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LETTER TO THE EDITOR

Liquid interface tilt angle under thermal gradient: the surface-tension temperature coefficient

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Abstract. Under an applied horizontal temperature gradient, the free surface of a thin liquid layer gets tilted by a small angle proportional to the surface-tension temperature coefficient. We show the effect to be particularly large at the interface between two liquids with comparable densities. From the corresponding interfacial slope, measured optically under different boundary conditions, one could draw an accurate determination of the upper liquid free-surface coefficient.

A horizontal thermal gradient $G = \nabla T$, however small, applied to a liquid layer, sets it into convection. There are two driving forces: thermal expansion (at work, e.g., in meteorology) and the surface-tension temperature (Marangoni) coefficient [1, 3].

The former effect—bulk thermal convection, scaled by the dimensionless Grashof number proportional to the fourth power of layer thickness—dominates in relatively thick layers, while thermocapillary convection takes over at small thicknesses, h . In usual liquids under ordinary gravity, the crossover is at about one centimeter. In this work, we have in mind thin layers between 1 and 3 mm thick, for which thermocapillary convection is very largely dominant. Also, due to the large aspect ratios (length, or width, over thickness), we shall not have to bother with end effects. Similarly, we may neglect any surface curvature and any temperature 'stratification' [1, 3]: the temperature gradient is essentially uniform.

The temperature coefficient $\sigma' = d\sigma/dT$, of surface tension σ , is practically always negative, due to the entropy term in the surface free energy. As a result, surface convection drives the upper part of the liquid toward the cold end (as does volume convection usually). The thermocapillary-convection velocity profile is parabolic [1, 3].

Because of the pressure field associated with convection, the liquid surface gets slightly tilted [2, 3], by an angle:

$$\theta = \frac{2}{3}(G/\rho g)\sigma'/h \quad (1)$$

(g : gravity acceleration, ρ : mass density of the liquid); the layer thickness *increases toward the cold end*. We have pointed out recently [3] that this formula—which is readily extended, if necessary, to include thermal expansion—provides us with an original way of determining $\sigma' = d\sigma/dT$ (actually, this material parameter is not very well

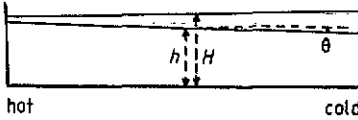


Figure 1. Geometry of the two-layer problem. 1(2): lower (upper) liquid; σ' and θ : interfacial-tension temperature coefficient, and slope; σ'_0 : free-surface temperature coefficient (this can be accurately deduced from two successive measurements of the relatively large angle θ , with and without an actual free surface). Note: thin layer (large aspect ratio) end effects may be ignored.

documented in the literature). The appropriate generalization, given below, of (1) for an interface indicates that the surface slope is spectacularly large in that case, and the accuracy on σ' correspondingly improved (with a reduced driving thermal gradient).

Let us derive the slope of the interface for two thin, superposed, immiscible liquid layers (1 and 2, with mass densities ρ_1, ρ_2 and viscosities η_1, η_2). The horizontal thermal gradient, G , and the resulting one-dimensional flow velocity, v_1 and v_2 , lie along Ox . The (linearized) Navier-Stokes equations are:

$$\partial p_1 / \partial x - \eta_1 (\partial^2 v_1 / \partial z^2) = \partial p_2 / \partial x - \eta_2 (\partial^2 v_2 / \partial z^2) = 0 \tag{2}$$

with the return-flow conditions:

$$\int_0^h v_1 dz = \int_h^H v_2 dz = 0 \tag{3}$$

where h and $(H - h)$ are the thicknesses of the lower (1) and upper (2) layers, respectively, figure 1. The boundary conditions are, (i) at $z = 0$ (lower horizontal wall): $v_1 = 0$, and, (ii) at $z = h$:

$$v_1 = v_2 \tag{4a}$$

$$\eta_1 (\partial v_1 / \partial z) - \eta_2 (\partial v_2 / \partial z) = d\sigma / dx = \sigma' G \tag{4b}$$

where σ is the interfacial tension (and σ' its temperature coefficient). For definiteness let the temperature decrease toward the right ($G < 0$): then, $\sigma'G$ is positive (since $\sigma' < 0$). The upper boundary condition, at $z = H$, is either: $v_2 = 0$, in the 'rigid' case where the system is bounded by walls both at $z = 0$ and $z = H$, or:

$$\eta_2 (\partial v_2 / \partial z) = d\sigma_0 / dx = \sigma'_0 G \tag{5}$$

in the 'free' case (σ_0 is the free-surface tension of upper liquid).

Consider first the rigid case. Let us formally decompose (4b) into:

$$\eta_1 (\partial v_1 / \partial z) = d\sigma_1 / dx \quad \eta_2 (\partial v_2 / \partial z) = -d\sigma_2 / dx \quad \sigma' = \sigma'_1 + \sigma'_2. \tag{6}$$

From (2)–(4) (and $v_1(z = 0) = v_2(z = H) = 0$), we get, for an unperturbed interface, the pressure gradients:

$$\Pi_1 = \partial p_1 / \partial x = 3G\sigma'_1 / 2h \quad \Pi_2 = \partial p_2 / \partial x = 3G\sigma'_2 / 2(H - h) \tag{7}$$

and the velocities at the interface ($z = h$):

$$v_1 = Gh\sigma'_1 / 4\eta_1 = v_2 = G(H - h)\sigma'_2 / 4\eta_2. \tag{8}$$

Hence σ'_1 and σ'_2

$$(\sigma'_2 = \sigma' [1 + (\eta_1 / \eta_2)(H - h) / h]^{-1} = \sigma' / (1 + \alpha))$$

and:

$$\Pi_1 = (3G\sigma'/2h)\alpha/(1 + \alpha) \quad \Pi_2 = [3G\sigma'/2(H - h)]/(1 + \alpha). \quad (9)$$

Now, this pressure field will induce a slight tilting of the interface, by an angle θ . Take two neighbouring points A and B on the interface, in the (x, z) plane $x_B - x_A = \delta x$; we may write

$$p_B = p_A + \Pi_1 \delta x - \rho_1 g \theta \delta x = p_A + \Pi_2 \delta x - \rho_2 g \theta \delta x$$

so that:

$$\theta = (\Pi_1 - \Pi_2)/g(\rho_1 - \rho_2) \quad (10)$$

(which is the straightforward generalization of $\theta = (\partial p/\partial x)/g\rho$, leading to (1) for a single layer). In the case of a *thin upper layer* ($H - h \ll h$), the pressure gradient there, Π_2 (equation (9)), is much larger than Π_1 (so much so that $\alpha = (\eta_1/\eta_2)(H - h)/h$ is small then, for comparable viscosities), and (10) reduces to:

$$\theta \approx -3G\sigma'/2g(\rho_1 - \rho_2)(H - h). \quad (11)$$

That is, the thin upper layer thickens at the cold end ($\rho_1 > \rho_2$ in our notation, and $G\sigma' > 0$ as we have seen). This is just the same trend as for a free-surface single layer (equation (1)), but the effect is much *enhanced*, both for geometrical $h/(H - h)$ and inertial $\rho/(\rho_1 - \rho_2)$ reasons.

A similar enhancement occurs in the 'free' case (free-surface upper layer with tension σ_0 , (5)). Then, the solution of (2)–(4) is particularly simple when $\eta_2 \gg \eta_1$ (viscous upper layer: the 'oil-on-water' case):

$$\theta = -\theta_0(\rho_2/(\rho_1 - \rho_2)) = -G(\sigma' + \sigma'_0)/g(\rho_1 - \rho_2)(H - h) \quad (12)$$

where θ_0 is the slope of the upper, free surface, and $\sigma'_0 = d\sigma_0/dT$. θ is negative while the smaller angle, θ_0 , is positive. The (now unconstrained) upper layer again thickens on the cold side. The same results holds, for comparable viscosities, in the case of a thin upper layer. Of course, we have assumed implicitly that the applied thermal gradient is weak enough for the relative variation of layer thicknesses along $0x$ (in (11) and (12) as well as in (1)) to remain small.

Thus, if one has access (direct optical access, say) to the interfacial slope, it should be easy to get an accurate measurement of σ' , the interfacial temperature coefficient. Furthermore, comparing (11) and (12), we may obtain the principle of a simple and accurate measurement of σ'_0 . For ordinary liquids under modest temperature gradients, G , of a few K cm^{-1} , (1) yields θ of order one degree. This is easily measurable by optical means (Poggendorff deviation, 2θ , of an incident light beam). We may, however, take advantage of the much larger interfacial tilt angles and deduce σ'_0 from (11) and (12) (with the thickness and viscosity restrictions discussed above). Using an underlying liquid (liquid 1), a first measurement, in a closed box (rigid case), of the interfacial angle leads to the interfacial temperature coefficient, σ' , through (11). A second measurement, with the same liquid couple but now in the free case, then yields the required σ'_0 via (12). It is essential of course that the two liquids be immiscible and the free surface itself non-contaminated. Due to the larger sensitivity, these measurements could be conducted by direct optical observations. Preliminary experiments on simple organic liquids, by Amrit and co-workers, confirm the feasibility of the method.

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