

**Supporting information**

**Shear-induced ordering of micellar arrays in the presence of  
single-walled carbon nanotubes**

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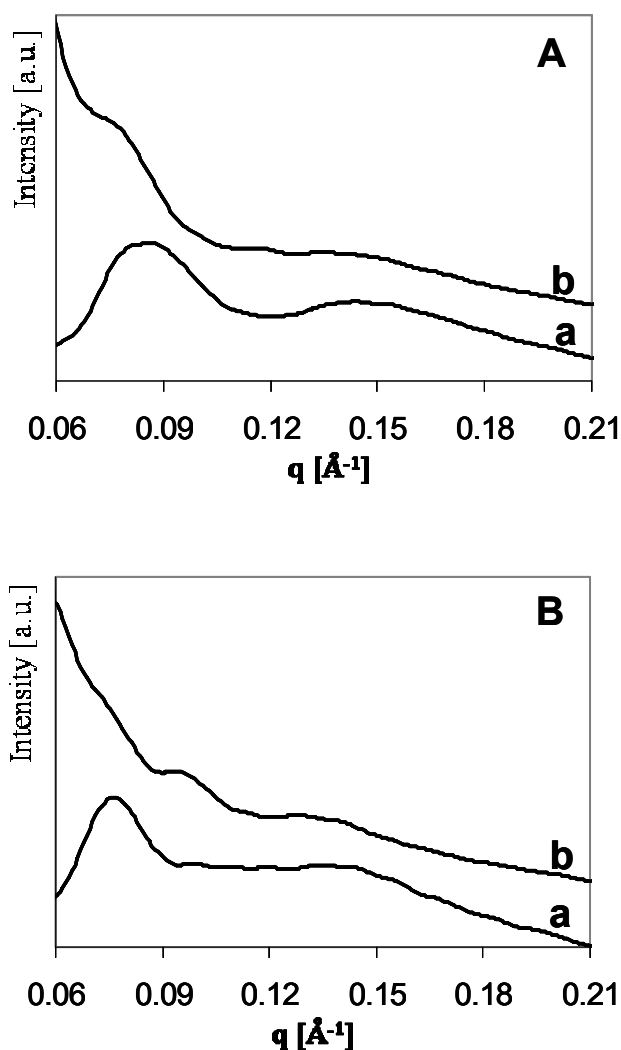
*Experimental*

Aqueous solutions of cetyl trimethyl ammonium bromide (CTAB) in concentrations of 5-20 wt% (CTAB in water) were prepared by dissolving the surfactant in water (Millipore water, resistance of 18.2 MΩ cm) at room temperature (25 °C). The solutions were incubated for at least two days before being used.

Liquid dispersions of single walled carbon nanotubes (SWNT), multi walled carbon nanotubes (MWNT) and carbon black (CB) were prepared by sonicating the powder of the raw material (typical concentration of 0.5-1.0 wt%) at very mild conditions (mini supersonic cleaner, Delta DG-1, 50W, 43KHz) for 30-40 minutes in 1 wt% CTAB solution. The dispersions were centrifuged (at 4500 rpm for 30 min) and the supernatant was decanted from above the precipitate. Dispersions in higher CTAB concentrations were prepared by adding a concentrated solution of CTAB to the dispersion.

*Techniques:* Small-Angle X-ray Scattering (SAXS) measurements were performed using a Ni-filtered Cu KR radiation of 0.15418 nm operated at 40 kV, 40 mA (Seifert ID 3000 generator). A linear position-sensitive detector (MBraun) was used to record the scattering patterns. Samples were prepared by filling thin glass capillaries (1.5 mm in diameter) and sealing them with epoxy.

In figure S1 we present SAXS scattering curves of CTAB solutions and dispersions of SWNT-CTAB.



**Fig. S1:** A) SAXS scattering curves of a) 15 wt% CTAB native solution and b) 15 wt% CTAB solution with SWNT. B) SAXS scattering curves of a) 12 wt% CTAB native solution and b) 12 wt% CTAB solution with SWNT.

The presence of the SWNT in the dispersions is indicated by the strong scattering in low  $q$  values. The wide peaks indicate the existence of interactions between CTAB micelles but not a long range order, as imaged by the cryo-TEM. The scattering curves of the (lower phase) of the dispersions (Fig. S1 A, B (b)) show a similar structure to that of the native solutions. In particular, we do not observe a transition to an ordered phase.

The dispersions were observed to de-mix into two macroscopic phases: an upper (transparent) phase and a lower (black) phase. The lower phase is enriched by well dispersed individual SWNT (as indicated by transmission electron microscopy, TEM and small angle x-ray, SAXS). The upper phase is depleted of the nanotubes. Similar de-mixing was observed in dispersions of multi-walled carbon nanotubes (MWNT), and carbon black (CB)) at a wide range of CTAB concentrations. We believe that the physical origin of the observed phase behavior is related to entropic interactions leading to osmotic depletion forces.<sup>1</sup> Thus de-mixing results in the co-existence of a slightly more concentrated (upper) and less concentrated (lower) phase of CTAB micelles). Several studies of depletion-induced phase separation have been reported in colloidal systems.<sup>2,3</sup> Excluded volume interactions that drive the segregation of the additives are known to scale as the ratio of the typical length of the additive,  $L$ , to the micelle diameter,  $d$ , as  $d/L$ . In our case the typical diameter of the CTAB micelles ( $d$ ) is about 4 nm. The length of a SWNT or a MWNT is of the order of a micron (or more), the diameter of CB particle is of the order of some tens of nanometers. Consequentially, in all cases small values of  $d/L$  favor de-mixing, while the actual concentration at which de-mixing takes place would depend on the value of  $d/L$ .<sup>1</sup>

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