Supporting information for: Dynamics of crystal structure formation in spincoated colloidal films

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Experiment videos

Video1.avi: experiment that demonstrates the transient dynamics on the long-range orientational order (configuration A of Figure 1):

1) pipeting suspension and von-Kármán spirals (*frames 1-101*)

2) 6-fold symmetry (*frames 101-308 - Phase I in text*)

3) inner transition from 6- to 4-fold (*frames 320-340 - Phase II in text*)

4) remaining outer 6-fold (*frames 340-390 - Phase II in text*)

5) inner 4-fold and 4-fold shrinking (*frames 340-480 - Phase II in text*)

6) loss of symmetry in fluid phase (> *frame 400 - Phase III in text*)

7) drying (> *frame 440 - Phase IV in text*)

Note: image contrast enhanced to properly visualize frames 340-440

Video2.avi: thinning dynamics in a similar experiment (configuration B of Figure 1):

1) pipeting suspension and von-Kármán spirals (*frames 1-90*)

2) asymmetry interference fringes (dissipation of pouring induced perturbation, *frames 90-115 - Phase I in text*)

3) symmetric concentric interference rings (*frames 115-262 - Phase I, II and III in text*)

4) drying (> *frame 235 - Phase IV in text*)

Both videos are jpeg compressed avi files at 25 frames per second (original frame rate: 1000fps). The resolution was reduced to yield files of suitable size.

Methodology for determining thinning rates from interference fringes (Figure 3(b))

Points were determined assuming for the suspension a mean refractive index of 1.42 ($\langle n_{\text{eff}} \rangle$). Standard Newton rings calculation gives a thickness change between consecutive rings (d) of:

$$d = \frac{\lambda}{2\langle n_{\text{eff}} \rangle} = \frac{540 \pm 40nm}{2 \times 1.42} = 190 \pm 14nm \tag{1}$$

For the calculation of thinning rates, the frequency at which these Newton rings pass through a particular radial distance was measured. This was done by overlapping a curve to a particular bright ring at a radius of interest. Then, the curve was fixed and the time until the next ring reached the curve was taken as the period. The inverse of this period multiplied by d gave the thinning rates.

Methodology for extracting dynamic thickness profiles (Figure 5(b))

In Figure 5 (b) the thickness profiles in the fluid phase permits the visualization of the two stages of the thinning process. For this, the ring (bright and dark) radial position was determined taking individual frames. Each ring has a difference in thickness of d/2: this gives the spatial gradient in thickness but not the absolute thickness. To obtain the absolute thickness, the situation when the outer part was dried and the inner part still presented interference rings was considered. In this case, a generalized bell function (h=$A+B/(1+(C/\rho)^2)$) was fitted to the points measured in the fluid region but forcing h to match the dried thickness (obtained experimentally *via* AFM) far from the center. The matching of static (dried) thickness and dynamic (fluid) thickness was only possible for a range of offsets to the fluid thickness, and the fluid absolute thickness was determined up to $\pm 0.3 \mu m$.