# Supplementary information
# Gravity-induced phase phenomena in plate-rod colloidal mixtures

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## I. SUPPLEMENTARY NOTE 1: BIFURCATION OF STACKING SEQUENCES

We discuss here the analogue to thermodynamic bulk coexistence for the stacking diagram. Two phases coexist in bulk along a binodal line and three phases coexist at a triple point. Fine tuning the interparticle potential, e.g. changing the colloidal shape in hard models, it is even possible to find higher order points at which more than three phases coexist in binary systems [1, 2]. In the stacking diagram, the analogue of bulk two-phase coexistence occurs along any of the boundary lines of the stacking diagram. At these lines, and within our LDA approach, one stacking sequence bifurcates from another one. That is, one layer of the sequence either starts to grow or vanishes when the boundary line is crossed in the stacking diagram (depending on which direction the line is crossed).

There are points in the stacking diagram at which three sequences bifurcate. These points arise from paths that are tangent to a bulk binodal and that simultaneously start or end at the point of tangency. At these points a sedimentation binodal of type II (paths tangent to a bulk binodal) bifurcates from a sedimentation binodal of type I (paths starting or ending at the bulk binodal). These points are marked by blue triangles in the stacking diagrams of Fig. 6 of the main text.

Bifurcation of four stacking sequences occurs at two types of special points in the stacking diagram. First, there are points where three sedimentation binodals and a terminal line meet. This occurs whenever a path either starts or ends at the triple point, which is always possible if a triple point exists in bulk, see the yellow squares in the stacking diagrams of Figs. 4 and 6 of the main text. The second special point occurs if two sedimentation binodals cross each other (violet squares in the stacking diagrams of Figs. 4 and 6 of the main text), which happens whenever a path simultaneously starts and ends at bulk binodals in the bulk phase diagram. See examples of such paths in Fig. 6b of the main text. Hence, the occurrence of these points highly depends on the topology of the bulk phase diagram, the precise curvature of the bulk binodals, and both the slope and the length of the sedimentation path. In the present system the number of such special points depends on the slope of the path. For $s = 3$ two sedimentation binodals cross at a point only once vs twice for the case $s = 0.5$. Due to the decreased slope of $s = 0.5$ and the curvature of the binodal there is an additional special point generated by the path that both start and end at the $I - N_\mathrm{p}$ binodal in bulk, see Fig. 6b of the main text.

Five sequences bifurcating from one point is also a possible scenario for finely tuned combinations of the sample height and the slope of the path. For example, for a specific value of the height $h \lesssim 50\,\mathrm{mm}$ and the slope $s = 0.5$ the sequence $IN_\mathrm{p}IN_\mathrm{r}$ in Fig. 6f of the main text disappears, leading to the bifurcation of five stacking sequences. This highly special point corresponds to the path that is tangent to the $I - N_\mathrm{p}$ binodal and that both starts at the $I - N_\mathrm{p}$ binodal and ends at the $I - N_\mathrm{r}$ binodal. In general, bifurcation of six stacking sequences from a point in the stacking diagram is also possible if three boundary lines intersect in the stacking diagram.

According to our local density approximation, the bifurcation of a sequence into another sequence occurs via the formation of a new infinitely thin layer. However, surface effects might prevent the formation of such a layer due to the energetic cost of the new interfaces that must be formed. Considering surface effects two scenarios are possible: The creation of a new layer can proceed continuously (via the formation of microdroplets instead of a complete layer) or discontinuously (a complete new layer appears abruptly once the thickness is large enough to compensate for the surface tensions of the associated interfaces). A set of sedimentation-diffusion-equilibrium experiments performed near the bifurcation points or boundary lines of the stacking diagram would be very valuable to shed light on this fundamental process. Also, a study of the time evolution from an initially homogeneous sample to the final equilibrium sequence can help to understand how stacking sequences evolves into each other.

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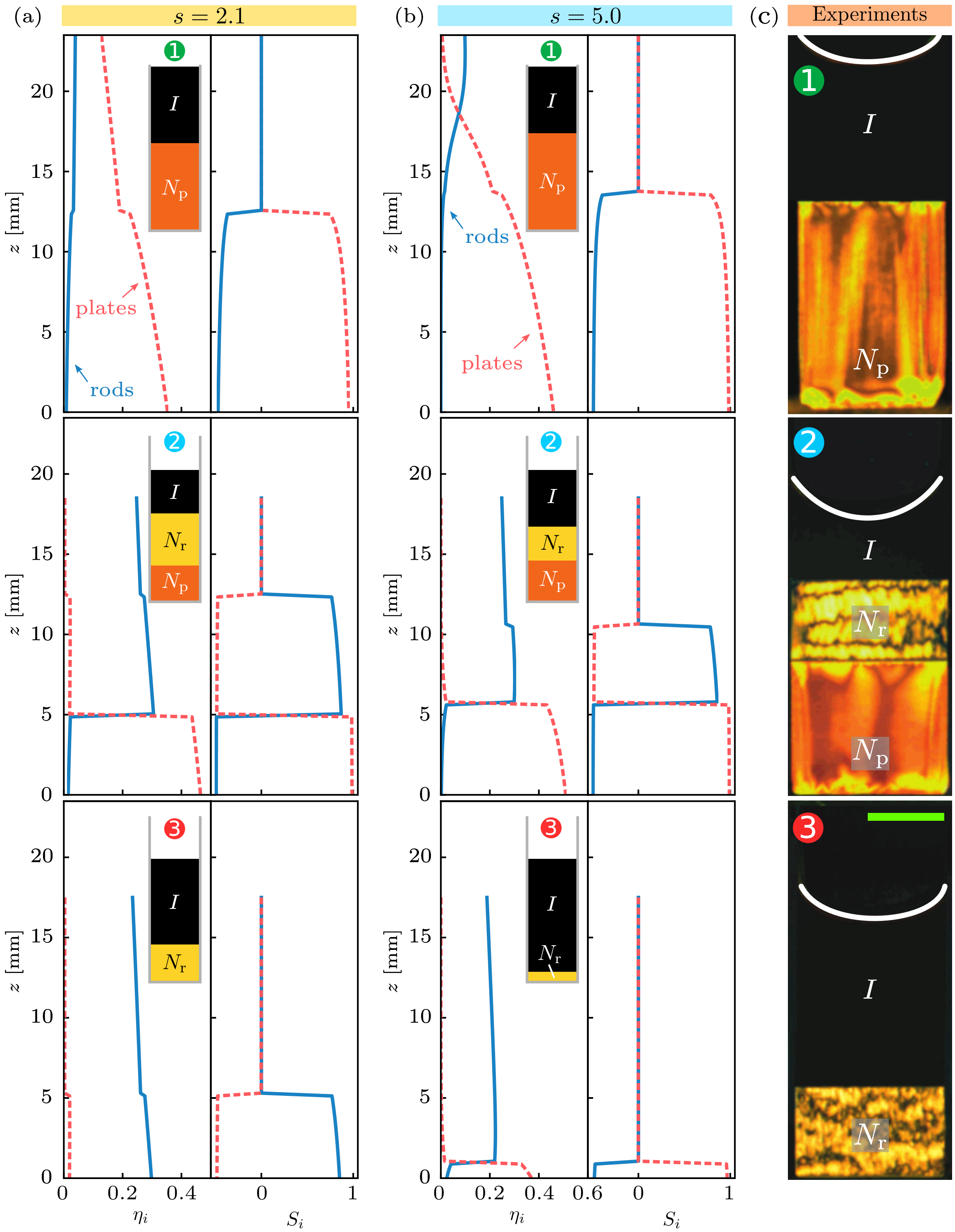


Supplementary Figure 1. Vertical profiles of the packing fractions $\eta_i$ and the nematic order parameters $S_i$ (with $i = \{\mathrm{r}, \mathrm{p}\}$) for rods (solid-blue lines) and plates (dashed-red lines) in three different samples, labeled as 1,2, and 3 (as indicated by the colored circles). The layers are isotropic ($I$), rod-rich nematic ($N_\mathrm{r}$), and plate-rich nematic ($N_\mathrm{p}$). The buoyant mass ratio is fixed to $s = 2.1$ in (a) and $s = 5.0$ in (b) which are the limiting cases of the experimental uncertainty. In both cases we change $s$ by varying the gravitational length of the plates $\xi_\mathrm{p}$ while keeping that of the rods constant, $\xi_\mathrm{r} = 5.5\,\mathrm{mm}$. Schematics of the samples are represented in the insets. The amount of solvent evaporated (only parameter used to find the best agreement between theory and experiments) is 22% (a) and 30% (b) in sample 1, 48% (a) and 47% (b) in sample 2, 60% (a) and 50% (b) in sample 3. The corresponding experimental samples (pictures taken between crossed polarizers) by van der Kooij and Lekkerkerker [3, 4] are shown in (c) (adapted with permission from [4], Copyright 2000 American Chemical Society). The meniscus is highlighted with a white line for clarity. The bright areas are due to light being polarized in layers with orientational order. The scale bar (green) is 5 mm.