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Stability of thin liquid films subjected to ultrasonic vibration and characteristics of the resulting thin solid films

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ABSTRACT

Thin solid films have ubiquitous presence in many existing and emerging technologies. Solution-processed thin solid films may be fabricated by casting a thin liquid film followed by a drying step. It has been shown that by imposing a low-amplitude ultrasonic vibration on the substrate, the physical and structural characteristics of the resulting thin solid films is improved, significantly. Therefore, in this study, to investigate and rationalize the aforementioned findings, the evolution and stability of thin and ultrathin films of Newtonian liquid solutions, subjected to vertical and horizontal ultrasonic vibration is studied, using the long-wave approximation and negligible inertia forces. An explicit criterion is obtained for the film instability. Consistent with established theories, it is found that the vertical vibration tends to destabilize the thin liquid film, although for ultrasonic vibrations with low-amplitude, the term contributing to the perturbation growth rate decays rapidly with time and the film may remain stable. However, the vertical ultrasonic vibration is found as a significant destabilizing force, if the film thickness is near a critical value in which case the destabilizing van der Waals and stabilizing gravity and surface tension forces balance one another. To validate the model, experiments on thin liquid films of dilute polymeric solutions are performed. It is found that while imposing ultrasonic vibration may potentially destabilize and breakup the thin film, imposing a low-power vibration can significantly improve the homogeneity, electrical properties, and uniformity of the film, whereas a large-amplitude vibration may have a detrimental effect, because of excessive mixing and agitation of the liquid film or cracking of the resulting thin solid film.

1. Introduction

The field of thin liquid films and its associated phenomena, such as the formation of surface waves and patterns, wetting/dewetting and instability, is indeed one of the most fascinating areas of fluid mechanics with practical applications (Bonn et al., 2009; Craster and Matar, 2009; Oron et al., 1997). The application of thin liquid films is now extended to emerging solution-processed thin film devices, such as polymer and perovskite thin film solar cells, transistors, sensors, and displays, which incorporate functional thin and ultrathin solid films. The layers of such devices are usually deposited using a casting method to form thin or ultrathin liquid films, which subsequently dry to form functional thin solid films (Eslamian, 2016). However, the common issue in solution-processed thin solid films is the occurrence of undesired phenomena, such as dewetting and pinhole formation, which arise during casting and drying of thin liquid films. At small thicknesses required for such applications, the intermolecular forces may contribute to instability of thin liquid films. In an effort to improve the intactness, uniformity and nanostructure of the solution-processed thin solids films, Eslamian et al. (Zabihi and Eslamian, 2015; Wang and Eslamian, 2016) imposed ultrasonic vibration on thin films of dilute polymer solutions, where remarkable improvement was achieved in almost all characteristics of the resulting thin solid films, as well as in the performance of the solar cells incorporating such thin films (Eslamian and Zabihi, 2015; Xie et al., 2016; Zabihi et al., 2016). This improvement was rather surprising, given that it is known that the vertical vibration has a destabilizing effect on liquid films, albeit under certain conditions such as amplitudes larger than a critical value, on the basis of the Faraday's experiments (Faraday, 1831), also revisited by others in various liquid solutions from inviscid flows to non-Newtonian liquid solutions (Benjamin and Ursell, 1954; Edwards and Fauve, 1994; Miles and Henderson, 1990; Kumar, 1996, 1999; Kumar and Matar, 2002; Matar et al., 2004; Raynal et al., 1999; Sharma, 1993). Then, further experiments showed that indeed if the amplitude of the imposed vertical vibration is excessively large, the resulting thin solid film will be ruptured (Habibi et al., 2016). Others have also used ultrasonic vibration to improve the process or device performance, e.g. Kim et al., 2010; Diemer et al., 2013. Beside vertical

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Nomenclature

A'	Hamaker constant
A_{01}	Lateral vibration amplitude (m)
A_{02}	Vertical vibration amplitude (m)
a_x	Acceleration in lateral direction (m/s^2)
a_z	Acceleration in vertical direction (m/s^2)
c	Mean curvature of the interface (m)
c_1	Coefficient defined in Eq. (24)
c_f	Speed of sound in fluid (m/s)
C	Capillary number
$\overline{D_0}$	Cut-off distance (m)
E	Evaporation term (kg/s^2)
$\vec{f}$	Prescribed force at interface (Pa)
$\overline{F}$	Pull-off force (N)
G	Gravitational acceleration (m/s^2)
$g(t)$	A function defined in Eq. (24)
h	Film thickness (m)
h_w	Wet film thickness (m)
h_0	Mean film thickness (m)
h'_0	Amplitude of perturbation (m)
k	Wave number (1/m)
$\vec{n}$	Unit vector normal to interface
p	Liquid pressure (Pa)
r	Arc length along the interface (m)
$\overline{R}$	AFM probe tip radius (m)
Re	Reynolds number
s	Perturbation growth rate (1/s)
$\vec{t}$	Unit vector tangential to interface
T	Non-dimensional time
t	Time in Eq. (24)
$\boldsymbol{T}$	Stress tensor (Pa)
u	Lateral liquid velocity component (m/s)
w	Vertical liquid velocity component (m/s)
x	Lateral direction (m)
z	Vertical direction (m)

Greek Symbols

β	Slip length (m)
β_0	Dimensionless slip length
Δd	Distance between two transducers (m)
ϵ	Length scale Pressure function (Pa)
λ	Wavelength (m)
μ	Liquid viscosity (kg/ms)
Π	Normal external stress (Pa)
ρ	Liquid density (kg/m^3)
σ	Liquid surface tension (N/m)
Σ	Dimensionless surface tension
ϕ	General external body force potential function (N/m^2)
ϕ_b	Body force potential due to all forces except van der Waals force (N/m^2)
ω_1	Lateral vibration frequency (Hz)
ω_2	Vertical vibration frequency (Hz)
ω	Equal lateral and vertical vibration frequency (Hz)
Ω	Angular velocity or spin speed (rad/s) or revolution per second (RPM)
ν	Solution kinematic viscosity (m^2/s)

vibration, the problem of liquid film excitation by horizontal vibration has been studied, as well (Yih, 1968; Or, 1997), where using the longwave approximation, the domain of instabilities were determined. This work, therefore, focuses on theoretical and experimental study of thin liquid films subjected to horizontal and vertical vibration to elucidate the above-mentioned effects for practical application of ultrasonic vibration on ultrathin liquid films, relevant to emerging thin film devices, mentioned above. We will show that the application of low-amplitude, high frequency ultrasonic vibration results in further simplification of the governing equations.

Most of the works mentioned above on the effect of excitation of liquid film by vertical vibration based on the Faraday's experiment (Benjamin and Ursell, 1954; Edwards and Fauve, 1994; Miles and Henderson, 1990; Kumar, 1996, 1999; Kumar and Matar, 2002; Matar et al., 2004; Raynal et al., 1999; Sharma, 1993) are not usually applicable to traveling waves formed on ultrathin liquid films in which the intermolecular forces are as important as the vibration force. Here, the works that consider vertical or horizontal vibration, as well as some aspects of ultrathin liquid films, such as intermolecular forces and/or dewetting, are briefly reviewed, to justify the need for the present work. The first work that considered the analysis of vertical vibration in thin liquid films at the presence of van der Waals forces, which corresponds to sufficiently thin liquid films, was performed by Kumar and Matar (2002). Their non-linear equations based on the lubrication theory determined that for long-wavelength disturbances (small amplitudes), the vertical vibration does not lead to the formation of standing waves, but for other cases disturbances may be excited by vertical vibration leading to the formation of standing waves. Then later Kumar and Matar (2004) performed a linear analysis on an unbounded liquid films with arbitrary depth to account for surface tension variations due to the presence of an insoluable surfactant, and found that the surfactant has a damping effect, raising the critical vibration amplitude needed to excite the disturbances. In a subsequent work, Matar et al. (2004) considered the above-mentioned problem and included the Marangoni effect, as well, where it was found that the presence of the Marangoni forces makes it more difficult for the vertical vibration to excite the disturbances. In the above-mentioned works (Kumar and Matar, 2004, 2002; Matar et al., 2004), the vibration term made the evolution equation time-dependent, thus the time-dependent solutions based on the Floquet theory were sought and numerical results were presented and discussed. Later, Shklyaev et al. (2008) studied dewetting of thin liquid films subjected to moderate frequency and large-amplitude vertical vibration, compared to the film thickness. The lubrication approximation was used to derive the evolution equation. They concluded that there is a window of frequency-amplitude of vertical vibration which results in film stabilization and suppression of dewetting, even though the vibration amplitude is large. One of their conclusions was that the vertical ultrasound irradiation of the substrate is an effective method of film stabilization. The same authors also examined the application of longitudinal (horizontal) and tilted vibration on the film stability (Shklyaev et al., 2009), under similar conditions stated above in their previous work. Their conclusion was that the horizontal vibration makes the film unstable, unless the frequency is very high. Shklyaev et al. (2015) studied the effect of

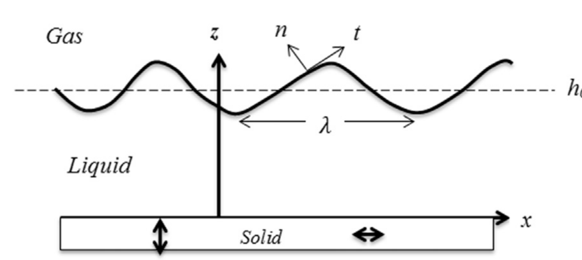

Fig. 1. Schematic of a two-dimensional laterally unbounded liquid film, assuming that the film behavior in the y direction is similar to that in the x direction (laterally infinite system). The solid substrate vibrates both vertically and laterally, imposing vibration on the entire film, as a body force. The film thickness and traveling disturbance wavelength are not to scale. The arrows on the substrate indicate the direction of vibration.