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Taylor instabilities in colloidal crystals

M. Joanicot and P. Pieranski

Laboratoire de Physique des Solides, Bât. 510, Université de Paris-Sud, 91405 Orsay Cedex, France

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Résumé. — Nous avons développé récemment un viscosimètre de Couette pour l'étude des cristaux colloïdaux sous cisaillement. Nous avons observé que, dans un premier temps, lors de l'accroissement du cisaillement, le cristal fond, puis, au-dessus d'un seuil critique se développent les instabilités de Taylor. Après l'arrêt de l'écoulement, la suspension recristallise en gardant mémoire des rouleaux de Taylor et ceci détermine la configuration de l'instabilité de Taylor lors de l'expérience suivante.

Abstract. — Recently we have developed a Couette viscosimeter for studying the shearing of colloidal crystals. We report here that with increasing shear rate the colloidal crystal melts (for ω_{crit}) and above a critical shear rate, ω_{TO} , Taylor instabilities develop. After stopping the shearing flow the suspension recrystallizes in a special texture which preserves the pattern of the Taylor rolls. This texture determines the configuration of Taylor instabilities in subsequent flow experiments.

1. Introduction.

Synthetic colloidal particles of polystyrene arranged in a crystal lattice are characterized by very low shear elastic modulus of the order of a few dyns per square centimeter [1]. For this reason the stress necessary to induce elastic deformations, creep flow, or melting is very low and can be obtained using a simple Couette viscosimeter [2].

Schematically, this viscosimeter is made of two coaxial glass cylinders of respective radii, $R_1 = 2$ cm and $R_2 = 3$ cm. The internal cylinder can rotate with an angular velocity ω which ranges between 0.01 tr/s and 5 tr/s. The corresponding torque, $\Gamma(\omega)$, is measured on the external cylinder which is suspended on pneumatic bearings. A typical stress-strain plot obtained with such an experimental set-up is shown in figure 1. We can distinguish between three different regions (Fig. 1) corresponding to :

- (a to b) plastic flow of the crystal,
- (b) melting of the crystal on the internal cylinder for ω_{crit} ,
- (b to c) coexistence of two phases; a melted phase and a plastic phase,
- (c to d) the flow of a completely melted suspension.

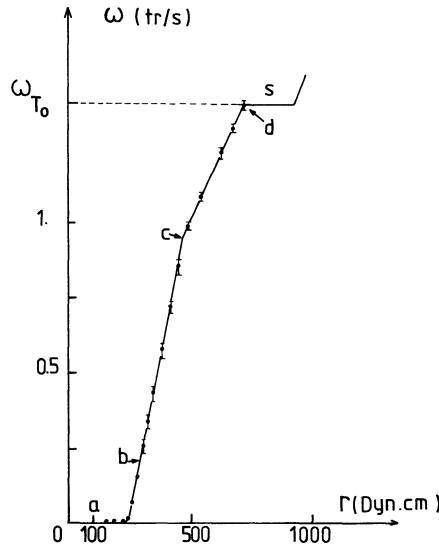


Fig. 1. — Stress-strain plot of colloidal crystals obtained with a Couette viscosimeter shows three different behaviours of sheared colloidal crystals : (a-b) plastic flow of the crystal ; (b-c) coexistence of two phases : a melted phase and a plastic phase ; (c-d) the flow of a completely melted suspension.

In the third region the crystal structure is lost and the suspension for this range of shear behaves like a Newtonian liquid of high viscosity, $\mu \approx 0.1$ poise.

2. Results and discussion.

2.1 THRESHOLD OF INSTABILITY. — Above a critical shear rate, ω_{T_0} , we observed the onset of Taylor instabilities which can be simply detected by observation of spiral trajectories of ion exchange resin grains carried within the suspension. The threshold of the instabilities corresponds to a step (s) in the last part of the stress-strain curve (Fig. 1) which corresponds to increased dissipation due to convection of momentum by rolls.

The transition to instability is also revealed by the appearance of narrow and ring shaped crystalline regions. These are disposed periodically along the tube. These rings correspond to regions where the flow converges between two rolls. The suspension recrystallizes in these regions because the shear rate drops below the critical shear value, S_{crit} , due to this locally convergent flow.

The critical experimentally measured angular velocity $\omega_{T_0} = 1.6$ Hz, is in good agreement with an estimate obtained numerically [3] for the case of a narrow gap between the cylinders :

$$\omega_{T_0 \text{ theor}} = \frac{41.3 \mu}{h \sqrt{R_2 h}} \approx 1.7 \pm 0.2 \text{ Hz}$$

where μ is the viscosity of the melted suspension determined from figure 1 ≈ 0.07 poise

$$h = R_2 - R_1 = 1 \text{ cm}$$

$$R_2 = 3 \text{ cm}.$$

2.2 MEMORY FLOW. — Figure 3 shows the current lines wound as tori which are piled up along the tube. The winding direction is reversed in adjacent tori. When the flow is stopped the melted suspension recrystallizes in a special texture which preserves the pattern of the periodic structures created by the flow. The orientation of crystallites in the recrystallized texture, as revealed by Bragg reflection of light, corresponds to the pattern of Taylor instabilities (Fig. 2).

This effect of crystal orientation due to shear flow is similar to previous observations of different flow geometries [4].

2.3 NEW INSTABILITIES. — When the colloidal crystal is subjected to a shear flow (induced by the rotation of the internal cylinder with an angular velocity ω_i larger than ω_{T_0}) the crystal melts. Also Taylor instabilities develop once again. We have observed, however, that the pattern of Taylor rolls in all (i -th) experiments depended on the value ω_{i-1} of the angular velocity determined in the previous experiment. Two different behaviours were observed following the value of ω_{i-1} with respect to the critical angular velocity ω_{T_1} ($> \omega_{T_0}$).

For $\omega_{T_0} < \omega_{i-1} < \omega_{T_1}$, the flow patterns in the i -th and ($i - 1$)-th experiments were the same, while for $\omega_{i-1} > \omega_{T_1}$, the i -th pattern of Taylor rolls was shifted by $\lambda/2$ with respect to the previous one ($i - 1$) (Fig. 3).

This unusual correlation between two patterns in two successive shear flow experiments attracted our attention to the intermediate texture of the colloidal crystal sample which was allowed to recrystallize between two flows.

Thus we considered in more detail the orientation of colloidal crystal by flow. First, it has been shown that bcc colloidal crystals are preferentially oriented with their densest plane (1, 1, 0) parallel to the container glass walls. The degeneracy with respect to the rotation around the

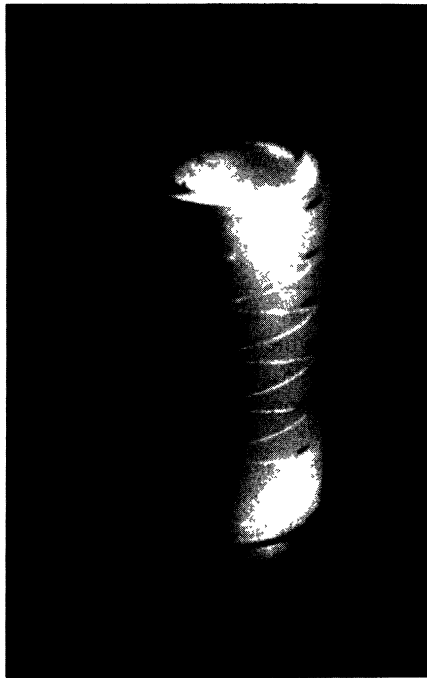


Fig. 2. — The Bragg reflection pattern of a recrystallized texture after a Taylor instability reveals the periodicity of this instability along the axis of the cylinders.

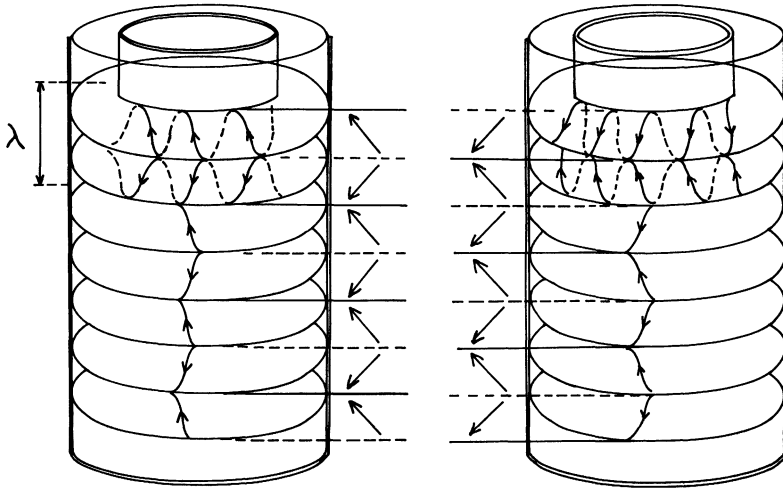


Fig. 3. — Pattern of Taylor instabilities in the $(i - 1)$ -th and i -th experiment, respectively, above the second critical angular velocity ω_{T_1} . Between the two patterns the rolls are shifted by $\lambda/2$, which is equivalent to an inversion of the winding direction of flow lines around the rolls as is shown by the arrows which indicate the direction of flow on the cylinder surfaces.

$[1, 1, 0]$ direction (perpendicular to the glass wall), is removed by shear flow. The $(1, 1, 0)$ crystal planes tend to orient with their $[1, 1, 1]$ rows parallel to the direction of the previous flow.

We have considered three typical experiments corresponding to different angular velocities ω_{i-1} :

- 1) $\omega_{i-1}^1 < \omega_{T_0}$,
- 2) $\omega_{T_0} < \omega_{i-1}^2 < \omega_{T_1}$,
- 3) $\omega_{i-1}^3 > \omega_{T_1}$.

The flow patterns are different in all these cases. Close to the container walls the velocity v will have mainly two components : a tangential one, v_θ , which, below the value ω_{T_0} , increases linearly with ω and the vertical one v_z , which only develops above the threshold (as shown in Fig. 4a). In the pattern of Taylor instabilities, v_z is a function of z and changes sign between two rolls wound in opposite directions. In one roll this component can be characterized by some average value, \bar{v}_z .

As a result, in one roll the average direction of the flow makes an angle ψ with the θ -axis and increases with ω above ω_{T_0} (Fig. 4b).

The texture of samples oriented by such flows was different in all three cases as evidenced in figure 4c. In bcc crystals, slip due to a shear stress occurs along $[1, 1, 1]$ axes. Therefore when such oriented samples are submitted to a tangential shear stress ($\Gamma_{\theta R}$), the initial plastic flow will occur along the $[1, 1, 1]$ direction which is different in each case :

— in the first case the slip direction (S_1) coincides with the direction of the stress ($\Gamma_{\theta R}$) (Fig. 4d(1)),

— in two other cases the slip will take place obliquely along the direction S_2 or S_3 , which corresponds to the largest component of stress in the $[1, 1, 1]$ directions (Figs. 4d(2) and 4d(3)).

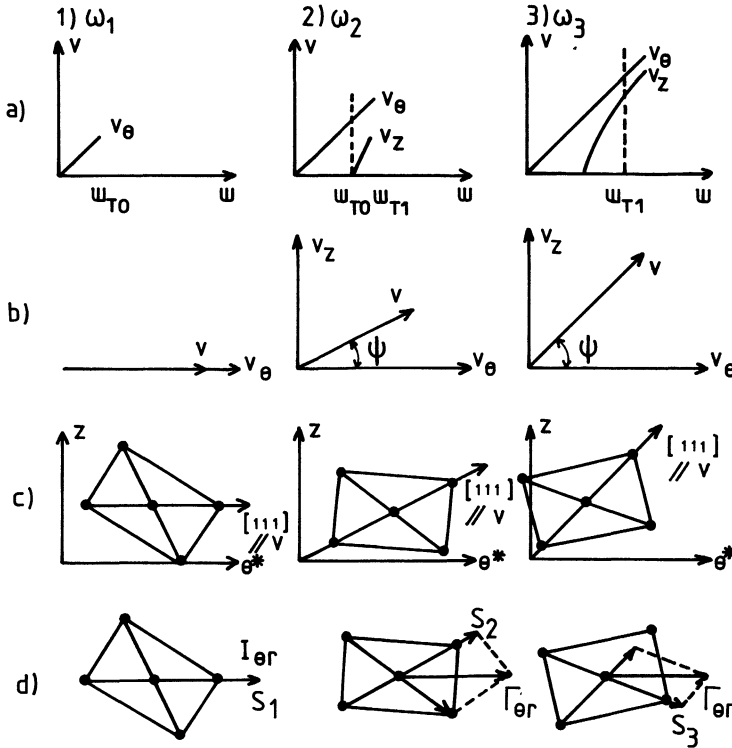


Fig. 4. — 1), 2) and 3) represent three different experiments with angular velocities ω_1 , ω_2 and ω_3 , respectively, in each experiment : (a) represents the relative compound of the flow velocity ; (b) shows the direction of flow on the surface of the cylinder ; (c) shows the orientation of the crystallites ; (d) shows the effect of stress on the texture due to rotation of the internal cylinder.

As the development of Taylor instabilities is very sensitive to fluctuations of V_θ , the roll pattern will follow preferentially the direction of the initial slip. As shown in figure 4d(2), the larger component of stress will induce the flow in the same direction (S_2) as the $(i - 1)$ -th one, but in the third case, the preferential direction of slip (S_3) will induce a flow which is wound in the opposite direction (Fig. 4d(3)).

In the third case, this exchange of winding directions in each torus looks like a shifting of the whole pattern by $\lambda/2$, as mentioned in the previous section (Fig. 2).

3. Conclusions.

In conclusion, in the present work we have reported new experiments on colloidal crystals subjected to large shear forces. We have shown that with increasing shear, plastic flow occurs first in the crystal, then above a first threshold ω_{crit} , the crystal melts and finally above a second threshold, ω_{T0} , Taylor instabilities develop in the melted crystal. These transitions, visible directly, have been shown to correspond to singularities on the stress-strain plot. We have also shown that the polycrystalline texture of a sample, allowed to recrystallize after a shear induced melting, keeps the pattern of the flow.

The melting of the crystal under shear and the orientation of recrystallized patterns are in agreement with similar phenomena observed in other geometries [5].

We report here, for the first time, that when Taylor rolls existed during one flow experiment, the recrystallized patterns is also periodic and in the next flow experiment it will influence the establishment of Taylor rolls.

Due to this effect the system of Taylor rolls can be shifted by $\lambda/2$ with respect to the previous one. This phenomenon was shown to occur above another threshold ω_{T_1} ($> \omega_{T_0}$). The nature of this third transition was qualitatively explained as due to the effect of previous patterns held by the « experienced » crystal.

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