Supplementary Information

Observation of Isotropic-Isotropic Demixing in Colloidal Platelet-Sphere Mixtures

Mingfeng Chen[1], Huawei Li[1], Ying Chen[1], Andres Mejia[2,3], Xuezhen Wang[2,3], Zhengdong Cheng* [2,3,4,]

[1] Soft Matter Center, Guangdong Provincial Key Laboratory on Functional Soft Condensed Matter, School of Materials and Energy, Guangdong University of Technology, Guangzhou, 510006, China

[2] Artie McFerrin Department of Chemical Engineering, Texas A&M University, College Station, TX, 77843-3122, USA.

[3] Mary Kay O'Connor Process Safety Center, Artie McFerrin Department of Chemical Engineering, Texas A&M University, College Station, TX, 77843-3122, USA

[4] Materials Science and Engineering, Texas A&M University, College Station, TX, 77843-3003, USA

The observation of different phases by SEM

As shows in Figure S1, SEM graph of different phases were obtained. Samples were taken out by syringe from the vial. Fig.S1 (a), (b) and (c) correspond to I_2, N and I_1, respectively, of the right vial shown in Fig 3b. The platelets were stacked due to drying. However, we can still notice that the I_2 phase has lesser proportion of silica spheres than either the N phase or I_1 phase, and the I_1 phase has the most of silica spheres among these three phases.

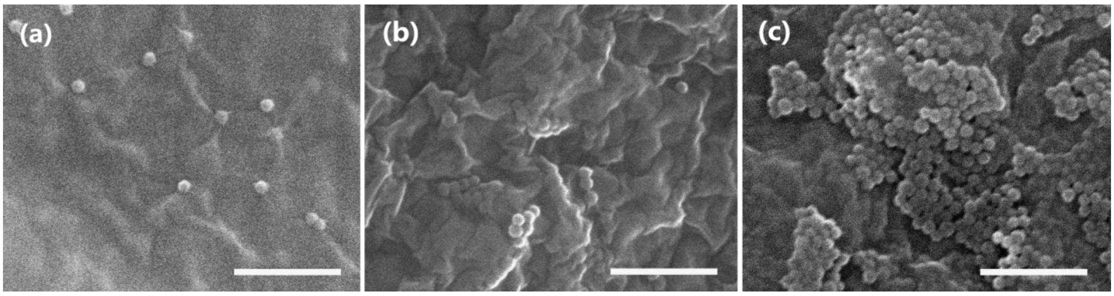

Fig.S1 SEM graph of different phases. (a) I_2 phase, (b) N pahse, and (c) I_1 phase. The scale bars in (a) (b) and (c) are 1 μm.

The influence of platelet's polydispersity on phase transition in pure platelet suspensions

Fig S2 presents the influence of platelet's polydispersity on I-N transition in a pure platelet system, which were obtained by Mejia.[1] It was found that for ZrP (TBA) system, the polydispersity causes the increasing of the I-N transition concentration and broaden the coexistence region of the transition (the polydispersity of ZrP platelets we used is 0.13 with an aspect ratio (L/D) at 0.0021). In consistence with previous data, we obtained an I-N transition at $\Phi_{I\text{-}N} = 0.0061 \backsim 0.0088$ for the pure ZrP platelets with a polydispersity of 0.13, which matched with the red dash curve in Fig. S2. However, for the platelet-sphere mixtures, neither theory nor systematic experimental study of the polydispersity influence is available yet.

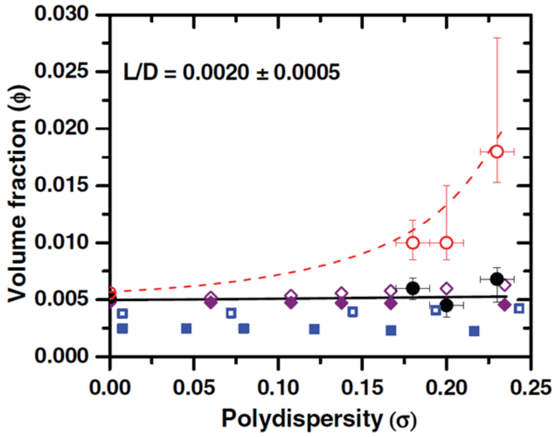

Fig. S2. (Color online) Volume fraction (Φ) (the solid black circles: isotropic and open red-gray circles: nematic) -of the cloud phases as a function of polydispersity (σ) at fixed aspect ratios of ZrP around 0.0020±0.0005. The concentrations at zero polydispersity for both graphs were obtained from simulations of Fartaria and Sweatman. The lines are guides for the eye. The solid and open blue-black squares are the theoretical ΦI and ΦN, respectively, from Sun et al. using a DFT calculation with Zwanzig approximation. The solid and the open purple-black diamonds are the theoretical ΦI and ΦN, respectively, from Bates and Frenkel for infinitely thin plates.

The observation of N-N (nematic-nematic) demixing

The phase transitions in mixtures with a size ratio of spheres over platelet at 0.013 were also observed. Increasing the concentration of silica to a relative high value, a nematic-nematic demixing occurred. Fig. S3(a) shows two series of samples with N-N demixing when silica sphere concentration reached 1.78 and 2.00 wt%. The different phases were indicated in Fig. S3(b).

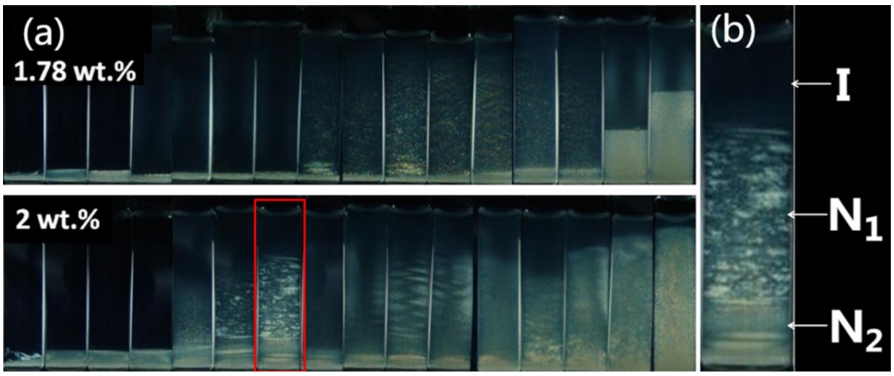

Fig.S3 (a) Photographs of ZrP/silica suspensions between crossed polarizers. From left to right, ZrP volume fraction is 0.0009, 0.0018, 0.0027, 0.0036, 0.0045, 0.0054, 0.0063, 0.0070, 0.0081, 0.009, 0.001, 0.0108, 0.0117, 0.0126, 0.0153, and 0.018, respectively. The error bar is ±0.00011. From top to bottom, silica concentration corresponds to 1.78, and 2.00wt.%, respectively. The error bar is ±0.003. (b) Labeling of the different phases. The volume fraction of ZrP and silica concentration of the sample are 0.0063 and 2.00wt.% respectively.

For a further analysis, N_1 was visualized under an optical microscope coupled with crossed polarizers and compared with N_2. The texture of N_1 observed between cross polarizers displays dark brushes having irregular curved shapes corresponding to extinction positions of the nematic liquid crystals. Disclinations of s = ± 1/2 and s = ± 1 are observable (Fig. S4a). The texture of N_2 displays a uniform domain and strong birefringence (Fig.S4b). As predicted by Lekkerkerker et al.,[2] the N_1-N_2 coexistence is originated from a competition among translational and orientational entropy (favoring mixed state) and depletion interaction (favoring demixing). N1 is a phase characterized of smaller liquid crystal domains in contrast to N2, which is a birefringent closed-packed phase.

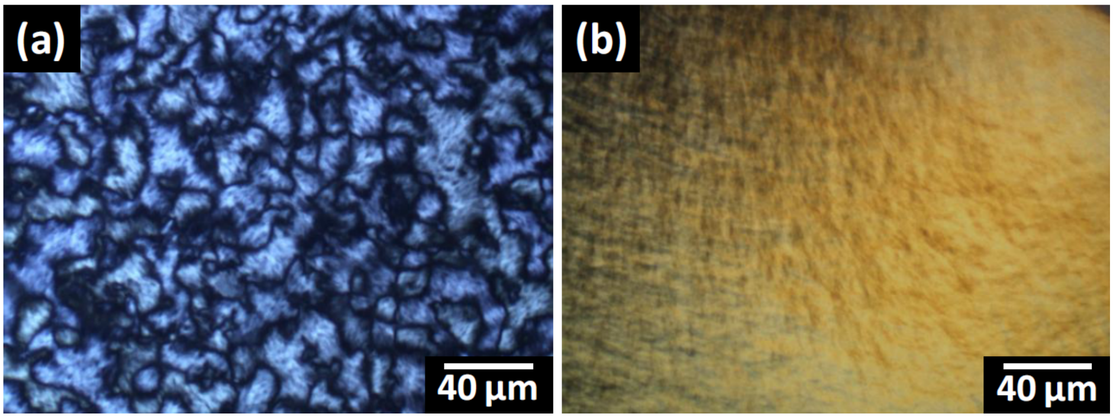

Fig. S4 Cross-polarized optical microscopic images of nematic Schlieren textures (a) N_1, ϕ_{plate} = 0.0081 (b) N_2, ϕ_{plate} = 0.018. Both silica concentration of the samples are 1.78 wt.%. The micrographs were taken using a Nikon microscope TE-2000U with crossed polarizers and 4× magnification.

References

[1] A. F. Mejia, Y.-W. Chang, R. Ng, M. Shuai, M. S. Mannan and Z. Cheng, Physical Review E, 2012, 85, 061708.

[2]F. M. van der Kooij and H. N. W. Lekkerkerker, Physical Review Letters, 2000, 84, 781-784