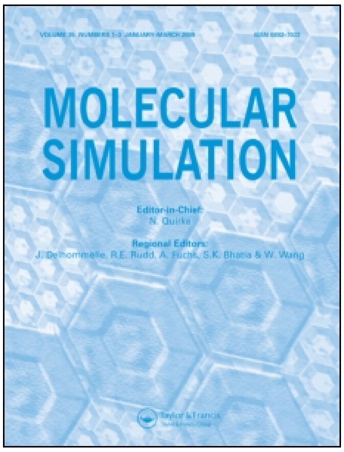

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Dynamic Light Scattering Studies in Silica/Ethylene Glycol Charged Colloidal System

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Dynamic Light Scattering Studies in Silica/Ethylene Glycol Charged Colloidal System

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Multiple scattering of light in colloidal suspensions can be suppressed by suspending the particles in a liquid with matching refractive index. We minimize the multiple scattering of light in charged silica suspensions by dispersing them in a mixed solvent of ethylene glycol/water (EGW). We report here the static and dynamic light scattering investigations carried out on these closely refractive index matched and deionised suspensions over a wide range of volume fractions ($\phi = 0.0006$–0.12). The suspensions are found to have a liquid-like order up to a volume fraction ϕ of 0.02 and a crystalline order beyond this ϕ value. A liquid-like ordered suspension with $\phi = 0.04$, when subjected to sudden compression by centrifugation, the suspension was found to freeze into a colloidal glassy state. The dynamics in liquid-like, crystalline and glassy states of these silica dispersions are analyzed and the results are compared with those reported for aqueous latex suspensions.

Keywords: Colloidal suspensions; Structural ordering and dynamics; Static and dynamic light scattering; Multiple scattering; Silica particles

INTRODUCTION

One of the most fascinating aspects of charged colloidal dispersions is the appearance of long-range order. For instance, in the case of deionised aqueous suspensions of polystyrene particles, the long-range order appears even in extremely dilute conditions i.e. volume fraction $\phi \sim 0.001$. These structures, commonly known as colloidal crystals, exhibit iridescence arising from the Bragg diffraction of visible light. In addition to the crystalline order, these dispersions exhibit other structural orders, viz. gas-like, liquid-like and even glass-like. Structural ordering in monodisperse charge stabilized colloidal dispersions can be tailored with ease by varying the range and strength of the interparticle interaction [1,2]. The dominant interparticle interaction in these systems is the screened Coulomb repulsion and its range and strength can be varied over a wide range by varying the suspension parameters such as the volume fraction ϕ, salt concentration C_s, charge on the particle Ze, and the dielectric constant ε of the medium [1,2]. The interparticle separation in charged colloids is of the order of the wavelength of visible light and the time scale of particle motion ranges from μs to ms. Hence static and dynamic light scattering techniques are appropriate for probing the ordering and the dynamics in colloidal suspensions and has been used extensively to study the phase behavior of aqueous suspension of polystyrene colloids [1–11].

When the refractive index difference, Δn, between the particle and the dispersion medium, is large (e.g. for aqueous suspension of polystyrene particles $\Delta n \sim 0.3$), multiple scattering of light becomes considerable even at intermediate volume fraction ($\phi \sim 0.01$). Hence the analysis and the interpretation of light scattering data becomes complex. Multiple scattering can be suppressed to a large extent by making Δn small. We achieve this in silica colloids by dispersing silica particles in a EGW mixture, where we tune the Δn by varying the ratio of ethylene glycol to water. The dielectric constant of the medium ε as well as charge on the particle gets altered by the variation of the ratio of EG to water in the EGW mixture. This results in the variation of the strength and range of screened Coulomb interaction between

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the like-charged silica particles. The silica-EGW charged colloidal system has not been investigated in detail for its phase behavior. Hence, we report here the static and dynamic measurements carried over a wide range of volume fraction ϕ ($\sim 0.0006-0.12$) in silica suspensions dispersed in EGW mixture with $\Delta n \sim 0.03-0.05$. The reduction of Δn by one order of magnitude less as compared to aqueous suspensions of polystyrene particles helped us to observe all the structural orders and investigate the dynamics over this wide volume fraction range.

STATIC AND DYNAMIC LIGHT SCATTERING

Consider light scattering from a scattering volume V_s containing N spherical colloidal particles dispersed in medium. The intensity of light scattered by the dispersion medium is negligible as compared to that scattered quasi-elastically by the particles. The time-averaged intensity using the Rayleigh-Gans approximation is given as [2,6,7]

$$I_s(Q) = \mathrm{A}P(Q)S(Q) \tag{1}$$

where $Q = (4\pi\mu_s/\lambda)\sin(\theta/2)$ is the scattering wave vector, θ is the scattering angle, μ_s is the refractive index of the dispersion medium and λ is the wavelength of laser light. The particle scattering form factor $P(Q)$ for a spherical particle of radius a is given as

$$P(Q) = \left[\frac{3[\sin(Qa) - Qa\cos(Qa)]}{(Qa)^3}\right]^2 \tag{2}$$

The interparticle structure factor $S(Q)$ in Eq. (1) is given by

$$S(Q) = 1 + \frac{1}{N}\sum_{i,j=1}^{N}\exp[iQ\cdot(\mathbf{r}_i - \mathbf{r}_j)] \tag{3}$$

where $\mathbf{r}_i$ is the position of the center of mass of the ith particle and A is a constant, which for a polarized light is given by

$$A = \frac{9\pi^2 n_m(\mu-1)^2 V_s n_p v_p^2 I_0}{\lambda^4(\mu+2)^2R^2}. \tag{4}$$

Here, $\mu = \mu_p/\mu_s$, μ_p is refractive index of the particle, n_p is the particle concentration and v_p is the volume of the particle, I_0 is the incident intensity of the laser and R is the distance between the scattering volume and the detector. $S(Q)$ is obtained by measuring $I_s(Q)$ as a function of Q and then correcting for $P(Q)$ using Eq. (2).

In a homodyne dynamic light scattering, experiment one measures the time averaged normalized intensity autocorrelation function $g^{(2)}(Q,t)$

$$g^{(2)}(Q,t) = \langle I(Q,0)I(Q,t)\rangle / \langle I(Q,0)\rangle^2 \tag{5}$$

and is related to the normalized electric field autocorrelation function $g^{(1)}(Q, t)$ by the Siegert-Relation [8].

$$g^{(2)}(Q,t) = 1 + |g^{(1)}(Q,t)|^2 \tag{6}$$

As the particle positions $\{\mathbf{r}_i(t)\}$ change due to Brownian motion, the electric field $E(Q,t)$ of the light single-scattered into the far field at scattering angle θ fluctuates randomly in time. These fluctuations can be characterized using the first order correlation function

$$g^{(1)}(Q,t) = \langle E(Q,0)E^*(Q,t)\rangle / \langle |E(Q,0)|^2\rangle, \tag{7}$$

which is related to the "dynamic structure factor" or the "intermediate scattering function" $F(Q,t)$ by

$$g^{(1)}(Q,t) = F(Q,t)/S(Q) \tag{8}$$

where

$$F(Q,t) = \frac{1}{N}\sum_{i=1}^{N}\sum_{j=1}^{N}\langle\exp(iQ\cdot(\mathbf{r}_i(0) - \mathbf{r}_j(t)))\rangle \tag{9}$$

and $S(Q) = F(Q,0)$. Information on the Brownian motion of colloidal particles is all obtained in the functional dependence of $F(Q, t)$ on Q and t.

EXPERIMENTAL DETAILS

The mother suspension of silica in EGW medium was prepared as follows. Aqueous dispersion of colloidal silica (Seahoster KE-P10W), stabilized by ammonium hydroxide, was purchased from Nippon Shokubai Co., Ltd (Tokyo, Japan). The particle diameter d, determined by dynamic light scattering and ultra-small-angle x-ray scattering method, was 0.12 µm. It was purified by dialysis against purified water, and then further deionised by coexisting a mixed-bed of anion- and cation-exchange resin beads (AG 501X8(D), Bio-Rad Labs, Hercules, U.S.A.). The suspension was then mixed with deionised 60% aqueous solution of EG with a ratio of 1:1 by volume, and water was carefully distilled out with stirring at 60°C under a reduced pressure. After the mixed-bed ion-exchange resin beads had been added, the suspension was kept standing at least for 10 days for further deionisation, and used as a mother suspension. The water content in the resulting suspension was 19%, which was estimated by Karl–Fisher method. Electrical conductivity κ of the dispersion was measured by using type DS-12 conductivity meter (Horiba, Japan) at 25°C. The counterion (H^+ ion in the present case) concentration C_c was calculated from κ using the molar conductivity of H^+ ion in 80% EG (28.5 Scm/mol at 25°C). The effective surface charge number and charge density of the particle estimated from C_c were 410/particle and 0.15 μC/cm^2, respectively.

Samples over a wide range of volume fraction ($\phi = 0.0006$–0.12) were prepared in 8 mm optical path length light scattering cells by diluting the mother suspension with EGW mixture. For further removal of ionic impurities, clean nylon net cloth bags containing the mixed bed ion exchange resins were hung from top of the cell and sealed hermitically. Static and dynamic light scattering measurements were carried out using Malvern (UK) 4700 multi-angle light scattering system with a multi-tau photon correlator and a mixed ion (Ar + Kr) laser. The multi-tau correlator helps in measuring $g^{(2)}(t)$ over 11 orders of magnitude in time with smallest delay time being 50 ns. All the light scattering measurements are carried out at room temperature ($T = 300$°K).

RESULTS AND DISCUSSION

We report here the results obtained from static and dynamic light scattering measurements carried on deionised silica suspensions dispersed in EGW mixture (96:4) with different volume fractions.

Non-interacting Suspension

This condition is realized by preparing a sufficiently diluted sample having a volume fraction of ($\phi \sim 10^{-4}$) from the mother suspension and adding a known amount of NaCl solution to screen the Coulomb interaction between the like-charged silica spheres. The structure factor $S(Q)$ measured from the scattered light intensity $I(Q)$ at different values of Q after correcting it to $P(Q)$ calculated using Eq. (2) for 0.12 μm size particle. It can be seen from Fig. 1 that the measured $S(Q)$ is independent of Q. This featureless $S(Q)$ suggests that the particles are spatially uncorrelated and it happens in a non-interacting (gas-like disordered) system of particles.

Since the particle positions and velocities of different particles are uncorrelated with each other in a non-interacting suspension, the cross terms ($i \neq j$) in Eq. (9) are absent. Then $F(Q,t)$ becomes the self-intermediate scattering function, $F_s(Q,t)$ and it can be shown that [5,6]

$$F_s(Q,t) = \langle \exp[-iQ\cdot(\mathbf{r}(t) - \mathbf{r}(0))] \rangle \qquad (10)$$

By assuming the distribution for random particle displacements $|\Delta\mathbf{r}(t)| = |\mathbf{r}(t) - \mathbf{r}(0)|$ to be Gaussian, one can write Eq. (10) as

$$F_s(Q,t) = \exp(-Q^2\langle \Delta r^2(t) \rangle / 6) \qquad (11)$$

For time $t \gg \tau_B$ (where $\tau_B = m/6\pi\eta a$ is the Brownian time τ_B), one obtains for free Brownian diffusion [5,6]

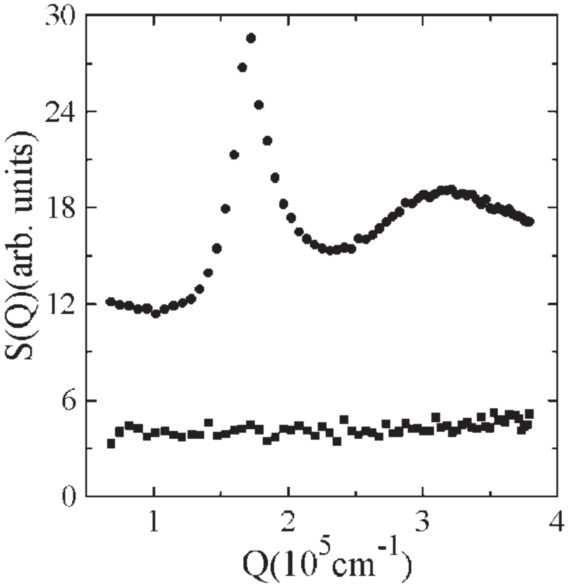

FIGURE 1 $S(Q)$ vs. Q for gas-like (■■■) and liquid-like (●●●) ordered suspensions with volume fraction $\phi = 0.0006$ and 0.013, respectively. EG to water ratio for gas-like and liquid-like ordered suspension is 99.5:0.5 and 96:4, respectively.

$$g^{(1)}(Q,t) = F_s(Q,t) = \exp(-D_0 Q^2(t)). \qquad (12)$$

The mean square displacement $\langle \Delta r^2(t) \rangle$ for a freely diffusing particle (from Eqs. (11) and (12)) is given as

$$\langle \Delta r^2(t) \rangle = 6 D_0 t \qquad (13)$$

and the free diffusion coefficient $D_0 = k_B T / 6\pi\eta a$, where a is radius of the particle of mass m, η is the shear viscosity of the medium and k_B is the Boltzman constant. For 0.12 μm diameter silica particle dispersed in EGW mixture (99.5:0.5) with $\eta = 15.1$ cp at T = 300°K, D_0 is estimated to be 2.42×10^{-9} cm^2/s.

Figure 2 shows the exponential decay of the field correlation function and the continuous line is the fit to Eq. (12) with $D_0 = 2.42 \times 10^{-9}$ cm^2/s. It can also be seen that the mean square displacement (see inset of Fig. 2) obtained from Eqs. (11) and (12) agrees well with that calculated using Eq. (12).

Interacting Suspensions

Deionised suspensions with volume fractions up to a volume fraction of 0.02 did not show iridescence for the incident visible light. The structure factor $S(Q)$ obtained from the angular dependence of the average intensity of the scattered light from these samples showed liquid-like ordering and Fig. 2 shows the $S(Q)$ for suspension with $\phi = 0.02$.

In a liquid-like ordered suspension, a given particle will generally find itself in potential well

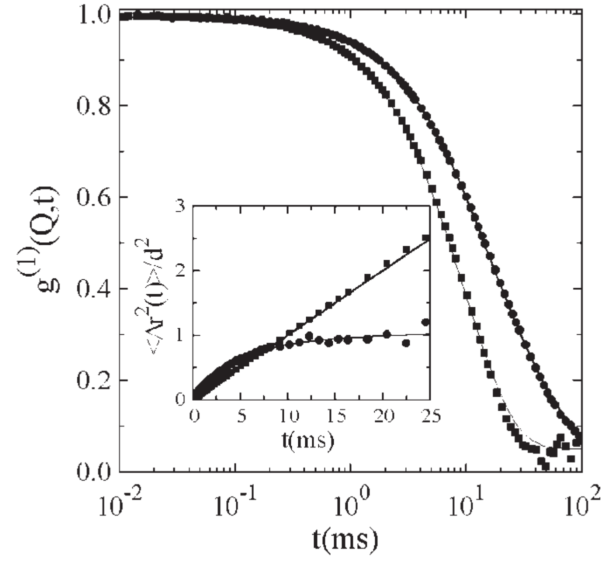

FIGURE 2 Correlation functions $g^{(1)}(Q,t)$ for gas-like (■■■) and liquid-like (•••) suspensions with ϕ same as Fig. 1 at $Q = 2.0 \times 10^5\,\text{cm}^{-1}$ and $3.8 \times 10^5\,\text{cm}^{-1}$, respectively. The continuous lines are a fit to Eq. (12) with $D_0 = 2.42 \times 10^{-9}\,\text{cm}^2/\text{s}$ for a gas-like ordered suspension and a cumulant fit with $K_1 = 756\ \text{s}^{-1}$ for liquid-like ordered suspension. Mean square displacement for the gas-like and liquid-like ordered suspensions calculated using Eq. (11) is shown as inset and the continuous lines are fit to Eq. (13) (gas-like) and Eq. (16) (liquid-like), respectively.

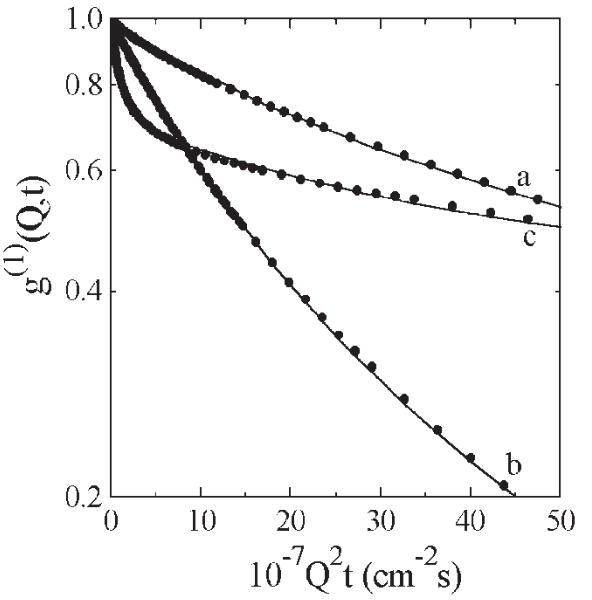

FIGURE 3 Correlation functions $g^{(1)}(Q,t)$ for the liquid-like ordered suspension at three different scattering vectors relative to the position $Q_{\max}$ of the peak of $S(Q)$: (a) $Q = 1.72 \times 10^5\,\text{cm}^{-1}$; (b) $Q = 3.8 \times 10^5\,\text{cm}^{-1}$ and (c) $Q = 1.02 \times 10^5\,\text{cm}^{-1}$.

formed by the instantaneous configuration ("cage") of its neighbors. Insight into the particle dynamics in liquid-like ordered suspension has been obtained by Pusey [5] using the Langevin equation. The particle can be viewed as free at short-times ($\tau_B \ll t \ll \tau_I$) but is temporarily trapped for $t > \tau_I$ in the "cage" formed by the instantaneous configuration of its neighbors. Here, τ_I represents the characteristic fluctuation time associated with the direct force on a particle due to the instantaneous configuration of its neighbors. It is roughly the time taken for the relative positions of a particle and its neighbors to change by a significant fraction of the average interparticle spacing $l(= n_p^{-1/3})$, e.g. $\tau_I \sim (0.1l^2)/6D_0$. Typical value of τ_I for a silica suspension in EGW mixture with $\phi = 0.01$ is about 15 ms. Pusey has showed that in an interacting suspension for $t \gg \tau_B$ the mean square displacement $\langle \Delta r^2(t) \rangle$ is given by

$$\langle \Delta r^2(t) \rangle = 6D_0 t - 6A\tau_1\left[t + \tau_I \exp(-t/\tau_I) - \tau_I\right]. \quad (14)$$

For short time ($\tau_B \ll t \ll \tau_I$)

$$\langle \Delta r^2(t) \rangle = 6D_0 t \quad (15)$$

and at long-times ($t > \tau_I$)

$$\langle \Delta r^2(t) \rangle = 6D_L^s t - 6A\tau_I^2 \quad (16)$$

where $D_L^s = D_0 - A\tau_I$ can be identified as the long-time self-diffusion coefficient of the particle. Fit to the Eq. (14) with $A = 5.4 \times 10^{-7}\,\text{cm}^2/\text{s}^2$ and $\tau_I = 4.36\,\text{ms}$ is shown as inset in Fig. 2.

For Q values much larger than the first peak position $Q_{\max}$ of $S(Q)$, $S(Q) \sim 1$ and the measured $F(Q,t)$ reduces approximately to $F_s(Q,t)$. Hence, one can obtain D_0 and D_L^s values in a liquid-like ordered suspension from the short-time and long-time behavior of $\langle \Delta r^2(t) \rangle$ calculated from the $g^{(1)}(Q,t)$ measured at $Q \gg Q_{\max}$.

Figure 3 shows measurements of $g^{(1)}(Q,t)$ as a function of t for three values of Q on a liquid-like ordered suspension with $\phi = 0.013$. It can be seen that correlation function shows a strong dependence on Q and is due to interactions in the suspension. Whereas like the non-interacting suspension (gas-like ordered) $g^{(1)}(Q,t)$ was found to be independent of Q. It can be seen from Fig. 3 that data shows curvature at shot times to a roughly linear behavior (on log scale) at longer-times. The diffusion coefficients D_s and D_L, which characterize the particle motion in a liquid-like ordered suspension at short and long-times, can be obtained from the following analysis of $g^{(1)}(Q,t)$.

In the absence of hydrodynamic interactions and in the limit of $\tau_B \ll t \ll \tau_I$ and at all values of Q, $\ln[g^{(1)}(Q,t)]$ can be fitted to a cumulant expansion [5] and the first cumulant of dynamic structure factor

$$K_I = D_0 Q^2/S(Q). \quad (17)$$

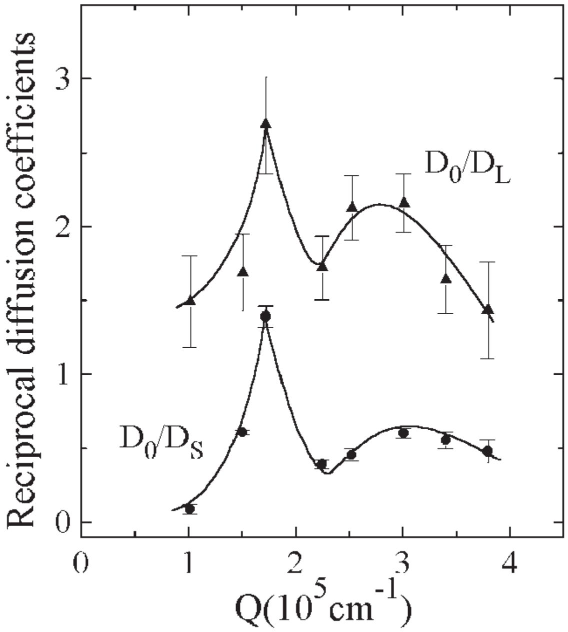

FIGURE 4 The reciprocal short time diffusion coefficient D_S^{-1} and the reciprocal long time diffusion coefficient D_L^{-1} in units of $D_0 (= 2.52 \times 10^{-9}\ \mathrm{cm}^2/\mathrm{s})$ for the liquid-like ordered suspension with $\phi = 0.013$. Lines drawn through the data points are guide to the eye.

The "effective diffusion coefficient" associated with K_1 can thus be written as

$$D_s = D_0/S(Q). \tag{18}$$

Figure 4 shows the D_0/D_s obtained by performing the cumulant analysis [4,5] on the measured $g^{(1)}(Q,t)$ and represents $S(Q)$. Hence the variation of D_0/D_s obtained for different values of Q is compared with $S(Q)$ measured from the angle resolved static light scattering experiment (see Fig. 1). We observed a good matching in the positions of first peak and second peak but not in the magnitudes of $S(Q)$ for all values of Q. Whereas the measurements on liquid-like ordered dilute aqueous suspension by Pusey and Tough [5] as well as by Grüner and Lehmann [4,9] showed a good agreement of these quantities. The effective diffusion coefficients D_L has been obtained from the long-time slopes of the plots in Fig. 3 and D_0/D_L are plotted against Q in Fig. 4. The D_L values are found to be 1.5–2.5 times smaller than D_0. In the case of aqueous latex suspensions D_L values were reported to be 3–5 times smaller than the corresponding D_0. This observation along with the disagreement observed between D_0/D_s and $S(Q)$ in our samples arises due to high viscosity (viscosity of EGW mixture is 15 times higher as compared to that of water) of the medium and also due to relatively high concentration of our samples. The latter one brings in the hydrodynamic interactions to be operative in present samples. Hence, extracting D_s and D_L from $g^{(1)}(Q,t)$ needs models, which accounts for the hydrodynamic interactions [5].

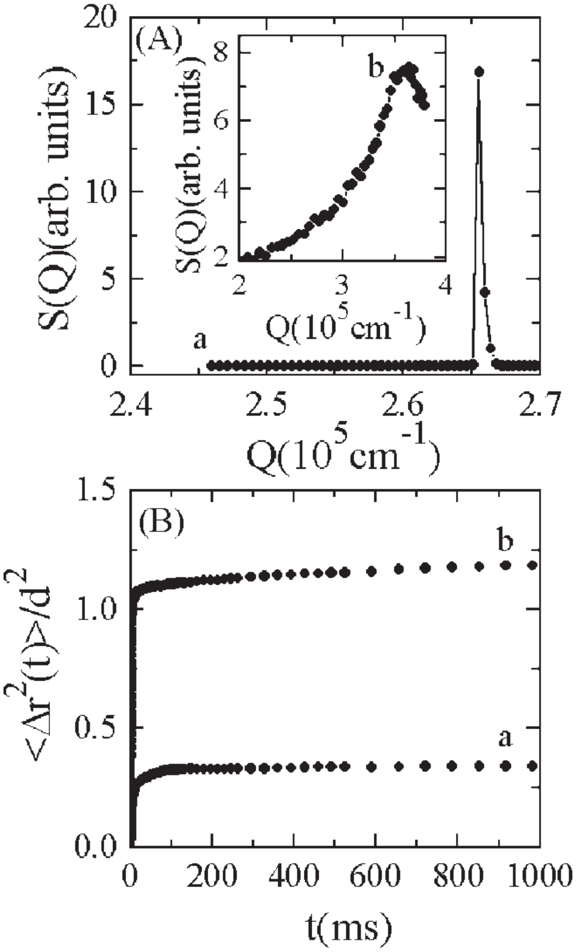

FIGURE 5 (A) Structure factors $S(Q)$ for suspension with crystalline order ($\phi = 0.048$) and the inset is for the glass-like ordered suspension with $\phi = 0.12$. (B) $\langle r^2(t)\rangle$ vs. t for crystalline (curve *a*) and glass-like (curve *b*) ordered samples. Lines drawn through the data points are guide the eye.

Samples with volume fraction $\phi \geq 0.02$ showed iridescence due to Bragg diffraction of visible light and also Bragg spots for the laser light. These observations suggest that the samples are crystalline and we could record the scattered intensity due to Bragg diffraction from some crystallites with (110) planes oriented perpendicular to the scattering plane is shown in Fig. 5(A). Since in a crystal as the long-time diffusion is arrested (i.e. $D_L = 0$), the mean square displacement $\langle \Delta r^2(t)\rangle$, calculated form Eq. (6), is expected to show saturation at long-times and indeed we observe saturation in $\langle \Delta r^2(t)\rangle$ vs. t (see Fig. 5(B)).

It is known from the lattice dynamics of colloidal crystals that all but the long-wavelength shear modes (transverse phonons) are strongly overdamped due to the viscous drag between the particles and the solvent in colloidal crystals [10,11]. By solving the Langevin equation for a harmonically bound Brownian particle, Pusey [5] has obtained an expression for mean square displacement in the strongly over damped limit and is given as

$$\Delta r^2(t) = \frac{6k_B T}{m}\left\{\frac{1}{\omega^2}\left[1-\exp\left(\frac{-\omega^2 t}{\beta}\right)\right] - \frac{1}{\beta^2}\left[1-\exp(-\beta t)\right]\right\} \quad (19)$$

where $\beta = 1/\tau_B$ and ω is angular velocity associated with harmonic potential. For $t > \tau_I$ the harmonic force retards the increase of $\langle \Delta r^2(t) \rangle$, hence $\langle \Delta r^2(t) \rangle$ saturates to a value

$$\langle \Delta r^2(t) \rangle\big|_{t \gg \tau_I} = 6k_B T/m\omega^2. \quad (20)$$

Using Eq. (20) we have estimated the frequency $\omega = 24.3$ kHz associated with the harmonic potential of the colloidal crystal for Q close to the $Q_{(110)}$ peak. We have also measured the phonon dispersion in large size (~3 mm) colloidal crystals [12].

Härtl *et al.* have observed glass-like ordering in index matched monodisperse charged colloidal suspensions at $\phi \sim 0.22$ [13]. Motivated by this observation, we concentrated a liquid-like ordered suspension with $\phi = 0.04$ by subjecting it to centrifugation for 4 h using a centrifuge having rotation speed of 6000 rpm. The measured $S(Q)$ in the concentrated regime of the sample showed a broad peak (see Fig. 5(A)) at $Q = 3.6 \times 10^5\,\mathrm{cm}^{-1}$ and corresponds to a volume fraction of 0.12. Though this value of $\phi > 0.02$, we did not observe iridescence in this sample. In order to find out whether the disorder observed in this suspension is liquid-like or glass-like, we have measured the $g^{(2)}(Q,t)$ at $Q = 3.6 \times 10^5\,\mathrm{cm}^{-1}$ and it is found to exhibit very slow decay extending up to several seconds. The estimated $\langle r^2(t) \rangle$ showed tendency towards saturation (see Fig. 5(B)). These observations confirm that the disorder present in the compressed suspension is glass-like. Colloidal liquid freezes into a glassy state because the liquid experiences sudden compression during centrifugation. Recently, Tata *et al.* [14] have shown that dilute charged colloidal liquid can be frozen into a colloidal glass by sudden compression and the present observation is in accordance with these simulations.

CONCLUSIONS

It has been shown that with the preparation of closely index matched silica colloidal system, it is possible to suppress multiple scattering of light and hence investigate the structure and dynamics over a wide concentration range using static and dynamic light scattering. In particular, the crystalline and glassy state can be investigated in more detail. By combining the static light scattering measurements with dynamic light scattering measurements we have unambiguously identified all the structural orders (gas-like, liquid-like, crystalline and glass-like) in this charged colloidal system. Our measurements show that the crystallization in the present system occurs at relatively higher volume fraction as compared to aqueous system of charged latex particles. Since the dielectric constant of EGW mixture is lower than water, the effective charge on the particles is lower due to the decreased dissociation of silanol end-groups on the surface of the particle. Unlike in polymer latex suspensions, the structure factor extracted from dynamics light scattering measurements is found to differ with that obtained from static light measurements. Though the origin for this disagreement is not clear, the relatively high viscosity and high particle concentration are responsible for this disagreement.

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