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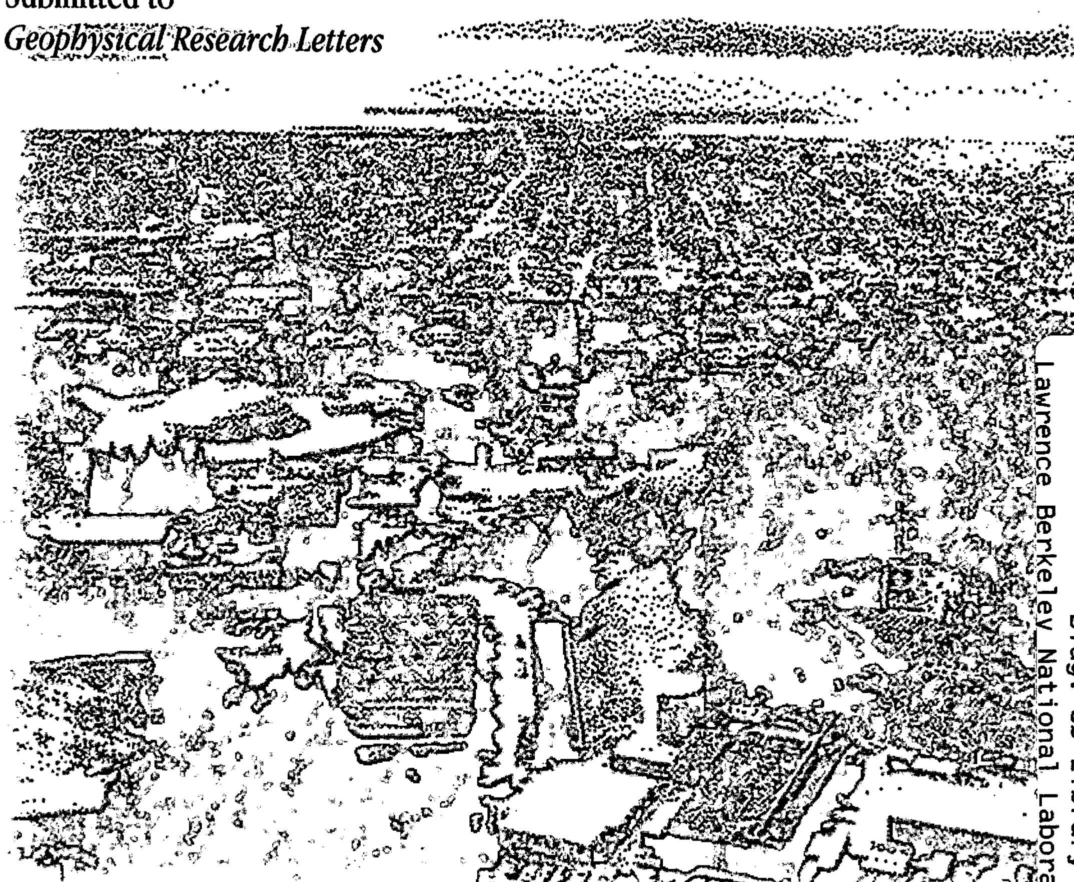

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A MODEL OF CHAOTIC EVOLUTION OF AN ULTRATHIN LIQUID FILM FLOWING DOWN AN INCLINED PLANE

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Abstract

This paper presents the derivation of a one-dimensional evolution equation describing the slow motion (small Reynolds numbers, $R \ll 1$) of a very thin liquid film down an inclined impermeable plane. In this equation, gravitational, capillary, and molecular forces are taken into account. The addition of the molecular force term leads to a highly nonlinear equation governing the spatial and temporal evolution of film thickness. In a weakly nonlinear limit, this evolution equation is rescaled to a canonical form. The latter predicts a chaotic hydrodynamic instability for the film surface. This chaotic behavior is illustrated using the attractors and diagnostic deterministic-chaotic parameters for the variations of the dimensionless film thickness along the coordinate and time.

1. Introduction

A number of laboratory experiments have shown that dripping water (Shaw, 1984) and water flow along a plane (Cheng et al., 1989) exhibit chaotic behavior. At very large Reynolds numbers ($R > 1{,}000$), the high-flow falling films exhibit turbulence (Floryan et al., 1987). At moderately high Reynolds numbers ($300 < R < 1,000$), gravity-capillary hydrodynamic instabilities appear (Chang, 1994). At zero flow rates, the stagnant, thin water film ruptures because of the effect of intermolecular forces (Williams and Davis, 1982). However, for film with a nonzero base flow (of a plain Couette type), Babchin et al. (1983a) showed that within certain limits of the interfacial shear stress, one-dimensional disturbances driven by the molecular forces will fail to break up the film.

Evidence of a chaotic fluctuation of liquid pressure was obtained from flow experiments conducted at Lawrence Berkeley National Laboratory (Berkeley Lab) on fracture replicas and water dripping through capillary tubes (Faybishenko, 1999; Faybishenko et al., 1999). In order to predict liquid flow behavior, one needs to develop adequate mathematical models describing the physics of flow. A better understanding of the physics of flow along the solid surfaces can be obtained by taking into account both molecular and capillary forces arising at the water-solid surface-air interface.

The goal of this paper is to derive a one-dimensional equation describing the slow motion of a very thin liquid film along an impermeable inclined plane, taking into account gravitational, capillary and molecular forces. The paper includes the derivation of a highly nonlinear equation governing the film surface evolution, which in a weakly nonlinear limit is rescaled to a canonical form. Chaotic behavior of the film surface is illustrated using the phase-space attractors and deterministic-chaotic parameters for a dimensionless film thickness.

2. Equation of the Film Surface Evolution

First, we consider a slow (low Reynolds number, $R \ll 1$) one-dimensional motion of a liquid film along the inclined (nonhorizontal) plane as depicted in Figure 1. For a long-wave slow evolution of the lubricating film with negligible inertial effects, we can write the following simplified Navier-Stokes equations as (Babchin *et al.*, 1983b; Frenkel *et al.*, 1987; Middleman, 1995):

$$\mu \frac{\partial^2 V}{\partial y^2} = \frac{\partial P}{\partial x} - \rho g \sin \alpha$$

$$\frac{\partial P}{\partial y} = 0 \qquad (1)$$

where P is liquid pressure (neglecting the hydrostatic pressure due to the normal component of gravity), accounting for molecular (P_m) van der Waals forces and surface tension (capillary forces, P_c), $P = P_m + P_c$; $V = V(x,y,t)$, with V being the velocity, and x and y Cartesian coordinates, t is time, μ is the liquid viscosity; ρ is the liquid density; g is the gravity acceleration; and α is the plane inclination from horizontal. From consideration of the molecular

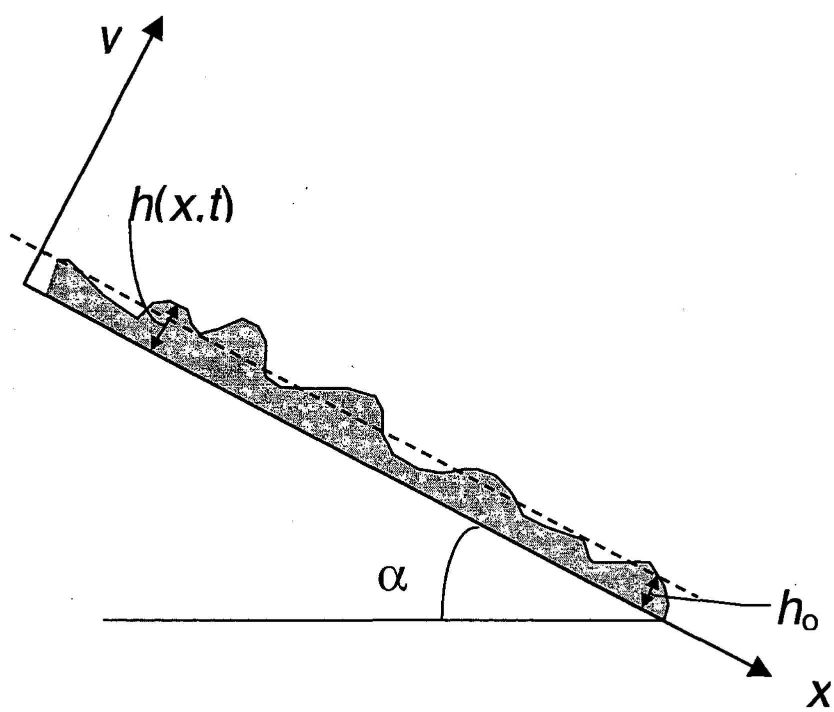


Figure 1. Inclined plane with a disturbed film

interactions between solid, liquid and air, one obtains the following expression for the molecular component of pressure:

$$P_m = \frac{A}{h^3} - \frac{B}{h^n} \qquad (2)$$

where h is the film thickness; n is an integer, $n > 3$; and A and B are coefficients of the Lenard-Jones molecular interaction potential.

The expression for the capillary pressure, P_c, is given by

$$P_c = -\gamma \left[\frac{\partial^2 h}{\partial x^2} \left(1 + \left(\frac{\partial h}{\partial x} \right)^2 \right)^{-3/2} \right] \tag{3}$$

where γ is the liquid-air surface tension, and the expression in brackets expresses the curvature of the liquid surface. For long-wave flow regimes, we have $(\partial h/\partial x)^2 << 1$, and Equation (3) becomes

$$P_c = -\gamma \frac{\partial^2 h}{\partial x^2} \tag{4}$$

Combining the expressions for the molecular (2) and capillary (4) components of pressure, we obtain

$$P = \frac{A}{h^3} - \frac{B}{h^n} - \gamma \frac{\partial^2 h}{dx^2} \tag{5}$$

Note that in Equation (1), $V(x,y,t) = f[h(x,t), y]$, and the boundary conditions for Equation (1) are

$V=0$ for $y = 0$,

and (6)

$\partial V/\partial y = 0$ for $y = h(x,t)$

As is readily verified by the direct substitution, the solution of (1) with boundary conditions (6) is

$$V(x,y,t) = \frac{1}{2}\left(\rho g \sin\alpha - \frac{\partial P}{\partial x} \right) \left(\frac{2hy}{\mu} - \frac{y^2}{\mu} \right) \tag{7}$$

Integration of (7) yields the liquid flux (the flow rate across the area of [1cm × width $h(x,t)$])

$$q = \int_0^{h(x,t)} V(x,y)dy = \left(\rho g \sin\alpha - \frac{\partial P}{\partial x}\right)\frac{h^3}{3\mu} \tag{8}$$

By differentiating Equation (5), we obtain

$$\frac{\partial P}{\partial x} = -\left(\frac{3A}{h^4} - \frac{nB}{h^{n+1}}\right)\frac{\partial h}{\partial x} - \gamma\frac{\partial^3 h}{\partial x^3} \tag{9}$$

Substituting this expression for $\partial P/\partial x$ in (8), we find

$$q = \left[\rho g \sin\alpha + \left(\frac{3A}{h^4} - \frac{nB}{h^{n+1}}\right)\frac{\partial h}{\partial x} + \gamma\frac{\partial^3 h}{\partial x^3}\right]\frac{h^3}{3\mu} \tag{10}$$

For an incompressible fluid, the equation of mass balance for film flow is

$$\frac{\partial h}{\partial t} + \frac{\partial q}{\partial x} = 0 \tag{11}$$

From (10) and (11), we obtain the following highly nonlinear equation of the evolution of the film surface:

$$\frac{\partial h}{\partial t} + \frac{\rho g \sin\alpha}{\mu} h^2 \frac{\partial h}{\partial x} + \left[\left(\frac{3A}{h^4} - \frac{nB}{h^{n+1}}\right)\frac{\partial h}{\partial x} + \gamma\frac{\partial^3 h}{\partial x^3}\right]\frac{h^2}{\mu}\frac{\partial h}{\partial x} +$$

$$\left[\gamma\frac{\partial^4 h}{\partial x^4} - \left(\frac{12A}{h^5} - \frac{n(n+1)B}{h^{n+2}}\right)\left(\frac{\partial h}{\partial x}\right)^2 + \left(\frac{3A}{h^4} - \frac{nB}{h^{n+1}}\right)\frac{\partial^2 h}{\partial x^2}\right]\frac{h^3}{3\mu} = 0 \tag{12}$$

Equation (12) is a further development related to the problem of flow-induced nonlinear effects on the instabilities of thin liquid films (Babchin et al., 1983a,b; Frenkel and Indireshkumar, 1996).

3. Derivation of a Canonical Equation

Equation (12) can be reduced to a canonical form using the following renormalization

$$h = h_o(1 + F) \tag{13}$$

where h_o is the mean film thickness. Assuming that F is small, $F \sim \varepsilon \ll 1$, and substituting (13) in (12), we obtain to the order of F^2

$$\frac{\partial F}{\partial t} + \frac{\rho g \sin \alpha h_o^2}{\mu}\frac{\partial F}{\partial x} + \frac{2\rho g \sin \alpha h_o^2}{\mu} F \frac{\partial F}{\partial x} + \frac{\gamma h_o^3}{3\mu}\frac{\partial^4 F}{\partial x^4} + \left(\frac{3A}{h_o^4} - \frac{nB}{h_o^{n+1}}\right)\frac{h_o^3}{3\mu}\frac{\partial^2 F}{\partial x^2} = 0 \tag{14}$$

In the frame of reference moving with an unperturbed film surface, a new coordinate z is given by

$$z = x - \frac{\rho g h_o^2 \sin \alpha}{\mu} t \tag{15}$$

so that Equation (14) becomes

$$\frac{\partial F}{\partial t} + \frac{2\rho g \sin \alpha h_o^2}{\mu} F \frac{\partial F}{\partial z} + \frac{\gamma h_o^3}{3\mu}\frac{\partial^4 F}{\partial z^4} + \left(\frac{3A}{h_o^4} - \frac{nB}{h_o^{n+1}}\right)\frac{h_o^3}{3\mu}\frac{\partial^2 F}{\partial z^2} = 0 \tag{16}$$

Equation (16) can be rescaled using the following dimensionless variables:

$$
\begin{aligned}
F / F_0 &= \phi \\
t / t_0 &= \tau \\
z / L_0 &= \tilde{x}
\end{aligned}
\tag{17}
$$

where F_0, t_0, and L_0 are the characteristic amplitude, time, and length, respectively, which are determined from the following system of equations:

$$\frac{2g\rho h_0^2 \sin\alpha}{\mu} \frac{t_0 F_0}{L_0} = 1 \tag{18a}$$

$$\frac{\gamma h_0^3}{3\mu} \frac{t_0}{L_0^4} = 1 \tag{18b}$$

$$\left(\frac{3A}{h_0^4} - \frac{nB}{h_0^{n+1}} \right) \frac{h_0^3}{3\mu} \frac{t_0}{L_0^2} = 1 \tag{18c}$$

Hence, Equation (16) for the surface evolution obtains the following canonical form (Sivashinsky and Michelson, 1980):

$$\frac{\partial \phi}{\partial \tau} + \phi \frac{\partial \phi}{\partial \tilde{x}} + \frac{\partial^2 \phi}{\partial \tilde{x}^2} + \frac{\partial^4 \phi}{\partial \tilde{x}^4} = 0 \tag{19}$$

Sivashinsky and Michelson (1980) were the first who indicated that the deterministic equation (19) leads to a chaotic behavior. The numerical solution of Equation (19) (see below) shows that ϕ is of the order of unity, as are the dimensionless characteristic time scale and length scale

$$\frac{\partial}{\partial \tau} \sim 1 \sim \frac{\partial}{\partial \tilde{x}}$$

With the characteristics scales (18) being known, every term in the full Navier-Stokes problem neglected in the derivation of Equation (19) can be estimated to be actually small as compared to the retained terms, provided the system parameters satisfy the constraints corresponding to the requirements $L >> h_0$, $F_0 << 1$, and (in order for the inertial forces to be negligible as compared to the van der Waals forces)

$$\frac{A}{h_0^7 \rho g^2 \sin^2\theta} >> 1$$

(A detailed discussion of such constraints, or the validity conditions for the derivation of the film evolution equations, can be found in Frenkel and Indireshkumar, 1996).

Assuming $A \sim 10^{-13}$ ergs (Williams and Davis, 1982), one can see that Equation (19) can be used to describe the evolution of a film with a thickness less than 10^{-4} cm for planes that are not too close to horizontal. For thicker films, the destabilizing effect of inertia forces (Frenkel and Indireshkumar, 1999) is more important than that of the van der Waals forces.

To take into account the normal component of gravity, the evolution equation has to be modified by adding the hydrostatic stabilizing term $\rho g \cos\Theta$ into every occurrence of the expression $(3A/h^4 - nB/h^{n+1})$ in Equations (12) and (16). For films with a thickness of 10^{-5} cm and thinner, the stabilizing effect of the normal component of gravity is negligible, even for films which are nearly horizontal. Under such conditions, Equation (19) would become inadequate, and, instead, the highly nonlinear equation (12) would have to be used.

It is well known that the solution of Equation (19), which describes nonlinear dynamics of film flow, exhibits chaos in both space and time (Manneville, 1990). Thus, the nonlinear effects of the base flow in cooperation with the destabilizing effect of the van der Waals forces and the stabilizing action of the surface tension lead to a chaotic behavior of the film surface. Furthermore, the film disturbance creates the spatio-temporal fluctuations of the liquid pressure inside the film.

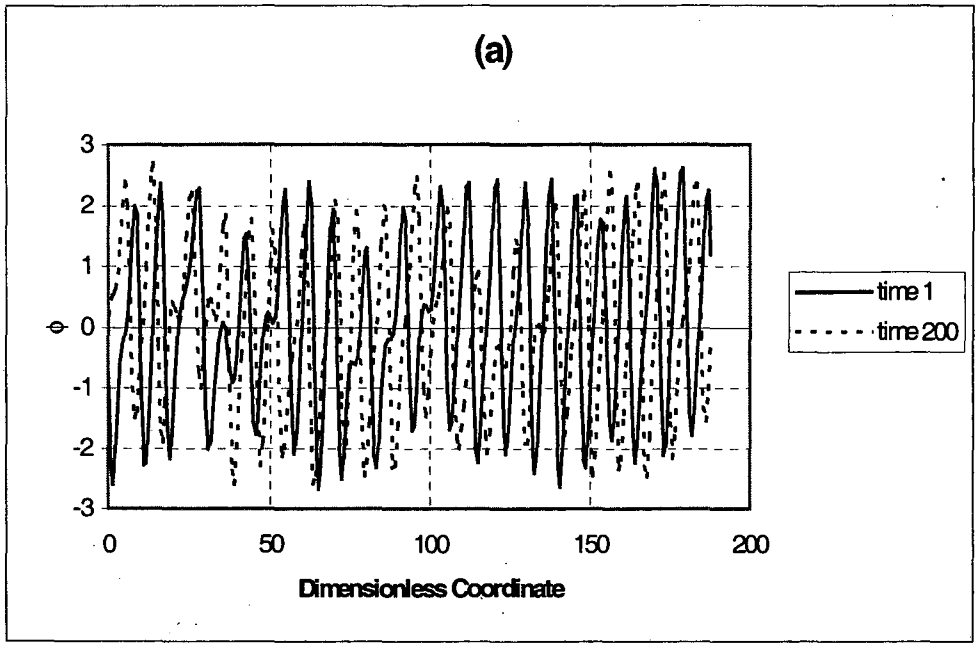
4. Illustration of the Chaotic Behavior Generated Using Equation (19)

Equation (19) was solved with periodic boundary conditions using a Fourier pseudo-spectral method. The spatial derivatives were computed using the discrete Fast Fourier Transform, and a third order explicit Adams-Bashforth method was used for advancing the solution in time. To illustrate the results, Figure 2a shows the variation of the relative film thickness for 256 points along the dimensionless coordinate x for two moments of time. Figure 2b shows the corresponding 3D attractors for the film thickness for these moments of time. It is interesting to note that, despite a significant difference in the changes of the film thickness along the coordinate, the attractors appear to be similar.

Figure 3a shows the variation of film thickness at two points along the dimensionless time coordinate; Figure 3b shows the corresponding 3D attractors. Table 1 summarizes the results of calculations of the time delay using the mutual information function, which was used in plotting the attractors shown in Figure 3b, Global Embedding Dimension, the Lyapunov Dimension, and Lyapunov exponents. Calculations were conducted using the CSPW code (Abarbanel, 1996). The negative value of the sum of Lyapunov exponents, the positive value of the largest Lyapunov exponent, and the presence of well-defined three-dimensional attractors (which have identical patterns in Figure 3b) confirm that chaotic behavior is inherent in the solution of Equation (19) that describes the one-dimensional spatio-temporal fluctuations of the film thickness.

Table 1. Characteristic Parameters of the Chaotic Time Variation of the Film Thickness

Parameters	Point 1	Point 2
Time Delay	6	3
Global Embedding Dimension	3	3
D_{Lyap}	1.154	2.39
Sum of Lyapunov exponents	–0.82	–0.37
Largest Lyapunov Exponent	0.1	0.35



(b)

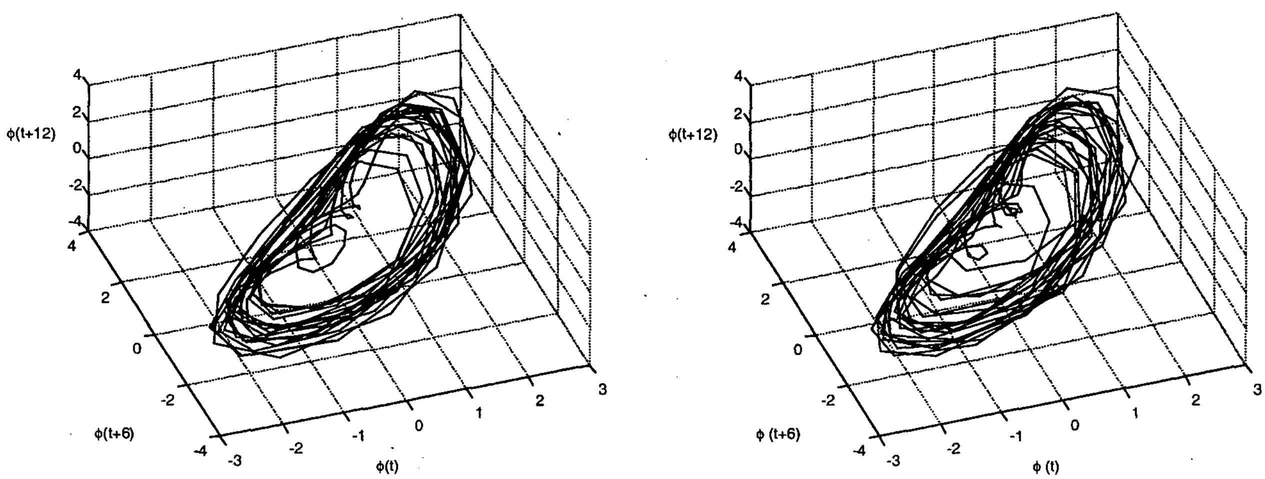
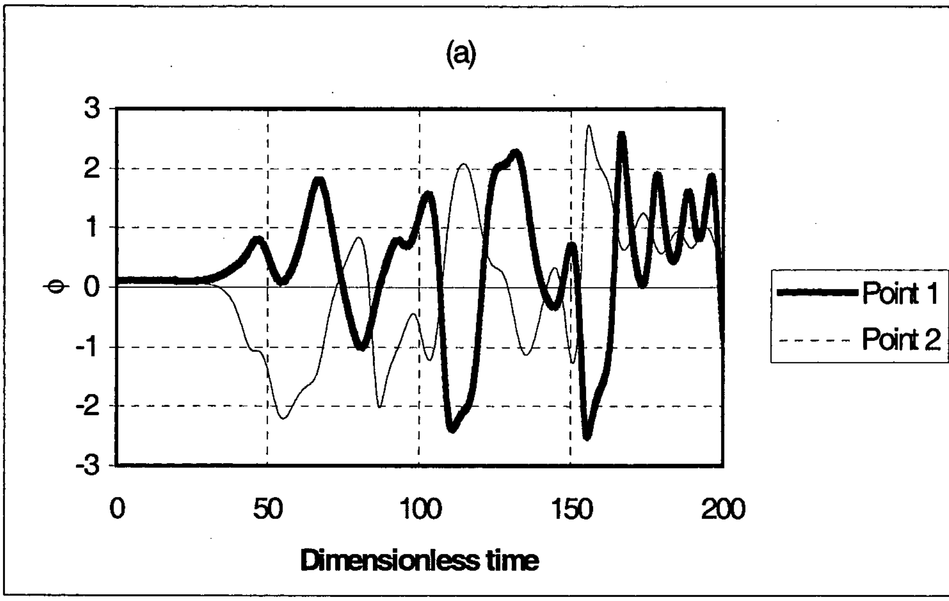


Figure 2. The results of the solution of Equation (19) illustrating (a) the fluctuations of the dimensionless film thickness (ϕ) along the dimensionless coordinate (x) for two dimensionless times, and (b) corresponding 3D attractors for Time 1 (left) and Time 2 (right).



(b)

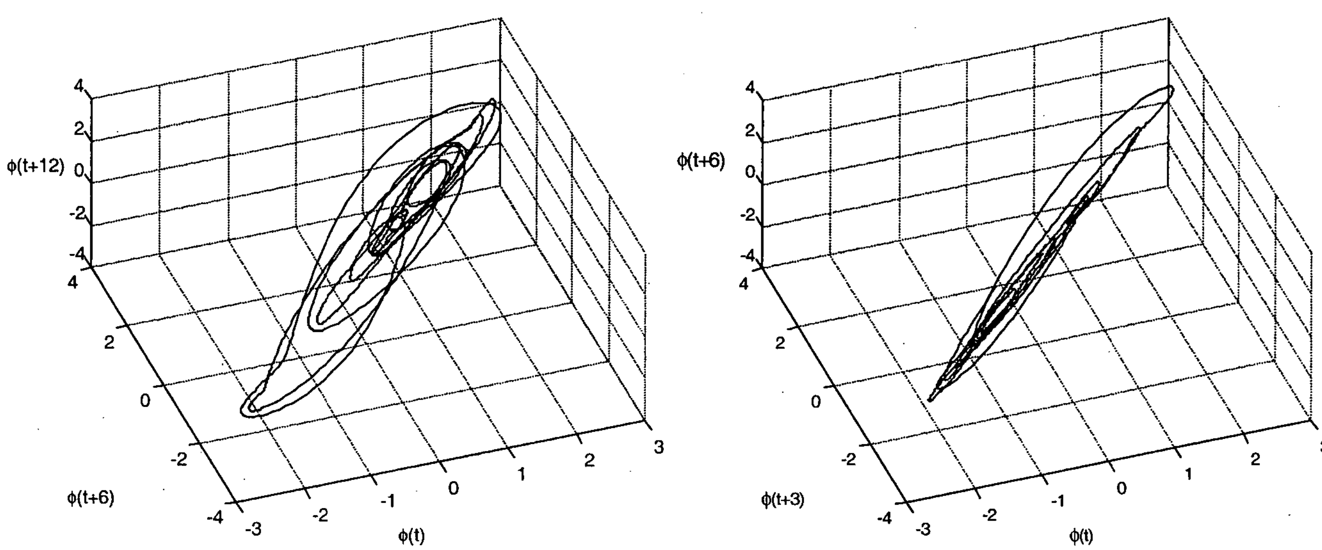


Figure 3. The results of the solution of Equation (19) illustrating (a) the fluctuations of the dimensionless film thickness (ϕ) for two points with dimensionless time, and (b) corresponding 3D attractors for Point 1 (left) and Poit 2 (right).

5. Conclusions and Directions of Future Investigations

The derivation of the evolution equation for the inclined film flow taking into account gravitational, capillary, and molecular forces leads to a canonical form given by Equation (19). The solution of this equation shows a chaotic spatial and temporal behavior of the thickness of the flowing liquid film. The presence of chaos is demonstrated by calculating diagnostic parameters of chaos for the solution of Equation (19). Future research in this area could be aimed at the derivation of (1) two-dimensional equations for the film thickness to describe the water channeling phenomena along the inclined surface, which was observed in fracture replica experiments (Faybishenko et al., 1999); and (2) an equation for a stratified flow of water and an immiscible viscous organic liquid using the approach given by Shlang et al. (1985).

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