NONLINEAR SIMULATION OF THIN LIQUID FILM FLOW ON INCLINED PLANES

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ABSTRACT

Stability and dynamics of a Newtonian liquid film flowing down an inclined plane is investigated using a nonlinear formalism. For thin supported film having thickness ≤ 100 nm, the long-range intermolecular forces assume significance over the viscous forces. A two-dimensional flow, modeled as a continuum is represented by Navier-Stokes equations, equation of continuity and associated boundary conditions. The excess force due to van der Waals attraction between the film fluid and the solid support is substituted for the body force term besides the gravity, in the Navier-Stokes equation. The governing equations are rescaled and simplified under long wave approximation to arrive at a strongly nonlinear equation of evolution for the film interface, h(x,t). A linear theory gave an estimate of the film rupture time. The nonlinear simulations revealed various interesting aspects of film dynamics and a true time of film rupture. Important findings are: For thin films ($h_0 \leq 100$ nm), van der Waals force controls the film dynamics while gravity controls flow in thick films ($h_0 > 100$ nm). Degree of inclination does not influence the rupture in thin films but it does affect thick films. The film rupture time increases with an increase in the film thickness and a decrease in the initial amplitude of perturbation. The linear theory may overestimate or underestimate the time of rupture by several orders of magnitude.

Keywords: *Thin falling film flows, long range intermolecular interactions, van der Waals force, nonlinear stability*

الملخص

ستتم دراسة استقرار وديناميكية طبقة من سائل نيوتوني يسيل على مستوى منحدر باستعمال نظريات لاخطية. من المعلوم نه بالنسبة إلى الطبقة الرقيقة المدعمة التي لايتجاوز سمكها 100 nm فإن القوى الطويلة المدى المتواجده بين الجزيئات تحتل أهمية اكبر من قوى اللزوجة. ستم دراسة سريان ثنائي الأبعاد من خلال نمذجة كوسط مستمر خاضع لعلاقـات Navier-Stokes بالإضافة إلى علاقة الاستمرارية والشروط الحدية المرافقة لهما. يتم تعديل العلاقـات بحيـث يـتم استبدال قوى الجسم والجاذبية بقوى الزيادة الناتجة عن تجاذب van der Waals الموجود بين طبقة السـائل والجسـم

الصاب الداعم. سيتم إعادة تقييس العلاقات الحاكمة وتبسيطها في اطار التقريب المعروف بالموجة الطويلة للحصـول على معادلة عالية اللاخطية لتطورسمك الطبقة h (x,t). ان النظرية الخطية تعطي قيمة تقريبية للزمن اللازم لتفكـك الطبقة. أما المحاكاة الغير الخطية فإنها أظهرت جوانب مهمة من دينامية الطبقة وايضا الحساب الدقيق لزمن التفكـك. ومن النتائج المهمة للبحث انه إذا كان السمك لايتعدى 100 nm فإن قوى van der Waals تتحكم في دينامية الطبقـة بينما قوى الجاذبية تتحكم في السريان في الطبقة السميكة (>100nm). ان درجة الانحناء لاتؤثر في التفكك في الطبقة الرقيقة بعكس الطبقة السميكة. ان زمن التفكك يزيد مع زيادة سمك الطبقة وينقصـان مـدى الاضـطراب الاولـي. ان النظرية الخطية قد تعطى قيما تقريبية لزمن التفكك تزيد او تنقص كثيراً عن الزمن الحقيقي للتفكك.

1. INTRODUCTION

The stability and dynamics of thin liquid films, in general are of immense scientific and technological importance. Their applications span a wide spectrum ranging from coatings, microelectronic deposition, flotation, foams, emulsions and thin film lubrications, to a wide range of nano-scale phenomena such as wetting, adhesion, interface heat and mass transfer, multilayer adsorption, heterogeneous nucleation and instability related to biological surfaces and membranes (Jameel, 1994; Sharma & Jameel, 1992; Sharma & Khanna, 1999). The stability of thin fluid films is essential in applications such as in coatings (paints), photographic films and in microelectronic devices -insulating layers (Reiter et al., 1999). Characteristics of thin fluid film are significantly different than its bulk phase because of its large surface-to-volume ratio, in which case interfacial properties become increasingly important (Reiter et al., 1999). When a liquid layer flows downwards, it is subjected to surface-wave instability, which causes thinning of the wave trough. Even in the absence of other form of instabilities, a thin wave trough is susceptible to long range van der Waals attractions, which may eventually lead to film breakup.

The isothermal falling film was first studied using linear theory by Yih (1955, 1963) and Benjamin (1957). They characterized the linear stability of film as a function of Reynolds number and the angle of inclination. A nonlinear approach was first employed by Benny (1966) for isothermal laminar flow on an inclined plane, and he derived a nonlinear equation of evolution for the film interface. The work was later extended by other investigators as reported by Joo et al. (1991) and Oron et al. (1997). Joo et al. (1991) studied the nonlinear stability of non-isothermal two-dimensional films flowing down an inclined plane. This is an extension of the previous work of Burelbach et al. (1988) on the stability of evaporating/condensing film on horizontal plane. Later Joo and Davis (1992a, 1992b) studied nonlinear stability of *three-dimensional* gravity flows in falling thin film. It is important to note here that works of Joo et al. (1991) and Joo and Davis (1992a, 1992b) did not account for the role of van der Waals forces in their model. We have shown here that van der Waals attraction assumes significance as the film thickness decreases during the course of evolution of the instability, and therefore can not be completely ignored.

In what follows, we address the role of long range van der Waals interaction on the stability characteristics of a two-dimensional liquid film flowing down an inclined plane. A strongly nonlinear equation of evolution of the film interface is derived assuming the disturbance wavelength much larger than the film thickness. The instability and rupture of the falling film is quantified using few selected results from numerical simulations and are compared with the linear theory results.

2. MODEL

Consider an isothermal Newtonian liquid film of constant density, ρ, and viscosity, μ, flowing down a plane inclined at an angle θ to the horizontal (Figure 1). The film of initial thickness, h_0 is bounded at the free surface by a passive gas and is laterally unbounded.

The flow in the film may be described by the Navier-Stokes(N-S) equation. The excess force due to long-range van der Waals interactions between the film fluid and the solid substrate is substituted for the body force in the N-S equation. For two dimensional (*X,Z*) motion in the liquid film, the modified Navier-Stokes equations, the equation of continuity, and the associated boundary conditions (Williams and Davis, 1982; Joo et al., 1991; Jameel, 1994; Atieh, 2001) can then be expressed in a *non-dimensional* Cartesian coordinate system (*x, z*). The following scale factors are used for the transformation: length ~ h_0 , time ~ h_0^2/ν , velocity ~ ν/h_0 , pressure ~ $\nu^2\rho/h_0^2$, where h_0 is the mean thickness of the liquid layer, and ν is the kinematics viscosity of the film fluid.

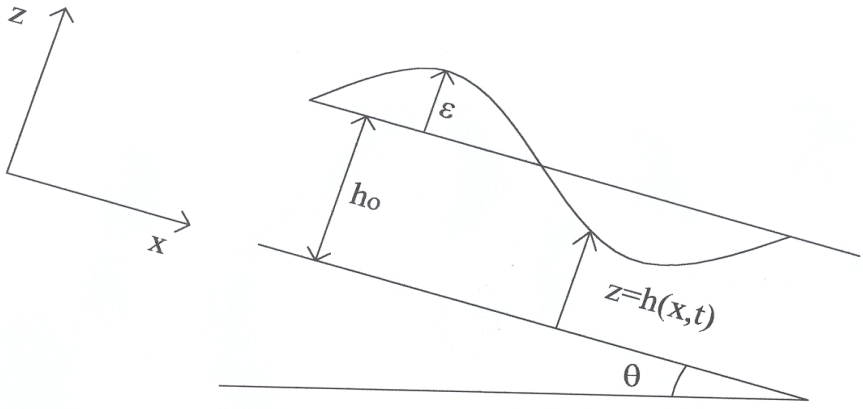

Figure 1: The physical configuration of thin layer flowing down an inclined plane.

N-S Equations:

$$u_t + uu_x + wu_z = u_{xx} + u_{zz} - p_x + G\sin\theta - \phi_x \quad (1)$$

$$w_t + uw_x + ww_z = w_{xx} + w_{zz} - p_z - G\cos\theta - \phi_z \quad (2)$$

where the suffix stands for the derivatives. u and w are the velocity components in the x and z direction, respectively, p is the pressure inside the film, and ϕ is the excess pressure due to intermolecular interactions; all in non-dimensional form defined by:

$$x=X/h_0 \ , z=Z/h_0 \ , t=T\nu/h_o^2$$

$$u=U\,h_0/\nu \ , w=Wh_0/\nu, \ p=(P-P_g)\,h_0^2/\nu^2\rho \ , \phi=\Phi\ h_0^2/\rho\nu^2$$

where the capital letters corresponds to dimensional quantities and Φ is the dimensional counterpart of ϕ .In Eqs. (1) and (2), G (non-dimensional) is defined as,

$$G=\frac{h_0^3 g}{\nu^2}$$

where g is the acceleration due to gravity (dimensional).

The potential function ϕ, due to the long range van der Waals attraction is proportional to the reciprocal third power of the local film thickness, $h(x,t)$,

$$\phi = Ah^{-3}$$

The constant A(non-dimensional) is related to the (dimensional) Hamaker constant A' by

$$A = \frac{A'}{6\pi h_0 \rho \upsilon^2}$$

Equation of continuity:

$$u_x + w_z = 0 \quad (3)$$

Boundary Conditions:

(i) No-slip at the solid-liquid interface:

$$u=w=0 \quad (4)$$

(ii) Kinematic condition at the free surface, $z=h(x,t)$:

$$w = h_t + uh_x \tag{5}$$

(iii) Shear stress condition at the free surface, $z=h(x,t)$:

$$(u_z+w_x)(1-h_x^2)-2h_x(w_z-u_x)=0 \tag{6}$$

(iv) Normal stress condition at the free surface $z=h(x,t)$:

$$p-2[u_x(h_x^2-1)-h_x(u_z+w_x)]/N^2 = -3Sh_{xx}/N^3 \tag{7}$$

where $h(x,t)$ is the local thickness of the layer, and $N=(1+h_x^2)^{1/2}$. The non-dimensional mean surface tension, S is defined as,

$$S=\frac{\sigma_0 h_0}{3\rho v^2}$$

3. LONG WAVE APPROXIMATION

In many practical situations, interfacial instability is locally generated and the disturbance wavelength, λ is much larger than the thickness of the layer, h_0, i.e., $\lambda >> h_0$. We, therefore, focus our interest on flows with a characteristic length in the x-direction proportional to the disturbance wavelength, λ. Then the governing equations can be rescaled consistent with a lubrication type approximation using transformations,

$$\varsigma = kx \ , \qquad \tau = kt \tag{8}$$

We follow the long wave reduction procedure of the governing equations as employed by previous workers (Williams and Davis, 1982; Burelbach et al., 1988; Joo et al., 1991; Sharma and Jameel, 1993; Jameel, 1994). This finally leads to the solution of the Navier-Stokes equation for velocity fields, which together with the kinematic boundary conditions yields the nonlinear equation describing spatio-temporal evolution of the film interface, $h(\varsigma, \tau)$:

$$h_\tau + Gh^2 h_\varsigma \sin\theta + \varepsilon\left[\frac{2G^2}{15}h^6 h_\varsigma \sin^2\theta - \frac{1}{3}Gh^3 h_\varsigma \cos\theta + S'h^3 h_{\varsigma\varsigma\varsigma} + \frac{A}{h}h_\varsigma\right]_\varsigma = 0 \tag{9}$$

which, in terms of original (nondimensional) coordinates (x,t), using Eq. (8) gives

$$h_t + \mathrm{Re}\, h^2 h_x + \left[\frac{2\,\mathrm{Re}^2}{15}h^6 h_x - \frac{1}{3}Gh^3 h_x \cos\theta + Sh^3 h_{xxx} + Ah^{-1}h_x\right]_x = 0 \tag{10}$$

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where Re is a Reynolds number defined as,

$$\mathrm{Re} = G\sin\theta$$

4. LINEAR STABILITY ANALYSIS

The initial (short time) evolution of the film profile is adequately described by linearized equation whenever the amplitude of initial disturbance is much smaller than the mean film thickness. Linearization of Eq. (10) about the mean film thickness, $h=1$, leads to the following solution,

$$h=1+\varepsilon\, sin(kx)\, exp(\omega t) \tag{11}$$

where ε is the non-dimensional initial disturbance amplitude. The resultant dispersion or characteristic equation, gives the relationship between the disturbance growth rate, ω, and the wave number, k, as

$$\omega = k^2\left[\frac{2}{15}G^2\sin^2\theta - \frac{1}{3}G\cos\theta - k^2 S + A\right] \tag{12}$$

Therefore, the film becomes unstable, viz., ω>0 only when $k<k_n$, where k_n is a critical wave number.

For neutrally stable wave (i.e., $\omega=0$), the critical wave number, k_n is given by,

$$k_n^2 = \frac{1}{S}\left[\frac{2}{15}G^2\sin^2\theta - \frac{1}{3}\quad G\cos\theta + A\right] \tag{13}$$

The maximum growth rate, ω_m of the linear waves occurs for the dominant wave number, k_m which is obtained by setting $d\omega/dk$ =0 from equation (12). Thus,

$$k_m^2 = \frac{k_n^2}{2} = \frac{1}{2S}\left[\frac{2}{15}G^2\sin^2\theta - \frac{1}{3}\ G\cos\theta + A\right] \tag{14}$$

Thus, the linear theory predicts unhindered growth of surface deformations (ω>0) upto the point of film rupture whenever $0<k<k_n$ as given by Eq. (13). It is obvious that the linear theory assumes a constant force during all stages of film deformation, whereas in reality, thinner and thicker portions of the film encounter different (nonlinear) force fields as deformations grow.

In order to assess the role of nonlinearities, we may take the linear theory seriously up to the point of film breakup, and determine a time of rupture by setting $h=0$ at $\sin(kx)=-1$ in the Eq. (11). This gives an estimate for the time of film rupture, for a disturbance of wavenumber, k

$$t_L = \frac{1}{\omega(k)} \ln \frac{1}{\varepsilon} \tag{15}$$

The shortest time of rupture from the linear theory corresponds to the dominant wave, i.e., $\omega=\omega_m$ *or* $k=k_m$.

5. NUMERICAL SOLUTION

The nonlinear equation of evolution [Eq. (10)] for the film interface, $h(x,t)$ is solved numerically using Finite Difference discretization in conservative form as part of an initial-value problem for spatially periodic solution on the fixed interval $0<x<2\pi/k$. Mid-point Crank Nicholson rule with forward difference in time and central difference in space are employed.

The following set of periodic boundary condition over a wavelength, $\lambda\ (=2\pi/k)$ are used:

$$(\partial^i h/\partial x^i)_{x=0} = (\partial^i h/\partial x^i)_{x=\lambda} \quad (i = 0,1,2,3); \qquad 0 \le x \le \lambda \tag{16}$$

and a space periodic initial condition was chosen as,

$$h(0,x)=1+\varepsilon \sin kx. \qquad (|\varepsilon|<1) \tag{17}$$

The resulting difference equations are nonlinear coupled algebraic equations, which are solved by an iterative procedure using IMSL subroutine DNEQNF and DNEQNJ. These subroutines employs Levenberg-Marquardt algorithm, a variation of Newton's method, for solving nonlinear algebraic equations. A finite difference approximation of the Jacobian is used in DENEQNF, and a user supplied Jacobian in DNEQNJ (IMSL Maths Library Manual, 1997).

6. RESULTS AND DISCUSSIONS

As noted earlier, the linear theory predicts that the film is unstable to infinitesimal perturbations with wavelength, $\lambda>\lambda_n$ or having wave-number, $k<k_n$. However, linear theory results are only good for small initial amplitudes, ε. True dynamics of film deformation and rupture is best represented by the numerical solution of the nonlinear equation of evolution [Eq. (10)]. Selected results from nonlinear simulations are discussed to elucidate the stability characteristics of inclined film flows. The results presented in the following are for water films on solid substrates.

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Linear theory predicts that the minimum time of rupture occurs at around wave-number of 0.707. There is no significant effect of film thickness and the angle of inclination of the plane (results not shown) on the parameters like dominant wave-number, k_m, maximum growth coefficient, ω_m and the rupture time, t_L, all from the linear theory, as evident from Tables 1 and 2.

Nonlinear simulations were carried out for a wide range of parameters. In the following, we present some selected results.

Table 1: Rupture time and growth rate as function of wavelength & wavenumber at $\theta=0$ & $h_o=10$ nm from linear theory.

k	Wavelength	T_L	ω_o
0.1	62.85714	232.58435	0.0099
0.2	31.42857	59.96315	0.0384
0.3	20.95238	28.11459	0.0819
0.4	15.71429	17.13233	0.1344
0.5	12.57143	12.28045	0.1875
0.6	10.47619	9.99386	0.2304
0.7	8.97959	9.21403	0.2499
0.8	7.85714	9.99386	0.2304
0.9	6.98413	14.7	0.1539
1.0	6.28571	341966.8	0.00001

$k_n=1.00000$ $k_m=0.70711$ $\omega_{om}=0.25000$ tr=9.21034

Table 2: Rupture time and growth rate as function of wavelength & wavenumber at $\theta=0$ & $h_o=100$ nm from linear theory.

k	Wavelength	T_L	ω_o
0.1	62.85714	232.61574	0.0099
0.2	31.42857	59.9714	0.0383
0.3	20.95238	28.11	0.0818
0.4	15.71429	17.135	0.1343
0.5	12.57143	12.282	0.187
0.6	10.47619	9.995	0.230
0.7	8.97959	9.2164	0.2498
0.8	7.85714	9.997	0.2303
0.9	6.98413	14.97	0.15379
1.0	6.28571	341966.8	0.00001

$k_n=0.99993$ $k_m=0.70708$ $\omega_{om}=0.\ 24997$ tr=9.21156

6.1 Effect of Inclination and Film Thickness

Nonlinear time of rupture (i.e., film rupture time from nonlinear simulations) are plotted as a function of wavelength for different angles of inclination and for different film thicknesses in Figures 2 through 4. There is no significant effect of inclination, θ, on the film rupture time, T_N for thin films ($h_0<100$ *nm)* as shown in Figure 2. However, for relatively thicker films ($h0\geq 100$ *nm),* rupture time, T_N increases with the increase in θ and h_0 as depicted in Figures 3 and 4. The increase in T_N with inclination and film thickness may be attributed to the corresponding increase in film Reynolds number, Re (= G sinθ), which in turn results into higher turbulence in the liquid film, thus delaying the appearance of dry spots, and hence the film rupture. In Figures 2 and 3, it may also be noted that the minimum time of rupture lies very close to $\lambda_m \cong 8.9$, the dominant wavelength from the linear theory. On the contrary, for thick films ($h_0 \geq 200$ *nm*), the minimum in rupture time shifts to a lower value of λ at around 6.9 (as shown in Figure 4 for $h_0=200$ *nm*). That is, nonlinearities select shorter waves for thick layers.

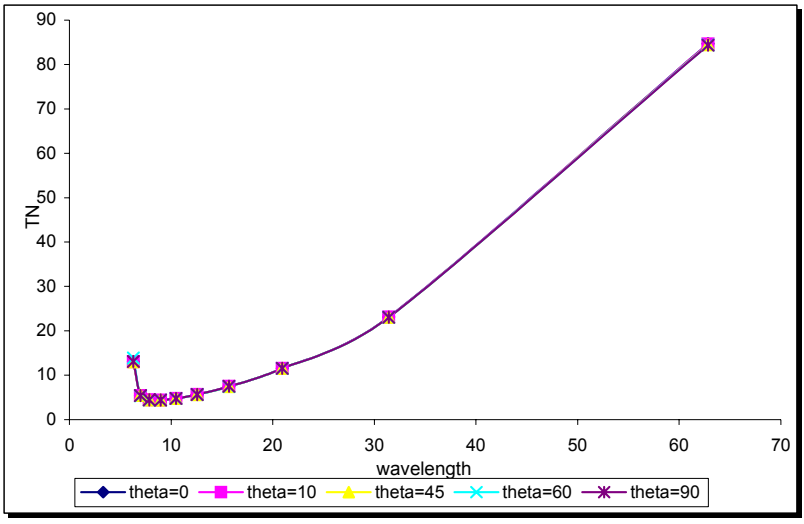

Figure 2: Rupture time as function of wavelength for h_o=50 nm and at different inclinations. Curve for all different θ values converge to a single curve.

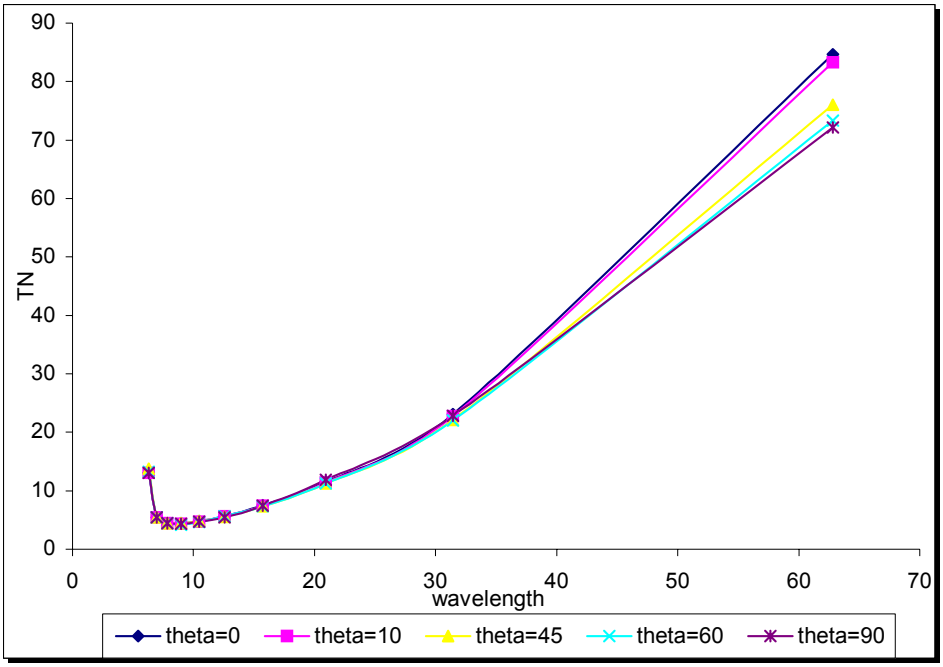

Figure 3: Rupture time as function of wavelength for h_o=100 nm and at different inclinations.

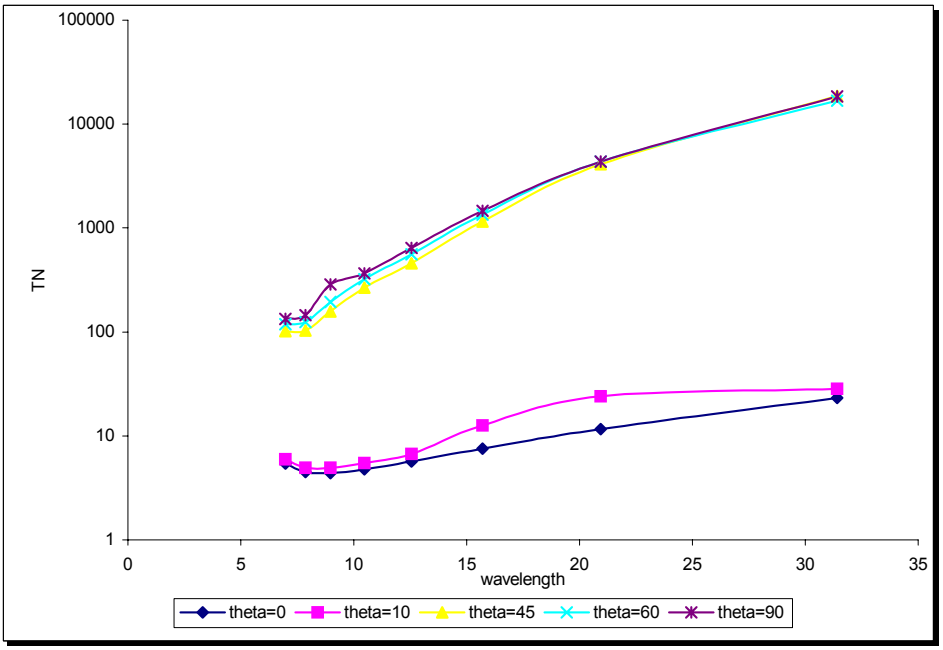

Figure 4: Rupture time as function of wavelength at h_o=200 nm and at different inclinations.

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6.2 Effect of the Amplitude of Disturbance on the Rupture Time

Invariably, the time of film rupture decreases with increasing amplitude of surface disturbance as shown in Figures 5 and 6. As depicted for a film of thickness 10 nm, the rupture time decreases gradually with increasing values of ε. It may be noted that even an infinitesimal perturbation such as of amplitude 1Å (ε =0.01 in Fig. 6) may engender film breakup in an unstable film. This instability is necessarily derived from the attractive van der Waals forces. The increased degree of inclination results into slightly higher rupture time. This is due to the fact that the increase in θ increases the Reynolds number leading to homogenization of fluid film and hence delay in the appearance of dry spots.

6.3 Comparison of Rupture Time from Nonlinear and Linear Theories

The variations of the ratio of rupture times from linear and nonlinear theories (T_N/T_L) are shown as a function of wavelength at different inclinations, θ (Figs. 7 and 8). Figure 7 shows that the minimum deviation between the two predictions occurs around λ_m, wavelength corresponding to minimum rupture time. Further, the influence of inclination becomes pronounced only at larger thickness, i.e., $h_0>50$ *nm*. Figure 8 shows T_N/T_L for thick film of 200 nm. For thick films, at low inclination, the linear theory overestimates the rupture time while at large inclination, linear theory greatly underestimates the time of rupture. This is due to the opposing nonlinear effects of van der Waals and gravity forces at high inclination. Gravity imparts stabilizing effect, which is zero for horizontal ($\theta=0$) film and is maximum for vertical film ($\theta=90°$). On the contrary, van der Waals attraction tend to rupture the film

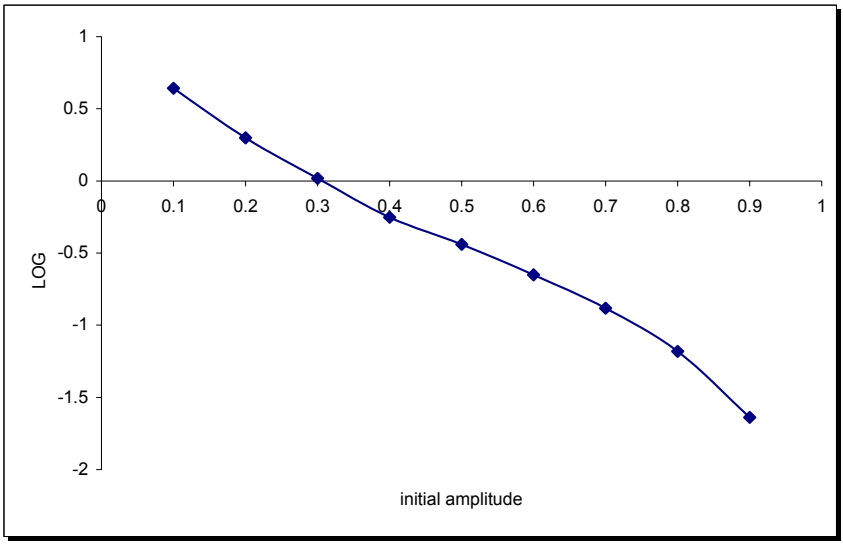

Figure 5: Rupture Time as function of amplitude of perturbation at $\theta=0^o$, h_o=10 nm and k_m=0.7

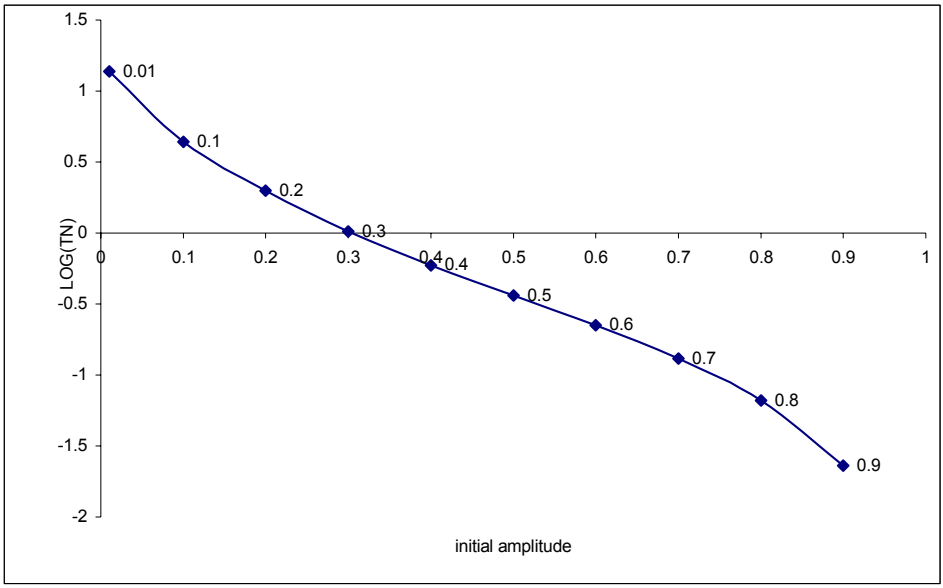

Figure 6: Rupture Time as function of amplitude of perturbation at $\theta=90^{o}$, $h_o=10$ nm and $k_m=0.7$

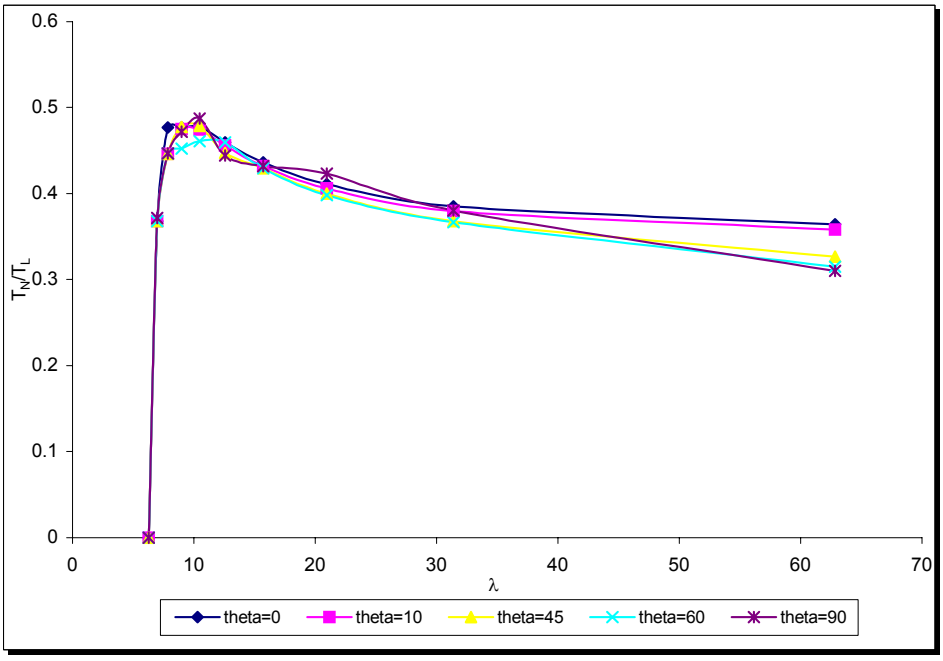

Figure 7: Ratio of rupture time as function of wavelength at $h_o=50$ nm and at different inclinations.

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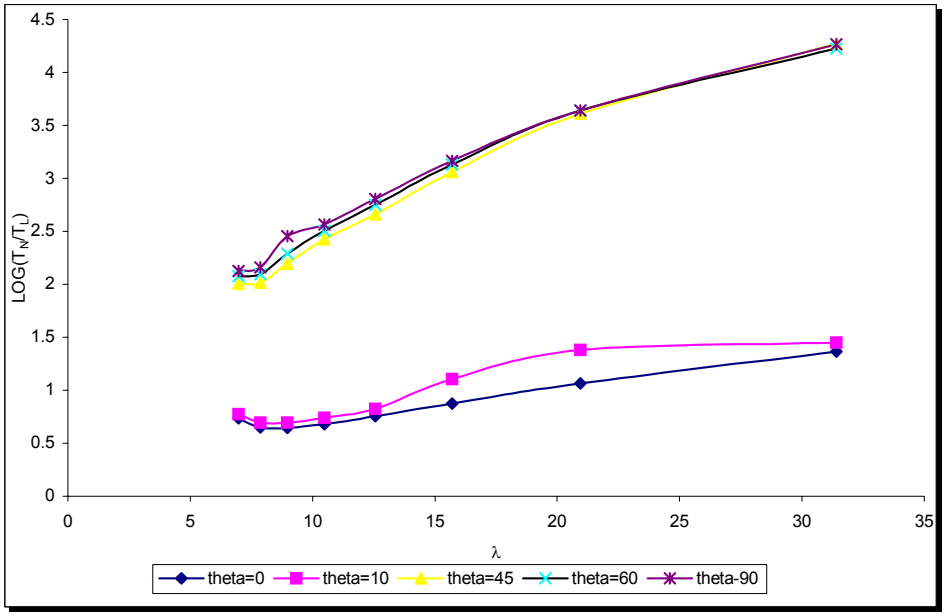

Figure 8: Ratio of rupture time as function of wavelength at h_o=200 nm and at different inclinations.

6.4 Evolution of Film Profile

Finally, Figure 9 shows the evolution of film interface with time. It may be noted that the approach to film rupture is catastrophic after initial (slow) growth of instability. That is, the rate of increase in the gradient of the film profile increases as the film approaches towards rupture and jumps sharply to very high values near the rupture. This behavior is reminiscent of the fact that as the wave troughs get thinner, the van der Waals force takes over the surface tension and gravity effects leading to sharp gradient as *h* decreases.

7. CONCLUSIONS

The role of long range van der Waals interactions on the stability of the gravity flow on an inclined plane has been investigated. Linear theory can predict the fastest growing surface wave and gives an estimate of the time of rupture. It has been shown that the body force due to gravity tends to be insignificant as the liquid layer approaches the thin film dimensions, i.e., $h_0 \leq 100$ *nm*, in which case long range van der Waals interaction controls the film dynamics. In this case, the rupture is rather catastrophic compared to thick films, primarily due to strong van der Waals attraction, which appear in the evolution equation as reciprocal fourth power to the film thickness. On the contrary, for relatively thicker films ($h_0>100$ *nm)*, the van der Waals force becomes insignificant and the force of gravity takes over the film dynamics.

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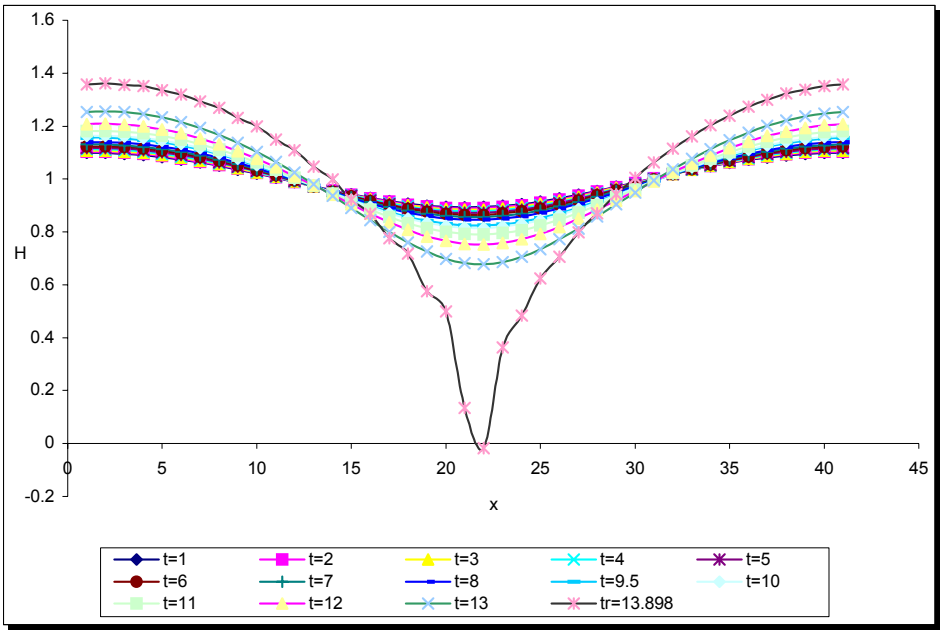

Figure 9: Film profile at different times for van der Waals and gravity system. The initial amplitude is 0.1, h_o=50 nm, θ=60^o & k=1.0. The rupture proceeds explosively at T_N= 13.898.

The fastest growing nonlinear waves are found to have wavelengths close to the predictions of the linear theory for all types of films. It appears that the degree of inclination does not affect the time rupture of the thin film, i.e., $h_0 \leq 100$ *nm.* However, Inclination does affect the growth of instability in thick films ($h_0 > 100$ *nm*). The film rupture time increases with increasing film thickness on inclined planes. The increased degree of inclination and the film thickness results into increased Reynolds number leading to homogenization of fluid film and subsequent delay in the appearance of dry spots. Invariably, the time of film rupture decreases with increasing amplitude of surface disturbance for all film thicknesses.

The deviation between the predictions of nonlinear and linear theories results is a minimum around dominant wavelength. The linear theory may overestimate or underestimate the time of rupture by several orders of magnitude depending upon film thickness. Similar trend was also reported for horizontal films (Jameel, 1994). In essence, linear theory is inadequate to capture all aspects of thin film dynamics, and therefore, a nonlinear formalism is inevitable for the study of thin film stability.

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