Electrically Modulated Dynamics of Thin Liquid Films in Microfluidic Confinements

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We demonstrate a new instability phenomenon governed by electrohydrodynamics of thin liquid films in narrow fluidic confinements. We show that under the action of axial electrical field, perturbations may be induced on the interfaces of thin corner films formed adjacent to the walls of a microchannel, leading to the inception of ordered lateral structures. These structures from the top and bottom walls intermingle to evolve into localized gas pockets in the form of bubbles. We further show that these bubbles do not remain static with changes in electric field, but undergo a sequence of elongation-deformation-breakup episode in a dynamically evolving manner, as a consequence of a complicated interplay of electro-mechanical forces and interfacial tension. This, in effect, leads to a new paradigm of controlled microbubble manipulation for on-chip applications, as governed by the underlying instability mechanisms over interfacial scales.

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Fascinating complex dynamics and pattern formations over small scales have triggered a wide range of scientific innovations addressing various physical facets of thin liquid films, primarily motivated by their spontaneous formations in nature and the roles played by them in a wide variety of transport processes in practical scenarios [1]. In reality, thin films are often characterized by several special features, including the formation of regular structures [2] (self-assembly), formation of shock-fronts [3] and periodic waves, and instabilities associated with the complex interplay of interfacial forces, leading to non-trivial dynamics over small scales. Applications of such dynamical aspects of thin liquid films [1] range from complex coating flows [4], optoelectronic display materials, engineering applications like heat exchangers, microfluidics [5][6] and microelectromechanical devices, gravity currents [1], granular and debris flows [3], snow avalanches [8], biophysical aspects like lung airways [9], tear-film flows, and bioadhesion [10], to name a few.

Thin films often act as precursors to create templates for patterned arrays of micro- and nano-structures. These patterns are obtained by exploiting the physical and energetic properties of the film. Alteration in these properties in tandem with other hydrodynamic effects results in instabilities (spontaneous or induced) in these films. Nevertheless, thicknesses of these films play crucial roles in determining the concerned instability phenomena [11][12]. In practice, thin film instabilities may be triggered by various mechanisms (for example, see [13][42]), including those of non-electrical as well as electrical origin.

In this Letter, we present a new physical paradigm of electro-hydrodynamically triggered thin film instability in microfluidic confinement. In contrast to other related reported investigations on thin film dynamics as triggered by externally applied transverse electric field alone, our present study focuses on thin film dynamics subjected to an intricate interplay of externally applied axial electric field, induced transverse electric field, and hydrodynamics over interfacial scales, in a confined microfluidic environment. We term the underlying newly unravelled phenomenon as electro-osmo-hydrodynamic (EOHD) instability. However, our scope of the present study is not merely restricted to demonstrate the mechanism of this EOHD instability alone. Rather, we also aim to bring out the new physics of controlled microbubble generation and their morphological evolution (essentially, a sequence of downstream events triggered by elongation-deformation-breakup dynamics) by exploiting the same underlying instability mechanism in conjunction with a dynamical interplay of interfacial tension and electromechanical forces over relevant spatiotemporal scales.

To demonstrate the new aspects of thin film morphodynamics, we consider rectangular microchannels with high aspect ratios. Such cross-sections are ubiquitous in several studies related to microfluidics [43]. A meniscus is formed (see Fig. 1a) by partially filling a glass microchannel [14] (width \( w \), depth \( H \) and length \( l \)) with borate buffer (typical concentration: 10 mM). For microchannel surfaces that are wetting in nature, the liquid meniscus extends in the form of precursor film over the channel walls, leaving a gas filled region even prior to the application of the electric field. A thin film, typically known as corner film (with typical thickness of order of 1 \( \mu \)m), is formed at the gutters (or corners) of the microchannel. Our central finding stems from the fact that under the action of an axial electrical field, interfaces of such thin corner films formed get perturbed, leading to the inception of lateral structures having corrugated topographies on highly wettable substrates (see Fig. 1b). This phenomenon may be perceived as a consequence of EOHD instability mechanism, as triggered by interplay between a charged interfacial layer (also known as electrical double layer) and thin film forces. The corrugated interfacial layers, formed at the opposite walls, tend to grow progressively with increments in the ap-
applied electrical voltage. Beyond a critical electric field, $E_{cr}$, which is a function of the channel dimensions for a given electrolyte-substrate combination, the progressively thickening interfacial films eventually intermingle. This intermingling, in turn, marks a transition from thin film regime to meniscus formation regime, resulting in localized gas pockets (see Fig. 1b) that evolve dynamically. From physical perspective, this critical field demarcates dominance of different electrical force regimes (electroosmotic versus excess electrical pressure) and consequently yields different instability wavelengths and growth rates. The gas pockets, thus formed, subsequently undergo intriguing elongation-deformation-breakup dynamics, leading to EOHD instability-driven bubble manipulation over local scales.

In an effort to make simple yet insightful theoretical estimates on the dynamical aspects of the thin corner films formed in the wall-adjacent interfacial layer, we first note that the electrostatics across this interfacial layer encompassing the thin corner films is faceted by the fact that the glass surface develops free surface charges because of ion adsorption or reaction in presence of the ionic liquid. This surface charging, in turn, leads to a charge density distribution in the solution, resulting in the development of electric double layer (EDL) [45]. Distribution of free charges within the EDL, governing the thin film dynamics, is essentially dictated by the Poisson-Boltzmann (PB) equation [45] as $\nabla^2 \psi = \kappa^2 \psi$ ($\psi$ being the potential induced in the EDL and $\kappa$ being the inverse of the Debye length describing a characteristic thickness of the EDL), consistent with Debye-Hückel approximation [46, 47].

Electrical effects of thin film under electroosmosis (see note [47]) may be quantitatively captured by considering the divergence of an electrical stress tensor ($T^E$) in the momentum equation, where $\nabla \cdot T^E = -\Pi + \epsilon_1 \sigma_0 (\mathbf{E}E - \frac{1}{2} \mathbf{E}^2 \mathbf{I})$. Here, $\Pi$ is the osmotic pressure, $\mathbf{E}$ is the resultant electric field, $\mathbf{I}$ is the identity tensor, and $\epsilon_1$ and $\epsilon_0$ are the permittivities of the medium and free space, respectively. The osmotic pressure may be calculated as: $\Pi = (n^+ + n^-) K_B T$; $n^+$ and $n^-$ being the number densities of the positive and negative ionic species, respectively, as described by the Boltzmann distribution [45]. The liquid pressure, $P_l$, alternatively interpreted as the effective pressure, is a combined consequence of the hydrodynamic pressure, osmotic pressure, electrical pressure and surface tension, expressed as: $P_l = P_g - \sigma \xi_{xx} + \Pi + \rho_{el}$ where the subscript $xx$ represents the double derivative of the local film thickness $h$ with respect to the axial coordinate $x$. Here, $P_g$ is the gas pressure, $\rho_{el} = \left( \frac{\varepsilon_0}{\varepsilon_1} (\varepsilon_1 - 1) (E_l^2 + \varepsilon_1 E_n^2) \right)$ denotes the pressure induced from the jump in the electrical stresses at the liquid-gas interface, $\sigma$ is the liquid-vapour surface tension coefficient, and $E_n$ and $E_l$ are the normal and tangential components, respectively, of the external electric field. While arriving at this expression, we also consider that at the gas-liquid interface, the electric field experiences the discontinuity of the displacement field $\left( \sigma_{el} E_n \right)_{||} = \left( \sigma_{el} E_n \right)_{\perp}$, where $\sigma_{el}$ is the electrical conductivity. For a perfect dielectric, it is consistent to replace $\sigma_{el} \xi_{xx}$ by $\varepsilon_1 \sigma_{el}$ [49]. Further, the tangential component of the electric field is continuous across the interface: $(E_l)_{||} = (E_l)_{\perp}$. The interfacial electric field, which acts solely in the tangential direction, however, varies axially, owing to the fact that the local film thickness also varies axially, under the constraint of a constant electric flux ($K$), so that one may write $E_l = \frac{K}{h}$ (see also note [50]).

The hydrodynamics in the thin film in steady state may be described by using the equation of momentum

\begin{equation}
\end{equation}
conservation as: \(-\nabla p + \nabla \cdot \mathbf{T} = 0\) (where \(p\) is an effective pressure which includes the effects of interfacial tension, osmotic pressure, and electrical stresses), along with the boundary conditions - (i) \(\mathbf{t} \cdot \mathbf{T} \cdot \mathbf{n} = 0\) at \(y = h\) (where \(\mathbf{T}\) is the resultant stress tensor, considering both hydrodynamic (\(\mathbf{T}^H\)) and electrical stress (\(\mathbf{T}^E\)), \(y\) being a transverse coordinate axis with origin at one of the microchannel walls) and (ii) the velocity vector \(\mathbf{u} = 0\) at walls \((y=0)\). The above, in conjunction with the continuity equation, yields the governing equation for the 2D evolution of the corner film as:

\[
\frac{\partial h}{\partial t} = \frac{\partial}{\partial x} \left( \frac{h^3}{3\mu} \frac{\partial}{\partial x} (-\alpha h_{xx} + p_{el}) \right) + U_e h_x \Theta \tag{1}
\]

where \(\mu\) is the dynamic viscosity of the liquid, \(U_e\) represents a characteristic electroosmotic velocity scale, given by: \(U_e = -\varepsilon_0 \varepsilon \varepsilon_0 (\kappa K) \tanh(\alpha)/\alpha \mu\), with \(\alpha = e \psi_0 / 4 K B T\) (here \(\psi_0\) is the surface potential) and \(\Theta = 1 - e^{-\kappa h} (1 + \kappa h/2)\).

The parameter \(h\), as appearing in Eq. (1), increase in response to the application of a progressively intensifying electric field, owing to the higher flux of ions in the thin film region, till the opposite films intermingle. We use a linear stability analysis to predict different instability regimes during dynamic evolution of the film as governed by Eq. (1). The base state solution \((h = h_0)\) satisfies the governing equation, and we check the stability of this base state with respect to small perturbations. We expand the base solution in normal modes with an ansatz: \(h = h_0 + \varepsilon e^{ikx+\omega t}\) where \(h_0\), \(k\), \(\omega\) and \(\epsilon\) are the mean film thickness, spatial growth rate, the temporal growth coefficient and the amplitude of the initial disturbance, respectively, with the assumption that \(\varepsilon \ll h_0\). The dispersion relation may be obtained correct to the first order of \(\varepsilon\) as: \(\omega = -h_0 k^2 (\partial p_{el}/\partial h)|_{h=h_0} + k^2 \sigma)/3\mu + \kappa k^2 = -3\mu U_e / h_0^3 - \partial p_{el}/\partial h)|_{h=h_0}.\) In the limit of \(U_e \to 0\), the inverse wavelength \((k)\) is a free parameter (similar to the behaviour described in Refs. [21, 24]), because the imaginary part solution is the trivial solution of the dispersion relation \(\partial p_{el}/\partial h)|_{h=h_0} = 0\). For such a case, the dominant wavelength and maximum growth factor corresponding to the fastest growing linear mode \((\partial \omega/\partial k = 0)\) may be obtained as \(\lambda_{max} = 2\pi/k = 2\pi(-\partial p_{el}/\partial h)|_{h=h_0}/(2\sigma + \kappa k^2)\) and \(\omega_{max} = h_0^3 \partial p_{el}/\partial h)^2|_{h=h_0}/12\mu \sigma\).

While assessing the relative importance of the electroosmotic term \(-3\mu U_e / h_0^3\) over the electrical pressure gradient term \(-\partial p_{el}/\partial h)|_{h=h_0}\) in the dispersion relation mentioned above, we first note that \(-\partial p_{el}/\partial h)|_{h=h_0} \sim \varepsilon_0 \varepsilon E^2 / h_0.\) In order to have the electroosmotic effects dominant over the gradient of electrical pressure, i.e., \(O(\mu U_e / h_0^3) \gg O(\partial p_{el}/\partial h)|_{h=h_0}\), it is thus essential to have \(|E| \ll |\psi_0| \tanh(\alpha)/\alpha\). With the typical values \(|\psi_0| = 75mV, \kappa^{-1} = 100nm, T=298K,\) it may be found that the wavelength of instability is electrosmotically dominated, when the magnitude of the electric field is less than \(|E| \sim O(10^4) V/m\), whereas the effects of the gradient of pressure from the jump in the electrical stresses at the liquid-gas interface \((p_{el})\) becomes significant beyond this threshold limit of the electric field. When \(O(\mu U_e / h_0^3) \ll O(\partial p_{el}/\partial h)|_{h=h_0}\), the system essentially behaves like a conductive fluid. Thus, below the threshold electric field, the wavelength \(\lambda\) and the time scale \(\omega^{-1}\) are dominated by the electroosmotic effects and scale as \(\lambda \sim E^{-1/2}\) and \(\omega^{-1} \sim E^{-3}\) respectively.

On the other hand, above the critical electric field, the effect of \(p_{el}\) becomes dominant, resulting in \(\lambda \sim E^{-1}\) and \(\omega^{-1} \sim E^{-4}\).

To obtain the instability wavelength corresponding to the dominant spatial frequency from an experimental perspective, we transform the thin film profiles (for details, see [51]) into Fourier space. The instability wavelength, which is a function of the electric field \((E)\), is shown in Fig. 2a, with the experimental and theoretical values compared. For \(E < E_{cr}\) (a critical value of the electric field for the films to just protrude into the channel centreline), spatiotemporal evolutions of the film thickness reveal that the perturbations recur in space. For \(E > E_{cr}\), the thin films intermingle with a rupturing of the film occurring at \(t = t_r\) (which is a function of the electric field; in particular: \(t_r \sim \omega^{-1} \sim E^{-4}\) for \(|E| > |E|_{cr}\)). For a visualization of the progressive transformation of the thin film regime into meniscus formation regime, see [51]. Experimentally, it was observed that the time to rupture decreases rapidly with the increasing electric field and scales as \(E^{-4.3}\), which is close to the scale obtained from the linear analysis of the inverse maximum growth rate. This rupture time scale hallmarks a transformation of the thin film regime to the meniscus formation regime, and results in the formation of localized gas-liquid pockets in the form of bubbles of different dimensions (for representative visualization, see [51]). Importantly, both \(t_r\) and \(E_{cr}\) are not constants, but are strong functions of the channel dimensions as well, as demonstrated in Fig. 2a.

Interestingly, the bubbles thus formed do not remain stationary but undergo a sequence of systematic and intriguing dynamical events with alterations in the electric field, as attributed to an intricate interplay of electro-mechanical forces and surface tension over relevant physical scales. This is completely counter-intuitive in a sense that most of the electric current and the flow is expected to escape through the corner films (flow bypass through the corner films would be equal to the flow behind the bubbles [43]) rendering the bubble to be stationary. However, we do observe the bubbles to be dynamic in nature, for completely wetting microchannel walls (where each bubble is covered by the thin film from all the sides). To elucidate the underlying dynamical aspects, one may note that electrical forces in the liquid film
\( P_d = \frac{\varepsilon_0}{2(\varepsilon_1 - 1)(K/\eta_0)^2} \) contribute to an excess liquid pressure \((P_t > P_g)\), while surface tension \((\pi R)\) where \(R\) is the radius of curvature of the thin film) resists the deformation of bubbles. The radius of curvature of the deformed configuration can be theoretically estimated to be of the order \(R \sim \frac{\sigma b_0^2}{(\varepsilon_0 - 1)\varepsilon_0 R^2}\). As the electric flux \((K)\) progressively decreases, \(R\) tends to increase, and in a limiting sense tends to infinity so that the thin film remains flat. Nevertheless, the curved corner film has to match to the hemispherical cap-ends of the front and rear end of the bubble. The dynamical events accompanying this feature may be analyzed analogous to the classical Bretherton problem of the bubble evolution in pressure driven flows \([52]\) where the thin film equation is governed by Eq. \((I)\) but the pertinent length scale is \(R\) instead of the Bretherton thin film length scale. Importantly, the asymptotic matching of Eq. \((I)\) in the limits of \(x \to -\infty\) and \(x \to +\infty\) are different, resulting in the radius of the front-cap to be smaller and the back-cap to be larger than the half width of the microchannel. This results in the pressure build-up behind the bubble to push it forward. Each bubble thus formed tends to adjust to the incipient stresses by reconfiguring itself into a peanut shape \([53]\). The deformation of the neck of each bubble, \(\varsigma\) (normalized by the channel width \(w\)), appears to be a function of the ratio of the electrical forces to that of the surface tension forces. This dependence may be expressed in terms of the electrical capillary number \(\chi = \frac{\varepsilon_0 E^2 H}{\sigma}\) where \(H\) is the depth of the microchannel \((H \ll R)\). Experimentally, \(\varsigma\) is found to scale linearly with \(\chi\). Further, \(\varsigma\) is also found to scale with \((L_0/L_b)^{-n}\), where \(L_0\) and \(L_b\) are the length of the deformed and undeformed bubble, and the exponent \(n\) may be obtained as 1.8 from a theoretical perspective \([51]\). A close analysis of the experimental data reveals that for \(n = 2.1\), all the experimental data collapse to a single master curve (see Fig. \((3)\)). Considering some idealization in the theoretical analysis, this agreement between theory and experiments appears to be reasonably close.

The bubble elongation-deformation described as above is by no means an eternal process. As such, the elongation of each bubble increases progressively, till the neck deformation \(\varsigma\) becomes zero, resulting in a bubble break-up. The numbers of new pockets formed are essentially dictated by the dominant wavelength \((\lambda)\) of the spatial disturbance of the thin film. The sizes of the daughter pockets (microbubbles thus generated) further scale linearly with the dominant wavelength, as evident from Fig. \((3)\), which implies that EOHD instability is responsible for the bubble splitting.

To summarize, in the present study, electroosmotic flow has been utilized to induce interfacial instabilities of the corner films in microfluidic channels, triggering non-trivial morphological dynamics. It has been demonstrated that the consequent electro-mechanical facets may essentially lead to the events of microbubble generation, elongation-deformation (see \([53]\) for Supplementary Movie) and break-up phenomena, which may be exploited towards realizing a new paradigm of integrated and controlled microbubble generation and manipulation for on-chip applications, with axial electric field as a tuning parameter. It is, however, also important to note that for partially-wetting or non-wetting substrates (unlike the cases of completely wetting substrates considered here), additional facets of thin film instabilities may feature, because of complicated interaction between corner film and surface film \((\sim 100nm)\) instabilities. The consequent dynamics holds the potential of obtaining fascinating surface patterns over miniaturized fluidic substrates, opening up scopes of interesting future investigations.

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FIG. 3. (a) Deformation of the bubble characterized by the neck \((\varsigma)\) (nondimensionalized by the width of the channel \(w\)) in presence of an electric field as a function of \((L_0/L_b)^{-2.1}\chi\) \([51]\), corresponding to different channel dimensions - 50µm x 10µm (○), 25µm x 12µm (□), 50µm x 25µm (◇) and 65µm x 30µm (×). Experimental errors are within 3.2% (errorbars not shown for clarity). The inset shows the optical micrograph of a bubble and its deformation with the application of electric field (b) The lengths of daughter bubbles generated after break-up \((L)\), as a function of \(\lambda\); both normalized by \(w\) (markers are not specific to any channel dimensions). The inset shows optical micrograph of the sequence of elongation-deformation-breakup events at different times \((t/t_b)\): 0.00, 0.03, 1.00, 1.02; where \(t_b\) is the time at the break-up.
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