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Spreading and atomization of droplets on a vibrating surface in a standing pressure field

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We report the first observation and analytical model of deformation and spreading of droplets on a vibrating surface under the influence of an ultrasonic standing pressure field. The standing wave allows the droplet to spread, and the spreading rate varies inversely with viscosity. In low viscosity droplets, the synergistic effect of radial acoustic force and the transducer surface acceleration also leads to capillary waves. These unstable capillary modes grow to cause ultimate disintegration into daughter droplets. We find that using nanosuspensions, spreading and disintegration can be prevented by suppressing the development of capillary modes and subsequent break-up. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4757567>]

Surface vibration can induce droplet to experience an upward motion on an inclined surface. If the vertical rocking motion is strong, the contact line will unpin and the motion will depend on the amplitude and frequency of the applied vibrations.¹ The migration of droplet due to surface vibration depends on the contact angle and adhesion force between the liquid and solid surface.^{1,2} A horizontal vibrating surface induces stationary waves on the droplet, which beyond a frequency and intensity, generates satellite droplets.^{3,4} When the initial forcing frequency is above the resonance frequency of the system, complete bursting of the droplet can occur.⁵

Droplet generation through interaction between liquid and acoustic waves has generated interest in large and small scale applications. High intensity acoustic wave induced oscillations in liquid films can create micron sized droplets.^{6–8} Standing acoustic waves can also be used to levitate droplets without disintegration.^{9–12} The standing wave supports the droplet against gravity by creating a non-uniform pressure distribution around the droplet. In the levitator, the acoustic pressure tends to stretch the droplet and viscosity provides the restoring force.

Thus, droplets interacting with a vibrating platform or an acoustic field display various modes of deformation that can lead to atomization. This letter integrates these two ideas for the first time and provides an analysis for droplet spreading. Instead of using a levitator to levitate the droplet, we place the droplet on the emitter surface off-axis. The droplet experiences vibration from the surface, however, it starts deforming and spreading due to the presence of the standing wave in the levitator. As the droplet spreads to the edge of the transducer surface, it begins to eject tiny micron-sized droplets. We also illustrate how changing the droplet viscosity using nanosuspensions^{13,14} allows for the control of the spreading rate of the droplets and enhancement or suppression of atomization. Such manipulation of droplet deformation is useful in enhancing spray-coating, ink-jet printing, and microfluidics. Controlled atomization is also beneficial in sprays, electronics cooling, and in spray drying for pharmaceutical and ceramic applications.

This letter analyzes spreading and disintegration of a droplet placed on the transducer surface of an acoustic levitator. We report observations of droplets on a vibrating transducer surface and verify the results with an analytical model. The circular vibrating surface (tantalum; radius 3.8 mm) is part of a single-axis acoustic levitator. The images of a 1 μ l volume droplet were captured (temporal resolution 50 μ s; spatial resolution 5 μ m) for a transducer frequency of 100 kHz. A vertical standing wave was established in the levitator. Due to the off-centric location of the droplet ($\approx$ 1.8 mm from the levitator axis), it was subjected to an asymmetric pressure field. Several fluid droplets were investigated: (a) water; (b) colloidal nanosilica suspension (particle diameter 20 nm) in water at volume fractions of 0.27 (NS27), 0.1 (NS10), and 0.05 (NS05); (c) glycerol; and (d) aqueous solution of glycerol-water (GW) at volume concentrations of 0.2 (GW2080) and 0.9 (GW9010). The fluid properties used are measured or taken from the literature.^{15–17}

Four regimes of droplet deformation in display [Figure 1] are: (1) small-scale oscillation regime; (2) droplet spreading regime; (3) two-lobe regime; (4) film-spreading/atomization regime. In regime 1, small-amplitude capillary waves corrugate the droplet surface, but the contact line remains pinned, featuring oscillations in the droplet centroid and contact angles. The peak amplitude of the waves is found to be 0.77% of the droplet base length in the case of water [Ref. movie 1 in Figure 2]. The competition between the energy supplied by the transducer surface and the droplet surface energy leads to these capillary waves. In regime 2, the droplet responds to the acoustic field by spreading to the right edge with pinned left contact point. Since the droplet is situated in the pressure antinode at the transducer surface, radial acoustic radiation force, F_r , pushes the droplet radially outwards and axial acoustic radiation force, F_z , exerts an upward pull.¹⁸ Hence, the rightward spreading of the droplet suggests that the droplet distortion is dominated by F_r .

The acoustic field is the main reason for the spreading phenomenon. During droplet spreading, the leftward liquid-solid adhesive force acting on the base of the droplet imposes a no-slip condition at the interface. Hence, the fluid layer adjacent to the surface remains static while the droplet free

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FIG. 1. Different regimes of partially wetting droplets on an oscillating surface in the presence of a standing wave at different time instants (ms) for water (t_W) and glycerol (t_G). $t = 0$ represents the onset of transducer oscillation. (a) Regime 1: Red and white arrows indicate the interfacial and acoustic forces, respectively. Inset (a) depicts the location of the droplet (4) relative to the standing wave (3) created between the transducer surface (1) and the reflector (2). The green and red regions represent pressure antinodes and nodes, respectively. (b) Regime 2: v denotes the radial velocity field within the droplet. (c) Regime 3. (d) Regime 4: High viscosity fluids. Arrow shows the direction of film spreading without atomization. (e) Regime 4: Low viscosity fluids. Left inset (e) demonstrates the amplitude, a and base width, w of a wave ligament. Middle inset (e) shows the low wavenumber capillary modes, which seldom result in atomization as they are dampened out quickly. Right inset (e) shows the high wavenumber modes whose capillary instability is the major mechanism for the generation of the fine atomized mist of droplets observed in (e).

surface starts to deform. When the right edge of the droplet reaches the transducer edge, regime 3 is initiated, where inertia forces the fluid rightwards, leading to two distinctive bulges due to the retracting surface tension force; the left contact point remains pinned. Subsequently, the liquid in the left lobe (under the action of F_r) moves to the right lobe. Regime 4 involves further flattening of the right lobe to thin films. Different behavior is observed for low viscosity fluids compared to the highly viscous glycerol in regimes 3 and 4. Atomization is initiated from the right lobe of lower viscosity droplets at the start of regime 3 and continues all the way into regime 4, seemingly with little transition [Ref. movie 2 Figure 3]. The mechanism of atomization is twofold. (i) The asymmetrically located droplet experiences an uneven external pressure distribution due to acoustic streaming which results in a higher average static pressure on the left side of the droplet. As the droplet spreads, the pressure non-uniformity increases, deforming the droplet to the extent that the surface tension force fails to balance the differential pressure force. (ii) Multi-modal capillary waves form on the right

lobe due to the kinetic energy imparted to the droplet by the streaming air flow and the transducer vibration (augmented by the thinning of the droplet). Low wavenumber modes are observed to attenuate while the high wavenumber modes grow and satellite droplets start pinching due to capillary instability culminating in catastrophic break-up [Figure 1]. High viscosity of NS27 and glycerol enables the droplets to dampen the surface disturbances and prevent atomization. Note that spreading of droplets as a precursor to atomization within short timescales (< 70 ms) has not been studied before.

The observed time duration of regime 2, $t_{2-actual}$, for identical spreading lengths is a maximum for glycerol, followed by GW9010 and NS27. Water, NS05, NS10, and GS2080 have similar low values (shown later). We simulate the droplet distortion in regime 2 by considering the different external forces acting on the droplet. Since the majority of the half of the droplet contact line near the center of the transducer surface is always in dynamic equilibrium, at any instant of the spreading regime the effective unbalanced interfacial force at the triple line (taking the rightward spreading direction to be positive) is $F_\sigma \sim (\sigma_{sv} - \sigma_{lv} \cos \theta - \sigma_{ls})l$, where σ , θ , and l denote the surface tension, dynamic advancing (right)

FIG. 2. A water droplet in regime 1 (enhanced online) [URL: <http://dx.doi.org/10.1063/1.4757567.1>].

FIG. 3. A water droplet progressing from regime 2 to 4 (enhanced online) [URL: <http://dx.doi.org/10.1063/1.4757567.2>].

contact angle, and instantaneous droplet base dimension, respectively. Subscripts s , l , and v represent the solid, liquid, and vapour phases, respectively. The force balance of a static droplet (before the transducer vibration is initiated) reveals that $\sigma_{sv} - \sigma_{ls} = \sigma_{lv}\cos\theta_0$, θ_0 is the static contact angle. Hence $F_\sigma \sim \sigma l(\cos\theta_0 - \cos\theta)$, where $\sigma \equiv \sigma_{lv}$.

The force due to the tangential viscous stress ($\sim \mu \dot{l}/h$, where $h \sim l\tan\theta$ represents the height of the droplet) acting on the solid-liquid boundary area ($\sim l^2$) is $F_\mu \sim -\mu \dot{l}/\tan\theta$. The negative sign ensures that the viscous force acts in the opposite direction of the spread rate, $\dot{l}$ as $\theta \in (0, \pi/2)$.

Figure 4 presents the time-series data of right and left contact angles normalized with the corresponding θ_0 (initial static contact angle) for a few fluids. In Figure 4, the maximum in the left contact angle occurs when the first crest of the surface wave, initiated by the advancement of the right contact point, reaches the left contact point. Subsequently, the rightward droplet bulk-motion decreases the left contact angle. The right contact angles show a continuous drop with time due to the sliding and seeping of the top mass of the droplet at the contact point. In the model, this temporally varying right contact angle is approximated by a linear function of the droplet base dimension change, $\delta (=l - l_i)$, subscript i represents the initial value in regime 2), i.e., $\theta = \theta(l - l_i)$. This function is obtained using the data for water since all the fluids show similar values and variation of contact angle as can be seen from Figure 4, where the individual curves do not vary much from the average curve.

For a droplet of mass m , undergoing spreading, the law of conservation of linear momentum can be written combining all the force terms [viscous force F_μ , surface force F_σ , external acoustic force F_r] as follows:

$$\ddot{l} = -C_\mu \mu \dot{l} / (m \tan\theta) + C_\sigma \sigma l (\cos\theta_0 - \cos\theta) / m + C_F (\tilde{F}_r) / m, \quad (1)$$

where $\tilde{F}_r$ denotes the normalized space-averaged acoustic force on the droplet. C_σ and C_μ are fit coefficients. In regime 2, the effect of transducer vibration on the droplet is not direct but through a pseudo-steady acoustic force given by the RMS

FIG. 4. Smoothened data of experimental non-dimensional contact angle versus non-dimensional time, T . The scatter of raw data (before smoothening) is represented by the error bars ($\pm 5\%$) for water at only a few selected points. The other data curves exhibit similar range of scatter. The average is taken across all the fluids.

potential field of the standing wave.¹⁸ Besides the radial acoustic force, other possible horizontal forces acting on the droplet are the friction force of the transducer surface, the force originating from the transducer surface acceleration and the force due to Poisson effect of F_z . These effects are absorbed in the factor C_F . In order to take into account the variation of the acoustic force during the spreading stage, the last term on the right hand side of Eq. (1) is obtained by taking the average of initial and final values of normalized unit radial acoustic force at the transducer surface averaged over the droplet base-length, i.e., $\tilde{F}_r \propto \int_{r_L}^{r_L+l} \sin(k_r r) dr / l$ where k_r is the radial wave number of the standing wave and r_L the distance of the pinned left contact point of the droplet from the axis of the wave. Although the acoustic field is modified due to scattering from the droplet surface and added impedance from the liquid layer, the functional variation of the acoustic force with the radial coordinate, r , is assumed to be the same as in Ref. 18.

Equation (1) is solved numerically using Runge-Kutta fourth-order method with the initial conditions: $l = l_i$ and $\dot{l} = 0$ at time, $t = 0$ and $C_\sigma = 0.1$, $C_\mu = 0.9$, and $C_F = 0.15 \times 10^{-4}$. The model is capable of predicting the droplet spreading characteristics of partially wetting droplets ($0.5 < We_a < 0.7$) for a wide range of viscosities ($0.01 < Re < 100$). Here acoustic Weber number $We_a = 2p\gamma M^2 l_i / \sigma$ (p : atmospheric pressure, γ : air specific heat ratio, M : acoustic Mach number) and Reynolds number $Re = \rho \dot{l} l_i / \mu$. In Figure 5, $\eta = \delta / (l_f - l_i)$ (subscript f denotes the final value in regime 2) and $T = t/t_2$ are non-dimensional droplet base-length change and the time normalized by the theoretical/experimental time-scales respectively. This figure shows that the shapes of the theoretical curves agree with the experimental profiles satisfactorily for all the fluids. The nature of $\eta - T$ curves of high viscosity fluids is different from that of the low viscosity ones. The agreement shows that the model is able to capture the spreading behavior of the fluids with widely varying properties reasonably well.

Timescales from the model are of a similar order to experimental values (Figure 5 caption). The discrepancies ($\sim 15\%$) are due to transducer surface contamination, uncertainty in viscosity and other fluid properties, the assumption of averaged forces due to standing waves, and the additional friction on the droplet due to internal motion due to liquid recirculation. Figure 3 suggests that recirculation is clockwise, and more vigorous for low viscosity, contributing to an over-prediction in the time-scale. However, the shape is relatively unaffected due to proper scaling. For glycerol and GW90-10, the ambient temperature does not affect the spreading rate significantly (Figure 5(a)).

In order to gain more physical insight into the spreading phenomenon, we derive the linear form of Eq. (1) by substituting $l = l_i + \delta$ in the surface tension force term, approximating $l \sim \bar{l} = (l_i + l_f)/2$ in the viscous force term and disregarding the temporal variation of the contact angle

$$\ddot{\delta} + C'_\mu (\mu \bar{l} / m) \dot{\delta} + C'_\sigma (\sigma / m) \delta = F' / m. \quad (2)$$

This equation represents the droplet as a simple spring-mass-damper system. This allows for the droplet damping coefficient to be defined as $\zeta = C'_\mu \mu \bar{l} / (2(C'_\sigma \sigma m)^{1/2})$ and yields the decay time, t_d for under-damped droplets as $(\zeta \omega_0)^{-1}$ and for over-damped case (corresponding to the faster of the

FIG. 5. η versus T plots: (a) Water-glycerol family, (b) water-nanosilica family. Viscosity normalized by water viscosity at 25 °C ($\mu_{\text{water}} = 0.893$ mPa s). Time scales of regime 2 are given as ($t_{2\text{-actual}}$: experimental time in ms, $t_{2\text{-predicted}}$: theoretical time in ms). Time scales: Water (8.5, 12.95); GW2080 (7, 14.15); GW9010 (28.9, 23.85); Glycerol: (69.35, 60.05); NS05 (8.25, 11.3); NS10 (8.8, 15.6); NS27 (19, 18.1). Uncertainty estimate (error bar) is about 2%.

two decaying exponents) as $((\zeta + (\zeta^2 - 1)^{1/2})\omega_0)^{-1}$, where $\omega_0 = (C'_\sigma \sigma / m)^{1/2}$ is the droplet natural frequency. Figure 6(a) shows that t_d decreases with Ohnesorge number, $Oh = \mu / (\rho l_i \sigma)^{1/2}$ (ρ is the liquid density). The very low value of t_d for glycerol and other high viscosity fluids implies that the external disturbances attenuate due to viscous dissipation, as dictated by the high value of Oh . This trend will remain consistent in the case of the actual non-linear equation too. Depending on whether $\zeta >$ or < 1 , the model results in the over-damped or under-damped solution (Figure 5).

Analysis for NS27 was handled differently as it is a non-Newtonian fluid. The temporal viscosity variation is obtained by measuring the shear rate, $\xi = u/y_C$, (y_C : distance from transducer surface to droplet centroid) as shown in Figure 6(b). Using ξ and the NS27 viscosity profile,¹⁹ the temporal viscosity was fitted to obtain the function $\mu = \mu(\xi)$ which was provided to the model. Initial viscosity is much higher (0.4 Ns/m²) till 1 ms and thereafter reduces with increase in

shear rate due to shear-thinning. The shape and the predicted theoretical time scale, $t_{2\text{-predicted}}$, are of the same order (Figure 5 caption) as in the experiment, suggesting that the non-Newtonian behavior is captured well. When the profile is modified to be exponential, the time scale prediction is excellent (18.1 ms), while the shape prediction remains the same (Figure 5(b)). A fluid-specific function $\mu = \mu(\xi)$ or $y_C \sim l^* \tan \theta$ can be provided to predict the non-Newtonian effects of spreading using our model.

Using non-dimensional variables η and $\tau = t\bar{u}/l_i$ ($\bar{u}$ is the average spread-rate) and approximating $m \sim \rho l_i^3$ and assuming external force, F scales as the inertia force one may obtain the dimensionless form of Eq. (2)

$$\frac{d^2 \eta}{d\tau^2} + \frac{C'_\mu d\eta}{Re d\tau} + \frac{C'_\sigma}{\kappa} \eta = \frac{F'}{\rho \bar{u}^2 l_i^2}, \quad (3)$$

where $\kappa = \rho \bar{u}^2 l_i / \sigma$ is the modified Weber number (ratio of kinetic energy to surface energy). Using the experimental data, we propose $\kappa = 0.054 Re^{0.413}$ [Figure 6(c)], which states that a lower Reynolds number fluid has lower average kinetic energy due to viscous damping. Hence glycerol has the least spread-rate. On the other hand, for a low viscosity fluid, the process is dominated by inertial forces. The scatter at high Reynolds number is partly due to the fact that internal recirculation is more prominent for low viscosity fluids compared to high viscosity fluids.

In conclusion, the asymmetric droplet spreading to a thin film on a transducer surface of a levitator is dominated by the radial acoustic force of the ultrasonic standing wave. This finding allowed spread rate to be modelled by a non-linear ordinary differential equation with the forcing term in the form of a step function. One of the modes following droplet spreading is the growth of multi-modal capillary waves. This led to atomization if the viscosity of the droplet was below a certain limit. By suspending nanoparticles at different concentrations, we were able to suppress atomization and subsequent break-up of the droplet. It is clearly

FIG. 6. (a) Theoretical decay time, t_d versus Ohnesorge number, Oh . (b) Shear rate (ξ) versus non-dimensional time (T) for NS27 and water. (c) Dependence of the modified Weber number, κ on Reynolds number, Re . The relative uncertainty in the experimental data shown is less than 2%.

shown that the higher viscosity is responsible for suppressing atomization and our single timescale for the non-Newtonian fluid compares well with the experiments.

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