## Spontaneous Pickering emulsion breaking by surface freezing of surfactant adsorbed films

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The adsorbed film of cetyltrimethylammonium chloride (CTAC) at the tetradecane (C14) – water interface undergoes a first-order surface transition from two-dimensional liquid to solid states upon cooling. In this study, we utilized this surface freezing transition to realize a spontaneous demulsification of Pickering emulsions stabilized by silica particles. In the temperature range above the surface freezing transition, the interfacial tension of silica laden oil-water interface was lower than CTAC adsorbed film, hence, stable Pickering emulsion was obtained by vortex mixing. However, the interfacial tension of CTAC adsorbed film decreased rapidly below the surface freezing temperature and became lower than the silica laden interface. The reversal of the interfacial tensions between silica laden and CTAC adsorbed films gave rise to Pickering emulsion demulsification by the desorption of silica particles from the oil-water interface. Similar behavior was also confirmed with the surface frozen film of CTAC and hexadecanol (C16OH), and fluorinated alcohols at dodecane-aqueous interfaces.