## **Supporting Information for**

Reverse Self-Assembly of Lipid Onions Induced by Gadolinium and Calcium Ions

Hee-Young Lee, Kaname Hashizaki, Kevin Diehn and Srinivasa R. Raghavan\*



**Figure S1.** Rheology at 25°C of a sample of 18.5 mM DMPC and 7 mM Gd<sup>3+</sup> in cyclohexane. The data are from a dynamic frequency sweep and show the elastic modulus G' and the viscous modulus G'' as functions of frequency. We note that G' and G'' are nearly independent of frequency and that G' > G''. Thus, the data reflect the gel-like (elastic) behavior of the sample.



**Figure S2.** Photographs of mixtures of 20 mM DMPC with varying concentrations of  $Ca^{2+}$  in cyclohexane. At 0 and 4 mM  $Ca^{2+}$ , the samples show precipitates. At 9 mM  $Ca^{2+}$ , the sample is gel-like. At 11 mM  $Ca^{2+}$ , the sample is of low viscosity and is mildly turbid (bluish), indicating reverse vesicles. At 13 mM  $Ca^{2+}$ , the sample is much more turbid, indicating larger reverse vesicles and/or lamellar structures. At 17 mM  $Ca^{2+}$ , the sample again shows a precipitate.

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**Figure S3.** DLS data at 25°C and analysis for samples containing 18.5 mM DMPC and varying concentrations of  $Gd^{3+}$  in cyclohexane. The data were analyzed using the Dynals software supplied by Photocor and the results are shown for each sample in terms of a particle size distribution. The average diameter and polydispersity index (PDI) are also shown for each sample.



**Figure S4.** TEM images (unstained) of a sample of 18.5 mM DMPC + 12 mM  $Gd^{3+}$  in cyclohexane. Multilamellar reverse vesicles (circled) are visible in these images. The bottom image shows a close-up of the concentric bilayers surrounding a couple of the vesicles.



**Figure S5.** TEM image (unstained) of a sample of 18.5 mM DMPC + 16 mM  $Gd^{3+}$  in cyclohexane. Fragments of multilamellar reverse vesicles (blue arrow) as well as lamellar stacks (red arrows) are seen in the image.



**Figure S6.** Additional TEM images (unstained) of a sample of 18.5 mM DMPC + 15 mM  $Gd^{3+}$  in toluene. Multilamellar stacks ("fingerprint pattern") are seen in these images.



**Figure S7.** SANS spectra (intensity *I* vs. scattering vector *q*) for samples in deuterated cyclohexane containing 20 mM DMPC and various concentrations of  $Ca^{2+}$ .