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## Instability of confined thin liquid film trilayers

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#### **Abstract**

The instability of a system in which three stratified thin liquid films are confined in a channel with parallel walls and the interior film is subject to van der Waals-driven breakup is examined in this work. We derive a model based on lubrication theory and consisting of a pair of nonlinear partial differential equations describing the position of the two liquid interfaces. A linear stability analysis is carried out to show that the effects of varying the boundary film thicknesses can be understood in terms of several known limits, including a supported monolayer, confined bilayer, and supported bilayer. Variation of the boundary film viscosities is shown in many cases to eliminate the supported-bilayer limit. The parameter regimes in which squeezing and bending modes dominate the initial growth are determined, and nonlinear simulations are used to show that the mode always switches to squeezing near rupture. It is also found that a multi-modal dispersion relation may be created by asymmetries in thickness ratio, but not viscosity ratio, even in the absence of asymmetric interfacial tensions. The results of this study are expected to be relevant to multiphase microfluidic systems and the lithographic printing process.

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Keywords: Thin liquid films; Instability; Trilayers

#### 1. Introduction

Theoretical studies of thin liquid film systems are prevalent due to their widespread industrial applications [1]. Originally, free films were studied due to their relevance to colloidal systems such as foams and emulsions [2,3]. Supported single films also gained attention for their applicability to paint and other liquid coatings [4,5]. More recent research has focused on liquid bilayers [6-9], which are relevant to multilayer coatings such as pressure-sensitive adhesives and magnetic media, and also tear films in the human eye [10,11]. The work presented here aims at understanding the dynamics of a related system in which three stratified thin liquid films are confined in a channel with parallel walls, and the interior film is subject to breakup via van der Waals forces. This system is relevant to microfluidic applications, where the breakup of a liquid phase into droplets within a second immiscible phase is of great interest [12], and also to the lithographic printing process, which is being explored for the mass-production of electronic devices [13,14].

The key step of the lithographic printing process is the transfer of an oil-like ink from a carrier substrate to a printing plate which carries its own thin layer of residual ink; the transfer occurs through a barrier film of water-like fountain solution. As a result of this step, a thicker ink layer with emulsified fountain solution will result on the plate surface. In order to better conceptualize this ink transfer step, the authors performed a set of experiments using the simplified model system illustrated schematically in Fig. 1a. During the experiment, a glass plate holding a film of oil ( $\approx$ 400 µm thick) is lowered onto another glass plate that holds a film of oil ( $\approx$ 200 µm thick) covered by a thicker layer of water. As the liquids are brought into contact, the water layer is thinned to a point where intermolecular forces are expected to become important, which triggers the rupture and subsequent retraction of the water film suggested by Fig. 1b. Fig. 2 shows pictures taken through the top oil layer of one such experiment. The retraction of the thin water film in Fig. 2a leads to the distribution of emulsified water droplets in Fig. 2b. The size and distribution of these droplets are key to determining print quality, so it is useful to understand how system parameters affect their formation. The experiments showed that the mechanism for droplet formation depends on

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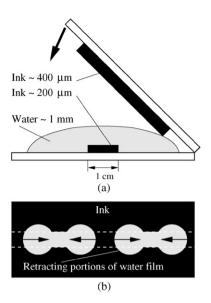
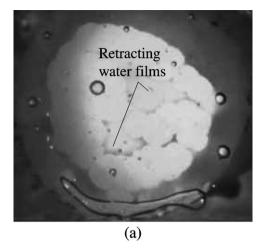


Fig. 1. Schematic of printing visualization experiment: (a) apparatus; (b) retraction of water film between ink layers.

the speed of water film retraction. When the retraction velocity was increased by decreasing the oil viscosity or increasing the oil—water interfacial tension, the retracting films tended to pinch off into smaller droplets [15]. The final droplet distribution, however, was still highly dependent on the frequency of initial rupture events. Such rupture events are difficult to systematically account for due to their small time scale and the occurrence of two different types of rupture: heterogeneous and what is referred to as spinodal [16]. Heterogeneities such as dust particles and nonuniform oil layers may result in the first type of rupture, while the amplification of interfacial disturbances through intermolecular forces will result in the second type [17].

Since heterogeneous ruptures are likely to occur independent of most system parameters, within this paper we set forth a computational study of the spinodal rupture mechanism. We present a model for the breakup of the inner layer of a confined trilayer system, in which all three layers are described by lubrication theory. Here, as in previous studies [1], we incorporate a van der Waals body force (or disjoining pressure) into the governing equations. We manipulate these governing equations in order to obtain a system of two coupled nonlinear evolution equations for the interfacial heights, and also perform a linear stability analysis to obtain a dispersion relation that describes the initial growth of interfacial disturbances. Our trilayer model serves to generalize several models previously offered for single and two-layer systems [5,7,18,19], which prove to be useful limits for understanding trilayer behavior. We examine the behavior of the inner layer subject to variations in the boundary film thicknesses and viscosities. To set a reasonable limit on the scope of this study, we consider only symmetric interfacial tensions. Several studies have previously shown in detail the effects of asymmetric interfacial tensions in a supported liquid bilayer [6,7,9], so although our trilayer model is generalized to include the possibility of such asymmetries, these analyses will not be repeated here. The problem setup is described in



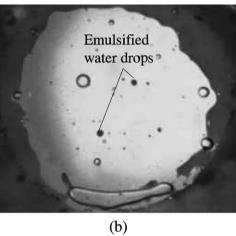


Fig. 2. Visualization with 8,000 cP oil viscosity: (a) water film retraction step 3 s after liquid contact; (b) final distribution of water droplets after 14 s.

Section 2, followed by results and discussion in Section 3. Conclusions are presented in Section 4.

#### 2. Problem formulation

Fig. 3 is a schematic of the confined trilayer problem setup. Since we are primarily interested in rupture of the inner layer, it is labeled 1 and its unperturbed thickness,  $\tilde{h}_1$ , is chosen as the characteristic vertical length scale. Thus the dimensionless boundary film thicknesses are the thickness ratios  $h_i = \tilde{h}_i/\tilde{h}_1$ , where i equals 2 or 3, and the tilde indicates a dimensional quantity. The horizontal length scale is chosen to be the wavelength of the disturbance that grows most rapidly,  $\tilde{\lambda}$ , which is taken to be much larger than  $\tilde{h}_1$ . This allows us to employ the lubrication approximation and define a small parameter  $\epsilon = \tilde{h}_1/\tilde{\lambda}$  for use in scaling. We also employ a velocity scale based on the viscosity of this inner layer. Our dimensionless spatial and velocity variables are then:

$$x = \frac{\epsilon \tilde{x}}{\tilde{h}_1}, \qquad z = \frac{\tilde{z}}{\tilde{h}_1}, \qquad u_i = \frac{\tilde{u}_i \tilde{\rho}_1 \tilde{h}_1}{\tilde{\mu}_1}, \qquad w_i = \frac{\tilde{w}_i \tilde{\rho}_1 \tilde{h}_1}{\epsilon \tilde{\mu}_1}, \tag{1}$$

where  $u_i$  and  $w_i$  are the liquid velocities in the x- and z-directions, and  $\tilde{\mu}_1$  and  $\tilde{\rho}_1$  are the viscosity and density of the

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