



Title	In situ Determination of Surface Tension-to-Shear Viscosity Ratio for Quasiliquid Layers on Ice Crystal Surfaces
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# Supplemental Material: *In situ* determination of surface tension-to-shear viscosity ratio for quasi-liquid layers on ice crystal surfaces

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## THE SPREADING DYNAMICS OF A QUASI-LIQUID LAYER

It is well-recognized that spreading of non-volatile liquids universally obeys the so-called Tanner's law [1]. In this case, the radial evolution of a sessile droplet (contact angle decay) follows a power law form as  $r \propto t^{1/10}$  ( $\theta \propto t^{-3/10}$ ), whose small exponent stems from the presence of a precursor film emerging ahead of the nominal contact line [2, 3]. For the spreading of QLLs, however, such a precursor film has not been confirmed in the early and intermediate stages even though our advanced microscopy is fully capable of visualizing the film itself. The absence of the precursor film leads to a change in the exponent in Tanner's law. In the followings, we derive a new equation describing the spreading dynamics without the precursor film under the lubrication approximation [4].

We revisit a force,  $F$ , acting at a contact line. During the spreading, the contact line of the droplet is stretched outward by  $F = \gamma_{SV} - \gamma_{SL} - \gamma \cos \theta$ , where  $\theta$  is the dynamic contact angle during the spreading (Here  $\theta \ll 1$ ), and  $\gamma_{SV}$ ,  $\gamma_{SL}$  and  $\gamma$  are the solid/gas, solid/liquid and liquid/gas interfacial tensions, respectively. Using the definition of spreading coefficient,  $S = \gamma_{SV} - \gamma_{SL} - \gamma$ , we can rewrite  $F$  as

$$F = S + \gamma(1 - \cos \theta) \sim S + \frac{1}{2}\gamma\theta^2. \quad (1)$$

Spreading means a transformation into the TLL state (a complete wetting state,  $S > 0$ ). Here due to  $\theta \ll 1$ , we focus on the case of  $F \sim S$ . This should balance the viscous force of the wedge,  $F_V = 3l\eta\theta^{-1}V$  ( $V$  being the velocity of the contact line) [3], which gives  $V$  as the following relation:

$$V = \frac{\theta S}{3l\eta}. \quad (2)$$

The volume of the droplet,  $\Omega = (\pi/4)\theta r^3$ , is conserved during the spreading. Thus, from the relation of  $d\Omega/dt =$

0, we obtain the following differential equation:

$$\frac{3}{r} \frac{dr}{dt} = -\frac{1}{\theta} \frac{d\theta}{dt}. \quad (3)$$

Using the relation of  $V = dr/dt$  and  $r = L\theta^{-1/3}$  ( $L$  being  $(4/\pi)^{1/3}$ ), Eq. (3) can be rewritten as

$$\frac{d\theta}{dt} = -\frac{S}{lL\eta} \theta^{7/3}. \quad (4)$$

Then we obtain as its solution the temporal decay of  $\theta$  during the spreading, as

$$\theta = \left( \frac{4}{3} \frac{S}{lL\eta} (t + t_0) \right)^{-3/4}, \quad (5)$$

where  $t_0 = 3lL\eta/4S\theta_0^{-4/3}$  is determined by the initial condition of the droplet (here  $\theta_0$  being the contact angle at the start point of the spreading). Furthermore, because of  $r = L\theta^{-1/3}$ , we can easily obtain the radial evolution of the droplet as follows:

$$r = L \left( \frac{4}{3} \frac{S}{lL\eta} (t + t_0) \right)^{1/4}. \quad (6)$$

Finally, we note that the precursor film appears in some of the BLDs as TLL states only in the late stage. However, here we do not deal with the dynamics in the late stage due to the complex effects of supersaturation which significantly assists the radial growth of TLLs (see Fig. 2(c)).

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