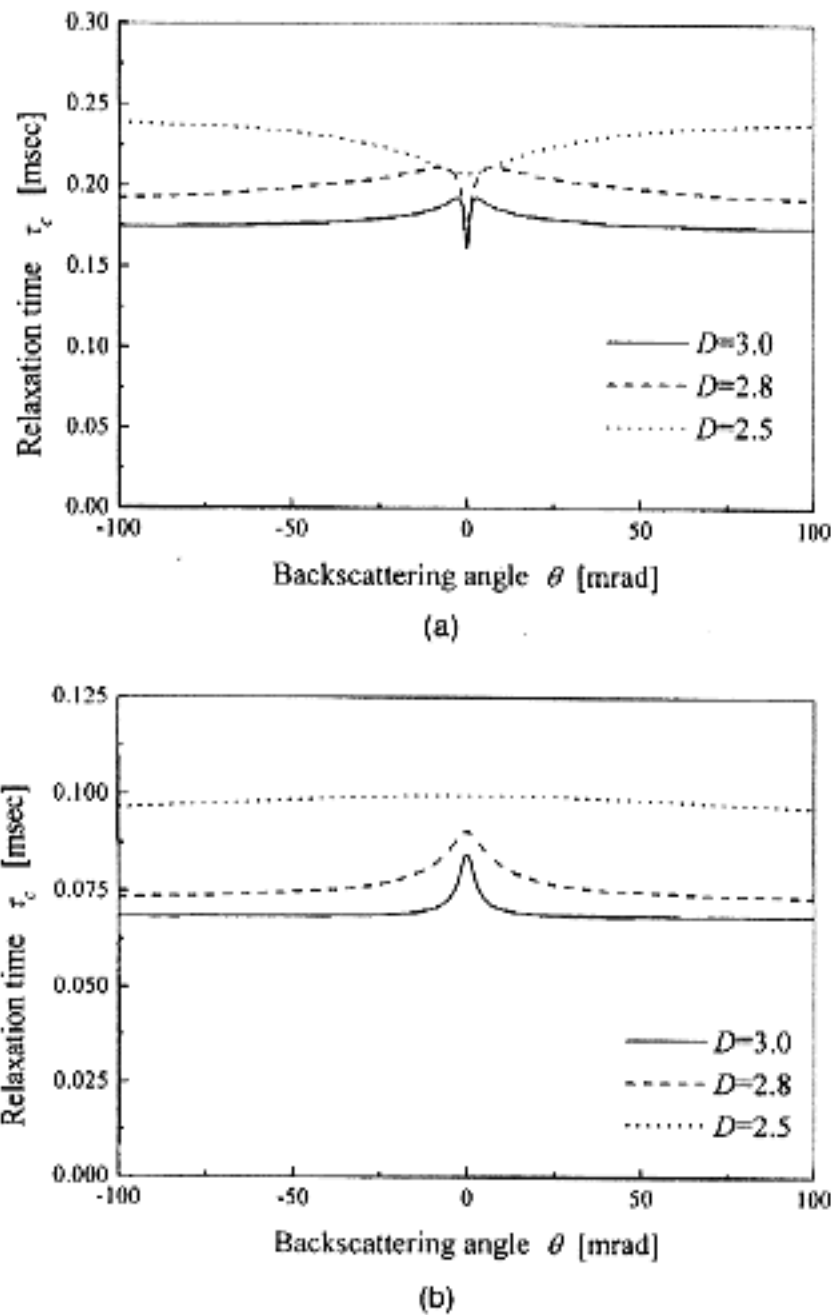
(a)

(b)

Fig. 4 Angular dependence of the relaxation time of (a) the co-polarized and (b) the cross-polarized components of light backscattered multiply from three different media with dimensions of $D = 2.5$, 2.8, and 3.0. The relaxation time is estimated to be $\tau_c = 2/A^2$ by fitting the TAF to $\exp(-A\sqrt{\tau})$.

the relations of $l_D^* \gg l_D$ and $n^* \gg 1$ are more enhanced with an increase of the particle size, the features of the TPDF and the SODF are generally held by replacing l_D and n with l_D^* and n^*, respectively. However, the relation of $p(s,\theta) = p(nl_D,\theta)$ breaks down in fractal media because the TPDF in the region of $s < l_D^*$ is no longer equivalent to the SODF in the region of $n < n^*$. Therefore, the dependence of the TAFs in $\tau > \tau_c$ on the fractal dimension comes primarily from that of the SODFs defined in the discrete version given by Eq. (4). In the region of $s > l_D^*$ where the scattering light contributes mainly to a decreasing behavior of the TAFs in $\tau < \tau_c$, the TAFs shown in Figures 1(a) and 1(b) decrease in accordance with $\exp(-A\sqrt{\tau})$. Therefore, the Laplace transform relation still holds between the TAFs and the TPDFs.

Figures 4(a) and 4(b) show the angular dependences of the relaxation time for the co- and cross-polarized components of light backscattered multiply from the fractal media, which show double peaks and a single peak around and in the direction

of retroreflection, respectively, and vary considerably depending on the dimensions of the media. This comes from the fact that a peak width of the enhanced backscattering spreads dramatically with a decrease in dimension but its dependence becomes weak in both directions of retroreflection and with considerably large backscattering angles.

We are concerned next in the TAF of multiply backscattered light with the relation between the fractal dimension and the absorption. The TAF of the multiply backscattered light from homogeneously random media is expressed by

$$\gamma(\tau) = \int_0^\infty p(s) \exp(-\mu_a s) \exp(-\tau s / \tau_0 l^*) ds,$$

(6)

when an independence between the scattering and absorption is assumed in the scattering process at the event.[18] Under the diffusion approximation, the integration of Eq. (6) yields

$$\gamma(\tau) = \exp(-A\sqrt{\tau + \tau_0 l^* \mu_a}) / \exp(-A\sqrt{\tau_0 l^* \mu_a}).$$

(7)

The TAF at the limitation of $\mu_a \rightarrow 0$ decays in accordance with $\exp(-A\sqrt{\tau})$. Equation (7) shows that the TAF in the long delay time decreases independently of the absorption while the effect of the absorption appears in the time shift toward the longer delay time proportional to a square root of the absorption coefficient. In this case, the time shift is transformed to the increase of the width of the TAF.

Figures 5(a) and 5(b) show the TAFs for the co- and cross-polarized components from the absorbent fractal aggregates of particles with $D = 2.8$ for five different absorptions with $\mu_a = 0$, $1/10l_D$, $1/5l_D$, $1/3l_D$, and $1/l_D$. The relaxation time of the TAF increases consistently with the absorption since the medium more strongly absorbs the light propagating along the longer paths that lead to a rapid decay of the TAF. However, a decreasing rate of the TAF is independent of the absorption in the region of the long delay time. The influence of the absorption appears only in the time shift of the TAF. This property agrees with that for homogeneously random absorbent media.

Figures 6(a) and 6(b) show the time shifts of the TAFs for the co- and cross-polarized components as a function of the absorption for five different fractal media with $D = 2.5$, 2.6, 2.7, 2.8, and 3.0, respectively. In Figure 6, the solid line stands for the theoretical curve of the time shift given by Eq. (7) for homogeneously random media. As can be seen, a variation of the time shift coincides with theory in the region of small absorption and the discrepancy increases consistently with the absorption. This is the reason why the contribution with the low scattering orders, especially the single scattering, becomes more dominant with an increase of absorption since the absorption coefficient $\mu_a = 1$ means
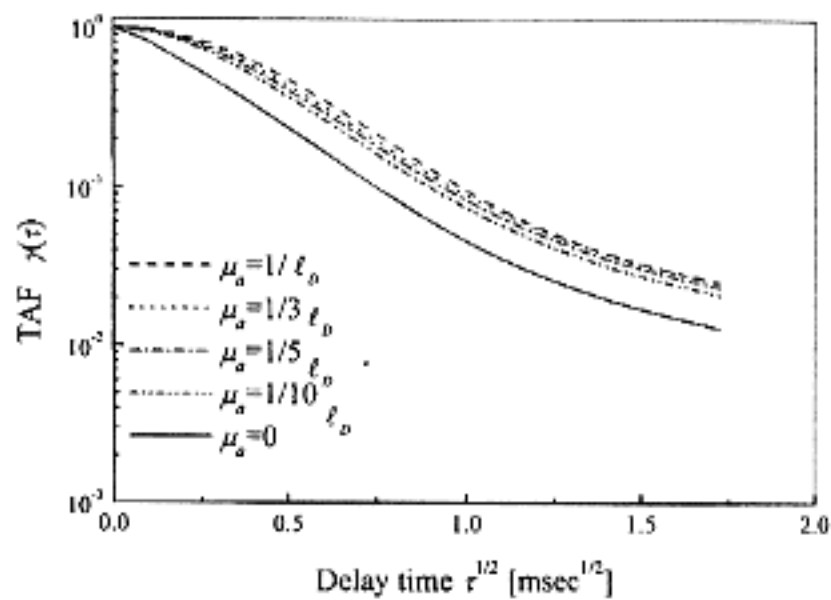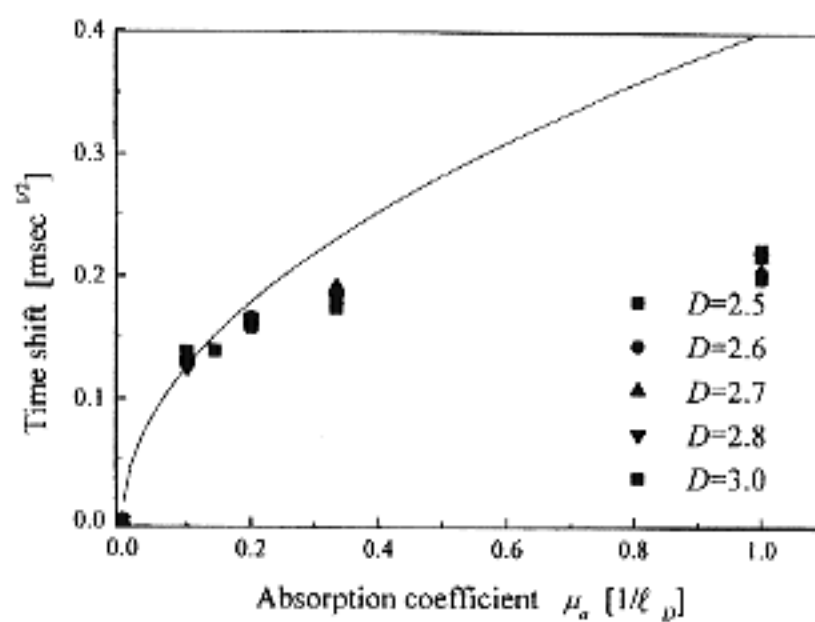
(a)
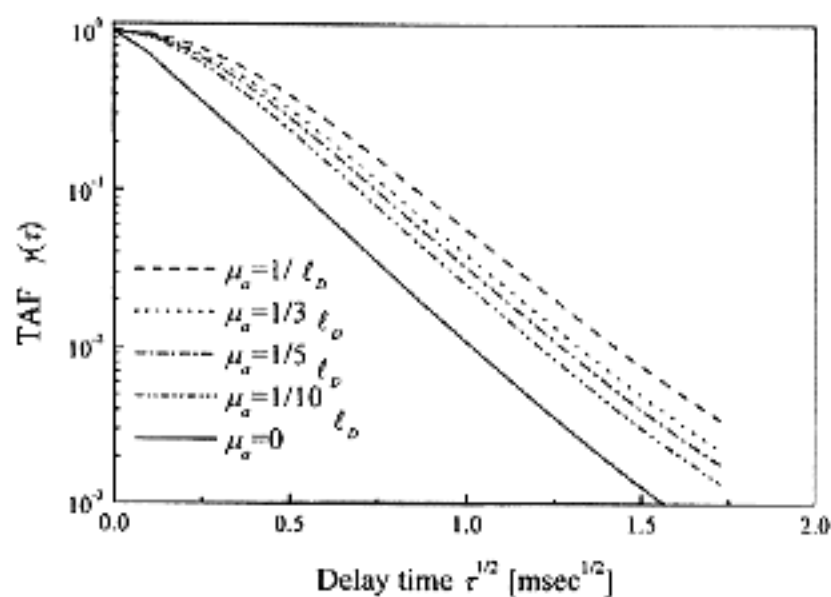
(b)

Fig. 5 TAFs of (a) the co-polarized and (b) the cross-polarized components of light scattered multiply in the direction of retroreflection from five different absorbent media of $\mu_a = 0$, $1/10l_D$, $1/5l_D$, $1/3l_D$, and $1/l_D$ with the dimension $D = 2.8$. The vertical and horizontal axes denote a logarithm of the TAFs and a square root of the delay time, respectively.

(a)

(b)

Fig. 6 Time shifts of the TAFs of (a) the co-polarized and (b) the cross-polarized components of light backscattered multiply from five different absorbent media with dimensions of $D = 2.5$, 2.6, 2.7, 2.8, and 3.0. The horizontal axis denotes the absorption coefficient normalized by the effective mean free path length l_D.
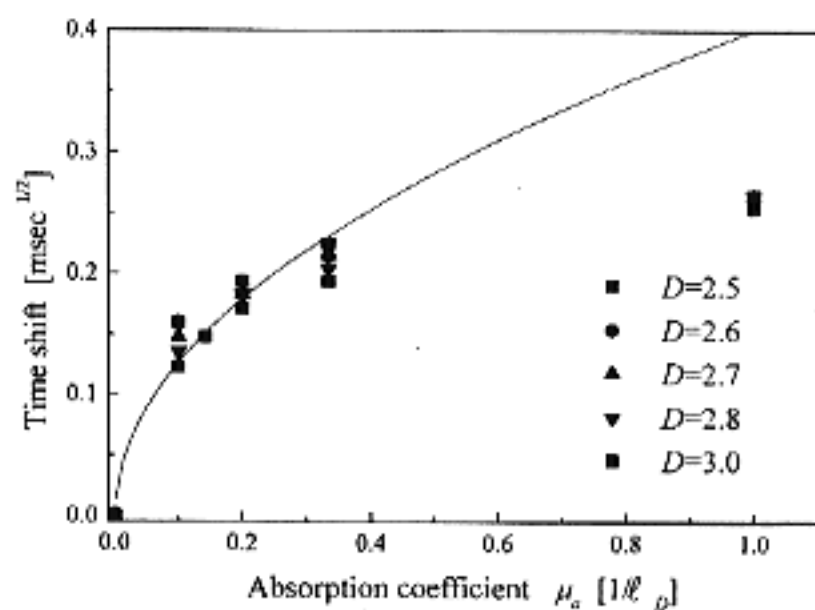
that the absorption length is equal to the mean free path length. It is confirmed by the fact that the discrepancy for the co-polarized component is larger than that for the cross-polarized one without the contribution of single scattering. Nevertheless, the results in numerical simulations indicate that diffusive wave spectroscopy has the capability of measuring the absorption of fractal media without the influence of the dimension by using the time shift of the TAF.

4 CONCLUSIONS

In this article, we have investigated the TAF of light backscattered multiply from the fractal aggregate of particles in an absorbent medium. The discussions were conducted by taking into account the discrete and continuous TAFs defined by the SODF and the TPDF, respectively. The temporal statistics of the backscattered light in the region of time longer than the relaxation time depend on the fractal dimension and are expressed as the discrete version of the TAF

by the SODF while, in the region of time less than the relaxation time, they follow a diffusive transport theory of photons under the diffusion approximation and are obtained by the Laplace transform between the TAF and the TPDF. The angular dependence of the TAF of light scattered multiply from fractal media varies dramatically depending on the fractal dimension in comparison with that from homogeneously random media. The influence of the absorption of media appears as the time shift of the TAFs in such a way that they vary approximately in accordance with a square root of the absorption coefficient and are independent of the fractal dimension. Consequently, we have demonstrated that diffusive wave spectroscopy has the capability of measuring the absorption of fractal media without dilution.

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