# Direct observation of oscillatory-shear-induced order in colloidal suspensions

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We use optical microscopy to directly study ordering in concentrated colloidal suspensions of model hard spheres subjected to oscillatory shear in a parallel plate geometry. At high strain amplitude the particles order quickly into hexagonal planes in the plane of shear, oriented so that a close-packed direction is parallel to the velocity axis, corresponding to previous scattering observations in other geometries. At low strain a polycrystalline structure forms, consisting of hexagonally ordered regions now with a distribution of different orientations, peaked around an orientation where a close-packed direction is now *perpendicular* to the velocity axis. Such polycrystallinity explains the evidence of disorder previously noted in scattering patterns. The ordered regions grow until ''grains'' fill the sample. Various examples of local disorder are observed in the shear-ordered structures, including vacancies, dislocations, and stacking faults. The observed motion of the planes during the cycle is complicated, but there is some evidence of a ''zigzag'' motion of planes at high strain, as has been proposed in previous studies. [S1063-651X(98)05006-5]

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## I. INTRODUCTION AND METHODS

The structural effects of the application of shear to colloidal suspensions are of substantial current interest. Shear-induced structure is important in rheological processing of suspensions, while the ability to control structure on the colloidal (i.e., optical) length scale may have substantial technological application, for instance, in the design of optical band gap materials. The structural effects of applied shear and their correspondence with the rheological behavior of colloidal suspensions and other ''soft matter'' are still not well understood. Colloidal systems, with their long time scales and weak interparticle interactions compared to atomic systems, are also good candidates for the study of generic crystallization and melting phenomena.

The rheological behavior of well-characterized concentrated colloidal suspensions has been extensively studied in recent years and scattering experiments have been used to investigate the correspondence between rheological behavior (e.g., shear thinning and shear thickening) and structural changes under shear [1–5]. In particular, many scattering experiments on systems consisting of ''model'' hard-sphere sterically stabilized polymethylmethacrylate (PMMA) colloids have been reported [2,6–8]. The zero-shear equilibrium phase diagram of PMMA suspensions has been shown to correspond closely to that expected for hard spheres [9]: Suspensions have a fluid structure below a freezing volume fraction $\phi_f \approx 0.494$ and a crystalline structure above a melting volume fraction $\phi_m \approx 0.545$. Between $\phi_f$ and $\phi_m$ fluid and crystal phases coexist. In PMMA suspensions at zero-shear homogeneous nucleation of crystallites leads to a polycrystalline, stacking-faulted hexagonally close-packed structure. Light-scattering studies of suspensions under shear in concentric cylinder and parallel rotating disk geometries have been interpreted as demonstrating that application of a shear field induces layering of the hexagonal planes of the lattice in the velocity-vorticity plane. Moreover, the observed scattering patterns indicate that the hexagonal layers are oriented by the shear [10]. In steady shear, the planes are oriented with a close-packed direction parallel to the velocity axis. Hoffman, in one of the earliest studies of a well-characterized system under steady shear, obtained micrographs showing hexagonally ordered planes of poly(vinyl chloride) particles [1].

In *steady* shear of a colloidal suspension between two parallel plates with a plate separation of $h$, strain $\gamma$ may be defined as $\gamma = A/h$, where $A$ is the relative displacement of the plates. In analogy, in *oscillatory* shear where the displacement of the plates is now an oscillating function of time we define the *peak to peak strain* $\gamma = A/h$, where $A$ is the peak to peak relative displacement of the plates. In steady shear of suspensions the ''high-strain'' regime $\gamma > 1$ is rapidly reached, while in oscillatory shear long-time experiments can be performed at strain $\gamma < 1$. In both steady shear and oscillatory shear experiments with concentrated colloidal suspensions at $\gamma > 1$, scattering experiments indicate that the shear-induced hexagonal planes are oriented with a close-packed direction parallel to the velocity axis [2,6,11] [Fig. 1(a)]. In contrast, in oscillatory shear at $\gamma < 1$ scattering ex-

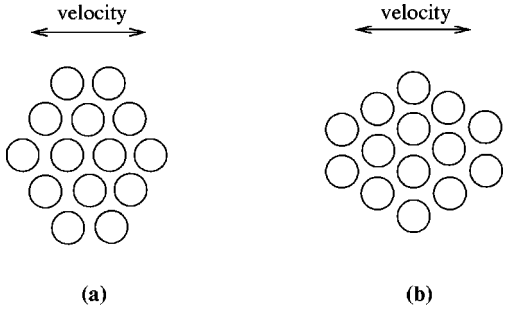


FIG. 1. Schematic picture of (a) ''high strain'' and (b) ''low strain'' orientations of a hexagonally ordered plane of particles. In (a) a close-packed direction in the plane is *parallel* to the velocity axis; in (b) a close-packed direction is *perpendicular* to the velocity.

 

periments indicate that the (111) planes are oriented with a close-packed direction *perpendicular* to the velocity axis [6,11] [Fig. 1(b)]. Especially in the low-strain oscillatory shear scattering experiments there remains also evidence of substantial *disorder* in the shear-generated structure, suggested origins of which are partly polycrystalline structures or hexatic order in the quasi-two-dimensional layers [6,11].

In this paper we present results from direct imaging of concentrated PMMA suspensions under oscillatory shear. Suspensions of sterically stabilized PMMA particles in *cis*-decalin at known volume fractions $\phi$ are made up as described in detail elsewhere [12]. In these experiments the particle diameter is 970 nm (as measured by static light scattering) with a size polydispersity of approximately 5%. We have carried out experiments on samples at $\phi=0.485\pm0.003$ (i.e., fluid at equilibrium), $\phi=0.515\pm0.005$ (coexisting fluid and crystal at equilibrium), $\phi=0.56\pm0.005$ (fully crystalline at equilibrium), and $\phi=0.59\pm0.007$ (colloidal glass). For each set of experiments at a given $\phi$ a ''stock'' suspension of a few milliliters is made up in a sealable cuvette, from which drops are taken and loaded into the shear cell. The sheared sample consists of a circular droplet, of typical diameter 12–15 mm, sandwiched between parallel glass plates. The lower plate (a large-area microscope slide) is held stationary while the upper plate (a large-area coverslip), connected via a frame to a linear electromagnetic vibrator, oscillates along the velocity axis. The plates are separated by strips of plastic shim. In these experiments the plate separation $h$ is typically 130 to 150 $\mu$m, though we have observed no dependence of the results on plate separation for separations up to 700 $\mu$m. The parallelism of the plates over the area of the sample is checked by measuring the upper and lower plate heights in the microscope. In the experiments discussed here we have used a triangular wave form to drive the vibrator; results for sinusoidal wave forms are qualitatively the same. Images of the sample are obtained while under shear by digitizing frames from a video camera via computer software. Note that given the frequencies and amplitudes considered here, it is possible to obtain images free of motional blur at the extremes of the oscillation cycle only. We use a 100× oil immersion phase contrast [13] objective with a working distance of 0.15 mm, further magnification being provided by a 2.5× or 5× ''eyepiece'' lens in front of the video camera. Vertical tracking through the sample during the experiment is carried out using a computer-controlled piezoelectric $z$ drive to move the microscope objective.

For parallel-plate oscillatory shear, the accelerated motion of the shearing (top) plate generates a shear wave through the sample, propagating in the velocity gradient direction (perpendicular to the plates). The velocity and peak to peak amplitude profiles $v(z)$ and $A(z)$ depend on the wavelength of the shear wave relative to the spacing between the plates [14]. Measurements of $A(z)$ carried out by tracking particles in dilute solution indicate that to a good approximation in the bottom half of the cell at least the amplitude gradient is linear for the frequencies and samples used in these experiments. Because of the need to resolve the particles in tracking experiments and therefore the need to use high magnification, it has not been possible to obtain a large enough field of view to accurately measure amplitudes closer to the moving plate. Approximate measurements of the velocity of particles closer to the moving plate (made by timing particles as they move across the field of view) do not, however, indicate any substantial nonlinearity in the velocity profile in our experiment. Additionally, under the assumption of the diameter of the drop being very much greater than the plate separation, the typical viscosities, frequencies, and plate separations used here are consistent with an approximately linear velocity profile [14].

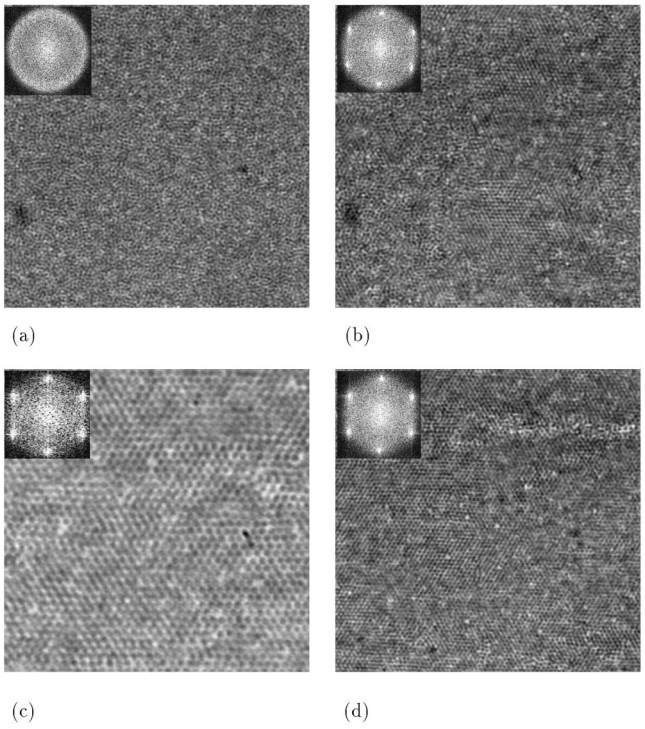


FIG. 2. Microscope images from a shear experiment at high strain, $\gamma=3.1$, 100× objective. The velocity direction is horizontal. The shear frequency is $f=0.2$ Hz. The particles have radius $r=485$ nm and the sample volume fraction is $\phi=0.56$. Times after the beginning of shear are (a) 0 (immediately before shear), (b) 2 min 50 s, (c) 7 min 30 s (magnification doubled using an ''eyepiece'' lens in front of the charge coupled device camera), and (d) 13 min 25 s. Images (a)–(c) are taken at height $z=35$ $\mu$m below the top plate. In (d), $z=10$ $\mu$m. The insets show the calculated Fourier transforms of the images.

## II. RESULTS

### A. High strain $\gamma>1$

We first discuss experiments at high strain amplitude $A=0.43\pm0.005$ mm in a cell with $h=0.14\pm0.003$ mm, giving $\gamma=3.1\pm0.1$. Figure 2 shows a sequence of images from an experiment at frequency $f=0.2$ Hz with a sample at $\Phi=0.56\pm0.005$, that is, a sample whose equilibrium state is fully crystalline. No order is visible in the sample immediately before starting the shear [Fig. 2(a)]. Note that without shear, equilibrium crystal order is observed only after about 3 h in this geometry. On commencing the experiment, within minutes we observe the appearance of regions of hexagonal order oriented with a close-packed direction parallel to the velocity axis [Fig. 2(b)]. In fact, within 4 min the visible plane is essentially completely ordered [Fig. 2(c)]. The large-

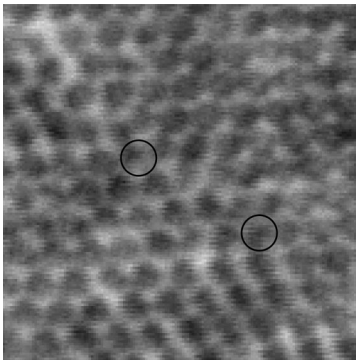

FIG. 3. Expanded detail from a microscope image (100× objective) from a shear experiment at ''high'' strain, $\gamma=2.42$, $f=0.2$ Hz, after 20 min of shear, in a sample at volume fraction $\Phi=0.515$. The plane shown is close to the bottom of the cell, at $z=122$ $\mu$m below the top plate, where the plate separation is $h=128$ $\mu$m. The circles show the positions of a pair of dislocations in the ordered (111) plane.

scale orientation of the planes is constant everywhere in the sample (unlike in the low-strain case; see below). This high degree of order is consistent with previous scattering experiments in cylindrical Couette geometry [6] and in rotating parallel disk geometry [11], as well as with our own scattering experiments in the parallel-plate geometry [15]. In the high-strain scattering experiments, a hexagonal pattern of sharp Bragg spots is observed. The insets in Fig. 2 show the calculated (two-dimensional) Fourier transforms of each microscope image. (Only the innermost set of Bragg peaks are shown in the transforms.) Before shear a bright ring characteristic of a disordered structure is observed. In the inset of Fig. 2(b) we can still see some evidence of this ''fluid'' ring, but now hexagonal peaks are also clear, corresponding to the partly ordered visible plane. Figures 2(c) and 2(d) show sharp hexagonal peaks indicating strongly ordered planes. However, in some regions of the sample various kinds of local disorder are apparent; Fig. 2(d) shows an example where there appears to be a narrow disordered band within the strongly ordered plane. One advantage of microscopy over scattering is that local disorder may be studied more easily; in the scattering patterns the effects of small amounts of disorder are usually swamped by the very bright Bragg peaks from the crystal regions. For example, Fig. 3 shows a magnified view of a portion of a crystal plane (this time from an experiment at $\Phi=0.515$ and $\gamma=2.42$; see below). The circles indicate the positions of two *dislocations* in the hexagonal lattice. In addition, in all experiments we have observed vacancies and some random disorder of particle positions within the lattice; an example image showing vacancies is given in Fig. 4. Direct observation by microscopy would seem to have great potential for the more detailed study of the type, degree, and evolution of disorder in shear-induced crystals.

The influence of the underlying equilibrium fluid-crystal phase coexistence in samples at $0.494\leqslant\Phi\leqslant0.545$ is not clear. In high-strain experiments at $\Phi=0.515$ fully ordered planes similar to those obtained at higher volume fraction are typically observed only near the bottom of the shear cell, as shown in Fig. 3. In the example experiment of Fig. 3 at $\Phi=0.515$ we see fully ordered layers up to about 40 $\mu$m above

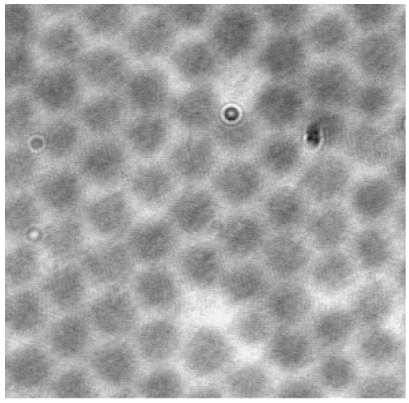

FIG. 4. Expanded detail from a microscope image (100× objective) from a shear experiment at low strain showing the presence of a pair of vacancies in the hexagonal plane. Vertical tracking up and down to neighboring planes demonstrates that these are ''real'' vacancies rather than simply particles too far out of the plane of focus to be imaged. In this experiment the strain amplitude $\gamma=0.5$, the frequency is $f=5$ Hz, and the volume fraction $\Phi=0.59$. The plane shown is at $z=30$ $\mu$m below the top plate, where the plate separation is $h=600$ $\mu$m. At this high frequency the shear must be stopped momentarily in order to obtain a blur-free image.

the bottom, i.e., in the bottom 30% of the sample. Above this, planes become less ordered with increasing height: We observe regions or ''islands'' of crystal order surrounded by disordered fluid, as shown in Fig. 5. These ''island crystallites'' are oriented with close-packed directions parallel to the velocity axis and therefore are clearly associated with the shear (no crystallites are visible in the sample before the application of shear). The fraction of the field of view occupied by the ordered regions decreases with increasing height in the cell. However, we do not observe these crystallites to grow or shrink within the plane appreciably over time. We do not yet understand these effects, though we have observed similar effects in many repeat experiments. This height-

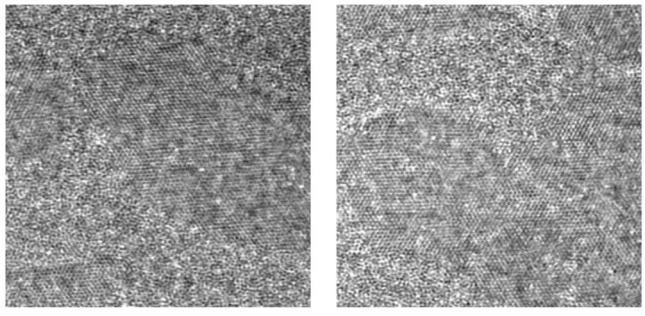


FIG. 5. ''Island'' crystallites, oriented regions of crystal order surrounded by disordered fluid, observed in experiments within the equilibrium coexistence region at high shear strain. These example images are from a shear experiment with strain $\gamma=2.42$, in a sample at volume fraction $\Phi=0.515$, at heights (a) $z=80$ $\mu$m and (b) $z=90$ $\mu$m below the top plate, where the plate separation is $h=128$ $\mu$m. The velocity direction is horizontal. Note that closer to the bottom plate the sample is much more strongly ordered; see Fig. 3, from the same experiment.

dependent disorder is *not* observed in samples at $\Phi > 0.545$, where the whole sample is always ordered by the shear. Furthermore, similar effects are *not* observed at *any* volume fraction in the low-strain experiments ($\gamma < 1$; see below) over comparable time scales, so that it does not seem likely that artifacts of the experiment or concentration gradients caused by sedimentation of the particles are responsible. Nonlinearity of the velocity profile $v(z)$ also does not seem likely to generate the observed features since we see a ''coexistence'' of ordered and disordered regions within a given plane at a single $z$. We also would not expect equilibrium phase separation (into coexisting crystal and disordered fluid) to occur within these time scales; typically, without shear, equilibrium crystal order is not observed in less than a few hours. It may be, however, that the *fast* formation under shear of *dense* crystal regions generates enough density variation *across* the sample (i.e., within the same plane) to enhance phase separation: If the shear-generated crystal regions are at an effective volume fraction *higher* than the original sample volume fraction then mass conservation would require a coexistence of dense crystal regions with regions of lower density. The disordered, less dense regions may then be at low enough density that the shear does not induce further crystal order. (For instance, in experiments on samples at lower volume fraction $\Phi = 0.485$, i.e., samples fluid at equilibrium, at the frequencies and strain amplitudes typically used here, we do not observe shear-induced ordering [16].) This could lead to a more or less stable coexistence of ordered and disordered regions. The more dense regions would furthermore be expected to sediment faster, possibly also explaining the apparent height dependence of the disorder. This is no more than a tentative explanation however, and in whatever case, these observations of order-disorder coexistence under shear require further study. It is worth pointing out that the effects of such order-disorder coexistence would be difficult to observe in scattering experiments due to the strong scattering by the highly ordered crystalline regions, though very careful and precise quantitative measurements of scattered intensity away from the Bragg peaks might demonstrate the existence of some degree of fluid disorder.

Assuming the structure of the sheared suspension to attain some approximately steady state, simple models of ''easy'' strains in close-packed crystals have been proposed by Loose and Ackerson [10] to explain the variations in the observed scattering with strain. Hexagonal planes aligned with close-packed directions parallel to the velocity are proposed to move easiest at high strain by carrying out a zigzag motion, each particle moving in the trough between the particles in the plane below, first to one side of the close-packed line and then back to the other side. We have made some preliminary studies of the motion of planes under shear by observing planes close to the stationary bottom plate of the cell where the velocity is low enough to obtain clear images. A regular sideways motion of the planes while under shear apparently consistent with the proposed zigzag has been observed. The zigzag motion does not become visible until the planes are strongly ordered and is not visible in experiments at low strain (see below). The number of zigzags corresponds approximately to that expected given the strain amplitude. It must be stated, however, that close examination of planes

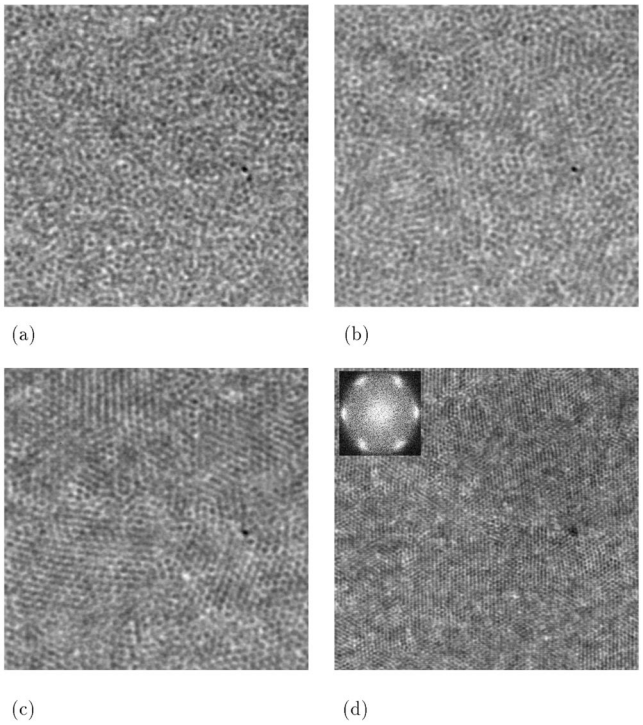


FIG. 6. Microscope images from a shear experiment at ''low'' strain, $\gamma = 0.53$. The shear frequency is $f = 0.625$ Hz and the velocity direction is horizontal. The particles have radius $r = 485$ nm and the sample volume fraction is $\Phi = 0.56$. Times after the beginning of shear are (a) 0 (immediately before shear), (b) 1 min 10 s, (c) 10 min, and (d) 34 min. Images are taken at height $z = 35$ $\mu$m below the top plate. In (d) the magnification is reduced to give a wider field of view. The inset in (d) shows the calculated Fourier transform of the image, demonstrating broad hexagonal ''lobes'' due to the polycrystalline particle ordering.

under shear often also demonstrates other more complex motion, for instance, apparent motion of planes in and out of the plane of focus (i.e., in the $z$ direction). In fact, given the degree of local disorder (dislocations, vacancies, stacking faults, etc.) we have observed, models of motion based on perfect hexagonal planes are likely to be too simple. Motion of a given plane does appear quite homogeneous and there is clearly some ''registered'' stacking of planes upon one another, though there seem also to be many stacking faults. The stacking of planes upon one another must play an important role in the plane motion under shear. Thus far we have not made a detailed study of the stacking of the shear-ordered planes. Meanwhile, more extensive microscopic study of plane motion is hampered by the necessity to observe slow-moving planes near the stationary bottom plate of the cell; motion very near the bottom plate may even be affected by the proximity of the surface. Studies of plane motion in the bulk at much lower strain (i.e., where velocities far above the bottom plate remain low enough for observation) are in progress.

### B. Low strain $\gamma < 1$

Next we discuss results obtained from experiments at *low strain* $\gamma < 1$. Figure 6 shows a sequence of images from an

experiment at $\Phi = 0.56$, with shear amplitude $A = 0.079 \pm 0.005$ mm, frequency $f = 0.625$ Hz, and plate separation $h = 0.149$ mm, giving $\gamma = 0.53 \pm 0.04$. As in the high-strain experiments, patches of hexagonal order can be seen within minutes [Fig. 6(b)], though at low strain there is now no clearly preferred orientation. As time progresses, more patches of order appear and existing patches grow larger [Fig. 6(c)] until the visible plane is filled. Unlike in the high-strain experiments the regions of order clearly have a distribution of different orientations. In fact, we observe a distribution apparently peaked with the close-packed direction *perpendicular* to the velocity axis [Fig. 6(d)], though orientations close to 30° outside this direction can occur. Note that, as stated above, similar order is visible at *all* heights within the cell, even at volume fractions within the equilibrium coexistence region. It has sometimes been postulated that shear-induced crystallization results from wall effects such as particle migration to walls or wall nucleation. We generally observe crystallization throughout the bulk of the sample (except for the coexistence samples at high strain as discussed in Sec. II A) and as far as we can tell ordering proceeds at the same rate at all heights. Furthermore, experiments with cells with plate separations up to 700 $\mu$m give qualitatively similar results [15].

Presumably, the low-strain shear generates particle collisions that are enough to encourage the particles to form layers in the shear plane and to begin to order, but not yet frequent or violent enough to ''melt'' nuclei whose orientations are reasonably close to but not necessarily exactly aligned with the shear direction. The inset in Fig. 6(d), the Fourier transform of the microscope image, compares well with scattering experiments [6,11]: We observe not sharp Bragg spots but spread-out ''lobes'' of intensity, consistent with a distribution of orientations of crystallites centred around a close-packed direction *perpendicular* to the shear direction (in contrast to high strain, when a close-packed direction is parallel to the shear direction). In our own scattering experiments [15] we have observed the angular spread of the scattering lobes to decrease as the experiment progresses while the intensity at the center of the lobe increases; this may be due to nucleation of more polycrystals with close-packed direction oriented more closely perpendicular to the velocity axis or possibly to ''annealing'' of already existing crystallites not exactly oriented. In some microscopy experiments we have observed a kind of ''flexing'' of crystallites, an apparent shifting motion of neighboring crystallites relative to each other in directions not parallel with the shear direction, facilitated by narrow bands of disorder at the border between the crystallites. Slow incorporation of separate crystallites into larger single crystallites has also sometimes been observed. However, a clear, general picture of the evolution of the large-scale crystallite structure is difficult to obtain both because we are not able to study a very large part of the sample (practical considerations mean we cannot move the shear cell laterally while under shear to look at different parts of the sample) and because microscopy gives a two-dimensional image and reconstruction of the three-dimensional large-scale structure can be a very complicated problem. For now we must therefore remain cautious about how the polycrystal structure evolves at later times. It is possible that at much later time the polycrystalline disorder observed in the low-strain experiments might be completely annealed out, leading to the formation of a macroscopic ''single crystal.'' Stacking faults might also be expected to disappear, given that the low-strain motion of particles in the troughs between particles in the plane beneath causes minimum disruption only in the absence of stacking faults. Thus we might expect the eventual generation of a perfect face-centered-cubic structure. However, observations have not been reported for times later than $\sim 40$ m after loading of the sample into the cell, since sedimentation of the particles may become significant at later times. Note that experiments using samples at different particle buoyancy (PMMA suspended in a mixture of *cis*-decalin and cycloheptyl bromide to vary the average solvent density with respect to the particle density) give qualitatively the same results, but a detailed examination of the effects of sedimentation has yet to be carried out. While a 1-$\mu$m-diam particle in a *dilute* sample would sediment appreciably over the time scale of our experiments, in concentrated suspensions sedimentation velocities are greatly reduced; in fact, observations of the concentration of particles at the top of the shear cell before and after shear experiments indicate that changes in concentration due to sedimentation of particles are not appreciable over the times we have discussed here.

## III. CONCLUSIONS

In conclusion, we have made direct optical microscope observations of shear-induced ordering in colloidal suspensions. Our findings are broadly consistent with models proposed on the basis of scattering experiments in cylindrical Couette and rotating disk geomoetries. Scattering experiments in the parallel-plate geometry [15] agree well with previous experiments in other geometries. We find that at low strain there is substantial polycrystallinity in the suspensions, explaining the disorder apparent in scattering patterns. At high strain, disorder in the orientation of crystals is far less evident and, macroscopically, the shear-induced crystal structure is closer to that of a single crystal. However, we do observe crystal planes to contain vacancies and dislocations and though we have not studied it in detail there are also stacking faults in the velocity gradient direction. Shear-induced crystal structures are thus locally more complex than assumed in models [10]. There is the possibility, however, that the crystals would evolve over much longer times to more perfect structures; experiments with buoyancy-matched particles are necessary before the long-time behavior can be studied.

The role of the underlying equilibrium structure of the suspension at high strain is not clear; there is evidence of some kind of coexistent order-disorder structure in the high-strain sheared suspensions at volume fractions within the equilibrium coexistence region. Finally, for samples at $\Phi \approx 0.59$, that is, samples in which the kinetic glass transition inhibits equilibrium crystallization [9], we find similar behavior to $\Phi = 0.56$: Crystallization is easily induced by the application of shear. The low-strain behavior of colloidal glasses and comparison with the behavior of ''soft'' glassy emulsions [17] are the subject of current work [15].

Models so far proposed for the structure of crystalline suspensions under shear start from the assumption of a single

well-ordered ''steady'' state; yet the mechanism of the generation of this order remains an important question. The nature of and interaction between two important effects, viz., the transition of the disordered fluid into well-defined layers in the shear plane and the nucleation of hexagonal crystals within these planes, are far from clear. Direct study of shear-induced crystallization phenomena in colloids by optical microscopy would appear to have great potential.

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