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Estimation of fractal dimension of colloidal gels in the presence of multiple scattering

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Colloidal dispersions of fluorinated polymer particles with a refractive index very close to that of water, have been used to investigate the effect of multiple scattering on the estimated fractal dimension of colloidal gels, at high-particle volume fractions. The extent of multiple scattering was varied by using cuvettes of different internal diameters, from 3 to 18 mm. Three gelation systems with different sizes and volume fractions of primary particles have been characterized by static light scattering SLS. The obtained results indicate that multiple scattering affects only the magnitude of the scattered radiation, but not the estimated fractal dimension of the gels. This result confirms the conclusion of the theoretical study reported by Chen *et al.* [Phys. Rev. B **37**, 5232 (1988)]. As a further confirmation, the same gels have been formed in a specially designed cell, with only 0.1 mm thickness (where multiple scattering is negligible) and characterized using small-angle neutron scattering (SANS). It is found that the fractal dimension estimated from SANS measurements, without multiple scattering, is the same as that estimated from SLS measurements, in the presence of substantial multiple scattering.

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I. INTRODUCTION

Under appropriate conditions, small particles of colloidal systems may come together to form large aggregates, which may exhibit complex structures. When the concentration of the small particles is sufficiently large, the aggregating system may form a gel, that is a state of a colloidal system with a semirigid three-dimensional network. These systems are the subject of great interest in the area of soft condensed-matter physics, also with respect to their industrial applications [1–12].

The structure of aggregates and gels is highly disordered, but there is experimental evidence that at certain length scales they exhibit sufficient self similarity to be described in terms of fractal objects [13–15]. In this case, we can experimentally quantify the structure of aggregates and gels using various fractal analysis techniques, which may be based on rheology, microscopy, image analysis, and most commonly, light, neutron, and x-ray scattering [16–21].

The rigorous scattering theory requires solving the Maxwell equations. Analytical solutions may be obtained only for very few simple systems. For fractal aggregates, approximated solutions may be obtained using properly simplified scattering theories, such as the Rayleigh-Debye-Gans (RDG) approximation, which has been successfully applied to predict the scattering behavior of diluted fractal aggregates systems of small particles [22–24].

Based on the RDG scattering theory of randomly oriented fractal aggregates, the average structure factor $S(q)$ has the following simple form:

$$S(q) \sim q^{-D_f} \quad \text{for} \quad qr_0 \ll 1 \ll q\xi, \tag{1}$$

where r_0 is the radius of primary particles, ξ is the maximum size of aggregates, D_f is the fractal dimension, and q is the wave vector defined as

$$q = \frac{4\pi n}{\lambda} \sin\left(\frac{\Theta}{2}\right), \tag{2}$$

where λ is the wavelength of incident electromagnetic radiation, n the refracting index of the medium surrounding the particles, and Θ the scattering angle. The range of q values in which Eq. (1) applies corresponds to the range of sizes in which an aggregate may be represented as a fractal. The fundamental requirement of the RDG theory is that the phase shift of the wave traversing each primary particle is negligible, which implies that

$$2r_0q(m-1) \ll 1, \tag{3}$$

where m is the relative refractive index of the system, i.e., the ratio of the refractive index of the particles to that of the surrounding medium. It follows that, in order to satisfy Eq. (3), the primary particle must have a refractive index very close to that of its surrounding medium, and its size should not be too large compared to the wavelength of radiation. Moreover, this theory neglects multiple-scattering effects, because it assumes that the radiation illuminating each particle in the aggregate is the incident radiation, totally unperturbed by the presence of other particles of the aggregate.

It is evident that for most systems, the above requirements cannot be satisfied. In particular, for concentrated systems, multiple scattering arising both from the particle-to-particle as well as from the aggregate-to-aggregate interactions is substantial, thus questioning the applicability of the above Eq. (1).

Several theoretical studies on the optical properties of aggregates in the literature investigate the limits of applicability of the RDG scattering theory [25–33]. Various techniques have been proposed to numerically solve the Maxwell equations, such as those reducing the integral form of the Maxwell equations into a set of linear algebraic equations through proper simplifications [31,32]. The obtained results, however, are not conclusive. For example, Chen and co-

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workers [28] came to the conclusion that multiple scattering affects only the magnitude of the scattering intensity, while Eq. (1) remains valid, which implies that the fractal dimension may be measured also in the presence of multiple scattering. On the other hand, Frey and coworkers [29] showed that it is not possible to say clearly if Eq. (1) is valid or not when multiple-scattering effects are present. Nevertheless, Eq. (1) is widely used in the literature to obtain experimentally the fractal dimension D_f without accounting for multiple-scattering effects [1–6,9,16–21].

In the present paper, we address this problem experimentally, by investigating aggregating systems of rather high-particle volume fraction, which eventually lead to gel formation. In particular, we show that multiple scattering affects only the magnitude of the scattered intensity, but it does not affect the fractal dimension of gel aggregates, estimated from the slope of the log-log plot of the scattered intensity as a function of q.

II. EXPERIMENTAL SECTION

The colloidal systems used in our experiments are latexes of MFA® (Ausimont SpA, Italy), produced by emulsion copolymerization of tetrafluoroethylene and perfluoromethylvinylether. This fluorinated polymer has a refractive index (1.35) that is very close to that of water (1.33), leading to a low turbidity of its suspensions in water. Thus, it allows us to use scattering techniques to characterize the structure of aggregates in suspensions with high-particle volume fraction. Particles of two different sizes have been used: 75 nm (polydispersity ~ 0.08) and 42 nm (polydispersity ~ 0.1), determined by dynamic light scattering. Original latexes are diluted to the desired particle volume fraction using Milli-Q (Millipore) deionized water.

Aggregation of the colloidal systems has been induced by adding a certain amount of salt, [$Ca(NO_3)_2$]. Three aggregating systems have been prepared:

(1) particle size 75 nm; volume fraction 0.02; $Ca(NO_3)_2$ concentration 1.58×10^{-3} mol/l;

(2) particle size 75 nm; volume fraction 0.04; $Ca(NO_3)_2$ concentration 1.58×10^{-3} mol/l;

(3) particle size 42 nm; volume fraction 0.04; $Ca(NO_3)_2$ concentration 3.16×10^{-3} mol/l.

In all these cases, since the particle volume fraction is rather large, gels are eventually formed. The time for complete gelation is approximately 6–7 hours.

Two types of instruments, light and small-angle neutron scattering (SANS) were used in this study. The light-scattering instrument is a BI-200SM (Brookhaven), using an argon laser (Lexel 95-2) as the light source (wavelength $\lambda = 514.5$ nm), with an angular range of the goniometer from 15 to 150 °C, while the SANS instrument is located at the Paul Scherrer Institut, Villigen, Switzerland.

The light-scattering instrument has been used to investigate the effect of multiple scattering by considering five quartz cuvettes of different sizes (internal diameter×wall thickness, in mm): 3×1, 8×1, 10×1, 13×1, 18×1. The gel samples were prepared starting from a high-concentration latex, i.e., 0.054 volume fraction for the 75 nm particles and 0.064 volume fraction for the 42 nm particles. The latex was diluted to reach the desired final concentration with an aqueous solution of $Ca(NO_3)_2$, where the salt concentration was such as to reach the desired concentration in the final gelling system. Dilution occurred by pouring the latex in the salt solution, without mechanical mixing of the system, since the shear stress produced by stirrer would effect the aggregation process and the structure of the final gel. The concentration of salt in the water solution was low enough to avoid that, so, during latex addition, aggregates could form. Each gelling system was prepared in large amounts (200 cc) in order to minimize experimental errors and each analyzed sample was taken from the same gelling system, and put in the cuvettes immediately after preparation.

During static light scattering (SLS) measurements of gels, the sample cuvette was rotated by a motor at a constant speed of 3 rpm, so that an average intensity over the whole section of the sample was measured, in order to smooth local inhomogeneities of the gel. The effect of the rotation speed has been checked by repeating the same SLS measurements at rotation speeds of 1, 2, and 4 rpm. No significant difference has been found. SLS measurements have been repeated several times for each sample, and the obtained data have been averaged before further treatment. Some of them have been repeated several times at the same conditions, and excellent reproducibility has been found in all cases.

The SANS instrument was used only to independently confirm the results of SLS analysis. In particular, a specially designed cell, with the possibility of varying the sample thickness from 0.1 to 4 mm, has been constructed. The variation of the sample thickness is obtained through a screw mechanism that moves one of the quartz windows forward and backward with no rotating action, i.e., with no rotational shear stress acting on the sample. This is particularly important due to the very small sample thickness. To reduce the radial shear stress generated by the liquid flow during the reduction of the sample thickness, we have moved the quartz window very slowly (with a speed of 0.1 mm/s). In addition, the cell includes a sample injection hole, an exit for excess material, and an internal reservoir for reducing the effect of evaporation.

The gel samples for the SANS experiments were taken from the gelling solution immediately after the addition of the latex, and carefully injected in the cell (set at the largest sample thickness) through the sample injection hole. Then the thickness was reduced down to 0.1 mm, the inlet and outlet holes were closed, and the sample was left alone to undergo the gelation process. SANS measurements were taken only after complete gelation occurred, which was determined by observing gelation in the remaining solution outside the sample.

III. RESULTS AND DISCUSSION

A. Form factor of particles

Based on the RDG theory for a system containing particles of equal size, the intensity of the scattered radiation is given by the following expression [25]:

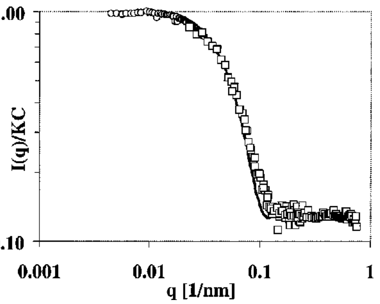

FIG. 1. Form factor of the 75 mm particles measured from a latex of 0.0002 volume fraction. Circles: light scattering data; squares: neutron scattering data; line: calculated using Eqs. (5) and (6) with $r_0 = 37.5\,\mathrm{nm}$.

$$I(q) = KCP(q)S(q), \tag{4}$$

where K is a constant, which depends on the equipment setup and the contrast between solvent and particles, C is the particle concentration, $P(q)$ the form factor of the particles, and $S(q)$ the structure factor of the system. Thus, in order to estimate the structure factor $S(q)$, one first needs to determine the particle form factor. This may be done by performing scattering experiments with very dilute systems, where the structure factor contribution vanishes, i.e., $S(q) \to 1$, and then intensity is proportional only to the form factor.

Accordingly, samples at 0.0002 volume fraction of both the 75 and the 42 nm particles have been prepared, and both SLS and SANS measurements have been taken. Figure 1 shows the values of $I(q)/KC$ as a function of q in the case of 75 nm particles, where the circles and squares indicate results from SLS and SANS, respectively. It is seen that the data given by the two types of measurements are very consistent. In the same figure, a curve is also shown, obtained by computing the form factor through the following equation [25]:

$$P(q) = \left[3\,\frac{\sin(qr_0) - qr_0\cos(qr_0)}{(qr_0)^3}\right]^2, \tag{5}$$

where r_0 is the radius of a particle, and then correcting it for the background noise in SANS as follows [24]:

$$\frac{I(q)}{KC} = P(q) + K_B, \tag{6}$$

where K_B is the background constant. It is seen that the agreement between calculated and experimental results is satisfactory up to $q = 0.06\,\mathrm{nm}^{-1}$.

Similarly, Fig. 2 shows the values of $I(q)/KC$ as a function of q for the 42 nm particles, measured both by SLS and SANS, as well as the curve computed through Eqs. (5) and (6). Also in this case, a good agreement is obtained between calculated and experimental results up to $q = 0.1\,\mathrm{nm}^{-1}$. The results obtained above confirm the reliability of the SLS measurements in the range $0.003 < q < 0.03\,\mathrm{nm}^{-1}$, and indicate that in the following, we may use directly Eq. (5) to compute the form factor $P(q)$. In addition, it is also confirmed that the particle size measured by DLS is correct and that the size distribution is indeed very narrow.

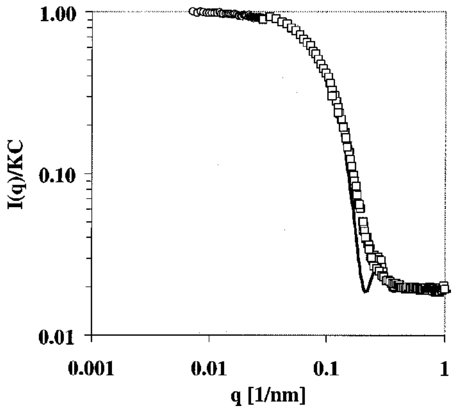

FIG. 2. Form factor of the 42 nm particles measured from a latex of 0.0002 volume fraction. Circles: light scattering data; squares: neutron scattering data; line: calculated using Eqs. (5) and (6) with $r_0 = 21\,\mathrm{nm}$.

B. Multiple scattering in concentrated latexes

Before investigating the effect of multiple scattering in aggregated systems, let us observe the behavior of multiple scattering in concentrated MFA latexes. For this purpose, we performed SLS experiments for the latex with 75 nm particles at 0.04 volume fraction, using different cuvettes with different internal diameter. The measured intensity profiles of the scattered light are shown in Fig. 3 as a function of q. Note that in order to compare the intensities of the scattered light at the same reference level, the intensity of the incident light before each experiment has been calibrated, using a 27 mm cuvette containing Decaline.

The results in Fig. 3 indicate that for a given q value, the intensity decreases as the cuvette diameter increases. This phenomenon, which is not observed with volume fractions smaller than 0.005, as shown in Fig. 4, is to be attributed to multiple scattering.

In Fig. 5, the intensity profiles of the different cuvettes relative to the smallest one, i.e., the 3 mm cuvette, are

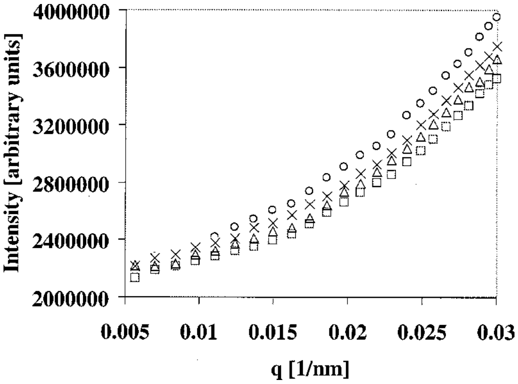

FIG. 3. SLS intensities for the 0.04 volume fraction latex with 75 nm particles, using cuvettes with various internal diameters. Circles: 3 mm; crosses: 8 mm; triangles: 13 mm; squares: 18 mm.

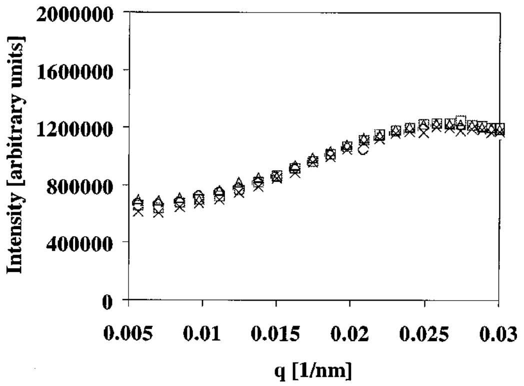

FIG. 4. SLS intensities for the 0.005 volume fraction latex with 75 nm particles, using cuvettes with various internal diameters. Circles: 3 mm; crosses: 8 mm; triangles: 13 mm; squares: 18 mm.

shown. It is seen that the corresponding intensity ratios decrease as a function of the wave-vector q. This indicates that the effect of multiple scattering is stronger at larger scattering angles. Since typically, intensity profiles of highly concentrated latexes exhibit a peak, which is out of the range of q values measurable by SLS, multiple scattering tends to broaden and smooth such a peak. This observation is in agreement with the results reported in previous investigations [34,35].

C. Multiple scattering in gels

The same experimental procedure described above in relation to concentrated latexes has been used to investigate multiple scattering in gels. Figure 6 shows the measured intensity profiles for the gel formed from 75 nm particles at 0.04 volume fraction in cuvettes of various internal diameters, divided by the form factor of the primary particles, computed through Eq. (5). Each curve represents the average over at least 12 sets of measurements. It can be seen that, similar to the case of concentrated latexes as discussed in the contest of Fig. 3, the intensity of the scatterd light for a given q value decreases as the diameter of the cuvette increases, indicating an increasing effect of multiple scattering.

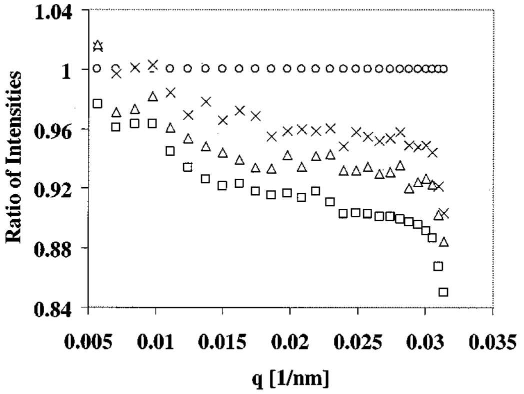

FIG. 5. Ratios between the SLS intensities for the samples in cuvettes of different size shown in Fig. 3 and the intensity corresponding to the cuvette of 3 mm. Circles: 3 mm; crosses: 8 mm; triangles: 13 mm; squares: 18 mm.

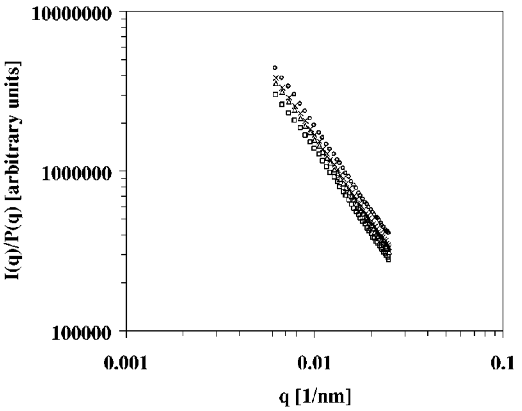

FIG. 6. Scattered intensities for gel originated from 75 nm particles at 0.04 volume fraction, measured in cuvettes with various internal diameters. Circles: 3 mm; crosses: 8 mm; triangle: 13 mm: squares: 18 mm.

Nevertheless, in the log-log plane, all the profiles of $I(q)/KC$ vs q are straight lines, and almost parallel. Let us evaluate the fractal dimension of the gel for each sample using the RDG theory.

According to Eq. (4), the quantity $I(q)/P(q)$ shown in Fig. 6 represents the product between the structure factor and the factor KC, which is constant for a given particle concentration. For a single fractal aggregate, the structure factor is related to the fractal dimension through the following equation [25,36]:

$$S(q)=1+\rho\int_0^\infty \frac{[g(r)-1]\sin(qr)}{qr}4\pi r^2\,dr, \tag{7}$$

where ρ is the number density of the particles in the system and $g(r)$ is the particle-particle correlation function given by [25]

$$g(r)-1=\frac{D_f}{4\pi\rho(r_0)^{D_f}}r^{D_f-3}h\left(\frac{r}{\xi}\right) \quad \text{with} \quad r>r_0, \tag{8}$$

where D_f is the fractal dimension, $h(r/\xi)$ is a cutoff function, taking into account the finite size of the aggregate ξ.

Among the various expressions for the cutoff function $h(r/\xi)$ proposed in the literature [25,37–39], the most commonly used is the stretched exponential

$$\ln\left[h\left(\frac{r}{\xi}\right)\right]=-\left(\frac{r}{\xi}\right)^{\beta}, \tag{9}$$

where the exponent β determines the sharpness of the cutoff function. Sorensen and Wang [39] recently examined different expressions for the structure factor and found that the gaussian cutoff, i.e., $\beta=2$ in Eq. (9), is the best choice. Using Eqs. (8) and (9) with $\beta=2$, the structure factor (7) becomes

$$S(q)=1+\frac{D_f}{r_0^{D_f}}\int_0^\infty r^{D_f-1}e^{-(r/\xi)^2}\sin\frac{(qr)}{qr}\,dr. \tag{10}$$

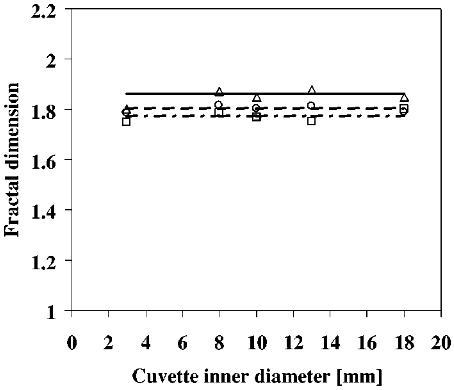

FIG. 7. Fractal dimensions of gels as a function of the cuvette diameter. Triangles: 0.02 volume fraction, 75 nm particles; circles: 0.04 volume fraction, 75 nm particles; squares: 0.04 volume fraction, 42 nm particles;

This integral has to be evaluated numerically, but it can be shown that Eq. (10) reduces to Eq. (1) in the limit of $q\xi \gg 1$, which is where Eq. (1) applies.

By allowing for an unknown proportionality factor, the data in Fig. 6 may be fitted with Eq. (10) in order to estimate the fractal dimensions D_f and the average size of aggregates ξ for each set of data at different cuvette internal diameter. The obtained values of the fractal dimension, which are plotted in Fig. 7 (circles) as a function of the cuvette internal diameter, are almost identical, with differences always smaller than 2%. In the same figure are also shown the values of the fractal dimension estimated from analogous scattering measurements taken on gels originated from 75 nm particles at 0.02 volume fraction (triangles) and from 42 nm particles of 0.04 volume fraction (squares). Again, it is found that the obtained fractal dimensions are independent of the cuvette size, and therefore not affected by the presence of multiple scattering.

The above findings indicate that, in aggregated colloidal systems, although multiple scattering does change the magnitude of the scattering intensity, it does not change the evaluated fractal dimension of the aggregates, thus confirming the conclusion of the theoretical work by Chen and coworkers [28]

It is worth mentioning that, in Fig. 7, for the gel formed from 75 nm particles at 0.02 volume fraction, the value of the fractal dimension obtained in the 3 mm cuvette is significantly smaller (5%) than the mean value obtained using the other cuvettes. This experiment has in fact been repeated several times but the same result was obtained. A possible explanation is the effect of sedimentation. It is known [40–42] that if the density of particles is substantially larger than that of the dispersing medium, sedimentation of the formed aggregates plays an important role in determining the structure of the gel. In particular, sedimentation may lead to a volume of the gel smaller than that of the original latex. In the case of MFA latexes, in fact, since the particle density is more than twice that of water (i.e., 2.15 g/cm^3), the volume of the gel formed from a latex with 0.02 volume fraction is 20% smaller than that of the original latex. However, such a difference in the volume does not occur in the 3 mm cuvette, i.e., the volume of the gel is practically the same as that of the initial latex. This leads to a less compact structure, and thus, to a smaller fractal dimension, with respect to that estimated in the cuvettes of larger size. A possible explanation of this observation may be the hindrance of sedimentation in the smallest cuvette due to capillary and hydrodynamic forces. This phenomenon has not been observed in the case of 0.04 particle volume fraction, since at these larger volume fractions, the volume of the gel is equal to that of the original latex in all cuvettes.

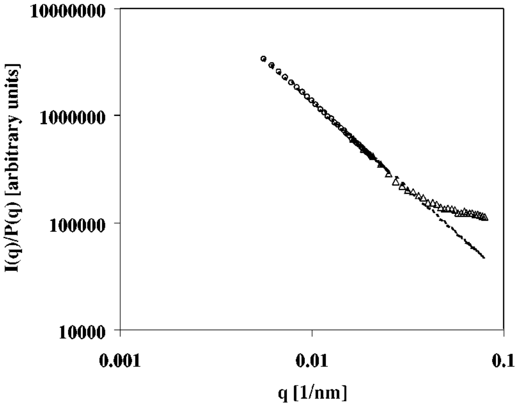

FIG. 8. SLS and SANS intensities divided by the form factor for the 75 nm particle gel at 0.04 volume fraction. Circles: SLS data obtained with the 18 mm cuvette; triangles: SANS data obtained with the 0.1 mm cuvette; line: Eq. (1).

In order to further confirm the conclusion that multiple scattering does not change the estimated value of the fractal dimension, gels have been prepared from both 75 and 42 nm particles at 0.04 particle volume fraction, in a specially designed cell of 0.1 mm thickness, as described in Sec. II, in order to minimize the effects of multiple scattering. The corresponding scattered intensities, measured by SANS and divided by the particle form factor computed form Eqs. (5) and (6), $I(q)/P(q)$, are shown as a function of q in Figs. 8 and 9, respectively. In the same figure, the corresponding SLS re-

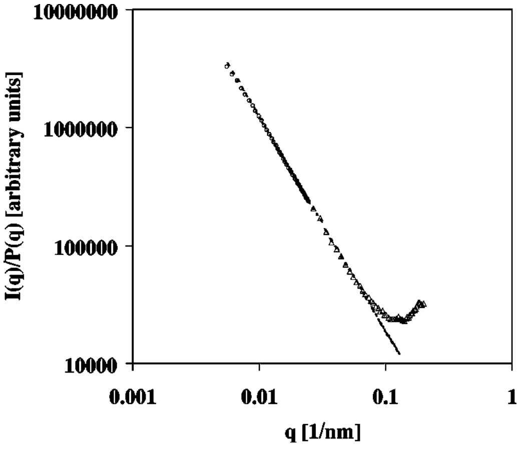

FIG. 9. SLS and SANS intensities divided by the form factor for the 42 nm particle gel at 0.04 volume fraction. Circles: SLS data obtained with the 18 mm cuvette; triangles: SANS data obtained with the 0.1 mm cuvette; line: Eq. (1).

sults with the 18 mm cuvette are also shown. It is worth noting that in order to overlap the results from SLS and SANS, the latter have been multiplied by a constant. It is seen that in the range $qr_0 \ll 1$ (i.e., $q > 0.03$ and 0.05 nm^{-1} in Figs. 8 and 9, respectively) the agreement between the results from SANS without multiple scattering and from SLS with multiple scattering is excellent, thus confirming that multiple scattering has a negligible effect on the evaluated fractal dimension of gels.

A final comment about the values of the fractal dimensions obtained in Fig. 7 is in order. It is found that gels originated from 0.02 particle volume fraction latexes exhibit larger fractal dimension than gels originated from 0.04 particle volume fraction latexes, which implies that the first ones exhibit a more compact structure. This result is difficult to justify, even though the same behavior has been found by Carpineti and coworkers [43] in the case of polystyrene aggregates, but with substantially lower-particle volume fraction. On the other hand recent Monte Carlo simulations contradict these findings. In particular, Gonzalez, Lach-hab, and Barojas [44] have found that the fractal dimension of gels simulated using a (DLCA) algorithm, increases as the particle volume fraction increases. Hasmy and Jullien [45], using a slightly different version of the DLCA algorithm, concluded that the fractal dimension that may be obtained from the structure factor of the gel is not the correct one and decreases as the particle volume fraction increases. The correct fractal dimension has been computed directly from the slope of the particle density correlation function. The fractal dimension so calculated increases with the particle volume fraction. Further studies are probably required to fully clarify this issue.

IV. CONCLUSIONS

The use of colloidal systems constituted of fluorinated polymer (MFA®) particles, which have a refractive index very close to that of water, allows us to investigate the effect of multiple scattering on the estimated fractal dimension of colloidal gels at high-particle volume fractions. Since in SLS measurements, the extent of multiple scattering increases with the thickness of the sample, various cuvettes with different internal diameter, from 3 to 18 mm, have been used.

Three polymer gels, prepared from latexes with different particle diameter and volume fraction, have been characterized using SLS. The obtained results indicate that multiple scattering affects only the magnitude of the scattering intensities, but not the estimated fractal dimension of the gels. This result confirms the conclusion of the theoretical study reported by Chen *et al.* [28].

In order to further confirm this conclusion, two of the gels considered above have been prepared in a specially designed cell with 0.1 mm thickness, where multiple scattering is negligible, and characterized using SANS. It is found that the fractal dimension estimated from these SANS measurements is the same as that estimated from the SLS measurements, although the latter are strongly affected by multiple scattering.

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