Test of Static Structure Factors for Describing Light Scattering from Fractal Soot Aggregates

C. M. Sorensen,* J. Cai, and N. Lu

Department of Physics, Kansas State University, Manhattan, Kansas 66506-2601

Received March 5, 1992. In Final Form: May 14, 1992

We compare several static structure factors for light scattering from fractal aggregates. The variation between these structure factors is a result of different cutoff functions for the cluster density correlation function. Light scattering data obtained from soot aggregates in a premixed CH_4/O_2 flame are fit with three representative structure factors. Fractal dimensions determined from these fits under the assumptions of a monodisperse size distribution are all unsatisfactory. Inclusion of a scaling cluster size distribution yields good fits to structure factors derived from correlation functions with cutoffs faster than exponential, the exponential structure factor yielding a poor fit. A Gaussian cutoff was found to work the best.

Introduction

The technique of in situ optical particle size measurement has a long and useful history. As with any technique, certain assumptions are usually made regarding the properties of the system being probed. Common to many early particle sizing efforts was an assumption regarding the shape of the particles, typically being taken as spherical. This assumption breaks down, however, if aggregation occurs because for solid particles aggregates are ramified, nondense clusters. The problem of description of such clusters has been, in the last decade, surmounted by the demonstration that these clusters are usually describable by the fractal concept.[1-3] A fractal is a self-similar object at various length scales with its degree of ramification describable by the fractal dimension, $D < 3$. Thus new vistas have opened for optical measurements which may now more accurately measure effective length scales and the fractal dimension for clusters of particles.

In order to take full advantage of our new knowledge regarding the fractility of clusters, we must have an accurate description of the static structure factor. The structure factor describes the intensity of radiation, for our purposes light, scattered from the aggregate or ensemble of aggregates all of equivalent size as a function of the scattering wave vector (see below). For the early pioneering efforts in this area, it was sufficient to know that for small wave vectors q such that the product $qR_g \ll 1$, where R_g is the radius of gyration of the cluster, the structure factor $S(qR_g)$ was a constant; while for $qR_g \gg 1$, $S(qR_g) \sim (qR_g)^{-D}$. Now, as we refine this technique, a detailed form for $S(qR_g)$ is required. To obtain this the exact form for the density autocorrelation function of the cluster is necessary since $S(qR_g)$ is the spatial Fourier transform of this function. The form for this correlation function is

$$g(r) = Ar^{D-3}h(r/\xi) \tag{1}$$

for three-dimensional space (A is a constant). The function $h(r/\xi)$, where $\xi \sim R_g$ is some characteristic length, describes the manner in which the power law correlation, r^{D-3}, characteristic of a fractal object, is cut off at the perimeter of the cluster.

It is here where current uncertainty arises. Although a few forms for the cutoff function $h(r/\xi)$ have been proposed, there is no universal agreement as to which, if any, of these functions is the correct one and no studies regarding its possible universality, i.e., whether it is dependent upon the nature of the aggregation process which yields the fractal cluster or not. One approach to this problem is to test the various cutoffs against scattering data through comparison of the data to the derived $S(qR_g)$. It is the primary purpose of this paper to describe such a test using in situ measurements on soot particles in a flame. When we do this, we find considerable diversity in the infered fractal dimension occurs depending on which form for the cutoff, hence which structure factor, is used. This occurs because the sizes of soot clusters are typically less than a couple hundred nanometers, hence $qR_g \lesssim 5$. This is not sufficiently large to obtain the asymptotic regime where $S(qR_g) \sim (qR_g)^{-D}$, which would allow an accurate measurement of D. Hence a proper analysis of the structure factor is particularly important for small clusters.

Unfortunately, such a test is not possible without some complications. In the problem of light scattering from soot aggregates in a flame, the most important complication is the polydispersity of the aggregates. Polydispersity will, of course, exist in any system undergoing aggregation, and hence its confrontation can be a valuable exercise. Indeed, in the work to be described, we show that a monodisperse assumption leads to an erroneous conclusion regarding $S(qR_g)$ and hence $h(r/\xi)$. Inclusion of a "proper" size distribution is necessary to draw an accurate conclusion regarding $h(r/\xi)$ and $S(qR_g)$.

Theoretical Review

The static structure factor for light scattered from a monodisperse ensemble of fractal aggregates has been calculated by several research groups using a few different cutoff functions. The most used and the least abrupt of these cutoffs is the exponential cutoff

$$h(r/\xi) = e^{-r/\xi} \tag{2a}$$

where

$$\xi^2 = \frac{2}{D(D+1)} R_g^2 \tag{2b}$$

Freltoft et al.,[4] Berry and Percival,[5] and Teixeira[6] have all calculated $S(qR_g)$ using eqs 2 and have arrived at equivalent

(1) Forrest, S. R.; Witten, T. A. *J. Phys. A* **1979**, *12*, L109.
(2) Family, F., Landau, D. P., Eds. *Kinetics of Aggregation and Gelation*; North Holland: Amsterdam, 1984.
(3) Stanley, H. E., Ostrowsky, N., Eds. *On Growth and Form*; Nijhoff: Boston, MA, 1986.
(4) Freltoft, T.; Kjems, J. K.; Sinha, S. K. *Phys. Rev. B* **1986**, *33*, 269.
(5) Berry, M. V.; Percival, I. C. *Opt. Acta* **1986**, *33*, 577.
(6) Teixeira, J. Reference 3.

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results. The result is

$$S(q\xi) = \frac{S(0)\sin[(D-1)\tan^{-1}(q\xi)]}{(D-1)q\xi(1+q^2\xi^2)^{(D-1)/2}} \quad (3)$$

The exponential cutoff structure factor can be approximated by the so-called Fisher–Burford form,[7] $S(q) = (1 + 2q^2R_g^2/3D)^{-D/2}$. In fact the comparison is exact at $D = 2$. The difference becomes significant in fits to data when $D \simeq 1.7$, a typical fractal dimension, so we will not use this form in our analysis.

In the first in situ optical measurement involving a fractal analysis for an aerosol, Hurd and Flower[8] proposed a cutoff derived from the overlap volume of the clusters. They found

$$h(r/\xi) = \left(\frac{4}{3}\pi\xi^3\right)(1 + r/4\xi)\left(1 - \frac{r}{2\xi}\right)^2 \quad r < 2\xi$$
$$= 0 \quad r > 2\xi \quad (4a)$$

where

$$\xi^2 = \frac{(D+2)(D+5)}{2D(D+1)}R_g^2 \quad (4b)$$

This cutoff is sharper than the exponential.

Mountain and Mulholland[9] performed computer simulations to create clusters. Their simulation was made to approximate the conditions typically found in a flame wherein the mean free path of the gas molecules is larger than the monomer size, by roughly a factor of 20 in their simulations. Both the density autocorrelation function and the structure factor, calculated directly using electromagnetic wave theory for scattering from the cluster, were calculated. The simulated $S(qR_g)$ agreed poorly with the Fisher–Burford form and hence one would infer the exponential cutoff would not apply to their clusters. This was shown to be true since they found from direct analysis of $g(r)$ that the cutoff was best described by

$$h(r/R_g) = e^{-c(r/R_g)^{2.5}} \quad (5)$$

The exponent in this modified exponential was described as somewhere between 2 and 3. When this exponent is 2.5, $c = 0.2$. One can show numerical Fourier transforms of eq 1 with eq 5 agree well with the $S(qR_g)$ calculated directly by Mountain and Mulholland.

We are also aware of two instances in which structure factors have been synthesized without recourse to the density correlation function. Dobbins and Megaridis[10] used the pragmatic concept that $S(qR_g)$ should obey the universal Guinier form for small qR_g and then go as $(qR_g)^{-D}$ thereafter. The two regimes are connected by forcing continuity in both $S(qR_g)$ and its qR_g derivative at the junction of the two regime. Thus

$$S(x) = e^{-(qR_g)^2/3}, \quad \text{small } qR_g \quad (6a)$$
$$= C(qR_g)^{-D}, \quad \text{larger } qR_g \quad (6b)$$

where C and the exact boundary between small and large qR_g was chosen to obtain continuity.

Lin et al.[11] have also simulated particles and directly determined the structure factor. For colloidal diffusion limited cluster aggregation they found a best fit polynomial

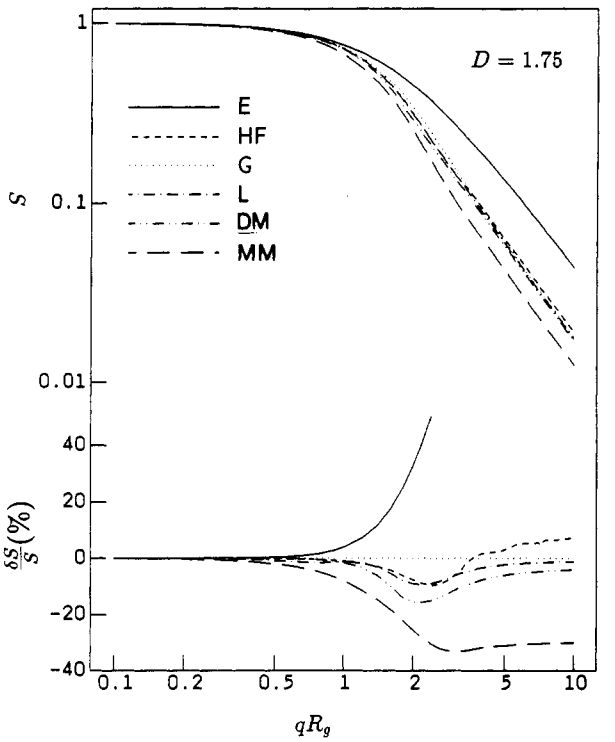

Figure 1. Comparison of various theoretical forms for the structure factor $S(qR_g)$: upper section shows the structure factors, lower section shows the normalized deviation from the Gaussian structure factor ($\delta S = S - S_G$). Key: E = exponential; HF = Hurd and Flower; G = Gaussian; L = Lin et al.; DM = Dobbins and Megaridis; MM = Mountain and Mulholland.

to their numerical results as

$$S(qR_g) = (1 + \sum_{s=1}^{4} C_s(qR_g)^{2s})^{-D/8} \quad (7)$$

with $C_1 = 8/3D$, $C_2 = 2.50$, $C_3 = -1.52$, and $C_4 = 1.02$.

We begin our critique of these structure factors with the work of Nelson[12] who compared Berry and Percival's structure factor derived using an exponential cutoff to simulated data. The comparison was not good. Nelson found that if a Gaussian was used instead in the Berry and Percival calculation, significant improvement was obtained.

To obtain the structure factor for a Gaussian cutoff

$$h(r/\xi) = e^{-(r/\xi)^2} \quad (8)$$

one can analytically Fourier transform eq 1 with eq 8. The result is

$$S(qR_g) = e^{-(qR_g)^2/D}\,{}_1F_1\left(\frac{3-D}{2}, \frac{3}{2}; \frac{(qR_g)^2}{D}\right) \quad (9)$$

where ${}_1F_1$ is the confluent hypergeometric series and

$$\xi^2 = \frac{4}{D}R_g^2 \quad (10)$$

We now wish to compare all these structure factors graphically. Figure 1 plots the $S(qR_g)$ for the exponential cutoff (eq 3), Gaussian (eq 9), Mountain and Mulholland's modified exponential (Fourier transform of eq 1 with eq 5), Hurd and Flower (Fourier transform of eq 1 with eq 4), Dobbins and Megaridis (eq 6), and Lin et al. (eq 7) all for $D = 1.75$. The upper part of Figure 1 shows $S(qR_g)$ vs qR_g, the lower part shows the deviation from the Gaussian cutoff

(7) Schaefer, D. W.; Martin, J. E.; Wiltzius, P.; Cannell, D.S. *Phys. Rev. Lett.* **1984**, *52*, 2371.
(8) Hurd, A. J.; Flower, W. L. *J. Colloid Interface Sci.* **1988**, *122*, 178.
(9) Mountain, R. D.; Mulholland, G. W. *Langmuir* **1988**, *4*, 1321.
(10) Dobbins, R. A.; Megaridis, C. M. *Appl. Opt.* **1991**, *30*, 4747.
(11) Lin, M. Y.; Klein, R.; Lindsay, H. M.; Weitz, D. A.; Ball, R. C.; Meakin, P. *J. Colloid Interface Sci.* **1990**, *137*, 263.
(12) Nelson, J. *J. Mod. Opt.* **1989**, *36*, 1031.

$S(qR_g)$. We somewhat arbitrarily chose this as a reference because Nelson found reasonable agreement with simulated data.

Figure 1 shows the exponential cutoff leads to a structure factor which stands much greater than any of the rest in the $qR_g > 1$ regime. At the other extreme lies the Mountain and Mulholland structure factor, and then fairly much clustered within 10–15% of each other between these extremes lay the others. Similar results, with some shifting, are found for $D = 1.5$ and 2.0. The Mountain and Mulholland and Lin et al. $S(qR_g)$ are empirically derived from computer simulations and hence one would hope that they would agree better. However, these simulations represent different coagulation regimes: that of Mountain and Mulholland in a regime where the gas molecule mean free path is significantly larger than the monomer size while the Lin et al. result is the inverse of this, monomer size is much greater than the mean free path. Hence the difference is not surprising.[13] Regardless of this disparity, we conclude the exponential cutoff does not accurately represent the simulated data.

We now turn to experiment to see if it can help us decide between these different structure factors.

Experimental Method

Our experimental setup was the same as used in previously reported work.[14,15] The flame was supported on a cooled porous frit burner obtained from McKenna Products. The premixed gases passed through a frit 6 cm in diameter. This frit was surrounded by an annular sheath region 0.5 cm wide through which nitrogen was passed. A steel stagnation plate 15 cm in diameter was placed above the burner surface to stabilize the flame. The burner was mounted on a translation stage for adjustment.

The gases used were methane and oxygen premixed before the burner. Their flows were controlled by critical orifices. The cold gas velocity, uniform across the frit, of the mixture was 6 cm/s. The nitrogen sheath flow was also critical orifice controlled to a velocity of 5 cm/s. This arrangement yielded a quasi-one-dimensional flame with the only major variable being the height above the burner h, i.e., the flame was "flat". The fuel-to-oxidizer ratio of these flames is described by the ratio of carbon atoms to oxygen atoms (C/O) in the gas mixture. We used C/O = 0.75.

Our light-scattering apparatus used an argon-ion laser operating at $\lambda = 488$ nm as a source. The vertically polarized light was focused by a 50-cm focal length lens into the flame. The burner was mounted on an xyz-translation stage over the pivot of our goniometer which was carefully aligned as described earlier. An optical rail 1 m long rotated above the pivot on a flat optical table. Mounted on the rail was a 10-cm focal length collection lens that imaged the incident beam onto an iris diaphragm with unity magnification. This gave us a scattering volume 1 mm long at 90°. Light passing through this iris diaphragm then passed through a 488-nm notch filter to eliminate the blackbody light from the hot soot. Detection was made with an ITT FW130 photomultiplier tube. Its analog output was converted to a digital signal, which was read by a personal computer. Values of the scattered light were sampled once per second with 1-s integration times. The variation between samples was usually <1%. Eleven scattering angles, which ranged between 10 and 110° ($0.087 \leq \sin\theta/2 \leq 0.84$), were used in these experiments. The intensity as a function of θ was measured sequentially through the 11 angles. This took ~1 min and was repeated 3 times. The values of the intensity were then averaged and corrected for the change in scattering volume as θ varied by multiplying by $\sin\theta$. The apparent random error of the intensities at any given angle was usually ~1%.

Earlier work[14] on this flame has shown that the scattered light intensity vs q is fairly featureless, indicating small R_g, until we are high in the flame. Hence our data were taken at a height of 20 mm above the burner surface when the stagnation plate was 30 mm above the surface. This yielded $R_g = 134$ nm; hence a maximum $qR_g = 2.9$. This stagnation plate height and run are identical to some of our earlier work.[14,15] To obtain an even larger R_g, we raised the stagnation plate to 40 mm, as far as we could and still keep the flame stable, and scattered light at 30 mm above the flame. This yielded $R_g = 188$ nm for a maximum $qR_g = 4.1$. These data, referred to as run 1 and 2, respectively, were used in our analysis below.

Results

To begin our analysis we determine the cluster radius of gyration R_g for our two runs by a method which is independent of the form of the structure factor. For small qR_g, one has for the scattered light intensity

$$I(q) = I(0)\left(1 - \frac{1}{3}q^2\langle R_g^2\rangle\right) \tag{11}$$

where $\langle R_g^2\rangle$ is an average radius of gyration averaged over the cluster size distribution and the scattered light intensity from a single cluster.[15] In the small qR_g regime, one can show that $\langle R_g^2\rangle \sim M_{2+2/D}/M_2$,[15] where M_i is the ith moment of the distribution. We have shown how a graph of $1/I(q)$ vs q^2, which by eq 11 has a slope of $\langle R_g^2\rangle/3I(0)$ and an intercept of $I^{-1}(0)$, can be used to determine both the root mean square value of R_g (hereafter referred to as R_g) and $I(0)$.[14] We used this analysis here as well and found R_g values of 134 and 188 nm for runs 1 and 2, respectively.

With R_g and $I(0)$ determined independent of the exact form of the structure factor, and hence the cutoff function, we can now use D as the sole variable and fit our data to various structure factors and test their efficacy. We do this in two stages: with an assumed monodisperse cluster size distribution, and then with a scaling distribution. We used three different structure factors: the exponential structure factor (E), eq 3; the Mountain and Mulholland structure factor (MM), representing MM simulation and the modified exponential cutoff, eq 5; and the Gaussian structure factor (G), eq 9, representing the Gaussian cutoff and to some extent similar to the Hurd and Flower, Lin et al., and Dobbins and Megaridis structure factors. The fits are shown in Figures 2–4 for run 2. All the fits are fairly good, although a similar systematic deviation is seen for the MM and G fits, and their quality cannot be used as a definitive guide for choosing the best $S(qR_g)$. A similar conclusion can be made for run 1. Values for the fractal dimensions obtained for the fits for both runs are given in Table I. Similar results are obtained for each run, and we see average fractal dimensions are $D(E) \simeq 2.1$, $D(\mathrm{MM}) \simeq 1.2$ and $D(\mathrm{G}) \simeq 1.2$. None of these values is consistent with those expected from a growing wealth of data now available which suggest D should be in the range $1.6 \lesssim D \lesssim 1.9$. Previous measurements by us on this flame under conditions identical to run 1 yielded $D = 1.79 \pm 0.10$ via a completely different, although somewhat novel, optical method.[15] This method determined D by comparing radius of gyration and volume equivalent sphere measurements. Close examination of Figures 3 and 4 show the data falling off faster than the fit at large qR_g suggesting the fractal dimensions for the MM and G fits are too small. Because of these reasons, we are suspicious of all these attempts to describe the data with a monodisperse size distribution.

A form for the size distribution must be picked. The previously most common choices, the log normal and zero

(13) Weitz, D. A., private communication. Also note that Figure 2 of Lin et al., ref 11, is in error. The MM result does not agree with the Lin et al. result and the structure factor for the exponential cutoff is misplotted.

(14) Gangopadhyay, S.; Elminyawi, I.; Sorensen, C. M. *Appl. Opt.* **1991**, *30*, 4859.

(15) Sorensen, C. M.; Cai, J.; Lu, N. *Appl. Opt.* in press.

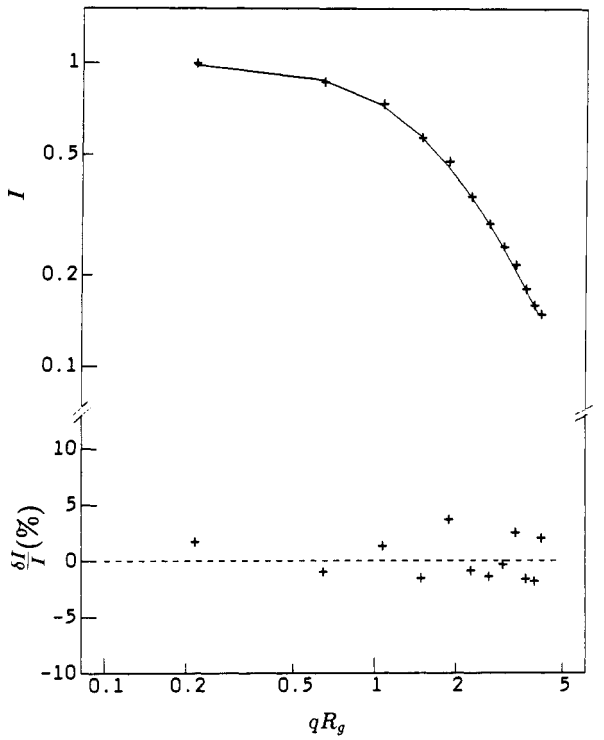

Figure 2. Fit of the exponential structure factor with monodisperse cluster size to the scattered intensity data for run 2. $\delta I = I_{data} - I_{fit}$.

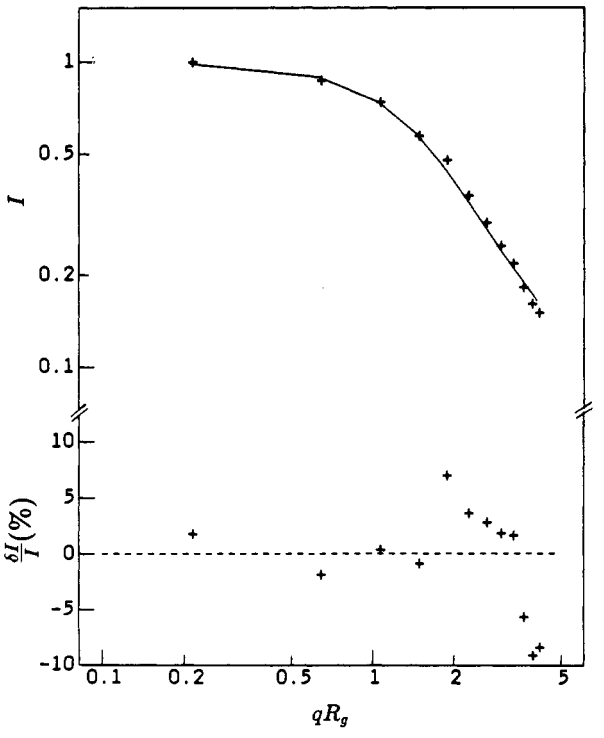

Figure 3. Fit of the Mountain and Mulholland structure factor with monodisperse cluster size to the scattered intensity data for run 2.

order log normal, are not suitable. By comparison to exact self-preserving solutions, we have shown the moments of the distribution obtained from the self-preserving distributions and the LN or ZOLD do not agree for moments of higher order than the second.[15] This is important in our work since as described above the measured radius of gyration is related to $M_{2+2/D}/M_2$, where M_i is the ith moment. Because of this, we use the scaling ansatz[16] to

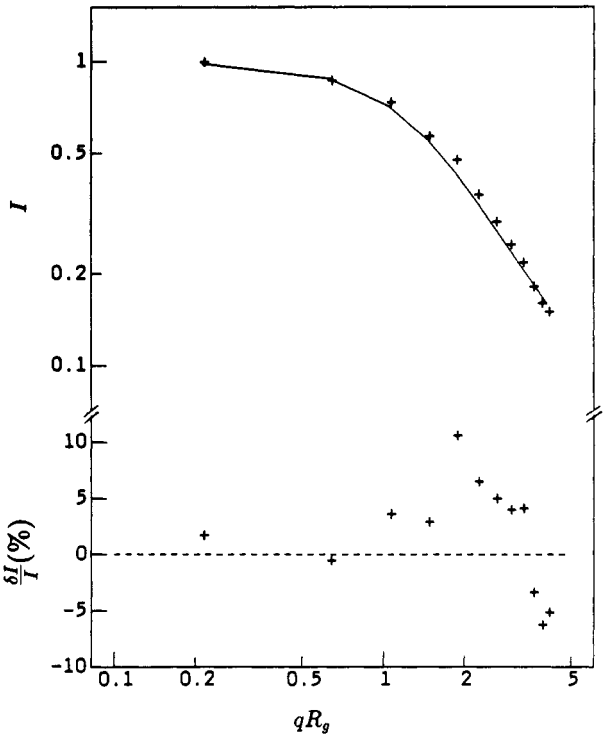

Figure 4. Fit of the Gaussian structure factor with monodisperse cluster size to the scattered intensity data for run 2.

Table I. Fractal Dimensions D Obtained from Fitting Various Structure Factors to the Data with either a Monodisperse or Scaling Size Distribution

	exponential			MM			Gaussian		
		scaling			scaling			scaling	
run	mono	$\tau = 0$	$\tau = 2/3$	mono	$\tau = 0$	$\tau = 2/3$	mono	$\tau = 0$	$\tau = 2/3$
1	2.11	2.51	2.61	1.18	1.52	1.59	1.19	1.71	1.82
2	2.04	2.38	2.48	1.20	1.51	1.59	1.26	1.68	1.78

describe the size distribution $n(N)$, where N is the number of monomers per aggregate

$$n(N) = M_1 s_1^{-2} \phi(x) \tag{12a}$$

$$x = N/s_1 \tag{12b}$$

$$s_1 = M_1/M_0 \tag{12c}$$

and

$$\phi(x) = Ax^{-\tau}e^{-\alpha x} \tag{12d}$$

where $A = \alpha^{\alpha}\Gamma^{-1}(\alpha)$, $\alpha = 1 - \tau$, and Γ is the gamma function.

In recent work[15] we used the scaling distribution in its simplest form with $\tau = 0$. This was motivated by the fact that moments calculated from this distribution agreed well with those calculated from the size distribution determined from numerical computation by Graham and Robinson[17] for coalescing particles in the free molecular regime. Our particles are not coalescing but Mulholland et al.[18] simulated noncoalescing, free molecular aggregation and showed the resultant size distribution was quite similar to that obtained with coalescence. Thus the $\tau = 0$ scaling distribution appears to be a good approximation for our soot system, certainly better than the log-normal distributions, and we shall use it here.

(16) van Dongen, P. G.; Ernst, M. E. *Phys. Rev. Lett.* **1985**, *54*, 139.
(17) Graham, S. C.; Robinson, A. *J. Aerosol Sci.* **1976**, *7*, 261.
(18) Mulholland, G. W.; Samson, R. J.; Mountain, R. D.; Ernst, M. H. *Energy Fuels* **1988**, *2*, 481.

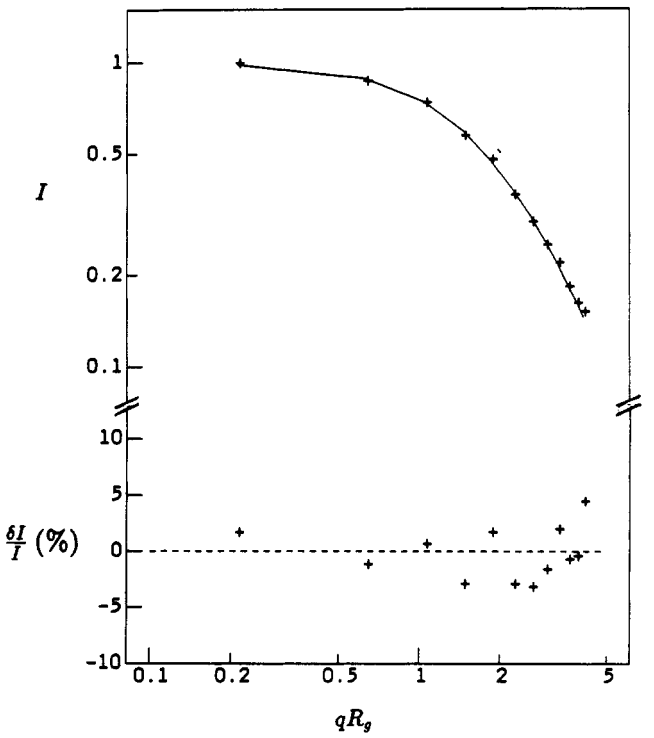

Figure 5. Fit of the exponential structure factor with a scaling cluster size distribution with $\tau = {}^2/_3$ to the scattered intensity data for run 2.

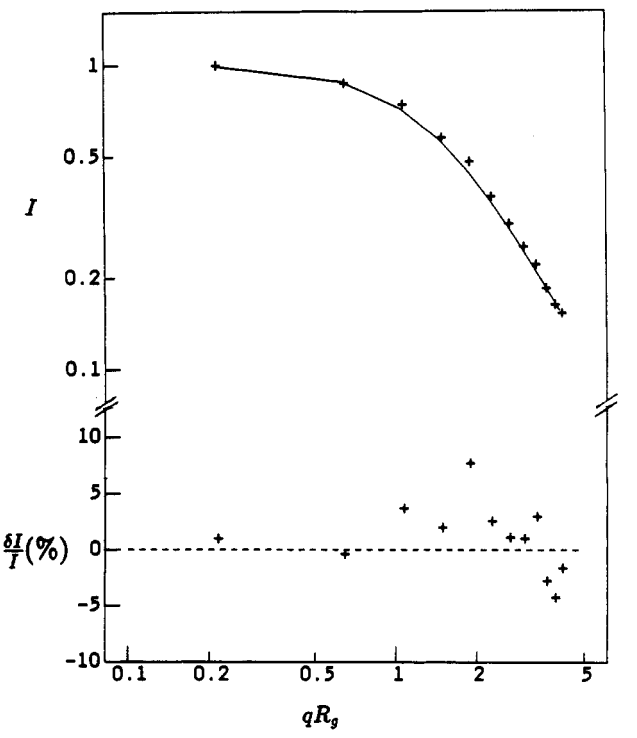

Figure 6. Fit of the Mountain and Mulholland structure factor with a scaling cluter size distribution with $\tau = {}^2/_3$ to the scattered intensity data for run 2.

We also want to use the scaling distribution for nonzero τ because this may be more reasonable for noncoalescing, fractal aggregates in the free molecular regime. Mulholland et al. have shown how τ is a function of D. There is some ambiguity near $D = 2$ which has not been worked out theoretically, but a reasonable estimate of $\tau = {}^2/_3$ for D in the range $1.7 < D < 2$ can still be obtained from their work. It is our opinion that theory indicates the $\tau = {}^2/_3$ may be a better estimate than $\tau = 0$ for free-molecular, noncoalescing aggregation.

With eq 12, one can show

$$\langle R_g^2 \rangle = a^2 k_o^{-2/D} s_1^{2/D} \alpha^{-2/D} \Gamma(\alpha + 2 + 2/D)/\Gamma(\alpha+2) \quad (13)$$

In eq 13 a is the monomer particle radius, k_o is defined by $N = k_o(R_g/a)^D$, and Γ is the gamma function. This allows us to measure R_g and use this value to determine s_1 necessary for the description of the size distribution.

The data are now fit to

$$I(q) = \int N^2 n(N) S(qR_g) \, dN \quad (14)$$

This equation uses $N = k_o (R_g/a)^D$ and is normalized to the experimental $I(0)$ so the values of a and k_o need not be known. Since R_g, hence s_1, is determined from the small qR_g behavior, the only fit parameter is D, as in the monodisperse case. We again perform fits using the E, MM, and G structure factors for both $\tau = 0$ and $\tau = {}^2/_3$. The results for $\tau = {}^2/_3$ are shown in Figures 5–7 for run 2.

All fits are good with comparable χ^2 values and no systematics, in contrast to the monodisperse fits. The values of D, however, vary systematically with the cutoff; for $\tau = {}^2/_3$ D(E) = 2.48, D(MM) = 1.59, and D(G) = 1.78.

Table I tabulates these results for both runs and both τ values. Similar results are seen for each run. The average fractal dimension for the exponential cutoff fit, D(E) $\simeq$ 2.5, is unrealistically large compared to other results. Both the Gaussian and modified exponential cutoffs (MM) yield

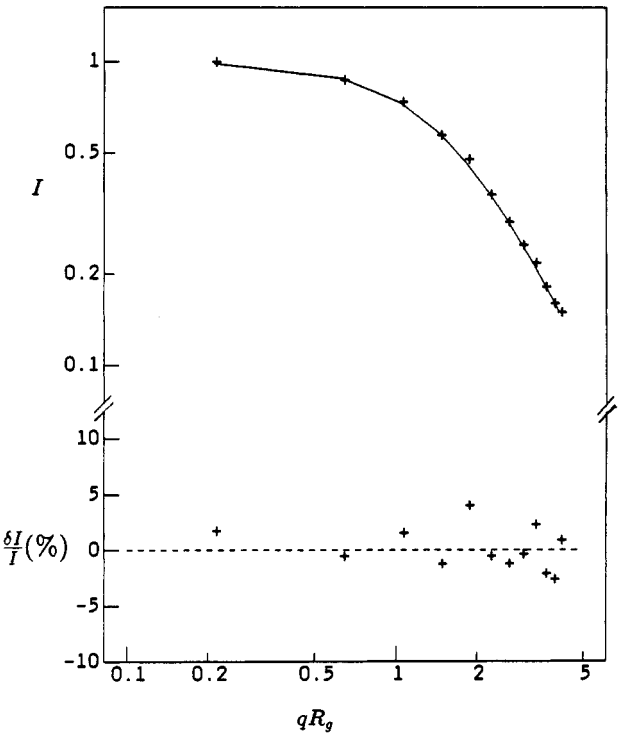

Figure 7. Fit of the Gaussian structure factor with a scaling cluster size distribution with $\tau = {}^2/_3$ to the scattered intensity data for run 2.

reasonable values for D, significantly improved from the monodisperse case. The Gaussian fit yields the best value, $D \simeq 1.7$–1.8 when compared to earlier work on this flame using a different optical method to measure D. Variation with τ from zero to ${}^2/_3$ is only about 5%.

Discussion and Conclusion

If the clusters are assumed to be monodisperse in size, none of the structure factors can fit the data to yield a

reasonable value of D. Certainly the value obtained from the exponential fit is not very bad but still beyond the range expected and previously measured. Of course the monodispersity assumption is a bad one given the statistical nature of aggregation and the great body of previous knowledge concerning the cluster size distribution. Hence we eliminate these fits.

Our results indicate that the effects of polydispersity must be included in a fit of the theoretical structure factors to our soot data. However, inclusion of polydispersity in the exponential structure factor fit leads to poor values of D. Hence this structure factor is eliminated. Without polydispersity, systematic deviation of the fits for MM and G occur near $qR_g \sim 2$. Lin et al. made a similar conclusion. They showed that inclusion of polydispersity broadened the bend in $S(qR_g)$ in the region of $qR_g \sim 2$ and this was necessary to fit their colloidal data. Whether this behavior near the bend in the structure factor could be used to obtain information regarding the nature of the size distribution is a worthwhile topic which we intend to pursue in the future.

Our results also indicate that a structure factor corresponding to a cluster density correlation function with a cutoff sharper than exponential is necessary to describe our soot data. Both the structure factors derived from the Gaussian and the modified exponential cutoff of Mountain and Mulholland do an adequate job of fitting our data when polydispersity is included, although the fractal dimension determined from the Gaussian fit is closer to the value expected and the value determined from previous measurements on this flame (run 1, D = 1.82 vs 1.79). We would expect from Figure 1 that the Hurd and Flower, Lin et al., and Dobbins and Megaridis structure factors would do an equally satisfactory job of fitting the data with a reasonable D. Of course, all three "exponential" cutoffs belong to the same family of $h(x) \sim \exp[-x^\alpha]$ where $\alpha(\mathrm{E}) = 1$, $\alpha(\mathrm{G}) = 2$, and $\alpha(\mathrm{MM}) = 2.5$. Here again we recall Mountain and Mulholland claimed that their precision implied the range $2 \leq \alpha \leq 3$, which is consistent with our results. Future work will include comparisons of light scattering to visually analyzed TEM micrographs of collected soot particles. However, even barring perturbations incurred by soot particle collection from flames, we are uncertain how much more precision can be developed for our current conclusion that, for soot particles, $\alpha \simeq 2$.

Acknowledgment. This work was supported by the National Institute of Standards and Technology and NSF through Grant CTS9024668.