Multilamellar structures induced by antagonistic salt added to a binary mixture of water and organic solvent

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Binary mixtures of water and organic solvent have been used extensively to study universal aspects of critical behavior and phase separation dynamics. However, not enough attention has been paid to unique ion effects in such mixtures, where preferential hydration around each ion affects the nano-scal structures. In this study, we investigated the effect of adding an antagonistic salt with hydrophilic cation and hydrophobic anion ($NaBPh_4$) to the mixture of D_2O and 3-methylpyridine (3MP). When 85 mM of $NaBPh_4$ is dissolved in water-rich solvents, emergence of spherical droplets with a size of 10 μm emerge was observed by optical microscope with decreasing temperature below 318K. These droplets showed the Maltase cross pattern under polarized light with crossed Nicole. This result suggests that onion morphologies are formed in these droplets. In order to clarify the nano-meter scale structure, small-angle neutron scattering experiments were performed at SANS-J in JAEA, Japan and NG7-SANS at NIST. Figure 1 shows the temperature dependence of SANS profile for the ternary mixture with 91 vol% of D_2O and 85 mM of $NaBPh_4$. When T < 318 K, a sharp Bragg peak is observed around Q=0.04 $Å^{-1}$ together with higher order peaks. These multi-peak profiles can be well explained by the model function for the randomly oriented lamellar-membrane structure [1]. This result indicates that lamellar / disorder phase transition is induced with decreasing temperature [2].

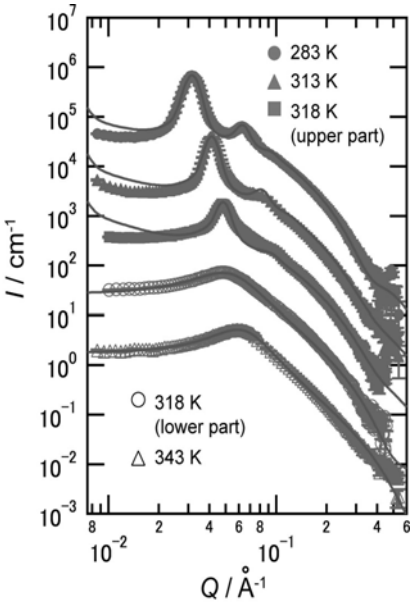

Fig. 1: SANS profile from the mixture with D_2O, 3MP and 85 mM of $NaBPh_4$. The solid line indicates the numerical fitting by the model function for lamellar structure [1] (below 318 K) and disordered structure (above 318 K).

[1] F. Nallet, R. Laversanne and D. Roux, *J. Phys. II France* **3**, 487 (1993).
[2] K. Sadakane, A. Onuki, K. Nishida, S. Koizumi and H. Seto, *Phys. Rev. Lett*., **103**, 106873 (2009).