

Relaxation of a moving contact line and the Landau-Levich effect

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(received 14 December 2000; accepted in final form 7 May 2001)

PACS. 68.08.Bc – Wetting.

PACS. 68.08.-p – Liquid-solid interfaces.

PACS. 05.40.-a – Fluctuation phenomena, random processes, noise, and Brownian motion.

Abstract. – The dynamics of the deformations of a moving contact line is formulated. It is shown that an advancing contact line relaxes more quickly as compared to the equilibrium case, while for a receding contact line there is a corresponding slowing down. For a receding contact line on a heterogeneous solid surface, it is found that a roughening transition takes place which formally corresponds to the onset of leaving a Landau-Levich film. We propose a phase diagram for the system in which the phase boundaries corresponding to the roughening transition and the depinning transition meet at a *junction* point, and suggest that for sufficiently strong disorder a receding contact line will leave a Landau-Levich film immediately after depinning.

When a drop of liquid spreads on a solid surface, the *contact line*, which is the common borderline between the solid, the liquid, and the corresponding equilibrium vapor, undergoes a rather complex dynamical behavior. This dynamics is determined by a subtle competition between the mutual interfacial energetics of the three phases, dissipation and hydrodynamic flows in the liquid, and the geometrical or chemical irregularities of the solid surface [1].

A most notable feature of contact lines, which is responsible for their novel dynamics, is their anomalous elasticity as noticed by Joanny and de Gennes [2]. For length scales below the capillary length, which is usually of the order of 1 mm, a contact line deformation of wave vector k , denoted as $h(k)$ in Fourier space, will distort the surface of the liquid over a distance $|k|^{-1}$. Assuming that the surface deforms instantaneously in response to the contact line, the elastic energy cost for the deformation can be calculated from the surface tension energy stored in the distorted area, and is thus proportional to $|k|$, namely $E_{cl} = \frac{\gamma\theta^2}{2} \int \frac{dk}{2\pi} |k| |h(k)|^2$, in which γ is the surface tension and θ is the contact angle [2].

The anomalous elasticity leads to interesting *equilibrium* dynamics, corresponding to when the contact line is perturbed from its static position, as studied by de Gennes [3]. Balancing $\frac{dE_{cl}}{dt}$ and the dissipation, which for small contact angles is dominated by the hydrodynamic dissipation in the liquid nearby the contact line, he finds that each deformation mode relaxes

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to equilibrium with a characteristic frequency (inverse decay time) $\omega(k) = c|k|$, in which $c = \gamma\theta^3/(3\eta\ell)$, where η is the viscosity of the liquid and ℓ is a logarithmic factor of order unity [3]. The relaxation is, thus, characterized by a dynamic exponent z , defined via $\omega(k) \sim |k|^z$, which is equal to 1. The linear dispersion relation implies that a deformation in the contact line will *decay* and *propagate* at a constant velocity c , as opposed to systems with normal line tension elasticity, where the decay and the propagation are governed by diffusion. This behavior has been observed, and the linear dispersion relation has been precisely tested, in a recent experiment by Ondarcuhu and Veyssie [4].

As an interesting example for *nonequilibrium* cases, corresponding to when there is an overall relative motion between the liquid and the solid, Landau and Levich studied the wetting of a plate vertically withdrawn from a completely wetting liquid at a constant velocity $-v$ [5]. The case of partial wetting, where the liquid has a finite contact angle θ_e at equilibrium, has been studied by de Gennes [6, 7]. He finds that the velocity v and the dynamic contact angle θ are related as $v = c(\theta_e^2/\theta^2 - 1)/2$, and thus argues that a steady state is achieved in which the liquid will partially wet the plate with a nonvanishing dynamic contact angle for pull-out velocities less than c , while a macroscopic Landau-Levich liquid film, formally corresponding to a vanishing θ , will remain on the plate for higher velocities, as depicted in fig. 1 [6]. Note that at the transition there is a jump in the “order parameter” θ , from $\theta_e/\sqrt{3}$ to zero.

In the presence of defects and heterogeneities in the substrate, which could be due to (surface) roughness or chemical contamination, a contact line may become *rough* because it locally deforms so as to find the path with optimal *pinning* energy [1]. This is in contrast to the case of a perfect solid surface, where the contact line is *flat*. The roughness can be characterized as a scaling law that relates the statistical width W of the contact line to its length L , via $W \sim L^\zeta$. The so-called roughness exponent ζ is equal to 1/3 for a contact line at equilibrium on a surface with short-range correlated disorder [8], while for a contact line at the depinning threshold it could be significantly different from 1/3 [9]. The contact line is also pinned by the defects, which means that a nonzero (critical) force is necessary to set the contact line to motion, through a depinning transition [10]. It is also important to note that there may be numerous metastable states for the contact line due to the random disorder, leading to hysteresis in the contact angle [2, 11].

Here we study the dynamics of the deformations of a nonequilibrium (moving) contact line. Using a balance between the hydrodynamic dissipation in the deformed moving liquid wedge and the rate of change of the interfacial energies, we find that the dynamical relaxation is described by

$$\partial_t h(k, t) = -(c - v)|k|h(k, t) - \frac{1}{2} \int \frac{dq}{2\pi} \lambda(q, k - q)h(q, t)h(k - q, t), \quad (1)$$

in which v is the average velocity of the contact line, and $\lambda(q, k - q) = -(2v - c)q(k - q) + 3c|q||k - q| + (c - v)|k|(|q| + |k - q| - |k|)$ gives the leading nonlinear correction. As compared to the equilibrium case ($v = 0$), we thus find that the linear relaxation is faster for an *advancing* contact line ($v < 0$), while it is slower for a *receding* one ($v > 0$). In particular, for a contact line that is receding at the “terminal” velocity $v = c$, which coincides with the onset of leaving a Landau-Levich film, linear relaxation becomes infinitely slow and the dominant relaxation is thus governed by the nonlinear terms.

We also take into account the effect of surface disorder on the moving contact line dynamics, which appears as a stochastic term in the right-hand side of eq. (1), and attempt to systematically study the dynamical phase transition using renormalization group (RG) techniques. We find that the onset of leaving a Landau-Levich film formally corresponds to

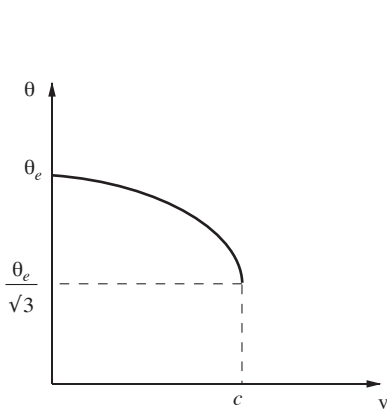


Fig. 1

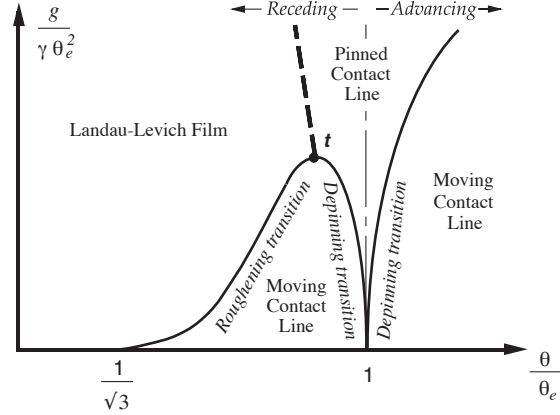


Fig. 2

Fig. 1 – Dynamic contact angle as a function of pull-out velocity [6].

Fig. 2 – The suggested phase diagram of a contact line on a disordered substrate. The depinning transition line is taken from ref. [11], corresponding to the hysteresis in receding and advancing contact angles. The asymptotic form for the roughening transition line is given in eq. (2).

a *roughening* transition of the (receding) contact line, which for a random substrate with strength g takes place at a critical velocity below c , corresponding to a dynamic contact angle

$$\frac{\theta_c}{\theta_e} = \frac{1}{\sqrt{3}} + \frac{(11\pi/3)^{1/3}}{2\sqrt{3}} \left(\frac{g}{\gamma\theta_e^2} \right)^{2/3}, \quad (2)$$

to the leading order. We combine our results with studies of the contact line depinning transition [10, 11], and propose a phase diagram for the system as depicted in fig. 2. In particular, we find that the phase boundaries corresponding to the dynamical phase transition and to the depinning transition meet at a junction point, and suggest that for sufficiently strong disorder a receding contact line will leave a Landau-Levich film immediately after depinning.

Let us assume that the contact line is oriented along the x -axis, and is moving in the y -direction with the position described by $y(x, t) = vt + h(x, t)$, as depicted in fig. 3. If a line element of length $dl = dx\sqrt{1 + (\partial_x h)^2}$ is displaced by $\delta y(x, t)$, the interfacial energy will be locally modified by two contributions: i) the difference between the solid-vapor γ_{SV} and the solid-liquid γ_{SL} interfacial energies times the swept area in which liquid is replaced by vapor, namely, $(\gamma_{SV} - \gamma_{SL})dl\delta y/\sqrt{1 + (\partial_x h)^2}$, and ii) the work done by the surface tension force, whose direction is along the unit vector \hat{T} that is parallel to the liquid-vapor interface at the contact and perpendicular to the contact line, as $\gamma\hat{T} \cdot \hat{y}dl\delta y$. Note that we are interested in length scales below the capillary length, where gravity does not play a role. The overall change in the interfacial energy of the system can thus be written as $\delta E = \int dx\sqrt{1 + (\partial_x h)^2} \left[\frac{\gamma \cos \theta_e}{\sqrt{1 + (\partial_x h)^2}} - \gamma\hat{T} \cdot \hat{y} \right] \delta y(x, t)$, in which we have made use of the Young equation: $\gamma_{SV} - \gamma_{SL} = \gamma \cos \theta_e$. Note that both “forces” should be projected onto the y -axis when calculating the work done for a displacement in this direction.

To calculate the dissipation, we assume that the contact angle is sufficiently small so that the dominant contribution comes from the viscous losses in the hydrodynamic flows of the

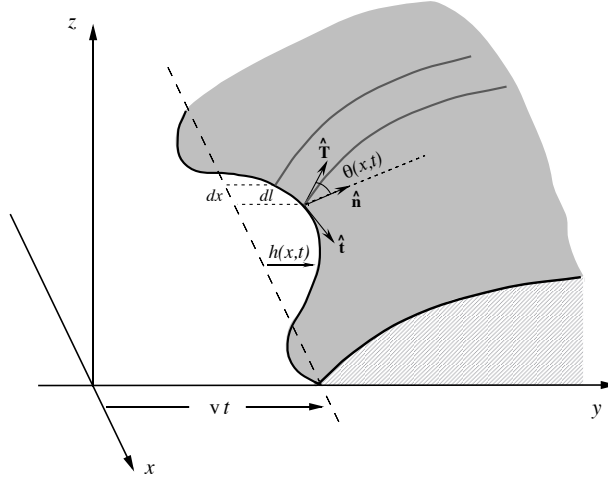


Fig. 3 – The schematics of the system.

liquid wedge [1]. For a slightly deformed contact line, we assume that the dissipation can be approximated by the sum of contributions from wedge-shaped slices with local contact angles $\theta(x, t)$, as shown in fig. 3. This is a reasonable approximation because most of the dissipation is taking place in the singular flows near the tip of the wedge [1, 3]. Using the result for the dissipation in a perfect wedge which is based on the lubrication approximation [1, 12], we can calculate the total dissipation as $P = \int dx \sqrt{1 + (\partial_x h)^2} \{3\eta\ell [v + \partial_t h(x, t)]^2 / \theta(x, t)\}$ [3].

Balancing the rate of change in energy and the dissipation as calculated above now yields the dynamical equation as [13]

$$v + \partial_t h(x, t) = \frac{\gamma}{6\eta\ell} \frac{\theta(x, t) [\theta_e^2 - \theta(x, t)^2]}{\sqrt{1 + (\partial_x h)^2}}. \quad (3)$$

The above equation might simply be recovered by locally applying the result of ref. [3] for straight contact lines, with the additional geometrical factor (which is needed when the direction of motion is not perpendicular to the contact line) taken into account. To complete the calculation, we need to solve for the profile of the surface and the corresponding angles as a function of h . Following ref. [2], we find $\theta(x) = \theta[1 + \int \frac{dk}{2\pi} |k| h(k) e^{ikx} + \frac{1}{2} \int \frac{dk}{2\pi} \frac{dk'}{2\pi} f(k, k') h(k) h(k') e^{i(k+k')x}]$, with $f(k, k') = |k + k'|(|k| + |k'|) - (k + k')^2 + kk'$ [14]. This can then be used in the above equation to yield eq. (1), and the relation between v and θ as described above, and depicted in fig. 1.

The linear relaxation of a moving contact line thus takes place at characteristic frequencies which obey the modified dispersion relation $\omega(k) = (c - v)|k|$, and as the onset of leaving a Landau-Levich film corresponding to $v = c$ is approached, the relaxation becomes progressively slower.

In addition to dissipation and elasticity, the dynamics of a contact line is also affected by the defects and heterogeneities in the substrate, which are present in most practical cases. If the interfacial energies γ_{SV} and γ_{SL} are space dependent, a displacement of the contact line is going to lead to a change in energy as $\delta E_d = \int dx g(x, vt + h(x, t)) \delta y(x, t)$, where $g(x, y) = \gamma_{SV}(x, y) - \gamma_{SL}(x, y) - (\bar{\gamma}_{SV} - \bar{\gamma}_{SL})$. Incorporating this contribution in the force balance leads to a noise term on the right-hand side of eq. (1) of the form $\eta(x, t) = \frac{\theta}{3\eta\ell} g(x, vt)$

to the leading order. Note that this major assumption is a good approximation provided we are well away from the depinning transition, and the contact line is moving fast enough [1,2,10,11]. Assuming that the surface disorder has short-range correlations (so that the correlation length is a microscopic length a) with a strength g , we can deduce the distribution of the noise as $\langle \eta(x, t) \rangle = 0$ and $\langle \eta(x, t) \eta(x', t') \rangle = \frac{c^2 g^2 a^2}{\gamma^2 \theta^4 |v|} \delta(x - x') \delta(t - t')$.

In the presence of the noise, the contact line undergoes dynamical fluctuations. These fluctuations can best be characterized by the width of the contact line, which is defined as $W^2(L, t) \equiv \frac{1}{L} \int dx (h(x, t))^2$. Using the scaling form of the two-point correlation function, one can show that $W \sim t^{\zeta/z}$ for intermediate times, while it saturates to $W \sim L^\zeta$ at long times. Similarly, we can study the fluctuations in the order parameter field $\delta\theta(x, t) = \theta(x, t) - \theta$, and find

$$\langle \delta\theta(x, t)^2 \rangle \sim 1 - B/t^{2(1-\zeta)/z}, \quad (4)$$

where B is a constant. Note that the order parameter fluctuations approach a finite limit at long times, provided $\zeta < 1$.

Keeping only the linear term in eq. (1), we calculate the width of the contact line as $W(L, t) \sim \sqrt{\ln[(c-v)t/a]}$ for $\frac{a}{(c-v)} \ll t \ll \frac{L}{(c-v)}$, and $W(L, t) \sim \sqrt{\ln(L/a)}$ for $t \gg \frac{L}{(c-v)}$, and, similarly, the order parameter fluctuations as $\langle \delta\theta(x, t)^2 \rangle \sim 1 - B/t^2$, for $t \gg \frac{a}{(c-v)}$. We thus find $\zeta = 0$ and $z = 1$ within the linear theory.

The nonlinear terms in eq. (1) will modify the above results only when it becomes appreciable at long length scales, as compared to the linear term. The ratio of the two terms in eq. (1) scales as $\frac{c(L/a)^{2\zeta-1}}{(c-v)(L/a)^\zeta} \sim \frac{ac}{L(c-v)}$, and is thus appreciable *only* when the smallest time scale in the linear theory $a/(c-v)$ becomes comparable to L/c . This happens near the onset of leaving a Landau-Levich film.

Let us now attempt to systematically study the dynamical phase transition, corresponding to leaving a Landau-Levich film, using an RG scheme. The dynamical equation, which can be generally written in d dimensions as $\partial_t h(k, t) = -\sigma |k| h(k, t) - \frac{1}{2} \int \frac{d^d q}{2\pi} \lambda(q, k-q) h(q, t) h(k-q, t) + \eta(k, t)$, with $\lambda(q, k-q) = -\lambda_1 q(k-q) + \lambda_2 |q||k-q| + \lambda_3 |k|(|q| + |k-q| - |k|)$, belongs to the general class of Kardar-Parisi-Zhang equations [15]. We take a noise spectrum given by $\langle \eta(k, t) \rangle = 0$ and $\langle \eta(k, t) \eta(k', t') \rangle = 2D(2\pi)^d \delta^d(k+k') \delta(t-t')$, and employ standard RG techniques following ref. [15] to calculate the RG equations describing the flow of the coupling constants. We find

$$\begin{aligned} \frac{d\sigma}{dl} &= \sigma [z - 1 - U], \\ \frac{d\lambda(q, k-q)}{dl} &= \lambda(q, k-q) (\zeta + z - 2), \\ \frac{dD}{dl} &= D \left[z - 2\zeta - d + \left(\frac{\lambda_1 + \lambda_2}{\lambda_2 + \lambda_3} \right) \frac{U}{2} \right], \end{aligned} \quad (5)$$

to the one-loop order, in which $U = \frac{\pi S_d (\lambda_1 + \lambda_2) (\lambda_2 + \lambda_3) D}{(2a)^{d+1} \sigma^3}$ and S_d is the surface area of a unit sphere in d dimensions.

To study the fixed-point structure of the above set of flow equations, we set $z = 1 + U$ and $\zeta = 1 - U$, and look at the flow equation for U : $dU/dl = -(d+1)U + [6 + (\lambda_1 + \lambda_2)/(\lambda_2 + \lambda_3)] \cdot U^2/2$, which has two stable fixed points at $U = 0$ (linear theory) and $U = \infty$ (strong coupling), as well as an intermediate unstable fixed point at $U = U^* \equiv \frac{2(d+1)}{6 + (\lambda_1 + \lambda_2)/(\lambda_2 + \lambda_3)}$.

For $U < U^*$, the nonlinearity is irrelevant and the exponents are given by the linear theory, *i.e.* $\zeta = 0$ and $z = 1$, while for $U > U^*$ the behavior of the system is governed

by a strong-coupling fixed point which cannot be studied perturbatively. The fixed point at U^* corresponds to a roughening transition of the moving contact line. The exponents at the transition are $z = 1 + U^*$ and $\zeta = 1 - U^*$, which turn out to be nonuniversal. The strong-coupling fixed point should presumably describe the Landau-Levich film.

We can also study how the transition is approached by linearizing the flow equation near the fixed point. Setting $U = U^* + \delta U$, we find $d\delta U/dl = (d + 1)\delta U$ that would imply divergence of the correlation length near the transition as $\xi \sim |\delta U|^{-\nu}$, with $\nu = 1/(d + 1)$. The correlation length corresponds to the typical size of rough segments in the contact line, which should diverge as the transition is approached.

Using the equation for the phase boundary $U = U^*$ with $d = 1$, and the values of the coupling constants that we can read off from eq. (1), we map out the phase diagram of the system as depicted in fig. 2, with the depinning transition line taken from ref. [11] (corresponding to the hysteresis in receding and advancing contact angles). In particular, we find the asymptotic form of the phase boundary for weak disorder as reported in eq. (2) above, and the nonuniversal exponents $z = \frac{17}{11} + \frac{15}{121}(\frac{11\pi}{3})^{1/3}(\frac{g}{\gamma\theta_e^2})^{2/3}$ and $\zeta = \frac{5}{11} - \frac{15}{121}(\frac{11\pi}{3})^{1/3}(\frac{g}{\gamma\theta_e^2})^{2/3}$ to the leading order, and $\nu = 1/2$. The order parameter fluctuations at the transition are given by eq. (4) with the above choice for z and ζ , and remain finite since $\zeta < 1$.

We find that the roughening transition phase boundary asymptotes to a maximum with zero slope that is located at $\theta_t/\theta_e = 0.887$ and $g_t/(\gamma\theta_e^2) = 0.138$, which is well within the weak-disorder limit where perturbation theory is applicable. We identify this point as a junction point, where the depinning transition line and the roughening transition line meet. For stronger disorder, the pinned contact line presumably goes directly into the Landau-Levich phase: the edge of the liquid drop is pinned so strongly that it remains still when the liquid wedge starts to move upon decreasing the contact angle, and thus a film is left behind. In this sense, at the dashed line in fig. 2 nothing really happens to the contact line itself, while the body of the liquid drop is “depinned” [16].

One can use a scaling argument to account for the power law in eq. (2). If we take the above KPZ equation and make the scale transformations $t \rightarrow \sigma t$, and $h \rightarrow \sqrt{D/\sigma}h$, the coefficients of the linear terms as well as the strength of the noise term will be set to one, and the only remaining coupling constant (the coefficient of the nonlinear term) will have the form $\lambda D^{1/2}/\sigma^{3/2}$. We expect the roughening transition to take place when this coupling constant is of order unity, which yields eq. (2) when the values $D \sim g^2$, $\sigma \sim \left(\frac{\theta_e}{\theta_c} - \frac{1}{\sqrt{3}}\right)$, and $\lambda \sim \text{const}$ are used.

A plausibility argument can be made to explain why the onset of leaving a Landau-Levich film should correspond to a roughening transition. At the threshold of the onset, the edge of the liquid drop suddenly prefers to stand still and stick to the substrate, as opposed to moving with the liquid, and it thus has to roughen to minimize its energy on the disordered solid. Note, however, that this is only a crude picture and one needs to know more about the microscopic features of the film and its coexistence with the moving liquid [16].

Due to the specific form of the noise term η , our approach is only valid when the system is away from the depinning transition, and thus it is not clear if we can trust the prediction of our theory in the vicinity of the junction point. In particular, how the two phase boundaries merge at the junction point, and the nature of the third boundary (dashed line in fig. 2) remain to be clarified [16]. Furthermore, the above calculation is only restricted to the case of small contact angles where the dissipation is dominated by the singular hydrodynamic flow near the tip of the liquid wedge, and also to length scales below the capillary length. Should other mechanisms of dissipation become important [17], and for the length scales that exceed the capillary length, the above picture would be altered [16, 18].

We finally mention that this work can hopefully motivate two types of experiments. One can try to probe the relaxation of a moving contact line, similar to the Ondarcuhu-Veyssie experiment on a static contact line [4], and measure the velocity dependence of the dispersion relation. This dependence could be used to determine the dominant dissipation mechanism [18]. It is also interesting to systematically study the onset of leaving a Landau-Levich film for receding contact lines on a disordered substrate using video microscopy techniques. In particular, it would be interesting to look for a roughening of the contact line before the Landau-Levich film is formed, and measure the corresponding roughness exponents.

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We are grateful to J. BICO, R. BRUINSMAN, P. G. DE GENNES, M. KARDAR and D. QUÉRÉ for invaluable discussions and comments. This research was supported in part by the National Science Foundation under Grants No. PHY94-07194 and DMR-98-05833.

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Europhys. Lett., **57** (2), p. 304 (2002)

Erratum

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(*Europhys. Lett.*, **55** (2), pp. 228–234 (2001))

PACS. 68.08.Bc – Wetting.

PACS. 68.08.-p – Liquid-solid interfaces.

PACS. 05.40.-a – Fluctuation phenomena, random processes, noise, and Brownian motion.

In our work, we should have used only the dissipation due to the *normal* component of the velocity to balance the work done by interfacial forces, as first noted by Blake and Ruschak [1]. To correct for this error, we should use

$$P = \int dx \sqrt{1 + (\partial_x h)^2} \left(\frac{3\eta\ell}{\theta(x, t)} \right) \left[\frac{v + \partial_t h(x, t)}{\sqrt{1 + (\partial_x h)^2}} \right]^2$$

instead of the expression on line 6 of page 231. Consequently, we obtain $\lambda(q, k - q) = cq(k - q) + 3c|q||k - q| + (c - v)|k|(|q| + |k - q| - |k|)$ in eq. (1), a modified eq. (2) as

$$\frac{\theta_c}{\theta_e} = \frac{1}{\sqrt{3}} + \frac{(5\pi/6)^{1/3}}{\sqrt{3}} \left(\frac{g}{\gamma\theta_e^2} \right)^{2/3}$$

and a corrected governing dynamical equation (eq. (3)) as

$$\frac{v + \partial_t h(x, t)}{\sqrt{1 + (\partial_x h)^2}} = \frac{\gamma}{6\eta\ell} \theta(x, t) [\theta_e^2 - \theta(x, t)^2] .$$

The values of the exponents reported on line 15 of page 233 should now read $z = \frac{8}{5} + \frac{1}{10} \left(\frac{9\pi}{50} \right)^{1/3} \left(\frac{g}{\gamma\theta_e^2} \right)^{2/3}$ and $\zeta = \frac{2}{5} - \frac{1}{10} \left(\frac{9\pi}{50} \right)^{1/3} \left(\frac{g}{\gamma\theta_e^2} \right)^{2/3}$, and the location of the junction point reported on line 19 of page 233 is now given as $\theta_t/\theta_e = 0.911$ and $g_t/(\gamma\theta_e^2) = 0.0594$.

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