Size distribution effect on the power law regime of the structure factor of fractal aggregates

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We consider the large qR_g, where q is the magnitude of the scattering wave vector and R_g is the aggregate radius of gyration, part of the structure factor of fractal aggregates, and quantify the coefficient C of the power law, $S(q) \sim C(qR_g)^{-D}$, where D is the fractal dimension, for various structure factors proposed in the literature. With the aid of earlier work, we conclude the most accurate structure factors have $C=1.0$. We then calculate the effects of polydispersity on this coefficient, and show the effects are significant, enough so to allow a measurement of the distribution width. These concepts are accurately supported with scattering data from a diffusion limited aerosol and a reaction limited colloid. [S1063-651X(99)05112-0]

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I. INTRODUCTION

Scattering techniques can provide detailed information regarding the size and morphology of clusters of particles [1–6]. These techniques involve measurement of the scattered intensity $I(q)$ as a function of the magnitude of the wave vector q, which is dependent on the scattering angle. The basis for the interpretation of these measurements is a knowledge of the relationship between the structure factor, which is the $I(0)$ normalized scattering intensity, and the real-space structure of the aggregate. Furthermore, the process of aggregation which creates the aggregates in aerosols and colloids leads to a distribution of aggregate sizes, and the effects of this distribution on the shape of $I(q)$ vs q must be understood.

Aggregates formed by the destabilization of aerosols and colloids have a fractal morphology [7–9]. These self-similar clusters are well described by

$$N = k_0 (R_g/a)^D, \tag{1}$$

where N is the number of primary particles or monomers in the aggregate, k_0 is a constant of order unity, R_g is the radius of gyration of the aggregate, a is the monomer radius, and D is the fractal dimension. In this paper we first review past work [6–10] regarding the possible forms of the single cluster structure factor of fractal aggregates, show that the various forms differ in the large qR_g regime of the structure factor, and make a conclusion regarding the correct form. We then calculate the effects of polydispersity in aggregate size on the large qR_g regime, and find that these can be significant. Consequently, one important conclusion is that use of the single aggregate structure factor for analysis of scattering data from polydisperse ensembles, which is almost always the case, is erroneous. A beneficial consequence is that the large qR_g regime contains information regarding the aggregate size distribution. We demonstrate the facility of this concept with scattering data from both an aggregating aerosol, which creates diffusion limited cluster aggregates (DLCA), and an aggregating colloid, which creates reaction limited cluster aggregates (RLCA), and quantitatively measure the effective width of the distribution.

II. STRUCTURE FACTOR OF SINGLE AGGREGATES

The structure factor and the density autocorrelation function of the aggregate are Fourier transform pairs; thus

$$S(q) = \int e^{i\vec{q}\cdot\vec{r}} g(\vec{r}) d\vec{r}. \tag{2}$$

For a fractal aggregate the autocorrelation function has the form

$$g(r) \sim r^{D-d} h(r/\xi). \tag{3}$$

Here D is the fractal dimension, d the spatial dimension, and ξ a measure of the linear size of the aggregate proportional to the radius of gyration R_g. The function $h(r/\xi)$ is the cutoff function describing the perimeter of the aggregate. Its properties are that $h(r/\xi \lesssim 1) \simeq 1$, but for large r/ξ it falls off faster than any power law. With form (3), the Fourier transform of Eq. (2) leads to a structure factor, which in the power law regime is given by

$$S(q) = C(qR_g)^{-D}, \quad qR_g \gg 1. \tag{4}$$

This form for $S(q)$ has two parts: the power law $(qR_g)^{-D}$, which is due to the power law part of the autocorrelation function r^{D-d}; and the coefficient C, which is dependent upon the cutoff function $h(r/\xi)$. It is convenient to quantify the cutoff function by writing

$$h(r/\xi) = e^{-(r/\xi)^\beta}. \tag{5}$$

Large β yields a sharper cutoff. Nicolai *et al.* [11] showed that the sharper the cutoff, the smaller the value of C.

A variety of single aggregate structure factors has been proposed, some of which are based on Fourier inversion of the density autocorrelation function and some of which are empirically motivated. We have reviewed some of these structure factors in previous work [6,10]. A more complete review is given here in Table I. Also in Table I we now include the large qR_g coefficient C in Eq. (4). These C values are plotted as a function of the fractal dimension D in Fig. 1. Figure 1 shows that in the range $D=1.7-2.1$, which

TABLE I. Structure factors and cutoff functions.

Name	$h(r/\xi)$	ξ^2	$S(q)$	C	Reference
Exponential	$e^{-r/\xi}$	$\frac{2R_g^2}{D(D+1)}$	$\frac{\sin[(D-1)\tan^{-1}(q\xi)]}{(D-1)q\xi(1+q^2\xi^2)^{(D-1)/2}}$	$\frac{\sin[(D-1)\pi/2]}{D-1}\left[\frac{D(D+1)}{2}\right]^{D/2}$	[3,12]
Gaussian	$e^{-(r/\xi)^2}$	$\frac{4}{D}R_g^2$	$e^{-(qR_g)^2/D}\,{}_1F_1\left(\frac{3-D}{2},\frac{3}{2},\frac{(qR_g)^2}{D}\right)$ ${}_1F_1$ is the Kummer or hypergeometric function	$D^{D/2}\frac{\Gamma(3/2)}{\Gamma\left(\frac{3-D}{2}\right)}$	[10]
Mountain and Mullholland	$e^{(-r/\xi)^{2.5}}$	-	numerical	ca. 0.77 for $D=1.8$	[13]
Overlapping Spheres	$=\left(\frac{4}{3}\pi\xi^3\right)(1+r/4\xi)$ $(1-r/2\xi)^2$, $r<2\xi$ $=0, r>2\xi$	$\frac{(D+2)(D+5)}{2D(D+1)}R_g^2$	numerical	ca. 1.07 for $D=1.75$	[14]
Fisher-Burford	ca. $e^{-r/\xi}$	$\frac{R_g^2}{3}$	$\left(1+\frac{2}{3D}q^2R_g^2\right)^{-D/2}$	$(3D/2)^{D/2}$	[3,15]
Dobbins and Megaridis	-	-	$\exp[-(qR_g)^2/3]$, small qR_g $C(qR_g)^{-D}$, large qR_g join with continuous slope	$(3D/2e)^{D/2}$	[16]
Lin *et al.*	-	-	$\sum\left[1+\sum_{n=1}^{4}C_s(qR_g)^{2g}\right]^{-D/8}$		[17]
			DLCA: $C_1=8/3D$, $C_2=2.5$ $C_3=-1.52$, $C_4=-1.02$	$1-D/400$	
			RLCA: $C_1=8/3D$, $C_2=3.13$ $C_3=-2.58$, $C_4=0.95$	$1+D/160$	

is the range of DLCA to RLCA aggregates, a value of $C \simeq 1$ is obtained, except for the exponential and Fisher-Burford structure factors.

The relevant question now is what cutoff, hence what structure factor, best describes fractal aggregates. Previous work from this laboratory considered light scattering from soot fractal aggregates in flames [10]. It was found that structure factors derived from autocorrelation functions with roughly Gaussian, $\beta=2$, cutoffs gave the best fits to the data when the effect of the aggregate polydispersity was included. The structure factor derived from the exponential, $\beta=1$, cutoff did a poor job of fitting the data, as did the Fisher-Burford form which approximates the exponential structure factor. Subsequently we used TEM images of soot aggregates thermophoretically captured from a flame to compute $g(r)$ directly [18]. Again cutoffs much sharper than exponential, and well described by a Gaussian, were found. Lin *et al.* [17] created both DLCA and RLCA aggregates with computer simulation. These were Fourier transformed to $S(q)$ and then an average $S(q)$ was fit to a polynomial in qR_g. These are given in Table I. Nicolai *et al.* [11] fitted both these DLCA and RLCA $S(q)$'s to structure factors derived from Eqs. (2) and (3) with arbitrary β and found $\beta=2$ fit best. Recently, Haw *et al.* [19] also concluded a cutoff significantly faster than exponential is necessary to describe scattering data.

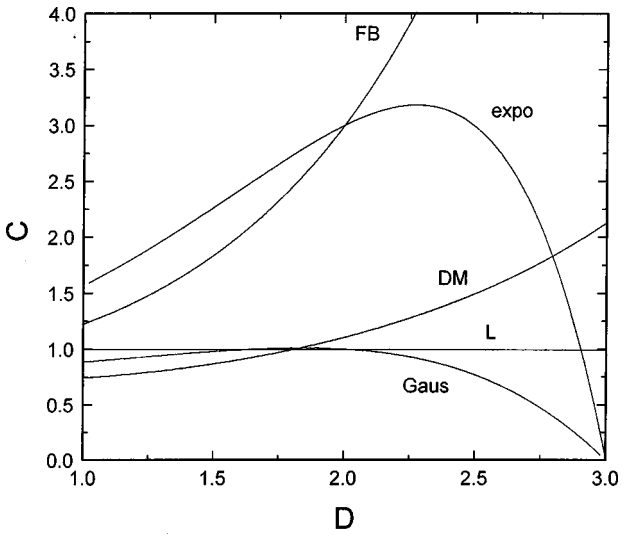

FIG. 1. Coefficient C of the large qR_g, power law regime of the single cluster structure factor $S(q)=C(qR_g)^{-D}$ as a function of D for various structure factors. Structure factors are: expo, exponential; Gaus, Gaussian; FB, Fisher-Burford; DM, Dobbins and Megaridis; L, Lin *et al.*

Given the experimental evidence in favor of a structure factor derived from a Gaussian cutoff and the agreement of the coefficient C in the relevant range of fractal dimensions with other empirically based structure factors, we shall take $C=1.0\pm0.05$ as the best value to describe the power law regime of the structure factor of a single aggregate with D in the range 1.7–2.1.

III. STRUCTURE FACTOR OF AN ENSEMBLE OF POLYDISPERSE AGGREGATES

Any real experiment detecting scattered radiation from an ensemble of fractal aggregates will involve a polydisperse

(in cluster size) ensemble. Aggregates are a result of aggregation which always gives a finite width to the cluster size distribution. This polydispersity causes the shape of the observed structure factor to be different than that of the single cluster structure factor. The single cluster structure factor, dependent on the cutoff and D, was described above. Now we consider how the shape is modified by a distribution in cluster sizes.

To determine the effective optical structure factor for an ensemble of aggregates, we will use the Rayleigh-Debye-Gans (RDG) approximation for light scattering from an aggregate. This approximation assumes no significant intracluster multiple scattering, hence the differential scattering cross section for the aggregate is related to the differential scattering cross section of the monomer by [6]

$$\frac{d\sigma^{agg}}{d\Omega} = N^2 \frac{d\sigma^{mon}}{d\Omega} S(qR_g). \tag{6}$$

If the monomer is in the Rayleigh regime specified by $ka = 2\pi a/\lambda < 1$, then [20]

$$\frac{d\sigma^{mon}}{d\Omega} = k^4 a^6 F(m), \tag{7}$$

where

$$F(m) = \left| \frac{m^2 - 1}{m^2 + 2} \right|^2, \tag{8}$$

and m is the index of refraction. This RDG approximation has been tested both experimentally [21] and theoretically [22], and found to be good to $\sim$10% for fractal aggregates with $D \simeq 1.8$.

In an experiment the structure factor is determined by measuring the scattered intensity as a function of the wave vector q, where

$$q = 4\pi\lambda^{-1} \sin\theta/2. \tag{9}$$

q is varied by changing the scattering angle θ. This scattering is normalized by the $q \to 0$ scattered intensity, i.e., the scattering intensity in the Rayleigh regime of the aggregates. Thus, in general, the effective structure factor for an ensemble of aggregates can be written as

$$S_{\text{eff}}(q) = \int N^2 n(N) S[qR_g(N)] dN / \int N^2 n(N) dN. \tag{10}$$

In Eq. (10), $n(N)$ is the size distribution, i.e., the number of clusters per unit volume with N monomers per cluster. The number of monomers per cluster and the cluster radius of gyration are related by Eq. (1).

Useful analytical solutions to the polydispersity problem in the power law regime can be obtained if we ignore the Guinier regime near $qR_g \sim 1$ and represent the single aggregate structure factor by its Rayleigh and power law limits

$$S(q) = 1, \quad qR_g \ll 1 \tag{11a}$$

$$= C(qR_g)^{-D}, \quad qR_g \gg 1. \tag{11b}$$

To compute the results of Eq. (10) applied to Eq. (11), we define the ith moment of the size distribution as

$$M_i = \int N^i n(N) dN. \tag{12}$$

Then with Eq. (1) we find

$$S_{\text{eff}}(q) = 1, \quad qR_g \ll 1 \tag{13a}$$

$$= \frac{M_1}{M_2} k_0 C (qa)^{-D}, \quad qR_g \gg 1. \tag{13b}$$

The structure factor, in its most economical form, is the Rayleigh normalized scattering as a function of the unitless variable qR_g. Thus we require $S(q)$ as a function of qR_g, where R_g is the ''average'' R_g measured by the experiment. Since the experiment is scattering, the average is determined by the scattering. The average R_g is best determined from analysis of scattering in the Guinier regime [1,2]. One can show by expansion of Eq. (2) that for $qR_g \lesssim 1$ the structure factor will have the form

$$S(q) = 1 - \tfrac{1}{3} q^2 R_g^2. \tag{14}$$

This is called the Guinier equation and is quite general, independent of the form of $g(r)$. It provides a convenient way to measure R_g, since a plot of inverse $S(q)$ versus q^2 will be linear with slope $R_g^2/3$. This is essentially the method proposed long ago by Zimm [23] for biophysical applications, and has also seen extensive use for sizing of fractal aggregates [24,25].

Scattering from a polydisperse system of aggregates will yield the effective structure factor in Eq. (10). Substitution of Eq. (14) into Eq. (10) yields, with the definition of moments in Eq. (12) and the fundamental scaling relation (1),

$$R_{g,z}^2 = a^2 k_0^{-2/D} \frac{M_{2+2/D}}{M_2}. \tag{15}$$

In Eq. (15) $R_{g,z}$ is the ''z-averaged'' radius of gyration, which is an average of R_g weighted by the second moment of the size distribution.

Recalling once again the laboratory, the experimentalist will measure an uncalibrated $I(q)$, and normalize it with the Rayleigh regime scattering $I(0)$ to obtain Eq. (13). He or she will then use the Guinier regime to determine $R_{g,z}$ [Eq. (15)]. Then he or she will plot $I(q)/I(0)$ versus $qR_{g,z}$ to obtain the structure factor of the ensemble. Thus one uses Eqs. (13) and (15) and substitute on $k_0 a^{-D}$ to obtain

$$S_{\text{eff}}(q) = 1, \quad qR_{g,z} \ll 1 \tag{16a}$$

$$= C \frac{M_1}{M_2} \left(\frac{M_{2+2/D}}{M_2} \right)^{D/2} (qR_{g,z})^{-D}, \quad qR_{g,z} \gg 1. \tag{16b}$$

The most notable result in Eqs. (16) is that the coefficient of the power law is modified by the polydispersity of the ensemble. If this modifying factor is significantly different than unity, then use of single cluster structure factors for analysis of scattering data could yield erroneous results, a

consideration that seems to have been missed in some previous work. The result also opens an opportunity to measure, to some degree, the polydispersity of the ensemble.

We define the polydispersity factor in Eq. (16b) as

$$C_p=\frac{M_1}{M_2}\left(\frac{M_{2+2/D}}{M_2}\right)^{D/2}. \tag{17}$$

Then Eq. (16b) becomes

$$S_{eff}(q)=CC_p(qR_{g,z})^{-D}, \quad qR_{g,z}\gg 1. \tag{18}$$

Given a size distribution, the polydispersity factor C_p can be calculated. It is well established that an aggregating system develops a self-preserving, scaling distribution [26,27] given by

$$n(N)=M_1 s_2^{-2}\phi(x), \tag{19}$$

where

$$\phi(x)=Ax^{-\tau}e^{-\alpha x}, \tag{20}$$

and x is the relative size

$$x=N/s_2. \tag{21}$$

In the equations above $s_2=M_2/M_1$ is a mean size, and A and $\alpha=2-\tau$ are constants determined by the normalization. The exponent τ is a measure of the width of the distribution with large τ, implying a broad distribution. It is equal to the coagulation kernel homogeneity for a certain class of kernels including DLCA. This scaling form is valid when $x>1$, the small x form being different and described in Ref. [27]. Since scattering strongly weights the large end, i.e., $x>1$, part of the distribution, the small x part has little effect on the properties of scattering from an ensemble of aggregates and hence can be ignored.

Other forms for the size distribution of aggregates exist but caution must be exercised in their use. For example the intuitive log normal distributions are frequently used in the literature. However, we have shown [25] that these distributions yield erroneous values for distribution moments higher than the second when compared to the exact scaling distribution. Since scattering involves higher moments, such as $M_{2+2/D}\simeq M_3$ for $D\simeq 2$, it would be erroneous to use these distributions for light scattering analysis.

With this scaling distribution, the moments of Eq. (12) are found to be

$$M_i=M_1 s_2^{i-1}A\alpha^{\tau-i-1}\Gamma(i+1-\tau), \tag{22}$$

where $\Gamma(x)$ is the gamma function. Substitution of Eq. (22) into Eq. (17) yields the polydispersity factor to be

$$C_p=\frac{1}{2-\tau}\left[\frac{\Gamma(3-\tau+2/D)}{\Gamma(3-\tau)}\right]^{D/2}. \tag{23}$$

In Fig. 2 we graph C_p as a function of the width parameter τ for a variety of fractal dimensions D. In general, we find C_p to be significantly greater than unity; thus our warning not to use single cluster structure factors for analysis of scattering experiments involving a polydisperse ensemble of aggregates is quite appropriate. In particular, for DLCA it is expected and well verified that $\tau=0$ and $D=1.75$; then from Eq. (23) or Fig. 2 we find $C_p=1.53$. Most importantly, Eq. (23) affords the opportunity to measure the distribution width parameter τ. Below we apply this to successfully measure τ in both the DLCA and RLCA regimes. Koylu [28] made an analysis of the power law regime to measure the size distribution width similar to ours. However, his method requires measurement of the absorption of the aerosol or colloid, and hence requires the particulate index of refraction.

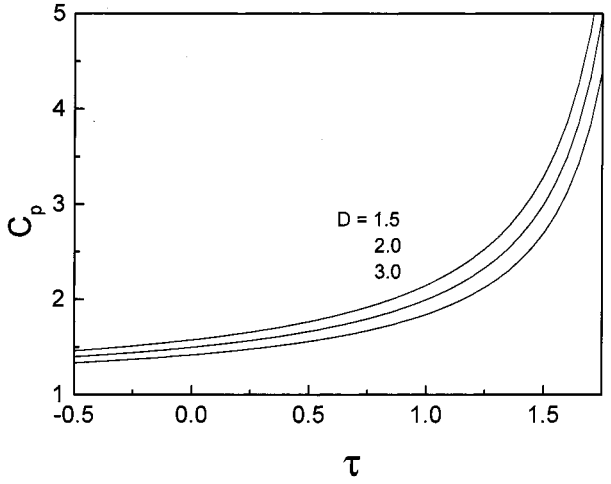

FIG. 2. Polydispersity factor C_p for the large qR_g, power law regime of the structure factor $S(q)=CC_p(qR_{g,z})^{-D}$ for an ensemble of clusters as a function of the width parameter τ of the scaling size distribution for three values of the fractal dimension D.

IV. EXPERIMENTAL METHOD

We performed static light scattering measurements on two fractal aggregate systems: A TiO_2 aerosol which created DLCA aggregates with $D=1.7$, and a polystyrene colloid which created RLCA aggregates with $D=2.15$.

The TiO_2 aerosol was generated by thermal decomposition of titanium tetraisopropoxide (TTIP), similar to the method reported by Okuyama *et al.* [29]. TTIP vapor was produced by heating the liquid TTIP to temperatures around 80 °C. This vapor was carried into a half meter long tube furnace at 400° by dry N_2 gas at a flow rate of 0.5 liter per minute. The decomposition of TTIP vapor took place inside the furnace to create spherical TiO_2 particles with a diameter of 70 ± 10 nm. A stainless steel cylindrical chamber (inside diameter 20 cm, height 35 cm) was used to contain the fresh aerosol. At the middle height of the chamber, a curved glass window allowed for light scattering measurements at scattering angles from $\theta=0°$ to 120°.

The colloid was made by diluting the original polystyrene sphere solution (uniform latex microspheres, diameter 28 nm, Duke Scientific Corp.) 10 000 times with distilled water. This resulted in an initial number density $9.6\times 10^{11}\,\mathrm{cm}^{-3}$. A glass container (inside diameter 8 cm, height 12 cm) was used to hold the colloid for the light scattering measurements. Addition of 0.07 mole NaCl into the colloid induced a reaction limited cluster aggregation.

A vertically polarized Ar ion laser with wavelength λ

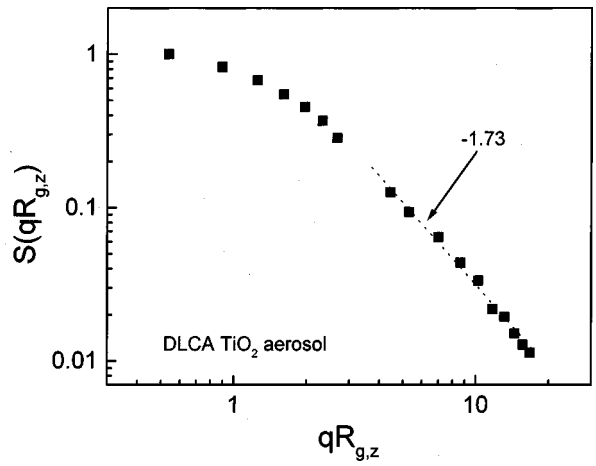

FIG. 3. Structure factor for a TiO_2 aerosol.

$=488$ nm was employed in this research. The experiments were normally started at $\theta=2°$ and ended at 120°, avoiding the spots of incident beam. This led to the wave vector range of $q=0.45$ to 22 μm^{-1} for the aerosol, and 0.59 to 29.6 μm^{-1} for the colloid.

V. RESULTS AND DISCUSSION

Figures 3 and 4 show the Rayleigh normalized scattered intensity, hence the structure factor of the ensemble of aggregates, for the aerosol and colloidal systems, respectively. These are plotted versus $qR_{g,z}$, i.e., the scattering wave vector times the z-averaged radius of gyration measured in the Guinier regime. Lines in these plots show the power law dependencies with fractal dimension of $D=1.7$ and 2.1, respectively.

The monomer sizes are $a=35\pm7$ and 14 ± 1 nm, and the aggregate radii of gyration are 790 and 430 nm, respectively. Thus N (with $k_0=1.3$ [18]) is in the range 260–2000, showing that each system is mature from an aggregation point of view. This is what we need to have a significant power law regime, which is apparent in both Figs. 3 and 4, so that we can now fit these regimes to Eq. (18). In fitting to Eq. (18), a criterion had to be used to determine the minimum value of $qR_{g,z}$ to be used in the fit. We did this by constraining the fit to $qR_{g,z}\geqslant5$ (see Fig. 1, Ref. [10]).

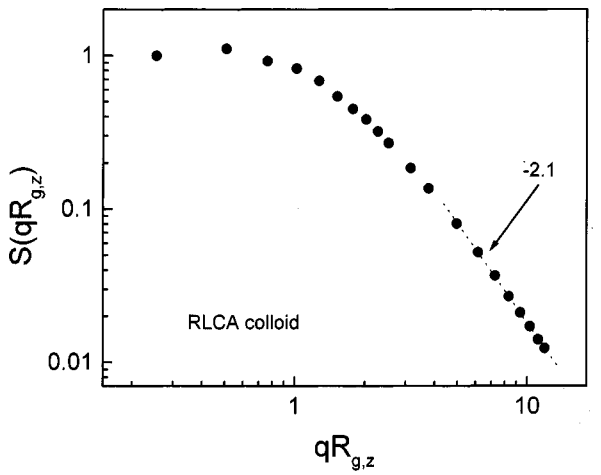

FIG. 4. Structure factor for a polystyrene colloid.

TABLE II. Results of fitting to Eq. (18).

Systems	D	$C_p\,(C=1)$	τ
Aerosol 1	1.66	1.57	0.08
Aerosol 2	1.65	1.71	0.46
Aerosol 3	1.71	1.69	0.42
Aerosol 4	1.73	1.64	0.31
Aerosol 5	1.72	1.53	−0.05
Colloid 1	2.11	2.6	1.38
Colloid 2	2.18	3.6	1.65

Five different aerosol and two different colloid structure factors were fit with Eq. (18). The results are contained in Table II. Both sets of fractal dimensions, $D=1.7$ for the aerosol and 2.15 for the colloid, are consistent with the expected DLCA and RLCA regimes of these two systems [9]. The values of the coefficient are significantly different from each other and from 1. To illustrate these differences, in Fig. 5 we plot the quantity $(qR_{g,z})^D S(qR_{g,z})$ vs $qR_{g,z}$. By Eqs. (16) and (18), such a log-log plot should rise for small $qR_{g,z}$ with slope D, and then level off for large $qR_{g,z}$ to a constant equal to CC_p. This proves to be true in Fig. 5 to dramatically demonstrate the difference between the DLCA and RLCA structure factors caused by the polydispersity (as opposed to the difference in fractal dimension caused by the structure). If we take the single cluster structure factor coefficient to be $C=1$, as concluded above, then the measured C_p values are those tabulated in Table II.

When these C_p values are compared to Fig. 2 or Eq. (23), values of the exponent τ can be determined, and these values are also given in Table II. For the aerosol the average τ of the five runs is $\langle\tau\rangle=0.24\pm0.23$. This is consistent with the expected value of $\tau=0$ for DLCA [27], although on the edge of the error range. For the colloid we find $\langle\tau\rangle=1.5\pm0.2$. This is in good agreement with previous work, both experimental [30–32] and theoretical [33,34], for RLCA aggregation.

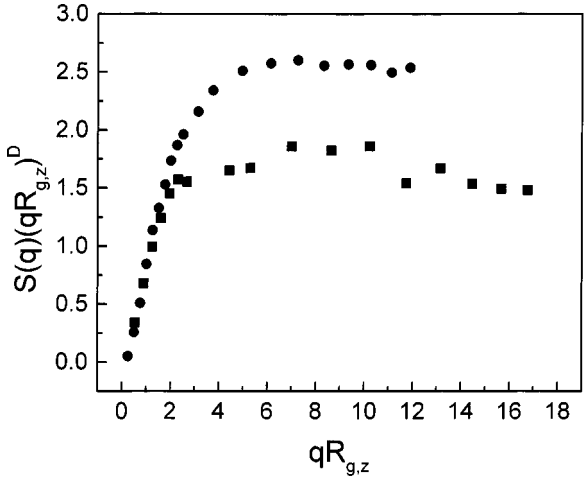

FIG. 5. $(qR_{g,z})^D S(qR_{g,z})$ plotted vs $qR_{g,z}$ for a DLCA aerosol and a RLCA colloid. The constant level at large $qR_{g,z}$ is, by Eq. (18), equal to CC_p.

VI. CONCLUSIONS

For a single aggregate the large qR_g power law regime carries information regarding the fractal dimension of the aggregate, in the power of qR_g, and, regarding the sharpness of its perimeter, in the coefficient of the power law. The coefficient is well described by 1.0 for DLCA and RLCA aggregates. Scattering from a polydisperse system of aggregates yields an effective structure factor which, except for systems with large polydispersity [11], retains the negative fractal dimension power law dependency of a single aggregate. The coefficient, however, is significantly altered. Our experiments demonstrate the validity of this and the usefulness for measuring the polydispersity. Moreover, proper analysis must include these polydispersity effects and not use the single cluster structure factor.

ACKNOWLEDGMENT

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