



Line tension of alkane lenses on aqueous surfactant solutions at phase transitions of coexisting interfaces



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ABSTRACT

Alkane droplets on aqueous solutions of surfactants exhibit a first-order wetting transition as the concentration of surfactant is increased. The low-concentration or “partial wetting” state corresponds to an oil lens in equilibrium with a two-dimensional dilute gas of oil and surfactant molecules. The high-concentration or “pseudo-partial wetting” state consists of an oil lens in equilibrium with a mixed monolayer of surfactant and oil. Depending on the combination of surfactant and oil, these mixed monolayers undergo a thermal phase transition upon cooling, either to a frozen mixed monolayer or to an unusual bilayer structure in which the upper leaflet is a solid layer of pure alkane with hexagonal packing and upright chains while the lower leaflet remains a disordered liquid-like mixed monolayer. Additionally, certain long-chain alkanes exhibit a surface freezing transition at the air–oil interface where the top monolayer of oil freezes above its melting point. In this review, we summarize our previous studies and discuss how these wetting and surface freezing transitions influence the line tension of oil lenses from both an experimental and theoretical perspective.

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1. Introduction

The line tension can be classified as the excess energy associated with the unit length of a contact line between either (a) two or (b) three coexisting interfaces [1]. For case (a), the line tension is regarded as a natural analog of the surface tension between two bulk phases and therefore, in this case, the line tension only takes positive values. When a first order phase transition occurs in two dimensional (2D) systems, the line tension produces an energy barrier for nucleation [2–4]. In cellular membranes, this stabilizes nanoscale domains called lipid rafts that are enriched in cholesterol and sphingolipids against their surrounding phase [5–9]. The size and shapes of the lipid raft domains

are determined by a competition between the line tension and electrostatic interactions between molecules within the domain. The line tension favors circular domains which minimize this line energy, whereas the electrostatic interactions favor smaller elongated domains [10–13]. Schwartz et al. recently demonstrated that some partially fluorinated compounds act as “linactants” which reduce the line tension between hydrocarbon-rich and fluorocarbon-rich phases and stabilize 2D molecular nanostructures [14,15].

For case (b), with three coexisting interfaces, the line tension can be negative due to the interplay between these interfaces. For a liquid droplet placed on a solid substrate, the line tension is generated at the three-phase contact between air, liquid, and solid phases and it becomes negative when an attractive surface force exists between air–liquid and liquid–solid interfaces [16–18]. Theoretical estimates for the magnitude of the line tension lie in the range of 10^{-10} to 10^{-12} N.

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In our latest paper [19], we have demonstrated that a positive line tension favors the minimization of the contact line through the coalescence of small oil lenses on the water surface, whereas, a negative line tension favors the creation of contact line by spontaneous lens fission. These phenomena indicate that, despite the small magnitude for the line tension, the line tension provides a substantial contribution to the free energy of microscopic systems. Therefore, the line tension plays a crucial role in microscopic phenomena at liquid surfaces such as colloidal adsorption of fine particles at the air–water interface [20–22], the thin-thick transition in foam films [23–25], the surface nucleation of droplets at liquid surfaces [26,27], and the condensation of a liquid on a substrate from its vapor [28,29].

In this review, we summarize our recent findings of type (b) line tensions observed for alkane droplets on cationic surfactant aqueous solutions. Our basic strategy to control the line tension is to change the physical state of the air–water and/or air–oil interfaces via surface phase transitions and their resultant wetting transitions. The contents of this review are as follows. In Section 2, we briefly introduce our experimental systems together with the surface phase transitions that occur upon them. The line tensions obtained for each experimental system are described in Section 3 where we also examine the spontaneous lens splitting and merging behavior. In Sections 4 and 5, we interpret the relationship between the physical states of the interface and their line

tension values for, respectively, long-range and short-range interactions using the interface displacement model of Indekeu et al. [16,17]. Line tension values are then reexamined from the view point of a “point contact mismatch” at the three phase contact line in Section 6. Finally, this review concludes with some closing remarks and a discussion of future experiments (Section 7).

2. Wetting and freezing transitions of alkane lenses on aqueous surfactant solutions

In this review, we discuss the line tension changes for three different systems: (i) a hexadecane droplet on dodecyltrimethylammonium bromide (DTAB) aqueous solution, (ii) a tetradecane droplet on tetradecyltrimethylammonium bromide (TTAB) aqueous solution, and (iii) an octadecane droplet on a decyltrimethylammonium bromide (DeTAB) aqueous solution. Fig. 1 provides a schematic summary of the wetting behavior and surface phase transitions observed in these systems [30,31]. When a liquid droplet is placed on a substrate, there are three possible outcomes which are called partial wetting, pseudo-partial wetting and complete wetting [32]. In the partial wetting state, the liquid does not spread on the substrate but instead remains in the form of a macroscopic lens. In contrast to this behavior, for complete wetting, the liquid spreads out to form a uniform film on the substrate.

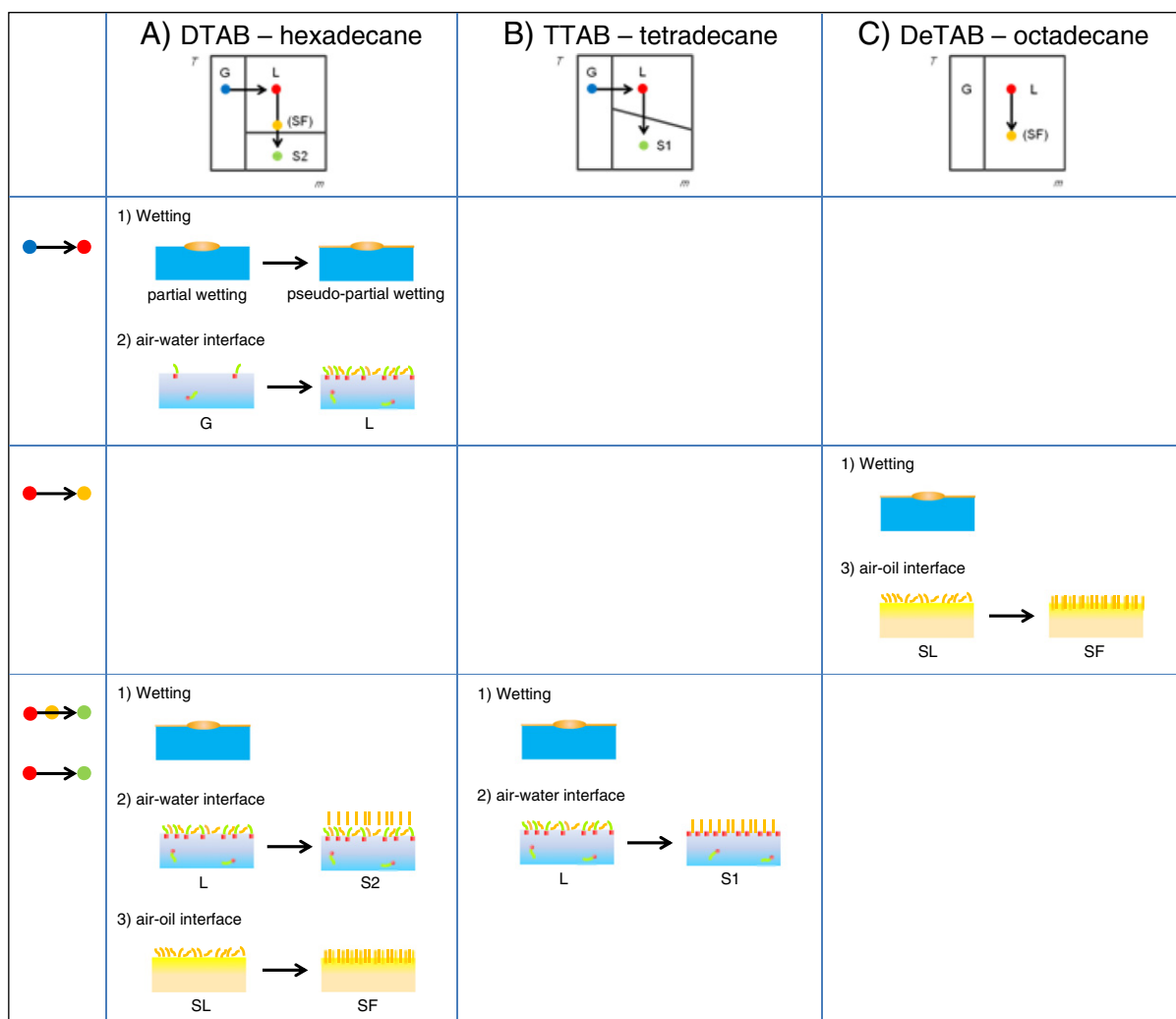


Fig. 1. Wetting and freezing transitions in (A) DTAB–hexadecane, (B) TTAB–tetradecane, and (C) DeTAB–octadecane systems. The top panel illustrates the schematic temperature–concentration diagrams at the air–water interface for each system [30,31]. The surface phases, G, L, S1, S2, SL and SF are explained in the text. 1), 2), and 3), in each cell, depict the macroscopic wetting behavior and the phase transitions at the air–water and air–oil interfaces, respectively, for the surface phase changes represented by the arrows in the left panel.

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