

Northumbria Research Link

Citation: Ruiz Gutierrez, Elfego and Ledesma Aguilar, Rodrigo (2019) Lattice-Boltzmann Simulations of Electrowetting Phenomena. *Langmuir*, 35 (14). pp. 4849-4859. ISSN 0743-7463

Published by: American Chemical Society

URL: <https://doi.org/10.1021/acs.langmuir.9b00098>
<<https://doi.org/10.1021/acs.langmuir.9b00098>>

This version was downloaded from Northumbria Research Link:
<http://nrl.northumbria.ac.uk/id/eprint/38412/>

Northumbria University has developed Northumbria Research Link (NRL) to enable users to access the University's research output. Copyright © and moral rights for items on NRL are retained by the individual author(s) and/or other copyright owners. Single copies of full items can be reproduced, displayed or performed, and given to third parties in any format or medium for personal research or study, educational, or not-for-profit purposes without prior permission or charge, provided the authors, title and full bibliographic details are given, as well as a hyperlink and/or URL to the original metadata page. The content must not be changed in any way. Full items must not be sold commercially in any format or medium without formal permission of the copyright holder. The full policy is available online: <http://nrl.northumbria.ac.uk/policies.html>

This document may differ from the final, published version of the research and has been made available online in accordance with publisher policies. To read and/or cite from the published version of the research, please visit the publisher's website (a subscription may be required.)



**Northumbria
University**
NEWCASTLE



UniversityLibrary

Lattice-Boltzmann Simulations of Electrowetting Phenomena

Élfego Ruiz-Gutiérrez and Rodrigo Ledesma-Aguilar*

*Smart Materials and Surfaces Laboratory, Northumbria University, Ellison Building,
Ellison Place, Newcastle upon Tyne, NE1 8ST, UK.*

E-mail: rodrigo.ledesma@northumbria.ac.uk

Abstract

When a voltage difference is applied between a conducting liquid and a conducting (solid) electrode, the liquid is observed to spread on the solid. This phenomenon, generally referred to as electrowetting, underpins a number of interfacial phenomena of interest in applications that range from droplet microfluidics to optics. Here, we present a lattice-Boltzmann method that can simulate the coupled hydrodynamics and electrostatics equations of motion of a two-phase fluid as a means to model electrowetting phenomena. Our method has the advantage of modelling the electrostatic fields within the lattice-Boltzmann algorithm itself, eliminating the need for a hybrid method. We validate our method by reproducing the static equilibrium configuration of a droplet subject to an applied voltage and show that the apparent contact angle of the drop depends on the voltage following the Young-Lippmann equation up to contact angles of $\approx 50^\circ$. At higher voltages, we observe a saturation of the contact angle caused by the competition between electric and capillary stresses, similar to previous experimental observations. We also study the stability of a dielectric film trapped between a conducting fluid and a solid electrode and find a good agreement with analytical predictions based on lubrication theory. Finally, we investigate the film dynamics at long times

19 and report observations of film breakup and entrapment similar to previously reported
20 experimental results.

21 Introduction

22 Electrowetting refers to the spreading of an electrically conducting liquid on a solid elec-
23 trode when a voltage difference is applied between the two¹. Because of its ability to
24 control the interaction of liquids with solid surfaces, electrowetting has triggered a num-
25 ber of applications, such as droplet-based microfluidic devices²⁻⁵, droplet actuation^{6,7} and
26 mixing⁸⁻¹¹, deformable optical apertures¹² and lenses^{13,14}, and electronic paper displays¹⁵⁻¹⁸.
27 Broadly speaking, there are two types of electrowetting setups: Electrowetting On Conductor
28 (EWOC), in which the conductive liquid is in direct contact with the solid electrode¹⁹, and
29 the more popular Electrowetting On Dielectric (EWOD), in which direct contact is removed
30 by coating the electrode with a dielectric layer²⁰⁻²².

31 The simplest electrowetting situation, used widely in many EWOC and EWOD setups,
32 is the spreading of a droplet of conductive liquid on a solid in the presence of an ambient
33 dielectric fluid²³. During the actuation, the ambient fluid forms a thin film underneath
34 the droplet that can become unstable and break up into small “bubbles” that remain in
35 contact with the solid^{24,25}. Such a transition introduces mobile contact lines^{26,27}, which
36 can drastically affect the friction force acting on its overall dynamics²⁸⁻³⁰. On the other
37 hand, the spreading of a droplet at high voltages can reach a saturation regime³¹, where the
38 apparent contact angle that the droplet forms with the solid settles to a limiting value^{32,33}.
39 At even higher voltages, the edge of the spreading droplet can become unstable, and trigger
40 the breakup of small droplets that form coronal patterns around the mother drop³⁴.

41 Despite these important advances, the rich phenomenology of electrowetting remains to
42 be fully understood. For this purpose, it is essential to develop computational methods that
43 capture the multiphase fluid dynamics and that resolve the effect of electrostatic interactions,

44 as these can help interpret experiments and inform theory. The Lattice-Boltzmann Method
45 (LBM) has proved to be a powerful tool to study multiphase fluid dynamics³⁵. To implement
46 electrowetting within the LBM, it has been proposed to prescribe the interaction energy of
47 the surface^{36,37}, which leads to an effective contact angle. Such an approach, however, does
48 not capture the underlying coupling between the hydrodynamic and electrostatic fields. As a
49 means to overcome this limitation, hybrid methods that solve the electrostatic field equations
50 separately have been developed³⁸, but these come at the expense of running and coupling
51 two numerical solvers concurrently.

52 Here we present a lattice-Boltzmann method capable of solving the coupled hydrodynamics-
53 electrostatics equations that govern electrowetting phenomena within a single algorithm. We
54 use the so-called free-energy approach as a starting point to model the multiphase fluid dy-
55 namics, and show that the effect of the electrostatic energy can be included explicitly in
56 the corresponding energy functional. We introduce a set of lattice-Boltzmann equations,
57 where the electrostatic potential field is determined by a new set of distribution functions.
58 We validate this “all-in-one” method by comparing the electrowetting-induced spreading of
59 a droplet to the classical theory of Young and Lippman³⁹. To illustrate the utility of the
60 method, we present results of the stability of the thin film separating a conducting droplet
61 and a solid electrode, considering both the linear and non-linear regimes.

62 **Theoretical model and lattice-Boltzmann algorithm**

63 **Diffuse-interface model of electrowetting phenomena**

64 Let us consider two incompressible, immiscible fluids: a perfect conductor, corresponding to
65 the spreading liquid, and a dielectric, corresponding to the surrounding phase. We describe
66 the two-fluid system using a diffuse-interface model that identifies each phase using an order
67 parameter, or phase field, $\phi(\mathbf{x}, t)$, where \mathbf{x} denotes the position vector and t denotes time.
68 Without loss of generality, we let $\phi > 0$ be the conductive phase and $\phi < 0$ be the dielectric.

69 The Helmholtz free energy of the fluid-fluid system can be defined as⁴⁰

$$F_{\text{th}}[\phi] := \int_{\Omega} \psi(\phi, \nabla\phi) d^3x + \int_{\partial\Omega} \zeta \phi dS. \quad (1)$$

70 The first term corresponds to the volumetric contribution to the free energy over the region
71 occupied by the fluid, Ω . This consists of the well-known energy density of a binary fluid^{41,42},

$$\psi(\phi, \nabla\phi) := \frac{3\gamma}{\sqrt{8}\ell} \left[\frac{\phi^4}{4} - \frac{\phi^2}{2} + \frac{\ell^2}{2} |\nabla\phi|^2 \right], \quad (2)$$

72 where the square-gradient term allows the coexistence of the two bulk phases, of equilibrium
73 phase-field values $\phi = \pm 1$, separated by a diffuse interface of thickness ℓ and surface tension
74 γ . The second integral in Eq. (1) corresponds to the surface interaction energy of the fluid
75 with the solid electrode, whose boundary is denoted by $\partial\Omega$, and where the constant ζ is
76 called the wetting potential⁴³.

77 In equilibrium, and in the absence of an electric field, the fluid-fluid interface is expected
78 to intersect the solid boundary at an angle θ_0 determined by the Young-Dupr e relation⁴³,

$$\gamma_{\text{sd}} - \gamma_{\text{sc}} = \gamma \cos \theta_0, \quad (3)$$

79 where γ_{sd} and γ_{sc} are the solid-dielectric fluid and solid-conductive fluid surface tensions.
80 This is a standard result that can be obtained from Eqs. (1)–(3), which yield a relation
81 between the wetting potential and the contact angle⁴⁴:

$$\zeta = \frac{3}{2} \gamma \operatorname{sgn}(\theta_0 - \pi/2) \{ \cos(\alpha/3) [1 - \cos(\alpha/3)] \}^{1/2}, \quad (4)$$

82 where $\alpha = \arccos(\sin^2 \theta_0)$. It can also be shown that, in such a limit, the pressure field,
83 $p(\mathbf{x})$, is uniform in each phase, but jumps across the interface satisfying the Young-Laplace

84 relation

$$\Delta p = 2\gamma\kappa, \quad (5)$$

85 where κ is the interface curvature⁴⁵.

86 To model the electrostatic behaviour of the fluid mixture we introduce the electrostatic
87 free energy:

$$F_{\text{el}}[V] := -\frac{1}{2}\varepsilon \int_{\Omega} |\mathbf{E}|^2 d^3x, \quad (6)$$

88 which quantifies the potential energy density of the electric field $\mathbf{E}(\mathbf{x}) = -\nabla V$, where $V(\mathbf{x})$
89 is the electric potential and ε is the electric permittivity^{46,47}.

90 Out of equilibrium, local differences in the total free energy, $F = F_{\text{th}} + F_{\text{el}}$, give rise
91 to capillary and electrostatic forces. On the one hand, changes in the phase field lead to a
92 chemical potential field

$$\vartheta(\mathbf{x}, t) := \frac{\delta F}{\delta \phi} = \frac{3\gamma}{\sqrt{8\ell}} [\phi(\phi^2 - 1) - \ell^2 \nabla^2 \phi], \quad (7)$$

93 and a corresponding capillary force density

$$\mathbf{f}_{\text{cap}} = -\phi \nabla \vartheta, \quad (8)$$

94 which reduces to Eq. (5) in equilibrium⁴⁵. On the other hand, changes in the electric potential
95 give rise to the electric charge distribution⁴⁷

$$\varrho_{\text{el}}(\mathbf{x}, t) := -\frac{\delta F}{\delta V} = -\varepsilon \nabla^2 V = \varepsilon \nabla \cdot \mathbf{E}, \quad (9)$$

96 and to the electric force density

$$\mathbf{f}_{\text{el}} = \varrho_{\text{el}} \mathbf{E}, \quad (10)$$

97 which is the Lorentz force in the absence of magnetic fields⁴⁷.

98 The chemical and electrostatic force densities, Eqs. (8) and (10), together with the local

99 pressure gradient, $-\nabla p$, change the momentum of the fluid. The resulting total force density
 100 can be written in terms of a generalised pressure tensor, $\mathbf{\Pi}$, i.e.,

$$-\nabla \cdot \mathbf{\Pi} := -\nabla p + \mathbf{f}_{\text{cap}} + \mathbf{f}_{\text{el}}. \quad (11)$$

101 This leads to the expression

$$\mathbf{\Pi} = (\phi \vartheta - \psi) \mathbf{I} + \frac{3\gamma\ell}{\sqrt{8}} \nabla \phi \nabla \phi - \varepsilon \left(\mathbf{E} \mathbf{E} - \frac{1}{2} |\mathbf{E}|^2 \mathbf{I} \right), \quad (12)$$

102 where the last term in brackets is the Maxwell stress tensor⁴⁷ and \mathbf{I} is the identity matrix.

103 The equations of motion of the fluids are obtained as follows. First, imposing the con-
 104 servation of momentum leads to the incompressible Navier-Stokes equations

$$\rho (\partial_t + \mathbf{u} \cdot \nabla) \mathbf{u} = -\nabla \cdot [\mathbf{\Pi} - \mu (\nabla \mathbf{u} + \nabla \mathbf{u}^T)], \quad (13)$$

105 where $\mathbf{u}(\mathbf{x}, t)$, ρ and $\mu(\mathbf{x})$ are the velocity field, density and dynamic viscosity of the fluid, re-
 106 spectively, and the superscript T denotes matrix transposition. To allow viscosity differences
 107 between the two phases we impose the local viscosity as

$$\mu(\phi) := \frac{1 + \phi}{2} \mu_c + \frac{1 - \phi}{2} \mu_d, \quad (14)$$

108 where μ_c and μ_d are the bulk viscosities of the conductive and dielectric fluids.

109 Imposing the conservation of the phase field leads to a convection-diffusion equation,
 110 often referred to as the Cahn-Hilliard equation⁴⁸:

$$\partial_t \phi + \mathbf{u} \cdot \nabla \phi = M \nabla^2 \vartheta, \quad (15)$$

111 where M is called the mobility.

112 To complete the formulation of the problem, we need to specify the electrostatic force

113 density, which is a function of the potential field, V . In the following, we assume that both
 114 phases are ideal, i.e., the conductor has a vanishing electrical resistivity, while the dielectric
 115 has a vanishing electrical conductivity. It then follows that, since the electric field in the
 116 conductor is zero, the potential is constant in the bulk of that phase, i.e.,

$$V = V_0 \quad \text{as } \phi \rightarrow 1. \quad (16)$$

117 On the other hand, for a perfect dielectric $\rho_{\text{el}} = 0$, so Eq. (9) reduces to

$$\nabla^2 V = 0 \quad \text{as } \phi \rightarrow -1. \quad (17)$$

118 The boundary conditions for the coupled set of PDEs, equations (13), (15) and (17),
 119 are specified as follows. For the velocity field we impose the impenetrability and no-slip
 120 boundary conditions:

$$\mathbf{u}(\mathbf{x}_b) = 0 \quad \text{for } \mathbf{x}_b \in \partial\Omega. \quad (18)$$

121 For the phase field, we impose the natural boundary condition

$$\hat{\mathbf{n}} \cdot \nabla\phi(\mathbf{x}_b) = -\frac{\sqrt{8}}{3\gamma\ell}\zeta(\theta_0) \quad \text{for } \mathbf{x}_b \in \partial\Omega, \quad (19)$$

122 where $\hat{\mathbf{n}}$ is the unit normal to the solid boundary, and which enforces the wetting behaviour
 123 of the fluid-fluid mixture. Finally, for the potential we impose

$$V(\mathbf{x}_b) = V_b \quad \text{for } \mathbf{x}_b \in \partial\Omega, \quad (20)$$

124 where V_b is the potential at the boundary.

125 **Lattice-Boltzmann algorithm**

126 In this section we formulate a lattice-Boltzmann algorithm capable of integrating Eqs. (13), (15)
 127 and (17), subject to the boundary conditions (18)-(20).

128 The lattice-Boltzmann method is a computational fluid dynamics solver that iterates the
 129 discretised Boltzmann equations

$$f_q(\mathbf{x} + \mathbf{c}_q, t + 1) = f_q(\mathbf{x}, t) + \mathcal{C}[f]_q \quad (21)$$

130 and

$$g_q(\mathbf{x} + \mathbf{c}_q, t + 1) = g_q(\mathbf{x}, t) + \mathcal{C}[g]_q, \quad (22)$$

131 where f_q and g_q are particle distribution functions that represent the average number of
 132 fluid particles with position \mathbf{x} and velocity \mathbf{c}_q at time t . Space and time are discretised, and
 133 the velocity space is sampled by a finite set of vectors $\{\mathbf{c}\}_{q=0}^{Q-1}$, where Q is the number of
 134 directions in which the particle populations can move. Here, we use the D2Q9 model, which
 135 consists of a two-dimensional square lattice with $Q = 9$ (see Supplementary Information I).

136 The time evolution of the distribution functions, given by Eqs. (21) and (22), consists
 137 of a collision step and a streaming step. The collision step, performed by the second term
 138 on the right-hand-side in each equation, relaxes the distribution functions local equilibrium
 139 values, f_q^{eq} and g_q^{eq} . Here we use the Multi-Relaxation Time scheme (MRT) to model the
 140 collision of the f_q , i.e.,

$$\mathcal{C}[f]_q := - \sum_{r=0}^{Q-1} \Lambda_{qr} [f_r - f_r^{\text{eq}}](\mathbf{x}, t), \quad (23)$$

141 where the coefficients Λ_{qr} determine the relaxation rate to equilibrium. We use the Gram-
 142 Schmidt orthogonalisation procedure to define the coefficients of the collision operator⁴⁹.
 143 For the collision of the g_q we use the single-relaxation time approximation,

$$\mathcal{C}[g]_q := -\Lambda[g_q - g_q^{\text{eq}}](\mathbf{x}, t), \quad (24)$$

144 where we set $\Lambda = 1$, which helps improve the stability of the numerical method without loss
 145 of generality⁵⁰.

146 The connection between the lattice-Boltzmann equations and the hydrodynamic equa-
 147 tions is done by relating the moments of the distribution functions to the hydrodynamic
 148 fields. The local mass, momentum and phase fields correspond to

$$\rho = \sum_{q=0}^{Q-1} f_q, \quad (25)$$

149

$$\rho \mathbf{u} = \sum_{q=0}^{Q-1} \mathbf{c}_q f_q \quad (26)$$

150 and

$$\phi = \sum_{q=0}^{Q-1} g_q. \quad (27)$$

151 The equilibrium distributions, f_q^{eq} and g_q^{eq} , are constructed to convey the thermodynamic
 152 behaviour of the fluid and to ensure the local conservation of mass and momentum. This
 153 is done by requiring that their moments satisfy the conditions: $\sum_q f_q^{\text{eq}} = \rho$, $\sum_q g_q^{\text{eq}} = \phi$,
 154 $\sum_q \mathbf{c}_q f_q^{\text{eq}} = \rho \mathbf{u}$, $\sum_q \mathbf{c}_q g_q^{\text{eq}} = \phi \mathbf{u}$, $\sum_q \mathbf{c}_q \mathbf{c}_q f_q^{\text{eq}} = \mathbf{\Pi} + \rho \mathbf{u} \mathbf{u}$ and $\sum_q \mathbf{c}_q \mathbf{c}_q g_q^{\text{eq}} = 2M\vartheta \mathbf{I} + \phi \mathbf{u} \mathbf{u}$.
 155 Suitable expressions of the equilibrium distributions have been reported before^{48,51}. For the
 156 f_q^{eq} , we use

$$f_q^{\text{eq}}(\rho, \mathbf{u}, \mathbf{\Pi}) = w_q \left[\frac{1}{c_s} \rho \mathbf{u} \cdot H_q^{(1)} + \frac{1}{2c_s^2} (\mathbf{\Pi} + \rho \mathbf{u} \mathbf{u}) : H_q^{(2)} \right] \quad (28)$$

157 if $q \neq 0$, and

$$f_0^{\text{eq}}(\rho, \mathbf{u}, \mathbf{\Pi}) = \rho - \sum_{q=1}^{Q-1} f_q^{\text{eq}}. \quad (29)$$

158 For the g_q^{eq} , we use

$$g_q^{\text{eq}} = w_q \left[\frac{1}{c_s} \phi \mathbf{u} \cdot H_q^{(1)} + \frac{1}{2c_s^2} (2M\vartheta + \phi \mathbf{u} \mathbf{u}) : H_q^{(2)} \right], \quad (30)$$

159 if $q \neq 0$, and

$$g_0^{\text{eq}} = \phi - \sum_{q=1}^{Q-1} g_q^{\text{eq}}. \quad (31)$$

160 In these expressions, the w_q are weighting factors determined by the geometry of the lattice,
 161 $H_q^{(n)} = H^{(n)}(\mathbf{c}_q)$ is the tensor Hermite polynomial of n -th degree, and $c_s = 1/\sqrt{3}$ is a
 162 constant that represents the speed of sound⁵² (see Supplementary Information I for a list of
 163 expressions).

164 Using a Chapman-Enskog expansion, Eqs. (21) and (22), together with Eqs. (23)–(31),
 165 reduce to the Navier-Stokes (13) and Cahn-Hilliard (15) equations. From the expansion, the
 166 viscosity, μ , is determined by the coefficients of the collision matrix, Λ_{qr} ⁴⁹ (see Supplementary
 167 Information I).

168 **The electric potential**

169 To model the effect of the electrostatic potential field, it suffices to introduce an algorithm
 170 that solves Laplace’s equation in the dielectric, whilst keeping the potential to a constant
 171 value in the conductor.

172 Hence, we take inspiration from the diffusive dynamics which arises from the LBM itself⁵³,
 173 and introduce a third lattice-Boltzmann equation in the following form,

$$h_q(\mathbf{x} + \mathbf{c}_q, t + 1) = h_q(\mathbf{x}, t) + \mathcal{C}[h]_q, \quad (32)$$

174 where we use a single-relaxation-time collision operator,

$$\mathcal{C}[h]_q := -\Lambda[h_q - h_q^{\text{eq}}](\mathbf{x}, t), \quad (33)$$

175 where $\Lambda = 1$.

176

This new distribution function is related to the local electric potential, V , by the relations

$$V = \sum_q h_q, \quad (34)$$

177 and

$$h_q^{\text{eq}} = w_q V. \quad (35)$$

178

Eq. (35) offers the advantage of setting the electric potential to a prescribed value, by fixing the right-hand side, and thus allows the modelling of a conducting liquid (for which the potential equilibrates to a constant).

181

We now analyse the long-time, large-lengthscale behaviour of Eqs. (32)-(35). First, we express Eq. (32) in terms of the equilibrium distribution, h_q^{eq} , using Eq. (35). This is done by writing the collision step as a differential operator acting on h_q^{eq} (for details, see Supplementary Information II), i.e.,

$$- [h_q - h_q^{\text{eq}}] = (\partial_t + \mathbf{c}_q \cdot \nabla) h_q^{\text{eq}} - \frac{1}{2} (\partial_t + \mathbf{c}_q \cdot \nabla)^2 h_q^{\text{eq}} + \dots \quad (36)$$

185

Applying the summation operator, \sum_q , to Eq. (36), and using Eqs. (34) and (35), we find

$$0 = \partial_t V - \frac{1}{2} \partial_t^2 V - \frac{1}{2} c_s^2 \nabla^2 V + \dots \quad (37)$$

186

where we identify $\epsilon = c_s^2/2$. During a relaxation process the first and second terms in Eq. (37) will asymptotically vanish, and thus, V will satisfy Eq. (17) at long times. In the context of electrowetting, one requires that this relaxation is faster than the typical timescales of the hydrodynamic fields, \mathbf{u} and ϕ .

190

To quantify the transient, let us investigate the solutions of Eq. (37). Since the equation is linear, we proceed in the standard way by proposing the *Ansatz* $V = X(\mathbf{x})T(t)$ ⁵⁴. This

191

192 leads to the ordinary differential equation for the temporal part,

$$2\frac{dT}{dt} - \frac{d^2T}{dt^2} + c_s^2 K^2 T = 0, \quad (38)$$

193 and a partial differential equation for the spatial part,

$$\nabla^2 X + K^2 X = 0, \quad (39)$$

194 where $K = \text{const.}$, is the eigenvalue that couples the system of equations.

195 For the temporal part, Eq. (38), we look for solutions that decay at long times, i.e.,

$$T(t) = \exp \left[(1 - \sqrt{1 + c_s^2 K^2}) t \right], \quad (40)$$

196 where the term in brackets is always negative for non-vanishing K .

197 To better understand the rate of decay of the transient, which is controlled by K , let us
 198 focus on the limiting case of a uniform dielectric phase in a rectangular domain of size
 199 $L_x \times L_y$. In such a case, Eq. (39) can be solved analytically⁵⁴, leading to the spectrum of
 200 eigenvalues

$$K^2 = \left(\frac{2\pi l}{L_x} \right)^2 + \left(\frac{2\pi m}{L_y} \right)^2, \quad (41)$$

201 where l and m are positive integers. Let us now define the transient period, τ_{trans} , as the
 202 characteristic decay time associated to the smallest eigenvalue,

$$\tau_{\text{trans}} := \max \frac{1}{\sqrt{1 + c_s^2 K^2} - 1}, \quad K \neq 0, \quad (42)$$

203 which, for the uniform rectangular domain, is

$$\tau_{\text{trans}} \leq \frac{1}{2} \left(\frac{\max(L_x, L_y)}{\pi c_s} \right)^2. \quad (43)$$

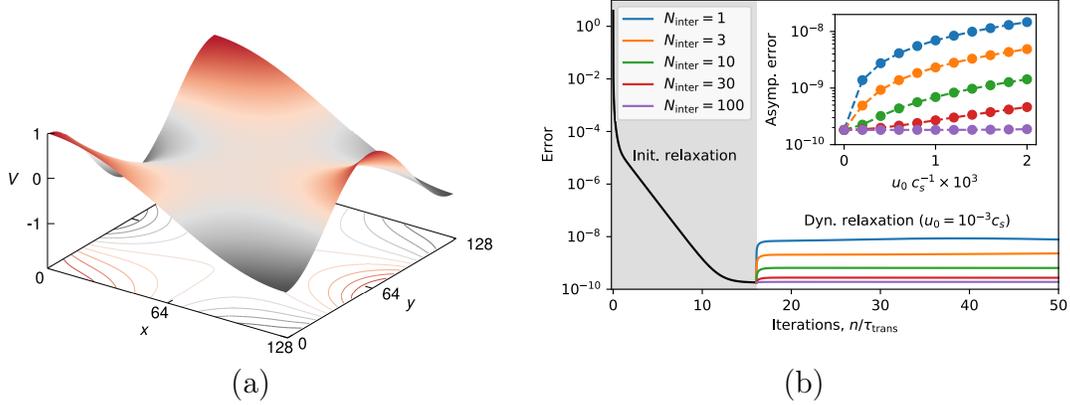


Figure 1: (Colour online) Convergence of the lattice-Boltzmann algorithm to model the relaxation of the electric potential field. (a) Surface plot of the electric potential obtained after an initial relaxation of $n = 16 \tau_{\text{trans}}$ iterations. (b) Semi-log plot of the evolution of the error as a function of the number of iterations. The grey area corresponds to the initial relaxation. Afterwards, evolution of the error is subject to the changes in the boundary conditions and it is represented by curves for different interleaved iterations, N_{inter} . The inset shows the asymptotic behaviour of the error with respect to the drift velocity of the boundaries, u_0 . The size of the simulation domain is $L_x = 128$ by $L_y = 128$.

204 The presence of a conductive phase will effectively reduce the domain of Eq. (39), and
 205 thus, will shift the spectrum of K to higher values. This implies that, Eq. (43) is an upper
 206 bound for the transient from arbitrary initial conditions to a steady state solution.

207 However, if the initial conditions for the electric field are close to a stationary solution, the
 208 transient number of iterations required to relax a small perturbation will be much smaller.
 209 For instance, introducing a perturbation of the order of one lattice unit to a stationary
 210 solution will lead to $K \approx 2\pi$. Hence, from Eq. (42), the transient reduces to

$$\tau_{\text{trans}} \approx \frac{1}{\sqrt{1 + (2\pi c_s)^2} - 1} < 1. \quad (44)$$

211 Such a fast relaxation can be particularly useful, for instance, when the bulk electrostatic
 212 potential V_0 is varied quasi statically to explore stationary wetting configurations, were a
 213 single iteration might be enough to update the electrostatic field.

214 To test the speed of convergence of the method to solve Laplace's equation, we carried out

215 a set of simulations of a 2D domain of dimensions $L_x \times L_y$ filled with a single dielectric fluid.
 216 We tracked the evolution of the electric potential field, $V(x, y, t)$, subject to the Dirichlet
 217 boundary conditions

$$V_b(x, y, t) = \cos \left[\frac{\pi x}{L_x} \right] \cos \left[\frac{2\pi}{L_y} (y - u_0 t) \right] \quad (45)$$

218 at $y \in [1, L_y]$ if $x = 1, L_x$ and at $x \in [1, L_x]$ if $y = 1, L_y$ (see Fig. 1a). To characterise
 219 dynamic changes in the potential field (e.g., due to a moving phase) we introduce the drift
 220 velocity u_0 .

221 As explained above, the algorithm solves Eq. (37), which is expected to converge to
 222 Laplace’s equation (Eq. (17)) at long times. Henceforth, we define

$$\text{Er} := \frac{[\sum_{\mathbf{x}} |\nabla^2 V|^2]^{1/2}}{L_x L_y |\max V - \min V|} \quad (46)$$

223 as the relative error per unit simulation node.

224 We first tested the speed of convergence of the algorithm from a random initial condition,
 225 which we allowed to relax for $n = 16 \tau_{\text{trans}}$ iterations. We set the drift velocity to $u_0 = 0$.
 226 Fig. 1a shows a surface plot of $V(x, y)$ after relaxation. The error, reported in Fig. 1b, drops
 227 below 10^{-4} after $n \sim \tau_{\text{trans}}$ and saturates below 10^{-9} for $n > 10\tau_{\text{trans}}$.

228 Introducing a finite drift velocity, u_0 , is useful to understand how quickly the algorithm
 229 “catches up” in dynamic situations. Hence, we tested the convergence of the error by allowing
 230 for a different number of interleaved iterations of the potential field within a single timestep,
 231 N_{inter} . Fig. 1b shows the convergence curves of the error for $u_0 = 10^{-3} c_s$, which saturate to a
 232 maximum error $\approx 10^{-8}$ for $N_{\text{inter}} = 1$. Hence, for a given value of the typical velocity in the
 233 simulations, the error can be reduced by increasing N_{inter} . For example, as shown in Fig. 1(b)
 234 inset, for $u_0 < 10^{-3} c_s$ (corresponding to a Mach number $\text{Ma} := u_0/c_s < 10^{-3}$), one interleaved
 235 iteration is enough to maintain $\text{Er} < 10^{-8}$. Typically, the motion of viscous conducting and
 236 dielectric fluids in electrowetting setups is damped by friction forces, corresponding low
 237 Reynolds and capillary numbers which also correspond to low Ma.

238 **Simulation setup: initial and boundary conditions**

239 We now describe the simulation implementation to model the dynamics in an EWOD setup.
 240 The electric potential and its corresponding distribution function are defined in a simulation
 241 box of size $L_x \times L_y$. The two-phase fluid and corresponding distribution functions are defined
 242 in a simulation box of size $L_x \times (L_y - 2d)$, where d is a gap used to accommodate for a solid
 243 dielectric layer. This has the purpose of isolating the conductive phase from the bounding
 244 electrodes on the finite domain, and thus, to avoid divergences in the electric field. The
 245 permittivity of the solid dielectric is set equal to the permittivity of the dielectric fluid.

246 The velocity field is set to

$$\mathbf{u}(\mathbf{x}, t = 0) = 0 \tag{47}$$

247 everywhere in the simulation domain. The phase field, is initialised to

$$\phi(\mathbf{x}, t = 0) = \phi_i(\mathbf{x}), \tag{48}$$

248 which we specify for the specific configurations reported in the following sections. The electric
 249 potential is initialised as follows.

$$V(\mathbf{x}, t = 0) := \begin{cases} V_0, & \text{if } \phi > 0, \\ V_0/2 & \text{if } \phi \leq 0. \end{cases} \tag{49}$$

250 At subsequent times, and in order to avoid discontinuities in electric field (which would
 251 lead to a diverging electrostatic force), we introduce a smoothing algorithm of the electro-
 252 static potential as follows,

$$V(\mathbf{x}, t) = \beta V_0 + (1 - \beta) \sum_q h_q(\mathbf{x}, t), \tag{50}$$

253 where β is an interpolation weight defined as

$$\beta(\phi(\mathbf{x})) := \begin{cases} 1 & \text{for } \phi > \phi_{\text{thr}} \\ \phi/\phi_{\text{thr}} & \text{for } 0 < \phi < \phi_{\text{thr}} , \\ 0 & \text{otherwise,} \end{cases} \quad (51)$$

254 where $\phi_{\text{thr}} = 0.9$, is a threshold value set to identify the bulk of the conductor. In this way,
 255 the potential is fixed to the prescribed value V_0 at the bulk of the conductive phase, whereas
 256 it evolves according to Eq. (34) in the bulk of the dielectric phase.

257 Using this setup, we found that the electric potential relaxes to a steady state typically
 258 after $L_x^2/8$ iterations. Nonetheless, since transient hydrodynamic flows are slow compared to
 259 the speed of sound ($|\mathbf{u}| \ll c_s$), we found that the distribution function h_q could be updated
 260 at the same pace as f_q and g_q , with only one iteration required to relax the electric potential
 261 field.

262 We impose periodic boundary conditions along the x and y directions, and fix the solid
 263 electrode at the top and bottom boundaries of the simulation domain. To implement the
 264 no-slip boundary condition at the solid surface we use the bounce-back algorithm⁵⁵. To im-
 265 plement the wettability of the surface, Eq. (19), we compute the gradient and Laplacian of
 266 the phase field at near-boundary nodes using finite differences to then fix the corresponding
 267 incoming distribution functions from the solid surface^{44,51}. Finally, to implement the bound-
 268 ary condition on the voltage, V_b , we follow a similar approach to that of Ledesma-Aguilar,
 269 et al.⁵³. We specify the distribution functions streaming from sites on the solid electrode,
 270 of position vector \mathbf{x}_b , to sites in the fluid near the solid boundary, of position vector \mathbf{x}_{nb} ,
 271 according to

$$h_{\bar{q}}(\mathbf{x}_{\text{nb}}, t) = w_{\bar{q}} V_b, \quad (52)$$

272 where the indices \bar{q} correspond to the distribution functions that stream away from the
 273 boundary. Specifically, $\bar{q} \in \{q : \mathbf{c}_q + \mathbf{c}_{q'} = 0, q \in \Gamma\}$, where $\Gamma := \{q : \mathbf{x}_{\text{nb}} + \delta\mathbf{c}_q = \mathbf{x}_b, 0 <$

274 $\delta < 1$ gives the indices of lattice vectors that stream towards the electrode.

275 In summary, the simulation algorithm is carried out as follows. The hydrodynamic be-
 276 haviour of the fluid is solved by the first lattice-Boltzmann equation, Eq. (21). The behaviour
 277 of the flow is subject to the stresses defined by the pressure tensor $\mathbf{\Pi}$, and are included by
 278 the equilibrium distribution function according to Eqs. (28) and (29). As a result, we obtain
 279 the velocity field at the next timestep by means of Eqs. (25) and (26). The second lattice-
 280 Boltzmann equation, Eq. (22), gives rise to the advection and diffusion of the phase field,
 281 ϕ . This is done by defining its equilibrium distribution function in Eq. (30) and (31), and
 282 by evaluating Eq. (27). In this way, the shape and position of the conducting and dielectric
 283 phases is obtained at the next iteration step. The third lattice-Boltzmann equation, Eq. (32),
 284 gives the electric potential. This is done by setting Eq. (35) and defining the voltage which
 285 is coupled by the phase field according to Eq. (50). Finally, by means of Eqs. (7) and (12),
 286 the phase field and electric potential sets the value of the chemical potential and pressure
 287 tensor, thus closing the iteration cycle.

Table 1: Parameters for the simulations of the spreading of a droplet and the dielectric film dynamics in Lattice-Boltzmann units.

Simulation parameter	Symbol	Value in §	Value in §
Simulation box	$L_x \times L_y$	512×288	$[418, 1256] \times 84$
Surface tension	γ	6×10^{-3}	8×10^{-3}
Interface width	ℓ	4	5
Contact angle	θ_0	$[160^\circ, 120^\circ]$	180°
Density	ρ	1	1
Dynamic viscosity	μ_c, μ_d	1/6, 1/6	1/600, 1/3
Mobility	M	1/10	1/10
Permittivity	ε	1/6	1/6
Dielectric thickness	d	2	2
Initial config.		$R_0 = 128$	$a = 1, H_0 = 20$

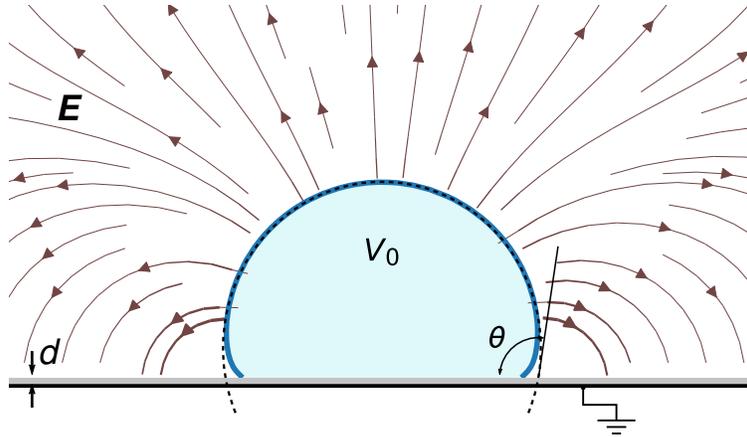


Figure 2: (Colour online) 2D LB simulations of a droplet in an EWOD set-up. A droplet of conducting liquid sits on top of a dielectric solid of thickness d (grey line). The droplet is set to an electric potential V_0 and, on the other side of the dielectric surface, the electric potential is set to zero (black line). The dielectric fluid surrounds the droplet where the electric field, \mathbf{E} is shown by the stream plot. The dashed line corresponds to the fitting circle that defines the new contact angle θ .

288 Results and Discussion

289 Electrowetting of a droplet

290 In this section we validate the lattice-Boltzmann algorithm by studying the electrowetting-
 291 driven spreading of a droplet in an EWOD setup. We start by reviewing the Young-Lippmann
 292 classical theory of electrowetting^{1,39}, before comparing to our simulation results.

293 Review of the Young-Lippmann Theory

294 Consider a droplet of a conductive liquid in an EWOD setup as shown in Fig. 2. As the
 295 potential difference applied between the droplet and the electrode is increased, the electric
 296 charges begin to gather at the interface of a conductive liquid with a higher density near the
 297 grounded electrode. This configuration corresponds to a capacitor. Therefore, and neglecting
 298 the charges that accumulate on the opposite side of the solid dielectric, the electrostatic
 299 energy, per unit surface area of the electrode, is $cV_0^2/2$, where c is the capacitance per unit
 300 area of the configuration⁴⁶. Because the droplet's surface is compliant, the electrostatic force

301 leads to a spreading of the liquid on the solid electrode.

302 The equilibrium configuration of the droplet will be determined by the balance between
303 the work done by the electric field against the increase in surface energy. Mechanically, an
304 infinitesimal radial displacement of the contact line, dR , results in a net radial force on the
305 interface of the droplet. Hence, mechanical equilibrium is achieved when

$$0 = (\gamma_{sd} - \gamma_{sc} - \gamma \cos \theta + cV_0^2/2)dR. \quad (53)$$

306 Using Eq. (3) and dividing by γ , Eq. (53) results in the Young-Lippmann relation¹,

$$\cos \theta(V_0) = \cos \theta_0 + \eta, \quad (54)$$

307 where

$$\eta := \frac{cV_0^2}{2\gamma} \quad (55)$$

308 is the electrowetting number.

309 Therefore, the contact angle of a droplet is reduced with increasing applied voltage.
310 Experimentally, Young-Lippmann's result has been verified over a range of voltages. However,
311 it has also been observed that at high voltages the contact angle reaches a saturation value,
312 beyond which the theory is no longer valid^{56,57}.

313 **Lattice-Boltzmann simulations**

The initial configuration of the system consists of a circular droplet of the conducting liquid suspended in the dielectric fluid. We impose the initial conditions in the simulations using Eqs. (47), (48) and (49); the initial phase field reads

$$\phi_i(\mathbf{x}) = \tanh \left(\frac{R_0 - |\mathbf{x} - \mathbf{X}_0|}{\sqrt{2}\ell} \right), \quad (56)$$

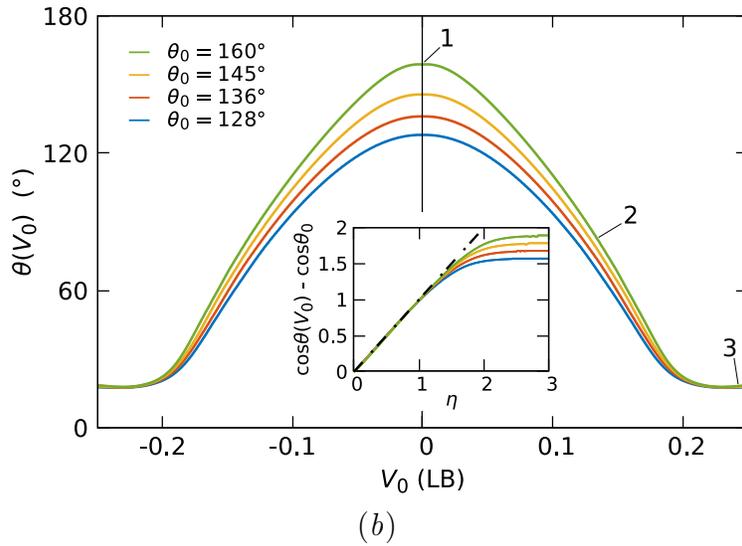
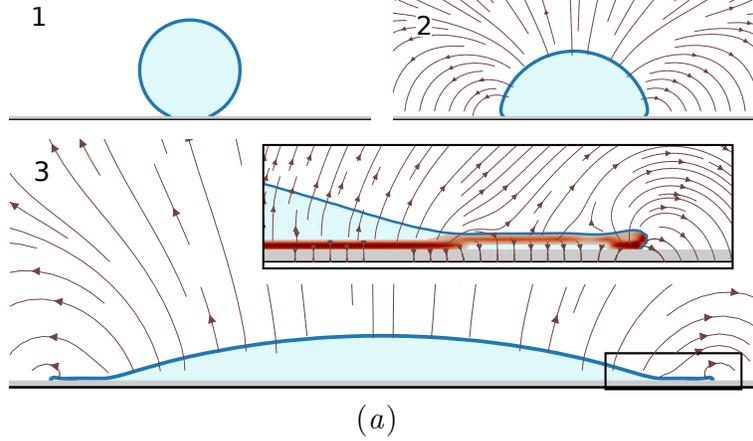


Figure 3: (Colour online) Simulations of droplet spreading using an EWOD setup. (a) Stationary droplet configurations at different applied voltage, V_0 . At $V_0 = 0$, the shape of a droplet is circular and intersects the solid dielectric at the equilibrium contact angle θ_0 . For $|V_0| > 0$, the shape of the droplet close to the solid wall is distorted by the electric field, leading to an apparent contact angle, $\theta(V_0)$. At high applied voltages, the droplet reaches a limiting configuration, where the main drop develops a lip that spreads away from its centre. The region around the lip shows strong fringe fields (inset) and the charge density (dark-red colour map). (b) Variation of the contact angle in response to the electric potential, V_0 , in lattice-Boltzmann units. The curves show a monotonic decrease in the contact angle with the increasing magnitude of the potential. The inset shows the expected universal collapse as a function of the electrowetting number, η , predicted by the Young-Lippmann relation (dotted-dashed line) at low electrowetting numbers and a later saturation.

314 where $\mathbf{X}_0 = (L_x/2, R_0)$, is the initial position of the centre of the droplet, and R_0 its initial
315 radius. The rest of the simulation parameters are summarised in Table 1.

316 We first set the potential within the conducting droplet to $V_0 = 0$ and allow the system

317 to relax for 2×10^5 iterations. As the droplet relaxes, it spreads on the surface and acquires
 318 a circular-cap shape intersecting the surface with the expected equilibrium contact angle, θ_0 ,
 319 predicted by Eq. (4). Then, we increase the voltage by an amount $0.01\sqrt{2\gamma d/\varepsilon}$ and allow the
 320 system to relax for a further 10^4 iterations. Once the relaxation has elapsed, the stationary
 321 configuration is recorded. The increment in the applied voltage is repeated until a maximum
 322 voltage $V_0 = 3\sqrt{2\gamma d/\varepsilon}$ is reached.

323 Fig. 2 shows a typical equilibrium configuration of the droplet subject to a non-zero
 324 potential. The upper part of the droplet conserves a circular shape that, extrapolated,
 325 intersects the surface at an apparent contact angle $\theta(V_0)$. However, near the solid surface,
 326 the inclination of the interface is closer to the prescribed equilibrium contact angle^{1,58}. As
 327 shown in Fig. 3b, the apparent contact angle decreases with increasing $|V_0|$. Note that
 328 reversing the polarity of the applied voltage leads to the same decrease in the apparent angle;
 329 this is expected, since Eq. (10) is invariant upon an inversion of the polarity of the electric
 330 potential ($V \rightarrow -V$). Therefore, the simulations capture the competition between electrical
 331 and capillary forces, as has been reported previously in experimental observations³².

332 Next, we carried out simulations to measure $\theta(V_0)$ for different values of the equilibrium
 333 contact angle, θ_0 . As shown in Fig. 3b, the $\theta(V_0)$ curves follow the same trend, with only a
 334 shift of the maximum to a value imposed by θ_0 . As shown in the inset, a plot of $\cos \theta(V_0) -$
 335 $\cos \theta_0$ shows a linear dependence on η , which is in agreement with the theoretical prediction
 336 of Eq. (54). Fitting the simulation data to a straight line gives $c \approx 0.66$.

337 As the voltage in the droplet is increased, the apparent contact angle reaches a saturation
 338 value $\theta \approx 18.43^\circ$. The saturation effect was found to be independent of the wettability of
 339 the surface, and begins to occur when the droplet reaches $\theta \sim 50^\circ$. From the simulations, we
 340 observe that at the onset of saturation the droplet develops two distinct regions. Close to
 341 its centre, the capillary forces smooth out the shape of the interface, which remains circular.
 342 However, the region close to the edge is subject to strong fringe fields, and deforms to take
 343 the shape of a ‘lip’, spreading away from the main drop (see panel 4 in Fig. 3(a)). The result

344 is that the bulk profile retains a limiting shape, characterised by the saturation contact angle,
345 while an increase in the voltage results in a further growth of the edge lip.

346 **Dynamics of a thin dielectric film**

347 In this section we illustrate the applicability of the lattice-Boltzmann algorithm to resolve
348 the dynamics of electrowetting liquids. Specifically, we study the stability of a thin dielectric
349 film confined between a solid charged wall and a conductive liquid layer. This problem is
350 relevant in many electrowetting setups, where the spreading conductive liquid often entraps
351 a thin film of dielectric fluid. As the dielectric film becomes thinner, it breaks up into small
352 droplets²⁵.

353 We start by formulating the problem analytically, which yields a prediction of the stability
354 of the film in the linear regime. We then report simulation results which we validate against
355 this prediction, and extend our study to report results of the dynamics of the film at long
356 times, including the regime of film breakup and droplet formation.

357 **Linear-stability theory**

358 We consider a thin, two-dimensional dielectric film of local thickness $H(x, t)$. The film lies on
359 top of a conducting solid electrode, located at $y = -d$ which is coated with a thin dielectric
360 solid layer of thickness d . At its top, the film is covered by a layer of conducting liquid of
361 negligible viscosity.

362 To model the dynamics of the thin dielectric layer in the presence of an electric field, we
363 use the lubrication equation⁵⁹,

$$\partial_t H - \partial_x \left(\frac{H^3}{3\mu_d} \partial_x p_{\text{film}} \right) = 0. \quad (57)$$

364 As shown by Eq. (57), the dynamics is driven by variations in the pressure within the film,
365 p_{film} . This is composed of a capillary contribution, $2\gamma\kappa$, and by a contribution due to the

366 electric stresses on the dielectric fluid, $-\frac{1}{2}\partial_H c V_0^2$. For a gently curved interface, $\kappa \approx -\frac{1}{2}\partial_x^2 H$.
 367 Hence,

$$p_{\text{film}} = -\gamma\partial_x^2 H - \frac{1}{2}(\partial_H c)V_0^2. \quad (58)$$

368 where we assume that the capacitance c for a dielectric film in contact with the dielectric
 369 solid layer is given by

$$c = \frac{\varepsilon}{H + d}. \quad (59)$$

370 We now study the stability of the dielectric film by analysing Eq. (57) using a perturbative
 371 approach. Let us consider the sinusoidal interface profile

$$H(x, t) = H_0 + a \cos(2\pi x/\lambda) \exp(t/\tau), \quad (60)$$

372 where H_0 is the average height of the film, a is the amplitude of the perturbation, λ the
 373 wavelength and τ is the characteristic growth time.

374 Substituting Eq. (60) into Eq. (57), and assuming $a \ll H_0$ gives the dispersion relation

$$\omega = \frac{1}{3}k^2 \left[2\eta \left(\frac{H_0}{H_0 + d} \right)^2 - k^2 \right], \quad (61)$$

375 where $\omega := \mu_d H_0 / \gamma \tau$ is the dimensionless growth rate, and $k := 2\pi H_0 / \lambda$ is the dimensionless
 376 wave number.

377 The first term in Eq. (61) corresponds to the destabilising effect of the electric field, which
 378 dominates for long-wavelength perturbations. This competes against the stabilising effect of
 379 surface tension, which dominates for short wavelengths. Setting $\omega = 0$, corresponding to the
 380 onset of instability, gives the separatrix

$$\eta = \frac{1}{2} \left(\frac{H_0 + d}{H_0} \right)^2 k^2, \quad (62)$$

381 which gives the minimum electrowetting number for which a perturbation of given wave

382 number leads to instability.

383 **LB simulations**

We impose the initial conditions in the simulations using Eqs. (47), (48) and (49); we introduce an initial perturbation to the interface between the conductive and dielectric fluids by imposing the phase-field profile

$$\phi_i(\mathbf{x}) = \tanh\left(\frac{y - H(x, 0)}{\sqrt{2}\ell}\right), \quad (63)$$

384 with corresponds to a sinusoidal perturbation of amplitude $a = 1$ and wavelength $\lambda = L_x$.
385 The rest of the simulation parameters are reported in the last column of Table 1. To allow the
386 thermodynamic relaxation of the phase field from the initial conditions, we let the simulations
387 run for 10^3 iterations, which we disregard.

388 Fig. 4(a) shows a typical instantaneous configuration of the film after the transient has
389 elapsed. Henceforth, we track the evolution of the fluid-fluid interface, whose location we take
390 as the level curve $\phi(x, y) = 0$. Once the location of the interface is determined, the amplitude
391 of the perturbation is found by fitting the instantaneous level curves to the sinusoidal function
392 $y(x) = c_0 + c_1 \cos(2\pi x/L_x)$, where c_0 and c_1 are fitting parameters. We then fit the measured
393 amplitude data, $c_1(t)$, to the exponential function $A(t) = c_2 \exp(t/c_3)$, where c_3 gives the
394 characteristic growth time. To obtain the dependence of the dispersion relation, for a given
395 electrowetting number, we repeat the simulation by varying the system length, L_x (see
396 Table 1).

397 Figs. 4(b) and 4(c) show the dispersion relations obtained from the simulations for $\eta = 0$
398 and $\eta = 0.03$. The data in the figures is reported in the dimensionless units of Eq. (61),
399 where μ_d , γ , H_0 are fixed using the values reported in Table 1. For $\eta = 0$, we observe the
400 expected power-law decay, $\omega \propto -k^4$, predicted by the linear stability analysis. For $\eta = 0.03$,
401 the dispersion relation shows a range of unstable wave numbers. In both cases, we find a

402 quantitative agreement with Eq. (61), which is superimposed to the simulation data as a
403 dashed line.

404 We measured the growth rate of the perturbation for 21×21 points in the η - k space.
405 Fig. 4(d) shows the simulation results, which we present as a contour plot of ω vs η and
406 k . The separatrix, corresponding to the curve $\omega(k, \eta) = 0$, was estimated from the data
407 using bilinear interpolation (solid line in the figure). Overall, there is a good agreement with
408 Eq. (62) (shown as a dashed line). We attribute the small discrepancy between the theory
409 and the simulation results to the charge distribution at the diffuse fluid-fluid interface, which
410 is dispersed in a region of the order of the interface thickness ℓ . This effect would then alter
411 the capacitance of the dielectric film. Indeed, by fitting the separatrix obtained from the
412 simulations to Eq. (62), we obtain an effective value for H_0 , which is displaced by a small
413 amount ($\sim 0.08\ell$) into the bulk of the conductive phase.

414 We now turn our attention to the growth of the perturbation at long times, when $a/H_0 \sim$
415 1. This regime, which is not accessible by the linear theory, is revealed in detail by the
416 simulations. As shown in Fig. 5(a), at large perturbation amplitudes inhomogeneities in the
417 electric field become apparent. The simulations capture the increase in charge density in
418 regions where the interface curvature is higher⁴⁷. This effect leads to a stronger electrostatic
419 attraction in regions of the interface which lie closer to the solid electrode. As a result, the
420 perturbation grows faster than predicted by the linear theory, and the interface is deformed
421 to an asymmetric shape.

422 At longer times, the troughs of the perturbation approach the solid surface. In this
423 regime, we found that the wettability of the solid has a strong effect on the dynamics. For
424 $\theta_0 < 180^\circ$ the fluid-fluid interface touches the solid surface, breaking the film into droplets.
425 The subsequent dynamics of the fluid-fluid interface is similar to the dewetting dynamics
426 observed by Edwards, et al.⁶⁰: the retracting edges collect fluid to form dewetting rims, which
427 eventually merge to form a single circular droplet (see Fig. 5(b)). As soon as the conducting
428 fluid reaches the solid surface the spreading begins (frame 2 in Fig. 5(b,c)). Initially, the

429 spreading occurs at a constant pace (frame 3) and ends in an asymptotic relaxation (frame
430 4). For $\theta_0 = 180^\circ$, the conducting fluid cannot wet the surface and, hence, the dielectric
431 film does not break up. Therefore, the film takes the shape of a series of ‘bumps’ which
432 remain connected by a thin film (of a thickness set by the range of the wetting potential
433 in the simulations). This situation is reminiscent of the oil entrapment regime reported by
434 Saticu et al.²⁵, who used an EWOD setup to spread water droplets immersed in silicone oil
435 on Teflon-coated electrodes.

436 Conclusions

437 We have introduced a lattice-Boltzmann algorithm capable of solving the coupled hydro-
438 dynamics and electrostatics equations of motion of a two-phase fluid as a means to model
439 electrowetting phenomena. Our method uses a set of distribution functions to solve the
440 electrostatics equations within the lattice-Boltzmann algorithm itself, eliminating the need
441 for concurrent methods to model the electric field, such as finite differences or finite element
442 methods. We have used a diffuse-interface approach to model the dynamics of immiscible
443 conducting and dielectric fluid phases, thus eliminating the need to track the fluid-fluid in-
444 terface at every iteration step. Instead, the electric potential field in our method is coupled
445 to the multiphase fluid domain, with electric stresses arising at the diffuse interface between
446 conducting and dielectric phases.

447 We have validated our algorithm by presenting numerical simulations of the electrowetting
448 of a droplet in an Electro Wetting On Dielectric (EWOD) setup. Our results reproduce the
449 dependence of the apparent contact angle of the droplet on the applied voltage predicted
450 by the Young-Lippman theory. We also observe a saturation of the contact angle at high
451 voltages. The saturation of the contact angle has been reported in experiments, and remains
452 an open question in the field of electrowetting. In the simulations, the effect is linked to a
453 saturation of the interface curvature, which triggers the formation of a ‘lip’ at the droplet’s

454 edge. Such a balance between the electric and capillary stresses in the simulations might
455 explain the saturation effect observed in experiments, but further experimental evidence is
456 needed to reach a conclusion in this regard.

457 We have also used our algorithm to study the stability and dynamics of a thin dielectric
458 film in an EWOD setup. For small perturbations, our simulations results agree well with
459 the prediction of lubrication theory. Beyond this small-perturbation regime accessible by
460 theory, we studied the long-time dynamics of the film. Our simulations show that as the
461 film is destabilised and the interface approaches the solid surface. On wettable surfaces, the
462 film breaks up and forms droplets that dewet from the surface. On non-wettable surfaces,
463 we observe the entrapment of the dielectric film and the stabilisation of mound-shaped
464 structures.

465 Here we have focused on droplet electrowetting as a situation of broad importance to val-
466 idate our model. However, our method is amenable to study other situations of both funda-
467 mental and applied interest. For example, the scheme presented here could be used to study
468 the stability of moving contact lines subject to electrowetting actuation, a situation that
469 has been reported to lead to the formation of micron-sized “bubbles” of the non-conducting
470 phase which remain adhered to the solid^{24,25}. Because of the ability of lattice-Boltzmann
471 to incorporate solid boundaries of arbitrary geometry, our model could also be used to un-
472 derstand the interplay between electrowetting actuation in patterned surfaces⁶⁰ or within
473 micro-channels. Here we have studied fluid phases of uniform density, whose dynamics can
474 be captured using an orthogonal decomposition method to determine the collision operator
475 in the lattice-Boltzmann equation. However, our method could also be tested with recently
476 developed non-orthogonal schemes⁶² to model situations where the fluids have different den-
477 sities. Such a situation is relevant to the electrowetting of a gas phase, e.g., to manipulate
478 bubbles. Finally, here we have restricted ourselves to study the electrowetting of a perfect
479 conductor surrounded by a perfect dielectric. However, by including a source in the equa-
480 tion for the electric potential, our method could be extended to model imperfect conductors,

481 where the charge density inside the conducting fluid is redistributed due to the osmotic pres-
482 sure and therefore inducing a non-vanishing electric field inside⁶¹, or imperfect dielectrics,
483 which exhibit breakdown or leakage currents³³.

484

485 **Supporting Information.** Details of the lattice-Boltzmann algorithm.

486 Acknowledgement

487 The authors acknowledge support from EPSRC Grant Nos. EP/P024408/1 and EP/R036837/1,
488 and from EPSRC's UK Consortium on Mesoscale Engineering Sciences (Grant No. EP/R029598/1).

489 References

- 490 (1) Mugele, F.; Baret, J.-C. Electrowetting: from basics to applications. *J. Phys.: Condens.*
491 *Matter* **2005**, *17*, R705.
- 492 (2) Wang, K.-L.; Jones, T. Electrowetting Dynamics of Microfluidic Actuation. *Langmuir*
493 **2005**, *21*, 4211–4217.
- 494 (3) Teh, S.-Y.; Lin, R.; Hung, L.-H.; Lee, A. P. Droplet microfluidics. *Lab Chip* **2008**, *8*,
495 198–220.
- 496 (4) Ren, H.; Fair, R. B.; Pollack, M. G.; Shaughnessy, E. J. Dynamics of electro-wetting
497 droplet transport. *Sens. Actuators, B* **2002**, *87*, 201–206.
- 498 (5) Fair, R. B. Digital microfluidics: is a true lab-on-a-chip possible? *Microfluidics Nanoflu-*
499 *idics* **2007**, *3*, 245–281.
- 500 (6) Baret, J.-C.; Decré, M.; Herminghaus, S.; Seemann, R. Electroactuation of Fluid Using
501 Topographical Wetting Transitions. *Langmuir* **2005**, *21*, 12218–12221.

- 502 (7) Schultz, A.; Papautsky, I.; Heikenfeld, J. Investigation of Laplace Barriers for Arrayed
503 Electrowetting Lab-on-a-Chip. *Langmuir* **2014**, *30*, 5349–5356.
- 504 (8) Mugele, F.; Baret, J.-C.; Steinhauser, D. Microfluidic mixing through electrowetting-
505 induced droplet oscillations. *Appl. Phys. Lett.* **2006**, *88*, 204106.
- 506 (9) Ko, S.; Lee, H.; Kang, K. H. Hydrodynamic Flows in Electrowetting. *Langmuir* **2008**,
507 *24*, 1094–1101.
- 508 (10) Lu, Y.; Sur, A.; Pascente, C.; Annapragada, S. R.; Ruchhoeft, P.; Liu, D. Dynamics of
509 droplet motion induced by Electrowetting. *Int. J. Heat Mass Transf.* **2017**, *106*, 920 –
510 931.
- 511 (11) Kudina, O.; Eral, B.; Mugele, F. e-MALDI: An Electrowetting-Enhanced Drop Drying
512 Method for MALDI Mass Spectrometry. *Anal. Chem.* **2016**, *88*, 4669–4675.
- 513 (12) Schuhladen, S.; Banerjee, K.; Stürmer, M.; Müller, P.; Wallrabe, U.; Zappe, H. Variable
514 optofluidic slit aperture. *Light Sci. Appl.* **2016**, *5*, e16005, Original Article.
- 515 (13) Berge, B.; Peseux, J. Variable focal lens controlled by an external voltage: An applica-
516 tion of electrowetting. *Eur. Phys. J. E* **2000**, *3*, 159–163.
- 517 (14) Supekar, O. D.; Zohrabi, M.; Gopinath, J. T.; Bright, V. M. Enhanced Response Time
518 of Electrowetting Lenses with Shaped Input Voltage Functions. *Langmuir* **2017**, *33*,
519 4863–4869.
- 520 (15) Hayes, R. A.; Feenstra, B. J. Video-speed electronic paper based on electrowetting.
521 *Nature* **2003**, *425*, 383.
- 522 (16) Kim, D. Y.; Steckl, A. J. Electrowetting on Paper for Electronic Paper Display. *ACS*
523 *Appl. Mater. Interfaces.* **2010**, *2*, 3318–3323.
- 524 (17) You, H.; Steckl, A. J. Three-color electrowetting display device for electronic paper.
525 *Appl. Phys. Lett.* **2010**, *97*, 023514.

- 526 (18) Lee, P.; Chiu, C.-W.; Lee, T.-M.; Chang, T.-Y.; Wu, M.-T.; Cheng, W.-Y.; Kuo, S.-W.;
527 Lin, J.-J. First Fabrication of Electrowetting Display by Using Pigment-in-Oil Driving
528 Pixels. *ACS Appl. Mater. Interfaces*. **2013**, *5*, 5914–5920.
- 529 (19) Lomax, D. J.; Kant, P.; Williams, A. T.; Patten, H. V.; Zou, Y.; Juel, A.; Dryfe, R.
530 A. W. Ultra-low voltage electrowetting using graphite surfaces. *Soft Matter* **2016**, *12*,
531 8798–8804.
- 532 (20) Berge, B. Electrocapillarity and wetting of insulator films by water. *C. R. Acad. Sci.*
533 *Paris, Ser. II* **1993**, *317*, 157–163.
- 534 (21) Lin, J.-L.; Lee, G.-B.; Chang, Y.-H.; Lien, K.-Y. Model Description of Contact Angles
535 in Electrowetting on Dielectric Layers. *Langmuir* **2006**, *22*, 484–489.
- 536 (22) Nelson, W. C.; Kim, C.-J. Droplet Actuation by Electrowetting-on-Dielectric (EWOD):
537 A review. *J. Adhes. Sci. Technol.* **2012**, *26*, 1747–1771.
- 538 (23) Quilliet, C.; Berge, B. Investigation of effective interface potentials by electrowetting.
539 *Europhys. Lett.* **2002**, *60*, 99.
- 540 (24) Kuo, J. S.; Spicar-Mihalic, P.; Rodriguez, I.; Chiu, D. T. Electrowetting-Induced
541 Droplet Movement in an Immiscible Medium. *Langmuir* **2003**, *19*, 250–255.
- 542 (25) Staicu, A.; Mugele, F. Electrowetting-Induced Oil Film Entrapment and Instability.
543 *Phys. Rev. Lett.* **2006**, *97*, 167801.
- 544 (26) Mugele, F. Fundamental challenges in electrowetting: from equilibrium shapes to con-
545 tact angle saturation and drop dynamics. *Soft Matter* **2009**, *5*, 3377–3384.
- 546 (27) Paneru, M.; Priest, C.; Sedev, R.; Ralston, J. Static and Dynamic Electrowetting of
547 an Ionic Liquid in a Solid/Liquid/Liquid System. *J. Am. Chem. Soc.* **2010**, *132*, 8301–
548 8308.

- 549 (28) Bhaumik, S.; Chakraborty, M.; Ghosh, S.; Chakraborty, S.; DasGupta, S. Electric
550 Field Enhanced Spreading of Partially Wetting Thin Liquid Films. *Langmuir* **2011**,
551 *27*, 12951–12959.
- 552 (29) McHale, G.; Brown, C.; Sampara, N. Voltage-induced spreading and superspreading of
553 liquids. *Nat. Commun.* **2013**, *4*, 1605.
- 554 (30) Hong, J.; Kim, Y.; Kang, K.; Oh, J.; Kang, I. Effects of Drop Size and Viscosity on
555 Spreading Dynamics in DC Electrowetting. *Langmuir* **2013**, *29*, 9118–9125.
- 556 (31) Klarman, D.; Andelman, D.; Urbakh, M. A Model of Electrowetting, Reversed Elec-
557 trowetting, and Contact Angle Saturation. *Langmuir* **2011**, *27*, 6031–6041.
- 558 (32) Buehrle, J.; Herminghaus, S.; Mugele, F. Interface Profiles near Three-Phase Contact
559 Lines in Electric Fields. *Phys. Rev. Lett.* **2003**, *91*, 086101.
- 560 (33) Drygiannakis, A. I.; Papathanasiou, A. G.; Boudouvis, A. G. On the Connection be-
561 tween Dielectric Breakdown Strength, Trapping of Charge, and Contact Angle Satur-
562 ation in Electrowetting. *Langmuir* **2009**, *25*, 147–152.
- 563 (34) Quilliet, C.; Berge, B. Electrowetting: a recent outbreak. *Curr. Opin. Colloid Interface*
564 *Sci.* **2001**, *6*, 34 – 39.
- 565 (35) Li, Q.; Luo, K.; Kang, Q.; He, Y.; Chen, Q.; Liu, Q. Lattice Boltzmann methods for
566 multiphase flow and phase-change heat transfer. *Prog. Energy Combust. Sci.* **2016**, *52*,
567 62–105.
- 568 (36) Li, H.; Fang, H. Lattice Boltzmann simulation of electrowetting. *Eur. Phys. J. Special*
569 *Top.* **2009**, *171*, 129–133.
- 570 (37) Clime, L.; Brassard, D.; Veres, T. Numerical modeling of electrowetting processes in
571 digital microfluidic devices. *Comput. Fluids* **2010**, *39*, 1510 – 1515.

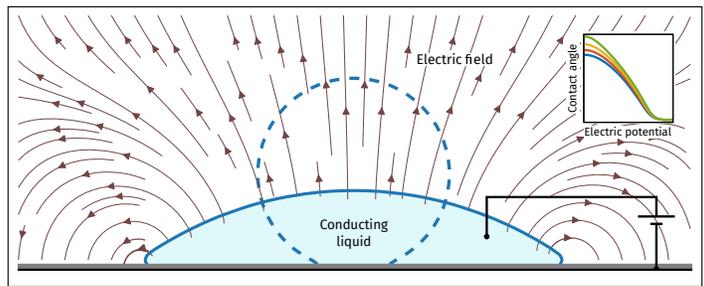
- 572 (38) Aminfar, H.; Mohammadpourfard, M. Lattice Boltzmann method for electrowetting
573 modeling and simulation. *Comput. Methods Appl. Mech. Eng.* **2009**, *198*, 3852 – 3868.
- 574 (39) Lippmann, G. Relations entre les phénomènes électriques et capillaires. Ph.D. thesis,
575 Gauthier-Villars, 1875.
- 576 (40) Landau, L. D.; Lifshitz, E. M. *Statistical Physics, Part 1*; Course of Theoretical Physics;
577 Butterworth-Heinemann: Oxford, 1980; Vol. 5.
- 578 (41) Bray, A. J. Theory of phase-ordering kinetics. *Adv. Phys.* **1994**, *43*, 357–459.
- 579 (42) Cahn, J. W.; Hilliard, J. E. Free energy of a nonuniform system. I. Interfacial free
580 energy. *J. Chem. Phys.* **1958**, *28*, 258–267.
- 581 (43) Cahn, J. W. Critical point wetting. *J. Chem. Phys.* **1977**, *66*, 3667–3672.
- 582 (44) Briant, A. J.; Wagner, A. J.; Yeomans, J. M. Lattice Boltzmann simulations of contact
583 line motion. I. Liquid-gas systems. *Phys. Rev. E* **2004**, *69*, 031602.
- 584 (45) Yang, A. J. M.; Fleming, P. D.; Gibbs, J. H. Molecular theory of surface tension. *J.*
585 *Chem. Phys.* **1976**, *64*, 3732–3747.
- 586 (46) Landau, L. D.; Bell, J. S.; Kearsley, M. J.; Pitaevskii, L. P.; Lifshitz, E. M.; Sykes, J. B.
587 *Electrodynamics of continuous media*; elsevier, 2013; Vol. 8.
- 588 (47) Jackson, J. D. *Classical electrodynamics*; Wiley, 1999.
- 589 (48) Swift, M. R.; Orlandini, E.; Osborn, W. R.; Yeomans, J. M. Lattice Boltzmann simu-
590 lations of liquid-gas and binary fluid systems. *Phys. Rev. E* **1996**, *54*, 5041.
- 591 (49) d’Humières, D.; Ginzburg, I.; Krafczyk, M.; Lallemand, P.; Luo, L.-S. Multiple-
592 relaxation-time lattice Boltzmann models in three dimensions. *Philos. Trans. R. Soc.*
593 *A* **2002**, *360*, 437–451.

- 594 (50) Krüger, T.; Kusumaatmaja, H.; Kuzmin, A.; Shardt, O.; Silva, G.; Viggien, E. M. *The*
595 *Lattice Boltzmann Method: Principles and Practice*; Springer, 2016.
- 596 (51) Desplat, J. C.; Pagonabarraga, I.; Bladon, P. LUDWIG: A parallel Lattice-Boltzmann
597 code for complex fluids. *Comput. Phys. Commun.* **2001**, *134*, 273–290.
- 598 (52) Qian, Y.-H.; Chen, S.-Y. Dissipative and dispersive behaviors of lattice-based models
599 for hydrodynamics. *Phys. Rev. E* **2000**, *61*, 2712–2716.
- 600 (53) Ledesma-Aguilar, R.; Vella, D.; Yeomans, J. M. Lattice-Boltzmann simulations of
601 droplet evaporation. *Soft Matter* **2014**, *10*, 8267–8275.
- 602 (54) Arfken, G. B.; Weber, H. J.; Harris, F. E. *Mathematical methods for Physicists, A*
603 *Comprehensive guide, Seventh Edition*; Elsevier, London, 2013.
- 604 (55) Yu, D.; Mei, R.; Luo, L.-S.; S., W. Viscous flow computations with the method of
605 lattice Boltzmann equation. *Prog. Aerosp. Sci.* **2003**, *39*, 329–367.
- 606 (56) Quinn, A.; Sedev, R.; Ralston, J. Contact Angle Saturation in Electrowetting. *J. Phys.*
607 *Chem. B* **2005**, *109*, 6268–6275.
- 608 (57) Peykov, V.; Quinn, A.; Ralston, J. Electrowetting: a model for contact-angle saturation.
609 *Colloid Polym. Sci.* **2000**, *278*, 789–793.
- 610 (58) Gupta, R.; Olivier, G.; Frechette, J. Invariance of the Solid-Liquid Interfacial Energy in
611 Electrowetting Probed via Capillary Condensation. *Langmuir* **2010**, *26*, 11946–11950.
- 612 (59) Oron, A.; Davis, S. H.; Bankoff, S. G. Long-scale evolution of thin liquid films. *Rev.*
613 *Mod. Phys.* **1997**, *69*, 931–980.
- 614 (60) Edwards, A.; Ledesma-Aguilar, R.; Newton, M.; Brown, C.; McHale, G. Not spreading
615 in reverse: The dewetting of a liquid film into a single drop. *Sci. Adv.* **2016**, *2*, e1600183.

- 616 (61) Fixman, M. The Poisson-Boltzmann equation and its application to polyelectrolytes.
617 *J. Chem. Phys.* **1979**, *70*, 4995–5005.
- 618 (62) Fei, L.; Luo, K.; Li, Q. Three-dimensional cascaded lattice Boltzmann method: Im-
619 proved implementation and consistent forcing scheme. *Phys. Rev. E* **2018**, *97*, 053309.

620 Graphical TOC Entry

621



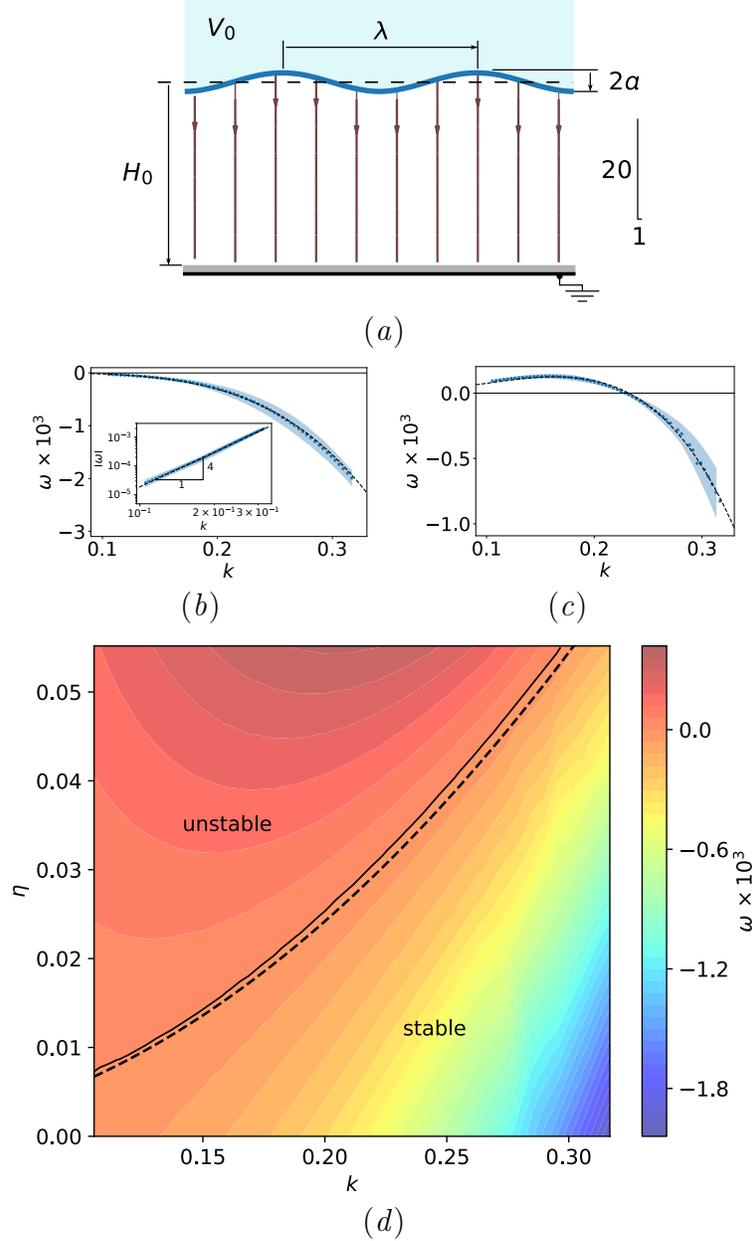


Figure 4: (Colour online) Simulation results of the linear stability of a thin dielectric fluid film in an EWOD setup. (a) Close-up of the interface configuration. The conducting liquid (light blue region) is kept at a constant voltage V_0 , whilst the solid electrode (grey rectangle) remains grounded. The thin dielectric fluid film (white region), of initial average thickness H_0 , is subject to a sinusoidal perturbation of amplitude a and wavelength λ . Direct contact between the dielectric fluid and the solid electrode is prevented by a thin dielectric film (black line). The stream lines depict the electric field. (b) and (c): Dispersion relations for $\eta = 0$ and $\eta = 0.03$, respectively. The solid symbols correspond to the simulation results. The dashed lines correspond to a fit to the analytical model. The shaded envelopes represent the error from the curve fitting analysis. The inset shows the expected $|\omega| \sim k^4$ scaling predicted by the linear theory. (d) Colour map of the growth rate as function of η and k . The solid line corresponds to the separatrix calculated from the simulation results using linear interpolation. The dashed line corresponds to the theoretical prediction.

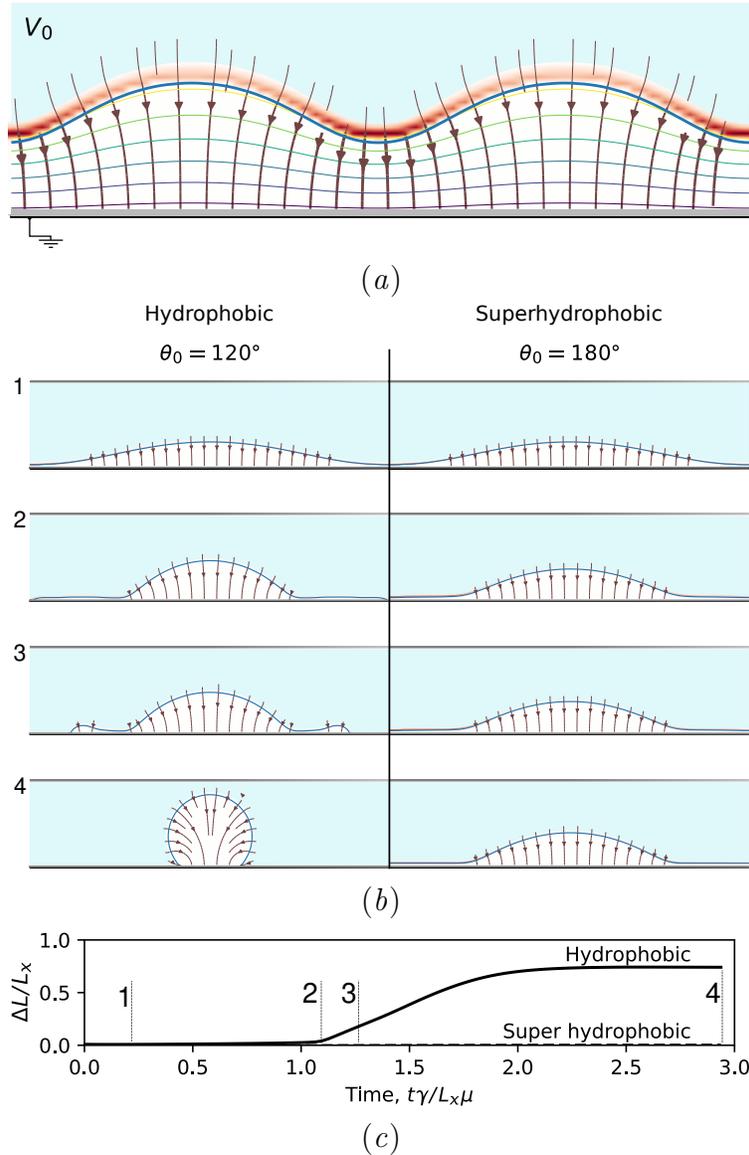


Figure 5: (Colour online) Entrapment and break-up of unstable dielectric films. (a) Instantaneous configuration of an unstable dielectric film at large perturbation amplitudes. The initial simulation parameters are $\lambda = 60$, $H_0 = 20$ and $\eta = 1.1$. The charge distribution, shown in dark red, is highest in the regions closer to the solid electrode, and the equipotential curves, perpendicular to the electric field, increase in density. (b) Long-time evolution of the dielectric film for $\lambda = 512$, $H_0 = 20$, and $\eta = 0.1$; and $\theta_0 = 120^\circ$ (left) and $\theta_0 = 180^\circ$ (right). On a wettable surface, the dielectric fluid breaks into isolated films that dewet to form droplets. On a non-wettable surface the wetting potential prevents the breakup of the film, leading to its entrapment. (c) Plot of the evolution of area fraction, $\Delta L/L_x$, covered by the conducting phase. The numbers represent the time of the snapshots in (a).