Colloidal gelation and non–ergodicity transitions

J. Bergenholtz
Department of Physical Chemistry, Göteborg University, 412 96 Göteborg, Sweden

M. Fuchs‡ and Th. Voigtmann
Physik-Department, Technische Universität München, 85747 Garching, Germany

Abstract.

Within the framework of the mode coupling theory (MCT) of structural relaxation, mechanisms and properties of non–ergodicity transitions in rather dilute suspensions of colloidal particles characterized by strong short–ranged attractions are studied. Results building on the virial expansion for particles with hard cores and interacting via an attractive square well potential are presented, and their relevance to colloidal gelation is discussed.

PACS numbers: 64.70.Pf, 64.75.+g, 82.60.Lf, 82.70.Dd

1. Introduction

Colloidal gelation is a non–equilibrium transition observed in dispersions where short ranged attractions exceeding a few $k_B T$ are present [1, 2, 3]. It is accompanied by the formation of (denser) domains, which coarsen and finally freeze when the gel line is traversed [4, 5]. The resulting gels are amorphous solids exhibiting finite elastic moduli [6, 7]. Often, the gel structure is fractal [6, 8], typically at low densities [8], and often low angle scattering peaks are observed, which are reminiscent of spinodal decomposition; in this case gelation manifests itself by an arrest of the time dependence of the peak position [4, 5, 6, 7, 8]. This explicit dependence on the time since quenching the suspension shows that colloidal gelation is a non–equilibrium phenomenon [8].

Colloidal gelation has been observed in colloid polymer mixtures [4, 5], in solutions of sterically stabilized colloidal particles if the solvent quality is decreased [3, 5, 7], in emulsions [2], in solutions of charge stabilized particles upon changes of the salt content [1], and in protein solutions [11]. In the last case, it prevents protein crystallization.

‡ To whom correspondence should be addressed
Colloidal gelation and non–ergodicity transitions

and thus is an important obstacle for the collection of protein structure information [12, 13, 14].

Previous theoretical explanations of colloidal gelation have focussed on the possibility of a dynamic percolation within a gas–liquid phase coexistence region which is metastable with respect to gas–solid coexistence [3]. The gel is assumed to form when the largest cluster spans the sample and the small angle scattering peak consequently arrests. Long ranged density fluctuations could arise from quenching below metastable ([15]) spinodal lines as argued in Refs. [16, 9, 6, 10]. Fine–tuning of the attraction strength allows for the observation of all aspects of spinodal decomposition in this systems without arrest and gelation [3, 5]. At low densities irreversible cluster–cluster aggregation provides a mechanism explaining the observed small wave vector structures [7]. A review of experiments and theoretical approaches up to 1997 has been given by Poon and Haw [3].

Recently an alternative explanation for the arrest of the dynamics at colloidal gel transitions has been put forward [18, 19]. Non–ergodicity transitions triggered by the local dynamics were suggested. Solutions of equations of the mode coupling theory (MCT) for structural relaxation [20, 21] were studied which exhibit a localization transition explaining a number of properties of colloidal gelation. At low densities but for strong short–ranged attractions, particles are tightly bound to (asymptotically infinite) clusters and large elastic moduli result [18, 19]. In this limit, the MCT–equations can be simplified to one–parameter models which capture a number of the pertinent physical mechanisms although this neglects other aspects of the obtained colloidal gels which are connected to their self–similar structure on mesoscopic length scales.

In the following, the asymptotic models connected to colloidal gelation shall be discussed further, where, especially, the existence of a divergent cluster size and its related dynamics is of interest. The strength of the required attractions at gelation is estimated also. Additionally, two unphysical aspects of the previously obtained MCT results for attractive Yukawa potentials and for Baxter’s adhesive hard sphere model (AHS) [22, 18] are reconsidered using the controlled low density virial expansion [23]. Adopting a very common model in colloid science, hard spheres interacting with a square well potential are studied [24].

2. Basic equations

The most simple MCT equations for the normalized time–dependent intermediate scattering functions at wave vector $q$, $\Phi_q(t)$, shall be studied, as appropriate for the description of the structural relaxation of colloidal suspensions (characterized by a Brownian short–time diffusion coefficient $D^s_q$) [25, 26, 21]:

$$\partial_t \Phi_q(t) + q^2 D^s_q \left\{ \Phi_q(t) + \int_0^t dt' m_q(t - t') \partial_{t'} \Phi_q(t') \right\} = 0 . \quad (1)$$

Here, the generalized longitudinal viscosity is given by a mode coupling functional, $m_q(t) = F_q([c], [\Phi(t)])$, which is uniquely specified by the equilibrium static structure as
given by the direct correlation function \( c_q \):

\[
\mathcal{F}_q([c], [f]) = \frac{\rho S_q}{2q^4} \int \frac{d\mathbf{k}d\mathbf{p}}{(2\pi)^3} \delta(\mathbf{k}+\mathbf{p} = \mathbf{q}) (\mathbf{q} \cdot \mathbf{p} c_p + \mathbf{q} \cdot \mathbf{k} c_k)^2 S^*_{k}S^*_{p} f_k f_p .
\]  

(2)

where \( \rho \) is the particle number density and \( c_q \) determines the static structure factor via, \( S_q = 1/(1 - \rho c_q) \).

Non–ergodicity transitions of MCT dynamical equations are obtained as bifurcation points of the non–linear equations for the \( q \)–dependent non–ergodicity or Edwards–Anderson parameters \( f_q \), which, in the idealized MCT, are defined as \( \Phi_q(t \to \infty) = f_q \):

\[
\frac{f_q}{1-f_q} = \mathcal{F}_q([c], [f]) .
\]  

(3)

Whereas vanishing non–ergodicity parameters, \( f_q = 0 \), indicate fluid or ergodic states, finite ones, \( f_q \geq f_q^* > 0 \), spring into existence at larger interactions and signal the arrest of density fluctuations into non–equilibrium states, which are amorphous solids like gels or glasses. The idealized MCT assumes a separation of time scales describing the arrested structures as truly non–ergodic states, although further slow transport processes may lead to a decay of the arrested structures at much longer times; see the reviews [20, 21] for discussions and extended theoretical approaches.

Whereas glass transitions have been studied extensively and are present in the studied colloidal suspensions at higher packing fractions also [4], non–ergodicity transitions into gel states exhibit novel phenomena and anomalies connected to the low packing fractions and the resulting tenuous ramified gel structures. The experimentally observed small–angle scattering peaks indicating domain formation after quenches into states far from equilibrium are one aspect, which however is neglected here, because short local length scales are considered where local equilibrium may be assumed. Another aspect, the existence and growth of tightly bonded clusters is signalled by the local dynamics; see below.

3. Virial expansion input

Specifying the static input, i.e. \( c_q \), the bifurcation lines and the resulting long–time dynamics can be determined as functions of the thermodynamic control parameters, viz packing fraction, \( \phi \), and temperature, \( T \). For colloidal particles characterized by steric repulsion and short–ranged attraction, the square well potential is widely used [24]:

\[
u(r) = \begin{cases} 
\infty & 0 < r < \sigma \\
-U_0 & \sigma < r \leq \sigma + \Delta \\
0 & \sigma + \Delta < r .
\end{cases}
\]  

(4)

The virial expansion provides a controlled approximation for dilute systems and determines the direct correlation function for low packing fractions as: \( c(r) = f(r) + \mathcal{O}(\phi) \), where \( f = e^{-u(r)/k_B T} - 1 \) is the Mayr cluster function [23]. The gel transitions can be studied in an asymptotic model of vanishing packing fraction and temperature,
which describes a dilute system of strongly interacting particles [18, 19]. Within the virial expansion this limit is obtained for:

\[
\phi \to 0 \quad \text{and} \quad A \to \infty, \quad \text{so that} \quad \Gamma_v = \frac{12 \Delta \phi A^2}{\pi^2 \sigma} = \text{constant},
\]

where \( A = e^{U_0/k_B T} - 1 \). Note that this results in \( S_q = 1 \), because of \( \phi \to 0 \), indicating that structural correlations can be neglected in explaining the gel non–ergodicity transitions. Thus, in this limit, glassy dynamics, due to caging at higher packing fractions, is absent as well as dynamics connected to critical phenomena, where long–ranged correlations are important; note that an increase in the compressibility (or \( S_{q=0} = 0 \)) would require lower temperatures than considered in Eq. (5) as the virial estimate indicates: \( S_{q=0} \to \infty \) for \( \phi A \Delta/\sigma = 1/24 + O(\phi, \Delta) \). Moreover, as expected on physical grounds, only attractive potentials can lead to the limit Eq. (5) and thus to gels, because \( A \) would remain bounded for pure repulsions; this is violated by the mean spherical approximation used in [18, 19].

4. Results for the gel structures

The results for the gel form factors \( f_q \) are of immediate interest as they provide information on the spatial correlations of the arrested solid–like structures.

\[\text{Figure 1. Critical non–ergodicity parameter } f_q^c \text{ of strongly interacting dilute fluids described by Eq. (5) for the well sizes } \Delta/\sigma = \frac{1}{4}, \frac{1}{2}, 1 \text{ and in the limit } \Delta \to 0; \text{ most curves are not shown below } q \Delta = 0.3 \text{ because of numerical inaccuracies. The inset shows the variation of the critical coupling parameter } \Gamma^c_v \text{ with well size } \Delta.\]

In the specified limit of a strongly interacting dilute colloidal fluid, the mode coupling functional simplifies to a two parameter model as its density and temperature dependence enters via \( \Gamma_v \) from Eq. (5) only: \( \mathcal{F}_q(c, [f]) \to \mathcal{F}_q(\Gamma_v, \Delta/\sigma, [f]) \). Figure 1
shows non-ergodicity parameters at the transition points for three different well sizes, \( \Delta/\sigma = 1, \frac{1}{2}, \frac{1}{4} \), and in the limit \( \Delta \to 0 \). At the gel transition, bonds of a length given by the attraction range are formed between the particles; the well size \( \Delta \) sets the length scale for \( f_q^c \). In the non-ergodic states, fast particle rearrangements are possible on shorter distances, causing the decay of \( \Phi_q(t=0) = 1 \) down to \( f_q \), but small wave vector fluctuations are progressively suppressed upon decreasing \( \Delta \). Only for bond-lengths or well sizes \( \Delta \geq 0.1 \), appreciable large distance temporal fluctuations of the gel structure exist.

The additional limit \( \Delta/\sigma \ll 1 \) is of interest, because the asymptotic model defined by Eq. (3) further simplifies and the results from Fig. 1 indicate that this limit qualitatively captures the gel structures (i.e. \( f_q^c \)) for \( \Delta \) below ca. 0.1. Moreover, the elastic constants of the gel increase strongly in this limit of short-ranged attractions, and thus observation of the non-ergodic gel states is more likely, as will be argued below.

Figure 2. Non-ergodicity parameter \( f_q^c \) (full red line) and critical amplitude \( h_q \) (long dashed red line) at the critical point of the virial model for the square well potential. The thin short dashed black, green and blue lines show the corresponding results from the AHS model using the wave vector cut-offs: \( k_{\text{max}} \sigma = 40, 80 \) and \( 160 \); the effective width is found as \( \Delta_{\text{eff}}^{\text{AHS}} = 3.68/k_{\text{max}} \).

A naive application of \( \Delta \to 0 \) leads to the AHS–virial result: \( c_q \to 4\pi A\Delta \sigma \sin q\sigma \). Because of its slow algebraic decay for \( q \to \infty \), the wave vector integration in Eq. (3) does not converge and the results depend on the chosen wave vector cut-off \( k_{\text{max}} \). In the AHS–model this holds for all temperatures and packing fractions as argued in [18]. However, first entering the direct correlation function into Eq. (3), and then performing the limit \( \Delta \to 0 \) leads to a different and almost everywhere convergent memory kernel:

\[
\mathcal{F}_q(\Gamma, [f]) = \frac{\Gamma_v}{q^2} \int d^3 \tilde{k} \left( \frac{\tilde{q} \cdot \tilde{k}}{qk^2} \right)^2 (1 - \cos \tilde{k}) f_{\tilde{k}} f_{|q-\tilde{k}|} \tag{6}
\]
where $\tilde{q} = q\Delta$ denotes the rescaled wave vector. Tight localization of the particles is predicted by the solutions to Eqs. (3) and (6) shown in Fig. 2, because the $q$-width of the Edwards–Anderson parameters $f_q$ is given by the inverse of the narrow well width $\Delta$. Binding to in this limit infinite clusters is described because the wave vector dependent longitudinal modulus diverges like $1/\tilde{q}^2$, even though it is connected to the total force acting among all particles, $q^2 F_q \propto \langle F_{\text{tot}}^*(t \to \infty) F_{\text{tot}}(0) \rangle$ for $q \to 0$, where the total force among all particles vanishes according to Newton’s actio–reactio principle, $F_{\text{tot}} = 0$. In the specified limit, however, the particles experience forces from infinitely removed particles belonging to the same cluster, and thus $q^2 F_q$ stays finite as the true $F_{\text{tot}}$ cannot be determined. Collective and single particle density fluctuations consequently become identical, $\Phi_q(t) = \Phi_q^s(t)$, leading e.g. to the limit $f_{\tilde{q}} \to 1 - (\tilde{q} r_s)^2$ for $\tilde{q} \to 0$, where the particle root mean square displacement, $r_s$, approaches (half) the well width, $\tilde{r}_c s = r_c s / \Delta = 0.48$, at the transition point; see also [18, 19].

Figure 2 shows the solution of Eqs. (3) and (6) (evaluated with 400 grid points spaced at $\delta \tilde{q} = 0.025$) at the critical point $\Gamma^c_v = 1.42 \ldots$, which corresponds to rather modest attractions; e.g. for $\Delta / \sigma = 1/100$ and $\phi = 0.1$ one finds $U_0/k_B T = 3.56$. Solutions from the AHS–virial approximation are also included for three different cut–offs $k_{\text{max}} \sigma = 40, 80$ and 160. One notices that for small wave vectors the solutions can be scaled onto the square well result; also the AHS critical coupling parameter scales as one would expect: $\Gamma^c_{\nu}^{\text{AHS}} \propto \frac{1}{k_{\text{max}}}$. However, as all wave vectors for example enter the determination of the exponent parameter $\lambda_{\nu}^{\text{AHS}} = 0.65$ compared to $\lambda_{\nu}^{\text{sq–W}} = 0.79$.  

5. Results for the dynamics

The bifurcations to non–ergodic solid–like states in Eqs. (3) and (6) also lead to anomalies in the long–time dynamics which provide the most detailed information about the gel–formation and the connected transport mechanisms. We focus on the limit of narrow attractions, $\Delta \ll \sigma$, as clustering is specific to the considered low–density transitions, and it is thus of interest to study the consequences of the small–$q$ divergence of the longitudinal modulus.

In the limit given by Eq. (3) and for $\Delta \ll \sigma$, introduction of a rescaled time, $\tilde{t} = t D_0 / \Delta^2$ with $D_0 = D_{q \to \infty}$, eliminates the transient parameters and leads to simplified equations of motion for the coherent (and identically for the incoherent) density fluctuations:

$$\partial_{\tilde{t}} \Phi_q(\tilde{t}) + q^2 \Phi_q(\tilde{t}) + \int_0^\infty d\tilde{\tau}' \tilde{m}_q(\tilde{\tau} - \tilde{\tau}') \partial_{\tilde{\tau}'} \Phi_q(\tilde{\tau}') = 0,$$

where the friction function is given by: $\tilde{m}_q(\tilde{t}) = q^2 F_q(\Gamma_v, [f])$, and the initial variation is $\Phi_q(t) = 1 - q^2 \tilde{t} + \ldots$. Figure 3 shows intermediate scattering functions for a range of interaction parameters and wave vectors. The two–step relaxation can be analyzed with the techniques developed for MCT–glass transitions [20, 21]. It exhibits the factorization
Colloidal gelation and non–ergodicity transitions

Figure 3. Intermediate scattering functions $\Phi_\tilde{q}(\tilde{t})$ determined from (7). For the interaction parameters $\Gamma_v/\Gamma_v^c = 0.9, 0.99, 0.999,$ and 1.01, corresponding to the short–, long–dashed, solid and dashed–dotted curves, solutions for the wave vectors $\tilde{q} = 0.8$ (red), 1.6 (blue) and 3.2 (green) are shown. The thin horizontal lines indicate the critical non–ergodicity parameters $f^c_\tilde{q}$ which give the long–time limits, $f^c_\tilde{q} = \Phi_\tilde{q}(\tilde{t} \to \infty)$ at $\Gamma_v^c = 1.3647$ for the chosen discretization, $\delta \tilde{q} = 0.08$ and 100 grid points.

Figure 4 shows the corresponding storage and loss moduli which, in the fluid, exhibit a near elastic plateau with power–law corrections predicted by the $\beta$–correlator $G^\lambda$. Using the initial variation of $\Phi_\tilde{q}(\tilde{t})$, a high–frequency divergence, $G'(\tilde{\omega}) \propto G''(\tilde{\omega}) \propto \tilde{\omega}^{1/2}$ for $\tilde{\omega} \to \infty$ results from the integration in Eq. (8) at high $k$. This result, which is familiar for colloidal hard spheres in the absence of hydrodynamic interactions [24], cannot be seen in Fig. 4 because of the chosen small wave vector cut–off.

Importantly, Eq. (8) predicts that large, $O((\sigma/\Delta)^2)$, elastic moduli result upon formation of the gels for $\Gamma \geq \Gamma_v^c$, if the particles interact via short–ranged attractions [27]. The elastic modulus at the transition, $G_c^e$, which follows from Eq. (8) and the critical gel structure factors, $f^c_\tilde{q}$, is included in Fig. 4.
Colloidal gelation and non–ergodicity transitions

The final decay of the intermediate scattering functions or of the shear modulus $G$ is described asymptotically by the second MCT scaling law, which entails the existence of a second set of divergent time scales and of $\Gamma_v$–independent and non–exponential relaxation functions [20, 21]. The small–$q$ divergence of the longitudinal modulus leads to a diffusive behavior of the final relaxation times, $\bar{\tau}_q \propto \bar{\tau}_q^\infty (\Gamma_v^c - \Gamma_v)^{-\gamma}$, with the $\Gamma_v$–independent amplitude $\bar{\tau}_q^\infty \propto 1/q^2$. Here the exponent $\gamma$ is determined by $\lambda$, and follows as $\gamma = 2.85$. The diffusive behavior is apparent in Fig. 3 and stresses that the final melting of the precursor gel structure in the fluid phase requires transport over large distances which scale like $\sigma/\Delta^2$ as follows from Eq. (6). Again this indicates the presence of infinite clusters which arrest and cannot melt anymore at and above the gel transition at $\Gamma_v^c$.

6. Conclusions

We have provided further support for the suggestion that colloidal gelation is caused by non–ergodicity transitions triggered on local distances [18, 19].

It was shown that non–ergodic structures characterized by strong elastic moduli are obtained in dilute solutions of colloidal particles interacting with short–ranged attractions upon lowering the temperature. Bonds with a length given by the attraction range are formed between the particles if the strength of the attraction increases to a few $k_B T$. These values are in qualitative agreement with phenomenological considerations for reversible gelation [10, 3]. We suggest, that this formation of long–lived bonds is the rate limiting step for (transient) gelation in colloidal suspensions.
The formation of (percolated) clusters is indicated by the long-ranged force correlations of the arrested structures which cause the small wave vector divergence of the mode coupling functional $F_q$. The description of the mesoscopic gel structure and of the related domain coarsening kinetics, however, has not yet been incorporated into the present approach.

The long-ranged force correlations at the gel non-ergodicity transitions cause that collective and single particle dynamics become identical for long times, and that one mode coupling functional determines all time dependent small wave vector quantities, like mean square displacement or moduli; such connections have already been exploited to measure the elastic response via light scattering techniques [28]. Moreover, a diffusive final relaxation of the colloid density fluctuations results close to the gel transitions, $\tau_q \propto 1/q^2$, which suggests that a generalized Gaussian description, $\Phi_q(t) = \exp\left\{-\left(\frac{q^2}{6}\right)\delta r^2(t)\right\}$, as suggested by Segrè and Pusey [29], can provide a reasonable description of the long time dynamics for all wave vectors; note that for colloidal suspensions at the glass transition this ansatz failed at small wave vectors because of the non-diffusive structural relaxation [29, 30].

Using the exact low density virial expansion, two at first sight unphysical aspects of previous MCT calculations were identified as artefacts of the approximate static structural input. First, the cut-off dependence found within the AHS model [22, 18] was analyzed for low densities. The interesting conclusion is that MCT predicts the AHS model to be in the non-ergodic state for any non-zero particle concentration. Second, it was clarified that gelation requires attractive interactions.

Dynamic light scattering measurements as preformed by van Megen and coworkers at the colloidal glass transition [31, 32, 33] or further viscoelastic measurements [7, 28] can provide crucial tests of our approach. First measurements of the non-ergodicity parameters support it [34]. Dynamic light scattering measurements of Krall and Weitz [35] also show intriguing connections and require further study.

7. Acknowledgments

Valuable discussions with Dr. P. N. Segrè, Professor D. A. Weitz, Professor M. E. Cates and Professor W. Götze are gratefully acknowledged. M. F. was supported by the Deutsche Forschungsgemeinschaft under Grant No. Fu 309/2.

8. References

[1] M. Carpinetti and M. Giglio, Phys. Rev. Lett. 68, 3327 (1992).
[2] J. Bibette, T. G. Mason, H. Gang, and D. A. Weitz, Phys. Rev. Lett. 69, 981 (1992).
[3] W. C. K. Poon and M. D. Haw, Adv. Colloid Interface Sci. 73, 71 (1997).
[4] W. C. K. Poon, J. S. Selfe, M. B. Robertson, S. M. Ilett, A. D. Pirie, and P. N. Pusey, J. Phys. II France 3, 1075 (1992).
[5] W. C. K. Poon, A. D. Pirie, and P. N. Pusey, Faraday Discuss. 101, 65 (1995).
[6] M. C. Grant and W. B. Russel, Phys. Rev. E 47, 2606 (1993).
[7] C. J. Rueb and C. F. Zukoski, J. Rheology 42, 1255 (1998).
P. Poulin, J. Bibette, and D. A. Weitz, Eur. Phys. J. B 7, 277 (1999).
H. Verduin and J. K. G. Dhont, J. Colloid Interface Sci. 172, 425 (1995).
N. A. M. Verhaegh, D. Asnaghi, H. N. W. Lekkerkerker, M. Giglio, and L. Cipelletti, Physica 242, 104 (1997).
M. Muschol and F. Rosenberger, J. Chem. Phys. 107, 1953 (1997).
P. R. ten Wolde and D. Frenkel, Science 277, 1975 (1997).
D. Rosenbaum, P. C. Zamora, and C. F. Zukoski, Phys. Rev. Lett. 76, 150 (1996).
W. C. K. Poon, Phys. Rev. E 55, 3762 (1997).
R. M. L. Evans, W. C. K. Poon, and M. E. Cates, Europhys.Lett. 38, 595 (1997).
A. J. Bray, Adv. in Phys. 43, 357 (1994).
F. Sciortino and P. Tartaglia, Phys. Rev. Lett. 74, 282 (1995).
J. Bergenholtz and M. Fuchs, Phys. Rev. E 59, 5706 (1999).
J. Bergenholtz and M. Fuchs, J. Phys.: Condens. Matter submitted (1999).
W. Götze, in Liquids, Freezing and Glass Transition, edited by J.-P. Hansen, D. Levesque, and J. Zinn-Justin (North-Holland, Amsterdam, 1991), p. 287.
W. Götze and L. Sjögren, Rep. Prog. Phys. 55, 241 (1992).
L. Fabbian, W. Götze, F. Sciortino, and P. T. F. Thiery, Phys. Rev. E 59, R1347 (1999).
J. P. Hansen and I. R. McDonald, Theory of Simple Liquids (Academic Press, London, 1986).
W. B. Russel, D. A. Saville, and W. R. Schowalter, Colloidal Dispersions (Cambridge University Press, New York, 1989).
U. Bengtzelius, W. Götze, and A. Sjölander, J. Phys. C 17, 5915 (1984).
W. Götze, Z. Phys. B 60, 195 (1985).
Note an error in [18], where $G$ corrected to $G \propto K_c^2 \phi_c^2 / b^3$ shows the same scaling behavior as the present result.
T. G. Mason and D. A. Weitz, Phys. Rev. Lett. 75, 2770 (1995).
P. N. Segrè and P. N. Pusey, Phys. Rev. Lett. 77, 771 (1996).
M. Fuchs and M. R. Mayr, Phys. Rev. E 60, in press (Nov. 1999).
W. van Megen and S. M. Underwood, Phys. Rev. Lett. 70, 2766 (1993).
W. van Megen and S. M. Underwood, Phys. Rev. E 49, 4206 (1994).
W. van Megen, T. C. Mortensen, J. Müller, and S. R. Williams, Phys. Rev. E 58, 6073 (1998).
W. C. K. Poon, L. Starrs, S. P. Meeker, A. Moussaïd, R. M. Evans, P. N. Pusey, and M. F. Soliva, Faraday Discuss. 112, 143 (1999).
A. H. Krall and D. A. Weitz, Phys. Rev. Lett. 80, 778 (1998).