Comment on "Hydrodynamic Behavior of Fractal Aggregates"

In a recent Letter,[1] Wiltzius reported careful measurements of the hydrodynamic behavior of fractal aggregates formed by reaction-limited aggregation of colloidal silica. He determined the average radius of gyration, $\bar{R}_G$, from the angular dependence of the static light scattering, and the average hydrodynamic radius, $\bar{R}_H$, from dynamic light scattering at small scattering wave vectors, q. Unfortunately, Wiltzius appears to have neglected the fact that these averages reflect different moments of the cluster mass distribution. Thus the ratio $\bar{R}_H/\bar{R}_G$ cannot be compared directly to the value $\beta=R_H/R_G$ calculated for a single fractal cluster of a given mass, M, and fractal dimension, d_f.

To make this correction, we assume that the cluster mass distribution has the power-law form $P(M)=M^{-\tau}F_M(M/M_c)$, expected for reaction-limited aggregation. Here F_M is the cutoff function of the distribution and depends on some characteristic cluster mass, M_c. We further assume that $\bar{R}_G$ is determined from the low-q form of the static light scattering, $I_M(q)\approx M^2(1-q^2R_G^2/3)$, which applies generally and is simpler in form than the rather arbitrary Fisher-Burford approximant used by Wiltzius. Finally, we assume fractal scaling of the clusters, $M\sim R^{d_f}$, for both R_G and R_H.

With these assumptions, the correct moments of the cluster mass distribution are readily calculated:

$$\langle R_G^2\rangle=\int M^2R_G^2P(M)\,dM\left[\int M^2P(M)\,dM\right]^{-1},$$

and

$$\langle R_H\rangle=\int M^2P(M)\,dM\left[\int M^2R_H^{-1}P(M)\,dM\right]^{-1}.$$

To account for these effects, we define

$$\langle R_H\rangle/\langle R_G^2\rangle^{1/2}=\gamma R_H/R_G=\gamma\beta.$$

Then a step-function form for F_M yields

$$\gamma_s=(3-\tau-d_f^{-1})(3-\tau+2d_f^{-1})^{1/2}(3-\tau)^{-3/2},$$

while an exponential form for F_M yields

$$\gamma_e=[\Gamma(3-\tau)]^{3/2}[\Gamma(3-\tau-d_f^{-1})]^{-1}\times[\Gamma(3-\tau+2d_f^{-1})]^{-1/2},$$

where Γ is the Γ function. Similarly, dividing all the arguments of the Γ functions in γ_e by 2 gives γ_g, for a Gaussian form for F_M.

TABLE I. Values of γ and β for different F_M and τ.

Cutoff function	$\tau=1.50$ γ	$\tau=1.50$ β	$\tau=1.85$ γ	$\tau=1.85$ β
Step	0.88	0.82	0.79	0.91
Gaussian	0.82	0.88	0.74	0.98
Exponential	0.74	0.97	0.67	1.08

We see that γ depends on both d_f and τ, in addition to the form of the cutoff function. In Table I, we tabulate γ for $d_f=2.1$, the value measured experimentally by Wiltzius, and for $\tau=1.5$ and 1.85, which spans the range found experimentally and theoretically. The correction is significant for all cases. Wiltzius measured $\langle R_H\rangle/\langle R_G^2\rangle^{1/2}=\gamma\beta=0.72$, and so we also present the corrected values of $\beta=0.72/\gamma$ for each case, which can be compared to calculations. For a spherically symmetric model of a fractal aggregate, with a mass distribution proportional to $g(r)F_r(r/R_G)$, where $g(r)\sim r^{d_f-3}$ and F_r is the cutoff function for a single cluster,[1,2] one finds $\beta=1.03$ for a step-function form of F_r, 0.61 for an exponential form, and 0.83 for a Gaussian form. The corrected results seem to rule out the exponential form for F_r, suggesting that the boundary of a fractal aggregate is rather sharp. Recent numerical calculations[3] of reaction-limited aggregation clusters suggest $\beta\approx0.97$, consistent with Wiltzius's value corrected with an exponential F_M and $\tau=1.5$. More precise comparison requires knowledge of τ and F_M, and careful consideration of the form of $I(q)$ used to determine R_G.

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[1]P. Wiltzius, Phys. Rev. Lett. **58**, 710 (1987).

[2]W. Hess, H. L. Frisch, and R. Klein, Z. Phys. B **64**, 65 (1986). Equation (4) in this paper must be corrected: The 4 should be replaced by 2.

[3]Z.-Y. Chen, P. Meakin, and J. M. Deutch, preceding Comment [Phys. Rev. Lett. **59**, 2121 (1987)].

