Wiltzius and van Saarloos Reply: In their Comment, Chen, Meakin, and Deutch[1] present values for the ratio of the hydrodynamic radius R_H to the radius of gyration R_G for fractal aggregates. The values have been obtained for two important computer simulation models, diffusion- and reaction-limited cluster-cluster aggregation. For the latter model, the value R_H/R_G is much closer to the experimental[2] result $R_H/R_G = 0.72 \pm 0.02$ than is the ratio 1.75 communicated in private earlier by Chen, Meakin, and Deutch.[1] Pusey *et al.*[3] show that if, in addition, the effects of polydispersity are taken into account, one obtains estimates for the ratio R_H/R_G that cluster around the experimental value. We are pleased with the contributions made in both Comments. Nevertheless, it is our opinion that the comparison of theory and experiment is somewhat more complicated than might appear from the preceding Comments.

It is well known in polymer theory[4] that R_H and R_G reach their asymptotic scaling behavior with slightly different powers of the degree of polymerization, giving rise to a notoriously slow crossover of R_H/R_G to the true asymptotic value.[5] This crossover is difficult to study within the Kirkwood-Riseman theory, but is naturally included in the porous-sphere model,[5] in which a polymer or aggregate is treated as a sphere with porosity inversely proportional to the density of monomers. For the clusters studied by Chen, Meakin, and Deutch,[1] the value $R_H/R_G \cong 1$ is in the range expected[6] in the porous-sphere model for clusters of size $N \lesssim 400$ and $d_f \cong 2.1$. Experimentally, the ratio of clusters of about the same size ($R_G \approx 400$ Å) is also around unity. Hence, if these relatively small clusters of the same size are compared, polydispersity is not needed to account for the data. In general, one has to be cautious to apply polydispersity considerations based on asymptotic power-law cluster-size distributions to small clusters with $N \lesssim 400$.

Chen, Meakin, and Deutch[1] unfortunately have no data for larger clusters. In the experiments, the measured value of R_H/R_G decreases with cluster size, to reach its "asymptotic" value around $R_G \approx 800$ Å. The porous-sphere model, however, predicts[6] an increase of R_H/R_G with cluster size; and for N of order 10^4, relevant for the largest clusters studied experimentally, R_H/R_G is predicted to be 20% *larger* than in the numerical simulations, whereas the experimental value is 30% *smaller*. Even when a reduction of order 20% to 30% due to polydispersity is taken into account, the theoretical estimates based on the porous-sphere model are somewhat larger than the value observed in this range.[2]

An aspect neglected in both the Kirkwood-Riseman scheme[2] and the Comments[1,3] is aggregate anisotropy. Large-scale computer simulations[7] of various aggregation models as well as number-fluctuation spectroscopy measurements on aggregating latex microspheres[8] have indeed shown that the long axis of large clusters is roughly twice as long as the short axis. Such an asymmetry could also introduce a systematic reduction[6] of R_H/R_G as measured[2] with light-scattering techniques in the range 500 Å $\lesssim R_H \lesssim$ 7000 Å. Additional complications for the theoretical treatment of the hydrodynamic behavior arise possibly from some flexibility in the periphery of the aggregates, although we do expect this effect to be small.

In conclusion, while we welcome the fact that the computer results[1] and the theoretical analysis of polydispersity[3] have brought the theory and the *in situ* experiments[2] on colloidal aggregates into much closer agreement than was previously believed,[2] we believe that the finite-size effects, polydispersity, and anisotropy of the clusters require further investigation, through, e.g., depolarized light scattering and sedimentation experiments.

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