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Coalescence of drops near a hydrophilic boundary leads to long range directed motion

EXTREME MECHANICS

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a r t i c l e i n f o

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a b s t r a c t

A new mechanism for the passive removal of drop on a horizontal surface is described that does not require pre-fabrication of a surface energy gradient. The method relies upon the preparation of alternate hydrophilic/hydrophobic stripes on a surface. When one side of this surface is exposed to steam, with its other surface convectively cooled with cold water, steam condenses as a continuous film on the hydrophilic stripes but as droplets on the hydrophobic stripes. Coalescence leads to a self-generated noise that in turn leads to a random motion of the center of mass of the fused drops on the surface, which are readily removed as they reach near the boundary of the hydrophobic and hydrophilic zones thus resulting in a net diffusive flux of the coalesced drops moving from the hydrophobic to the hydrophilic stripes on the surface. This phenomenon is, indeed, similar to that of the random walk of particles with an absorbing wall. This method of creating directed motion of drops does not require a pre-existing wettability gradient and may have useful applications in thermal management devices.

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1. Introduction

Liquid drops move on a flat surface if they are subjected to a surface tension driven unbalanced force. A widely studied method to generate such a motion is to place a drop on a surface that has a gradient of wettability [\[1–7\]](#page--1-0). Due to the difference in the intrinsic contact angles on two opposite sides of the drop, a curvature gradient is imposed upon it that causes a net motion of the drop from the less to the more wettable region of the surface. Recently, it has been found that a hydrophobic surface having a morphological [\[8–10\]](#page--1-1), a curvature gradient [\[11,](#page--1-2)[12\]](#page--1-3) and electro-wetting [\[13–16\]](#page--1-4) also induce such types of motions. Liquid drops can also move on a surface if a thermal gradient [\[2,](#page--1-5)[17–19\]](#page--1-6) is imposed that induces a motion due to unbalanced surface tension on the liquid surface and the drop usually moves from the hotter to the colder part of the gradient. Reactive spreading [\[20–23\]](#page--1-7) is

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<http://dx.doi.org/10.1016/j.eml.2014.11.007> 2352-4316/© 2014 Elsevier Ltd. All rights reserved. another mechanism of drop motion that relies upon the insitu created surface energy gradient on a surface.

The main detriment to the motion of liquid drops is related to pinning [\[3](#page--1-8)[,7,](#page--1-9)[24\]](#page--1-10) of the type that gives rise to the hysteresis of contact angle on any real surface. While it is not practical to produce a defect free surface, hysteresis can, however, be advantageous if the surface is subjected to a mechanical vibration [\[25–32\]](#page--1-11). The role of hysteresis can be viewed in two ways. Hysteresis on a heterogeneous surface can be asymmetric [\[27\]](#page--1-12), which, like a diode, rectifies an externally imposed structured noise that gives rise to a unidirectional motion. When a random noise is imposed, the non-linear friction due to hysteresis manifests in terms of a quasi-linear friction with an emergent relaxation time depending upon the noise strength and the magnitude of hysteresis [\[33–36\]](#page--1-13). The effective relaxation time (τ_l^*) of the of a drop on a surface can be expressed [\[36\]](#page--1-14) in terms of the Langevin relaxation time (τ _L) and the hysteresis (Δ) as:

$$
\frac{1}{\tau_L^*} = \frac{1}{\tau_L} + \frac{\Delta^2}{K}.\tag{1}
$$

With the above effective relaxation time, we can also define an effective diffusivity as $D\sim K\tau_L^{*2}$, which assumes the form $D \sim K^3/\varDelta^4$ in the limit of low noise strength. In the presence of an externally applied force $\bar{\gamma}$, the drift velocity of the drop [\[36\]](#page--1-14) is:

$$
V_d = \bar{\gamma}\tau * = \frac{\bar{\gamma}\tau_L}{1 + \Delta^2\tau_L/K}.
$$
\n(2)

In Eqs. [\(1\)](#page-0-1) and [\(2\)](#page-1-0) both the hysteresis force Δ and the externally applied force $\bar{\gamma}$ are expressed on the basis of unit mass of the drop, which, thus have the units of acceleration. The noise strength is defined as: $K=\left\langle \gamma ^{2}(t)\right\rangle \tau _{c}$, where $\gamma \left(t\right)$ is the value (m/s²) of the noise pulse and τ_c is its duration (40 μ s). *K* is a measure of power input per unit time.

The bias can be provided either by a chemical or a thermal gradient of surface energy. The force due to the chemical gradient is $\gamma_w \left(\frac{d \cos \theta}{dx} \right) A$, where γ_w is the surface tension of water drop, *A* is its base area and *d* cos θ/*dx* is the wettability gradient. When a liquid drop on a surface is under the influence of a thermal gradient, the main driving force arises from the gradient of surface tension of the liquid due to a gradient of temperature (*T*), which results in a Marangoni flow on the surface [\[2\]](#page--1-5) of the liquid drop causing it to move toward the colder side of the gradient. The magnitude of this force is \sim $\left(\frac{d\gamma_w}{dT}\right)\left(\frac{dT}{dx}\right)$ A, although this force would be randomized when subjected to vibration [\[19\]](#page--1-15). In both cases, however, the drop experiences a resistive force due to wetting hysteresis as its frontal side attempts to reach the local advancing angle, whereas its rear side tends to attain a local receding angle. This leads to a threshold force of magnitude: $\gamma_w b(\cos \theta_r - \cos \theta_a)$, where *b* is its width, whereas θ_a and θ_r are the advancing and receding contact angles on a given location on the surface respectively. Eq. [\(2\)](#page-1-0) predicts that the drift velocity of the drop increases with the noise strength till a limiting velocity is reached that is controlled only via Langevin relaxation time [\[36\]](#page--1-14) (see also the [Appendix A\)](#page--1-16). In the case of drops condensing on a surface, a self-generated noise can induce a random motion of the drops, the strength of which depends on the excess surface energy due to random coalescence.

Controlled transport of drops has many practical applications in biology [\[37\]](#page--1-17), as well as in unit operations involving water and thermal managements [\[4\]](#page--1-18). Thermal management is of considerable importance in micro-heat exchanger and heat pipe technologies. Vapor condenses as a thin continuous film on a hydrophilic surface that reduces the heat flux. While dropwise condensation can enhance the heat flux, an external force is needed to remove them. Furthermore, even though the dropwise condensation on an inclined surface can enhance heat flux on the vapor side of a heat exchanger, it is not effective on a horizontal surface as the drops coalesce and grow upon it to cover the entire surface in the absence of gravity. These issues were discussed in a previous publication [\[4\]](#page--1-18), where we demonstrated that the heat flux through a horizontal metal disc, one side of which is convectively cooled while the other side of which is exposed to steam, can be enhanced [\[4\]](#page--1-18) if a wettability gradient is designed on the surface of the disc that is in contact with the steam. As the steam condenses on such a gradient surface, the droplets are removed from the center to the edge of the disc by the gradient that enhances the net heat flux much more than that on a hydrophilic surface where steam condenses as a film. In the removal of the drops from a gradient surface, coalescence [\[4](#page--1-18)[,38](#page--1-19)[,39\]](#page--1-20) of the drops plays a very important role. On a homogeneous surface, the coalescence of two equal size drops causes the free edges of the drops to move toward their equilibrium position as the surface area and thus the surface energy of the coalesced drops is minimized. If two asymmetric drops coalesce, there is a net movement of the resulting center of mass on the surface, which results in a random motion that is reminiscent of a self-avoiding random walk. When such coalescence occurs on a surface possessing a mechanism to break the symmetry, a directed motion of the drops occurs on the surface. Combination of a heterogeneous wettability and hierarchical roughness can also be used to direct motion of drop on a condensing surface assisted by coalescence as was demonstrated in a recent nice study [\[40\]](#page--1-21). Our current paper draws inspiration from motion generated in out of equilibrium systems exhibiting large fluctuations [\[36\]](#page--1-14) in that the random coalescence of drops gives rise to a self-generated noise, the strength of which depends upon the degree of sub-cooling of the substrate, which, in turn, affects the density and the rate of nucleation as well as the random velocity of the motion of the coalesced drops. The similarity of a drop moving on a gradient surface due to an external noise and the heat flux resulting from the motion of the coalesced drops on a gradient surface is illustrated in the [Appendix A.](#page--1-16)

So far, a chemical or a morphological gradient has been explored to effectively remove condensed drops on a surface. Here we propose a new mechanism to remove condensed drops, the principle of which is illustrated in [Fig. 1.](#page--1-22) Consider a flat surface that has alternate stripes of hydrophilic and hydrophobic zones. Steam condenses as a film on the hydrophilic stripes but as drops on the hydrophobic stripes.

As the drops coalesce on the hydrophobic surface, the center of mass of the fused drops undergoes a random walk. When these drops occasionally come in contact with the edge of the condensed water film, they are readily pulled into the hydrophilic zone due to the difference in the Laplace pressure of the drop and the film. This scenario is remotely similar to a single component diffusion of a species near an absorbing boundary, in that a gradient of the concentration is created near the boundary of the two zones that creates a net diffusive flux of the coalesced drops from the hydrophobic to the hydrophilic sides of the surface. Once the liquid is collected on the hydrophilic channels, it can be removed by a wicking mechanism that can be designed on the steam side of a metal disc. In this paper, we intend to provide a proof of the concept based on the evidences reported below.

2. Experimental details

2.1. Preparation of striped surface

In order to create a hydrophobically modulated striped pattern on the silicon wafer, a plastic template is formed

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