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Periodic structures in a colloidal crystal submitted to an oscillatory flow

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Abstract. — A colloidal crystal retained in a cylindrical tube and submitted to an oscillatory flow exhibits under certain conditions a spatially periodic pattern. Optical observations of the colored flecks show the existence of a continuous modulation of the orientation of the crystallites at the tube surface. It is due to the presence of a transverse component in the velocity field. We also present new experimental results concerning the wavelength of this pattern as a function of frequency and motion amplitude.

1. Introduction.

Colloidal suspensions have been widely studied these last years, especially monodisperse latex spheres suspended in water. Under appropriate conditions of ionic purity of the solvent, these particles may acquire an electric charge sufficient to make the system crystallize. The polystyrene balls are then regularly disposed in the liquid matrix so as to form a BCC or FCC lattice structure. Such systems are known as colloidal crystals [1, 2]. The lattice constant is typically equal to few particle diameters (about 0.1 μm). Thus one expects Bragg scattering of visible light. Moreover, colloidal crystals are characterized by an elastic modulus \( k \approx 10^2 \) dyne/cm² (this is very small compared to ordinary solids where \( k \approx 10^{11} \) dyne/cm²). This low value of \( k \) implies that hydrodynamic flows have a great influence on the arrangement of the polymeric particles. Many experiments using different geometries [1, 3-6] have revealed this coupling. In many of them, the suspension behaves as a classical liquid with a macroscopic viscosity \( \eta \) (about 0.1 Poise) depending on the shear rate. The effect of the flow is then to modify the structure built by the particles inside the suspension. A low shear rate induces orientation of the crystallites with respect to the velocity field. Higher shear rates
produce structural transitions finally leading to melting. A new type of behaviour was first reported by Dozier and Chaikin [7]. They studied the response to an oscillatory pressure of a colloidal crystal retained in a capillary. Under certain conditions, it exhibits periodic structures on the tube wall surrounding a plug flow in the core. The period is typically given by the tube diameter. The striking feature in this experiment is that the crystalline structure at the tube surface is not destroyed even at high shear rate.

In this paper, we present new results concerning this phenomenon. The experimental set up is described in section 2. In section 3, we report some qualitative observations which led us to analyse (Sect. 4) the outer pattern as due to a continuous periodic modulation of the orientation around its normal axis of the densest crystalline plane (parallel to the glass surface). Experimental measurements of the period of the structure are presented in section 5.

2. Experimental set up.

The experimental set up we have used is very similar to that of Dozier and Chaikin [7]. A colloidal crystal is poured in a U-shaped glass tube connected to an oscillatory pressure source via a plastic pipe (Fig. 1). Experiments have been performed with tube diameters in the range 1 to 5 mm. The suspension is retained in the horizontal part of the tube. This geometry allows observations through an optical microscope. The crystalline structure has been determined using Bragg scattering of white light. A lamp S$_1$ embodied in the microscope provides light parallel to the direction of observation. Lateral lighting is supplied by a mobile source S$_2$. The method we have used is quite similar to the Laue method in X-ray crystallography. The main difference is that, in our case, the direction of observation is fixed (microscope) whereas we can vary the direction of incident light. An external pressure source generates an oscillatory motion of the suspension. The driving amplitude of the motion is measured by the peak to peak displacement of the meniscus: it can be varied between 0.1 and 10 mm while the frequency lies in the range 1-50 Hz. The samples are suspensions of monodisperse polystyrene spheres [8]. Their diameter measured by electron micrography is 0.105 μm, their mean charge (3 500) has been measured by conductometric titration using sodium hydroxide. The volume fraction of the suspension is 3.5%. This corresponds to a mean inter-particle distance of

Fig. 1. — Experimental set-up. The colloidal crystal is retained in a U-shaped tube. A periodic pressure supplied by a piston driven with a loudspeaker is applied at one side of the tube. Optical observations are made through an optical microscope with use of two different light sources S$_1$, embodied in the microscope and S$_2$, external and mobile.
0.3 \mu m and to a density of 0.5 \times 10^{14} \text{particles/cm}^3. At this concentration, the crystallographic structure is BCC [1]. The elastic modulus \( k \) (40 dyne/cm\(^2\)) has been determined using the shear waves method [9]. Nevertheless one must be aware that the quality of the sample (elastic modulus, viscosity...) in the experimental tube is very sensitive to the cleaning treatment of the glass vessels. Another essential point in these experiments is the importance of the memory effect in the suspension. When the alternative pressure is applied to the suspension, the pattern which appears can depend strongly on the one previously established. Therefore to destroy any remanent structure, we induce the melting of the colloidal crystal by making it flow over several centimeters. This revealed essential to obtain reproducible results.

3. Qualitative observations.

When the colloidal crystal is at rest, we observe colored spots due to the Bragg scattering of light on crystallites randomly oriented in the sample. Moreover a red stripe in the direction of the tube axis indicates that one of the densest planes (1\(\bar{1}0\)) is parallel to the glass wall [1, 3, 4]. When oscillatory motion starts, the visual appearance of the suspension changes. After some time (from one minute up to several hours according to the amplitude and frequency of the flow), a steady state is reached. We observe the « checkerboard pattern » described in [7]. This pattern concerns only a thin ordered layer located on the glass wall. No motion is observed there. On the other hand, the inner core flows rigidly as visualized by the motion of small particles such as resin grains or colloid aggregates. When the motion is stopped (Fig. 2a), the checkerboard pattern still remains and crystallites appear in the core.

3.1 Description of the outer pattern. — As mentioned in section 2, we can use two different light sources \( S_1 \) and \( S_2 \). Observation is done under microscope. The lamp \( S_1 \), embodied in the microscope, allows observation with incident rays parallel to the reflected ones. The scattering wave vector lies therefore in this common direction. With this lamp, we observe two types of Bragg scattering :

- a central band (red) which is similar to the one observed at rest. This band is due to scattering on high density planes (1\(\bar{1}0\)) which stay parallel to the glass surface;
- bright red spots laying on two lines located on each side of the central band. They are periodically distributed along the tube. We observe two alternate types of spots (Fig. 2b), the shape of which changes when varying the focus of the microscope. This pattern defines the period \( \lambda \) characteristic of the phenomenon.

The second lamp \( S_2 \) can easily be moved. The aspect of the outer pattern depends on its position. If the scattering plane is perpendicular to the tube axis (Fig. 3a), we observe a picture similar to the one obtained with \( S_1 \), that is periodic points lying on each side of a continuous band of the same color. Nevertheless the whole picture has turned in the direction of the scattering vector and the colour has shifted from red to green. When the scattering plane is not perpendicular to the tube axis, we observe colored segments instead of the previously described spots as shown in figure 3b. Their color is also shifted toward green. An important feature of this checkerboard pattern is that a symmetrical position of lamp \( S_2 \) leads to a mirror image (Fig. 3b, c). Notice also that the observed pattern does not change when the tube is rotated around its axis whatsoever the lightning is. This proves that the structure responsible for the Bragg scattering is axially symmetrical.

3.2 Description of the core. — At low amplitude, the core remains crystallized. In these conditions, we observe in this region a transverse spatial undulation. This modulation is periodic with the same period as the outer pattern (Fig. 2c). It appears in the core region as
The suspension has been driven until an equilibrium state was reached. At the tube surface, crystallites have been oriented by the flow, giving rise to a periodic pattern. When the oscillatory motion is stopped, this pattern remains and randomly oriented crystallites appear in the tube core. The colored spots are due to Bragg scattering on these crystallites. This picture has been taken with various lighting. The first one, perpendicular to the tube axis (Fig. 3a) gives the red spots and the continuous red line. The presence of this line indicates that the densest planes near the tube surface are parallel to it. The white stripe is due to the reflection of the light on the glass tube. A side-lighting (Fig. 3b) provides the yellow-green segments located between pairs of red spots. A symmetrical lighting on the other side (Fig. 3c) would provide a symmetrical picture. This can be seen in this picture, where we have used a third lighting which is not exactly symmetrical with the second one and which gives the green segments. (b) A variation of focus enables for clearly distinguishing two types of reflection spots corresponding to the red spots of figure 1a. (c) In the core the sample often exhibits a periodic undulation, especially at low amplitude. Its wavelength is the same as that of the outer pattern. (d) Near the surface crystallites remain stuck to the glass wall while the inner core flows quasi rigidly. The dark lines, which appear at small amplitude, are due to dislocations, the motion of which allows the core displacement by plastic deformation at least at small amplitude.

proved by the following experiment. When we make the sample slowly flow in the tube, the checkerboard pattern remains « stuck » to the glass surface whereas the modulation is displaced with the colloid. This obviously shows that this phenomenon appears in the core region. When we stop the oscillatory pressure, the undulation remains still more visible. At higher amplitudes, the core melts and it is not clear whether this modulation persists or not. When small motions are produced, regularly spaced ridges appear in the surface layer (Fig. 2d). This indicates the existence of dislocation loops, the motion of which allows the core displacement by plastic deformation.
Fig. 3. — Aspect of the sample under different illumination conditions. (a) Incident and reflected wave vectors $K_0$ and $K_1$ are in a plane perpendicular to the tube axis. The system exhibits periodically spaced spots. (b) The plane defined by $K_0$ and $K_1$ is no longer perpendicular to the axis. One observes the « checkerboard pattern ». (c) A symmetrical lighting produces a symmetrical pattern.

4. Structural analysis.

4.1 LOCAL ORIENTATION OF THE COLLOIDAL CRYSTALS. — Let us now proceed with the analysis of the colloidal crystal structure in the vicinity of the tube wall. In this region, the sample remains crystallized as described in section 3.1. As experimentally shown by several authors [1, 3, 4], crystallites in the vicinity of the glass surface usually align one of their planes of highest density (say (110) for the BCC structure) parallel to the glass surface. This is also observed here. Indeed any sample illuminated with the lamp $S_1$ always exhibits a continuous Bragg reflection stripe along the tube axis revealing that (110) planes are precisely aligned parallel to the glass surface. But this does not determine completely the orientation of the core.

Fig. 4. — If the core flows exactly along the tube axis, it aligns the crystallites on the tube surface so as to present the densest lines [111] parallel to the axis. Perpendicularly illuminated, such a structure produces three continuous lines (a) due to the scattering of light on the planes $1\overline{1}0$, $01\overline{1}$, $10\overline{1}$, located at 60° from each other (b).
colloidal crystal: rotations in this plane are still allowed. As reported by many authors [1, 3-5], in driven colloidal crystals the densest lines, say [111], tend to lie in the direction of the flow. Let us follow this hypothesis and suppose that the velocity field is parallel to the tube axis. It orientates the crystallites which are stuck to the glass wall. Perpendicularly illuminated, such a structure produces three continuous reflection stripes parallel to the tube axis (Fig. 4a) and of the same color. The stripe at the top of the tube is due to the reflection on (110) planes as already mentioned. But there exist two other families of planes containing the [111] direction and equivalent to the (110) family: (101) and (011) (Fig. 4b). At 60° from apart the central stripe, one of these two families of planes is perpendicular to the scattering vector and gives rise to the same colored stripe. However, experimentally, the lateral stripes are not continuous but dotted. To explain this, let us now suppose that the [111] direction does not lie everywhere along the tube axis but oscillates periodically around this axis (Fig. 5a), the (101) plane remaining parallel to the glass surface. These oscillations generate the buckling of (011) and (101) planes illustrated in figure 5b. These planes give Bragg reflection only when their normal is parallel to the scattering vector. This produces regularly spaced spots as shown in figure 5c. Their shape is linked to the way the planes are curved. Thus a slight displacement of the microscope focus alters in a different way the shape of two neighbouring points. On the other hand, the symmetry around the tube axis implies that, if the (110) planes are curved for instance upwards on one side of the tube (producing a spot say A), the (011) planes are curved downwards on the other side and generate a spot B (Fig. 6).

Fig. 5. — If the core velocity has a periodic azimuthal component (a), the lines [111] will oscillate around the tube axis leading to the buckling of high density planes (b). Under perpendicular illumination, the structure exhibits Bragg spots at points A and B. The sample then looks like c. By slowly rotating the scattering plane around K (Fig. 3), one observes a continuous displacement of the Bragg spots A and B towards A' and B' on one side of the tube, and towards A'' and B'' on the other side, until points A' and B' (resp. A'' and B'') meet at point C' (resp. C'').
It is now easy to understand what happens with lateral lighting. The scattering vector $q$ is no longer vertical. Its orientation is determined by two angles $(\tau, \varphi)$. $\varphi$ is the angle between the scattering plane and the plane perpendicular to the tube axis. $\tau$ is the angle made by $q$ with the direction of observation (see Fig. 3). $\tau$ is also the incidence angle of the light on the scattering planes. On the other hand, the orientation of a crystallite is defined by $\alpha$, the azimuthal angle, and $\psi$, the angle between its [111] direction and the tube axis (Fig. 5). A straightforward calculation shows that (101) and (011) planes satisfy the Bragg condition only when

$$\sin \psi = s \frac{2}{\sqrt{3}} \sin \tau \sin \varphi$$

where $s = +1$ for (101) and $s = -1$ for (011) planes. For given $(\tau, \varphi)$, this relation is twice satisfied per period (see Fig. 5a), at points $A'$ and $B'$ for (101) planes and at $A''$ and $B''$ for (011) planes. Experimentally, we observe segments instead of spots since the plane curvature is small. Increasing incidence angle $\tau$ shifts their color from red to green. When $\tau$ and $\varphi$ increase, $A'$ and $B'$ ($A''$ and $B''$) become closer to each other until they meet at $C'$ (resp. $C''$). This proves that the observed pattern is really due to a continuous modulation of the crystallite orientation.

4.2 DOES THE STRUCTURE REMAIN BCC ?. — This analysis is based on a BCC structure. In the same experiment done with larger spheres (0.22 mm diameter), Dozier and Chaikin [7] observed that the shear transformed the crystal structure from its BCC configuration to a FCC one with laser Bragg scattering studies. They interpreted the checkerboard pattern as due to the existence of two different orientations in the tube. The 111 planes remained parallel to the glass surface (Fig. 7a). In one type of domain, the [211] direction was aligned along the flow direction whereas it was the [121] direction in the other domains. Notice that these two directions lie symmetrically apart an easy shear direction [110]. This implies that the orientation of the crystallites is perfectly determined at a given point along the axis and therefore, that there is no local twinning.

One could have thought that a change of the structure would induce a variation of the color of the main spots. Unfortunately this variation (3 %) is too small to be observed. Nevertheless we can show, using crystallographic arguments, that the checkerboard pattern we have observed does not correspond to the one described in [7]. Let us admit that the red spots observed on both sides of the tube are due to Bragg scattering on densest planes not parallel to the glass surface but containing the direction of the tube axis. Following this hypothesis, if a [211] row was aligned along this direction, we should not observe lateral spots with lamp
In a FCC colloidal crystal the high density planes which align parallel to the glass surface are the (111) planes. A projection of the structure along this axis gives a succession of A, B, C hexagonal layers (a). The high density line is along the [101] row. As shown in (b), there exists only one other high density plane (111) which also contains this line.

If it was a [110] row and if we assume that the crystallites have the same orientation all around the tube, we would observe only one lateral red spot due to the (111) planes (Fig. 7b). On the other side, there should appear a spot due to (002) planes but with a different color. The existence of two lateral spots should therefore imply the coexistence at the same place of ABC and ACB crystallites oriented with the [110] row along the tube axis. This is not in agreement with the Bragg scattering data reported by Dozier and Chaikin.

Let us now suppose that the structure remains BCC. Our observations are then still compatible with a BCC twinning where the twinning plane contains the dense row [111] and is perpendicular to the glass surface. One could imagine that, if a structural transition BCC-FCC occurs, BCC twins could give rise to FCC or HCP crystallites the orientation of which depends on the orientation of the twins. This has already been observed by Ackerson and Clark [3, 4]. If a structural transition takes place after the modulation in the orientation of the twins, this would lead to the checkerboard pattern described in [7]. This transition would not be observed in our samples made of small spheres. Dozier and Chaikin have used twice larger polystyrene spheres and it is well known that such suspensions give more easily an FCC structure than suspensions of smaller spheres. This suggests that the essential point in the phenomenon is not the change of structure but in both cases the variation in the orientation of the crystallites [3, 4].

5. Experimental study of the wavelength of the periodic pattern.

5.1 Conditions of Observation. — When pressure oscillations are induced in a randomly oriented suspension, its appearance changes until a steady state is established. The time necessary to reach this state varies from one minute to several hours for small amplitudes. It has been impossible to detect any threshold in amplitude needed for the pattern appearing since the establishment time becomes prohibitively long at small amplitudes. We have often
noticed that the periodic modulation appears first near the meniscus either at the open end or at the other one. Then it extends slowly inside the tube. Sometimes two patterns with different wavelengths are generated from the two surfaces of the suspension. Two evolutions may occur: either the extension of one pattern against the other or a sort of compromise (one period at one side and the other one at the other side). This explains why two points \( \lambda \) are sometimes plotted corresponding to identical other parameters (Fig. 8). However, in

\[ \begin{align*}
\lambda & \quad \text{II} \\
5 & \quad \text{III} \\
2 & \quad \text{IV}
\end{align*} \]

Fig. 8. — Experimental measurements of the wavelength \( \lambda \) of the periodic pattern as a function of the driving amplitude \( A \) for a given frequency \( f \) and for different tube sizes (radius \( R \)). It decreases by steps with increasing amplitude.
many experiments, the period $\lambda$ is homogeneous in a great part of the tube (usually the middle part). All the measurements indicated in this article correspond to this region. We have also noticed a possible slow evolution in time of the value of $\lambda$ for a given pattern. When the pattern is well established, we wait about ten minutes to check that the variation of $\lambda$ is weak enough. In this study, different parameters can be varied. The length $L$ of the latex sample plays no role in the problem. The wavelength $\lambda$ is the same when $L$ is 5 or 50 mm (if $\lambda$ is smaller than $L$). This is easily verified by creating bubbles which separate different volumes of suspension. We have used different tube sizes: $R = 0.6$, 1.25, 1.5, 2 mm. We estimate that their cleaning treatment is good enough if the latex leads to reproducible results over at least two days. The amplitude of the flow $A$ depends on the tension applied to the loudspeaker. It is measured using the peak to peak displacement of the meniscus which is directly proportional to the total flux. The error on its value can reach $5\%$ particularly at low amplitudes or low frequencies. We also verify that $A$ is constant during the formation of the checkerboard pattern. The maximum amplitude we obtain without destroying the loudspeaker is typically $A \approx 12$ mm for $R = 2$ mm.

5.2 EXPERIMENTAL RESULTS. — The most striking results are obtained by fixing the frequency $f$ and varying the amplitude $A$. Typical measurements are reported in figures 8a, b, c. The following features should be emphasized:

- the wavelength $\lambda$ decreases with increasing $A$;
- this variation occurs step by step;
- on each step $\lambda$ remains nearly constant.

When we change the tube size, we observe that the wavelength varies in the same way as the radius. For a given frequency $f$, the steps appear, for different tube radii $R$, at the same value of the amplitude $A$. The wavelength $\lambda$ on each step is roughly proportional to the radius $R$ (Fig. 9). The different values of $\lambda/R$ we obtain experimentally are $0.59 \pm 0.1$, $2R \leq 4 \text{ mm}$, $2R \leq 3 \text{ mm}$, $2R \leq 2.5 \text{ mm}$.

Fig. 9. — Plot of the ratio $\lambda/2R$ versus the driving amplitude $A$ at a given frequency $f$. In different tubes, the steps appear at the same value of $A$. The wavelength on each step is roughly proportional to the radius of the tube.

Fig. 10. — Variation of the wavelength $\lambda$ with the frequency $f$. It decreases with increasing $f$ but it is not clear whether it is done step by step or continuously.
1.25 ± 0.15, 2.5 ± 0.3, 3.85 ± 0.3. The error on the value of \( \lambda \) on each step prevents from finding a simple law relating the wavelength on neighbouring steps. Varying \( f \) is not a well controlled experiment since it is difficult to reproduce a given amplitude at different frequencies. However it clearly appears that \( \lambda \) decreases as \( f \) increases (Fig. 10). We are unable to decide whether \( f \) decreases continuously or by steps. Nevertheless we have noticed (Fig. 11) that, at the transition between two steps, the relevant variables \( A, f, \lambda, R \) verify the relation

\[
\frac{Af\lambda}{R} = \text{constant}
\]

where \( A \) and \( f \) are measured at the transition point between two steps. When \( \lambda \) is the wavelength associated with the lower level of the step, the constant is about 2 cm s\(^{-1}\). When it is taken at the higher step level, it is equal to 4 cm s\(^{-1}\). This can be compared to the velocity of the transverse elastic waves in the medium given by \( \sqrt{(k/\rho)} \) which is about 6 cm s\(^{-1}\).

Let us yet mention that a strong increase of \( A \) or \( f \) divides each period \( AB \) (see Sect. 4) into two (or even four) periods \( AB \rightarrow ABAB \). The new wavelength is then half of the preceding one. The elastic modulus of the colloidal crystal probably plays a role in the appearance of the periodic pattern. This is not easy to verify since this elastic modulus should be measured when the suspension is inside the tube. Nevertheless we have observed lower values of the wavelength in a sample kept in the tube for few days. This is probably due to contamination of the suspension by the glass surfaces or by air. It could induce a decrease of the elastic modulus and therefore lead to lower values of \( \lambda \).

![Graph](image)

Fig. 11. — The steps appear for values of \( Af \) such as \( \lambda/R = C(Af)^{-1} \). The value of the constant \( C \) is 4 cm s\(^{-1}\) when \( \lambda \) is taken at the top of the steps, it is about 2 cm s\(^{-1}\) for the bottom of the steps.

6. Discussion.

In this article we report some new observations and measurements on colloidal crystals submitted to oscillatory motion in a capillary. Such a system exhibits a spatial periodical pattern. The study of the flecks of visible light has revealed to be an efficient and flexible method to determine the local orientation of the crystallites at the tube surface and even to confirm their structures. It has clearly appeared that the periodic pattern is due to a continuous modulation of the orientation of the colloidal crystal near the tube surface. This modulation is linked to the existence of a transverse component of the velocity in the core. The step by step decrease of the wavelength \( \lambda \) with increasing driving amplitude shows the non-linear feature of the origin of this pattern. The plug flow observed in this experiment is
not particular to colloidal crystals. Indeed a classical Newtonian fluid submitted to an oscillatory Poiseuille flow presents such a flow characterized by a penetration depth:

$$\delta = \frac{2 \mu}{2 \pi f}$$

where \( \mu \) is the kinematic viscosity of the fluid. For pure water, at \( f = 5 \) Hz, this length is about \( 300 \mu m \). It gives the size of the region in the fluid which separates the plug flow in the core and the tube surface.

For colloidal suspensions, the checkerboard pattern paradoxically appears in the boundary region where the shear rate is maximum (up to \( 1000 \) s\(^{-1} \)). This is possible if the boundary region is divided into two parts. A crown of crystallised suspension remains stuck at the tube surface with nearly no shear stress. The intermediate region between this crown and the core is submitted to high shear stress. At low driving amplitude the presence of dislocation walls reveals a plastic flow in this region. In the core crystallites are still visible. If the amplitude is too high the suspension probably becomes disordered in the intermediate region.

The periodic pattern at the tube surface as well as the periodic undulation of the core are probably due to the existence of a transverse component in the velocity field. Nevertheless transverse waves can occur in a viscous fluid but they are rapidly damped in the fluid over a distance \( \delta \). In colloidal crystals, this experiment proves that such waves can propagate especially for high longitudinal velocity. The main macroscopic difference between colloidal crystals and water is the visco-elasticity of this first medium. Therefore any theoretical approach of this effect [10] and particularly the behaviour of the period \( \lambda \) requires an analytical study of the non-linear elasto-hydrodynamics equations for such systems [11].

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