Universal Transient Dynamics of Electrowetting Droplets

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Electrowetting on substrates by electrowetting exhibits either of the two transient behaviours: one characterised by contact line oscillation, and the other one by slow spreading dynamics. The transition between these behaviours remains elusive due to the current limited understanding of the spreading dynamics on the hydrodynamical and electrical properties of electrowetting systems. To understand this transition we propose a model capturing the transition’s occurrence based on both the hydrodynamical and electrical parameters. We derive the critical viscosity at which the transition occurs and reveal its subtle and often hidden dependence on the electrowetting dynamics. We find and experimentally verify that the condition for minimization of droplets’ actuation time is only achieved at the transition. Particularly, the transition time as a function of damping ratio exhibits the general feature of Kramers’ reaction-rate theory.

A droplet resting on a flat electrode changes its contact angle if a voltage difference between the droplet and the electrode is applied. In this so-called electrowetting phenomenon, it is advantageous to electrically insulate the droplet and the electrode to prevent current and the resulting electrolysis1. Such setting is termed electrowetting-on-dielectric (EWOD)2, and has become increasingly important in diverse applications requiring active control of droplets such as fast response displays3, high-power energy harvesting4,5, digital microfluidics6, liquid lens7, light valves8, fast optical imaging9, optical films10, and tissue engineering11. In a typical EWOD setup (Fig. 1a), an electrically conductive substrate is coated with a thin insulating layer. A droplet deposited on the insulating layer is in contact with an electrode on top, while the conductive substrate is connected to another electrode. When a voltage $U$ is applied between the two electrodes, surface energy at the liquid-solid interface is changed, causing the droplet to deform and take another equilibrium state. The contact angle $\theta_0$ of the droplet at the new equilibrium state can be related to the initial contact angle $\theta$ using the well-known Young-Lippmann (Y-L) equation $\cos \theta_0 = \cos \theta_0 - \varepsilon \varepsilon_0 U^2/2d \sigma = \eta$, where $\eta$ denotes the so-called electrowetting number, $\sigma$ is permittivity of free space, and $\varepsilon, d, \sigma$ respectively are the dielectric constant, the insulating layer thickness, and the interfacial tension of the droplet’s liquid and the surrounding medium.

While the Y-L equation gives predictions in good accord with the measured changes in contact angles of a droplet under an electric field1, it only relates the equilibrated contact angles before and after the electric field is applied. Thus, it cannot be used to describe the transient dynamics between the two equilibrium states. Understanding of droplet characteristics during actuation, however, plays a critical role in applications utilizing EWOD for droplet manipulation. In particular, the relations between the system parameters, e.g., droplet size, liquid properties, applied voltage, and the resulting transient characteristics, e.g., the actuation time, are of both fundamental and engineering interests. The goal of this paper is to investigate these relations experimentally and analytically.

Results and Discussions

In our experiments, illustrated in Fig. 1a, we use aqueous glycerin solutions consisting of glycerol, DI water, and 0.125 M sodium chloride as working liquids for generating droplets. The electrical conductivity of the solutions of DI water and 0.125 M sodium chloride is $\approx 8.8$ EC. By adjusting the glycerol concentration, we vary the viscosity $\mu$ of the solutions from 1 mPa · s to 258.8 mPa · s. We generate droplets by dispensing liquid from a micro-needle and vary the droplet radius $R$ from 0.05 mm to 1.6 mm. The substrate is made of an indium tin oxide (ITO) glass slide covered by a fluoropolymer layer (Teflon AF-1600, DuPont) having dielectric constant $\varepsilon = 1.93$ and thickness $d = 2.5 \mu m$. The Teflon layer acts as a hydrophobic coating and electrically insulates the ITO layer. We immerse both the droplets and the substrate in silicone oils with viscosity $\mu_0$, varying in the range $1.75 \text{ mPa} \cdot \text{s} \leq \mu_0 \leq 98.9 \text{ mPa} \cdot \text{s}$.

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mPa·s; the oil's temperature is kept at 20 ± 0.5 °C to maintain consistent experimental conditions. We note that varying \( \mu_o \) in the explored range has a minute effect on the interfacial tension \( \sigma \) between the working liquids and the oils. However, \( \sigma \) decreases from 35.7 mN·m⁻¹ to 25.2 mN·m⁻¹ if \( \mu \) increases from 1 mPa·s to 258.8 mPa·s and \( \mu_o \) is fixed (Fig. 1, Supplementary Information).

**Figure 1.** (a) Schematic of an EWOD experimental setup. (b) Dependence of equilibrium contact angle \( \theta_e \) on applied voltage \( U \) showing that contact line saturation occurs at \( U = 115 \) V. (c) Snapshots showing deformation of an underdamped droplet (upper panel) and an overdamped droplet (lower panel) after a voltage of 100 V is applied. The scale bars represent 0.5 mm. The dashed lines mark the substrate level. (d) Spreading radius \( r \) versus time \( t \) for underdamped droplets (open squares), overdamped droplets (open diamonds), and droplets with transitional behaviour (open circles).
In order to apply a potential difference between a droplet and the ITO layer, we immerse one end of a tungsten wire (18 μm in diameter) in the droplet and connect the other end to the positive electrode of a power supply (IT6723G, ITECH) via a solid-state relay (SSR). The vertical distance between the substrate and the tip of the tungsten wire is roughly equal to the droplet’s radius. The ground electrode of the power supply is connected to the ITO layer by using the SSR to close the circuit momentarily, we generate an electrical pulse in the form of a step function between the wire and the ITO layer. We observe that the droplet reacts almost immediately after the circuit is closed, suggesting that its electrical response time is very small compared to its hydrodynamical time. The droplet radius \( r \) is only applicable for the case of electrowetting actuation without contact line saturation. In the overdamped regime, the TCL velocity is

\[
\tau_{\text{ct}} = \frac{\lambda \eta e}{\eta + \sigma},
\]

where \( \eta \) is the spreading radius \( R \), and \( \eta \) is the frictional coefficient and \( \eta_{\sigma} \) is the TCL velocity. The dimension of \( \lambda \) is dynamic viscosity, and those of both \( F_{\lambda} \) and \( F_{\eta} \) are forces per unit length. If we denote \( \tau_{\gamma} \), the characteristic timescale for the spreading motion in the overdamped regime, the TCL velocity is \( \eta_{\lambda} \approx \tau_{\gamma} \tau_{\eta} \), where \( \tau_{\gamma} = \beta R \) is the spreading radius at equilibrium. Here, \( \beta \) can be derived based on the assumptions that the shape of a droplet remains spherical at its equilibrium states and its volume is conserved; \( \beta = \frac{r_{\lambda}}{R} = \frac{1 - \cos^2 \theta}{1} \left( \frac{4(1 - \cos \theta)}{(2 + \cos \theta)^2} \right), \)

By balancing the friction with the driving force, we obtain \( \tau_{\gamma} = \lambda \eta e/\eta \). The friction coefficient \( \lambda \) is a function of both viscosities \( \mu \) and \( \mu_{\sigma} \), and can be measured experimentally (see Methods and Fig. 2, Supplementary Information). The electrode potential difference is varied from 0 V to 500 V to investigate the effects of the driving force and is controlled using a high-voltage pulse generator with a pulse width of 20 μs. In order to observe the transient dynamics, we perform a high-speed cinematography with a high-speed camera (SAX2, Photron) and periodically record the deformation of droplets (Fig. 1c). For a set of control parameters \( R, \mu, \eta \), we repeat the experiment 5 to 7 times and measure the spreading radius \( r \) as a function of time \( t \) (Fig. 1d).

We observe three characteristic transient behaviours of the spreading radius \( r \). For each set of \( (R, \mu, \eta) \), we categorise the corresponding behaviour as overdamped if \( r(t) \) varies monotonically (Fig. 1d, \( \mu = 258.8 \text{ mPa s} \)), and underdamped if overshootings are consistently observed in \( r(t) \) (Fig. 1d, \( \mu = 1.0 \text{ mPa s} \)). We note that the underdamped behaviour is always accompanied by capillary waves at the liquid-oil interface (see Fig. 1c). In the case that repetitive runs of the same parameters result in alternative characteristics, we categorise the behaviour as transitional (Fig. 1d, \( \mu = 17.6 \text{ mPa s} \)). In Fig. 2, we show the phase diagram of these behaviours for wide ranges of control parameters: 0.05 mm \( \leq R \leq 1.6 \text{ mm} \), 1.0 mPa s \( \leq \mu \leq 258.8 \text{ mPa s} \), and 0.35 \( \leq \eta \leq 1.18 \). The overdamped and underdamped regimes are clearly separated by the transitional regime. Among the control parameters, \( \eta \) has a negligible effect on the transition between these regimes. We attribute the extent of the transitional regime to irregularities in physical and chemical properties of the substrate; such effects could be minimised with better surface treatments. Thus, the overdamped-to-underdamped transition is possible with an uncertainty indicated by the extent of the transitional regime (Fig. 2, inset).

We now examine the transient dynamics in each of the overdamped and underdamped regimes. In both regime, the concentrated charge density along the liquid-oil interface at the vicinity of the three-phase contact line (TCL) causes a net force \( F_{\lambda} = \eta \sigma \) that pulls the liquid horizontally. We note that this driving force is only applicable for the case of electrowetting actuation without contact line saturation. In the overdamped regime, the dominant factor opposing to the driving force is the contact line friction, which is estimated as \( F_{\lambda} = \lambda \eta_{\lambda} \), where \( \lambda \) is the frictional coefficient and \( \eta_{\lambda} \) is the TCL velocity. The dimension of \( \lambda \) is dynamic viscosity, and those of both \( F_{\lambda} \) and \( F_{\eta} \) are forces per unit length. If we denote \( \tau_{\gamma} \), the characteristic timescale for the spreading motion in the overdamped regime, the TCL velocity is \( \eta_{\lambda} \approx \tau_{\gamma} \tau_{\eta} \), where \( \tau_{\gamma} = \beta R \) is the spreading radius at equilibrium. Here, \( \beta \) can be derived based on the assumptions that the shape of a droplet remains spherical at its equilibrium states and its volume is conserved; \( \beta = \frac{r_{\lambda}}{R} = \frac{1 - \cos^2 \theta}{1} \left( \frac{4(1 - \cos \theta)}{(2 + \cos \theta)^2} \right), \)

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In Fig. 3a, we show a log-log plot of \( \lambda \) versus \( \mu \) of working liquids. Datasets collected in air and on different insulating layers are shown for comparison. The solid line represents the scaling law \( \lambda \sim \mu^{1/2} \). (b) Log-log plot of \( \lambda \) versus viscosity \( \mu_o \) of the surrounding liquids (silicone oils). We only show data in the overdamped regime, i.e., for droplet actuation without capillary waves. The solid line represents the scaling law \( \lambda \sim (\mu \mu_o)^{1/2} \). Inset: Log-log plot of \( \lambda \) versus \((\mu \mu_o)^{1/2}\) for our experimental data. The solid line represents the scaling law \( \lambda = C(\mu \mu_o)^{1/2} \), where \( C = 32.9 \pm 3.2 \) is the fitting parameter.

Information). In Fig. 3a, we show a log-log plot of the measured values of \( \lambda \) versus \( \mu \). The presented data are consistent with the scaling law \( \lambda \sim \mu^{1/2} \) and other datasets obtained for spreading of glycerol droplets in air and on surfaces coated by Teflon, Silicon dioxide (SiO2) and Silane. The vertical shifts between datasets reflect variations in substrate properties and surrounding media. Similarly, the data shown in Fig. 3b indicate the scaling law \( \lambda \sim (\mu \mu_o)^{1/2} \), consistent with the data collected for aqueous sodium chloride droplets in silicone oils. Thus, this result suggests equal contributions of \( \mu \) and \( \mu_o \) to variations in \( \lambda \). In other words, the dependence of \( \lambda \) on the bulk viscosities can be described as \( \lambda = C(\mu \mu_o)^{1/2} \) for the tested ranges of \( \mu \) and \( \mu_o \), where \( C \) is a constant that depends on the roughness and chemical properties of the surface. Indeed, all of our data collapse to a single curve when plotting \( \lambda \) versus \((\mu \mu_o)^{1/2}\), as shown in the inset of Fig. 3b. A best fit to our data gives \( C = 32.9 \pm 3.2 \), a constant specific to the properties of our substrate. Here we emphasise that the scaling law \( \lambda \sim (\mu \mu_o)^{1/2} \) is applicable for electrowetting actuation of droplets immersed in silicone oils. An extrapolation of this scaling law to the case in which the outer medium is air \((\mu_o = 1.81 \times 10^{-2} \text{ mPa s})\) may require a substantial extension of experimental data towards the lower limit of \( \mu_o \) and merit a separate study. We therefore arrive at the expression for the characteristic timescale in the overdamped regime \( \tau_o = C(\mu \mu_o)^{1/2}/(r \sigma) \). Thus, the data collapse using \( \tau_o \) indicates that \( \tau_o \) characterises the transient dynamics in the overdamped regime.

In the underdamped regime, we assume that viscosity is negligible and the driving force is only resisted by the droplet’s inertia. Thus, by balancing the driving force and inertia, one finds that droplets in this regime oscillate with characteristic frequency \( \omega = (\eta \sigma / \rho R^2)^{1/2} \). As a result, the characteristic timescale for the droplets to reach maximum deformation is \( \tau_u = \pi (\rho R^2 / \eta \sigma)^{1/2} \). We use \( \tau_u \) to normalise the data of spreading radius \( r(t) \) in the underdamped regime and observe data collapse for all control parameters \((R, \eta, \mu, \mu_o)\) (Fig. 5, Supplementary Information). This strongly suggests that \( \tau_u \) is the characteristic time of the underdamped regime.

Let us discuss parameter relations at the overdamped-to-underdamped (O-U) transition. We argue that the characteristic timescales of the two regimes are comparable at the O-U transition. In other words, the condition for the O-U transition to occur is \( \xi = D \tau_o / \tau_u = 1 \), where \( D \) is a prefactor of unity order and is independent of the
control parameters. Here, the so-called damping ratio $\xi$ can be fully expanded as

$$\xi = (DC/\pi)(\beta \eta^{-1/2})(\mu \sigma R)^{1/2}(\rho \sigma R)^{-1/2},$$

revealing similar physical significance as the Ohnesorge number with an additional electrical term $\beta \eta^{-1/2}$.

The damping ratio is used to indicate whether the droplet behaviour is in the underdamped regime ($\xi < 1$), or in the overdamped regime ($\xi > 1$). The condition for the O-U transition to occur, $\xi = 1$, translates to a linear relation between the critical viscosity $\mu_c$, defined as the viscosity at the transition, and the droplet radius $R$:

$$\mu_c = \left(\frac{\pi}{CD}\right)^2 \frac{\eta \rho \sigma}{\beta^2} R.$$  \hspace{1cm} (1)

This relation is consistent with the O-U transition shown in the inset of Fig. 2; fitting $\mu_c$ to the data at the transition in the phase diagram gives $D = 1.49 \pm 0.21$ for all tested values of the electrowetting numbers $\eta$ (Fig. 2, inset). Moreover, we note that the composite term $\eta \beta^{-2}$ carries the dependence of $\mu_c$ on both the applied voltage and the inherent electrical properties of the system, e.g., the thickness and the dielectric constant of the dielectric layer. For $\eta$ varying from 0.35 to 1.18 in our experiments, the value of $\eta \beta^{-2}$ changes in a narrow range, from 0.55 to 0.62. Thus, we conclude that $\mu_c$ varies linearly with $R$ and depends weakly on $\eta$ for the explored ranges of parameters.

To obtain a quantitative description of the actuating motion of droplets under EWOD conditions, we measure the actuation time $t_a$, defined in practice as the duration for the spreading radius to reach 95% of the radius at equilibrium state after a voltage is applied. As illustrated in Fig. 1d, $t_a$ depends strongly on the viscosity. More generally, it suggests that $t_a$ is closely linked to the transient dynamics, i.e., overdamped or underdamped. In Fig. 4a, we show a plot of $t_a$ versus $\mu$ for a fixed applied voltage ($\eta = 0.98$) and various droplet sizes ($0.05 \text{ mm} \leq R \leq 1.6 \text{ mm}$). Indeed, different transient dynamics result in distinctive behaviours for the actuation time: $t_a$ decreases with $\mu$ in the underdamped regime, but increases in the overdamped regime. We explore this link, i.e., between $t_a$ and $\mu$, $R$, $\eta$ for different transient dynamics, by making an analogy between an actuating droplet and a mass-spring system. We take $\omega$ as the natural frequency and the ratio $\xi = D \tau_0 / \tau_a$ as the damping ratio of the analogous mass-spring system. We note that for a mass-spring system of natural frequency $\omega$ and damping ratio $\xi$, the actuation time, i.e.,

Figure 4. (a) Log-log plot of the actuation time $t_a$ versus $\mu$ for different droplet size and at $\eta = 0.98$. The colored areas are indicative of different transient dynamics. (b) Log-log plot of $\omega t_a / 4$ versus $\xi$. The minimum point $\omega t_a / 4 = 1$ occurs at $\xi = 1$. 
the duration for the system to reach an equilibrium state, is specified as $\tau = 4(\xi \omega) = (4\pi/DC)\mu R^3 \beta^3 \gamma (\mu/\mu_a)^{1/2}$. This suggests that $\tau$ is the characteristic timescale for the actuating motion of droplets in both transient dynamics. As a result, we nondimensionalise $t$ and $\mu$ as $t/\tau = \xi \omega/4$ and $\mu/\mu_a = \xi^2$, respectively. The link between $t$ and $\mu$, in a power-law form, is therefore equivalent to that of $\omega t/4$ and $\xi$. In Fig. 4a, we show a log-log plot of $\omega t/4$ versus $\xi$ for all the data shown in Fig. 4a. In both the underdamped and overdamped regimes, excellent data collapses confirm that $\tau$ indeed represents the actuation time of droplets regardless of transient dynamics. In addition, the minimum value of the cloud of data occurs at $\xi = (\mu/\mu_a)^{1/2} = 1$ and $\omega t/4 = 1$. We conclude that for fixed values of $R$ and $t$, minimisation of $t$ is always achieved at $\mu = \mu_a$. Remarkably, in the underdamped regime the transition time reduces as the damping ratio increases. This also indicates that the internal energy transfer from oscillation to spreading is facilitated by increasing the viscosity. In the overdamped regime, the energy dissipation plays an important role, thus leading to the slow-down of spreading with increasing damping ratio. This demonstrates that Kramers’ reaction-rate theory may be helpful to understand electrowetting dynamics.

Conclusions

The transient dynamics of actuating droplets exhibit either underdamped or overdamped behaviour; the overdamped-to-underdamped (O-U) transition is insensitive to the applied voltage $U$, but is strongly dependent on the droplet size $R$ and liquid viscosity $\mu$. This results in a linear relation between the droplet size $R$ and the critical viscosity $\mu_a$ at the O-U transition. The droplet actuation time $t_a$ does not only depend on the system’s control parameters, but also on the transient dynamics: it is minimised at the O-U transition. Interestingly, the weak dependence of the O-U transition on the driving force implies that our analysis may be applicable to the co-planar electrowetting setting in which the resistive forces remain, but the driving force is modified due to partial coverage of co-planar electrodes on the substrate. Based on similar force analysis, we anticipate that the O-U transition in the top-plate electrowetting setting may be determined by taking into account an additional resistive force: the capillary pressure gradient of the curved liquid surface between the two plates of the top-plate setting. Finally, the transition time shows the cross-over feature from under-to-over-damping as nicely described in Kramers’ reaction-rate theory.

Methods

Friction coefficient measurement. Suppose that the three-phase contact line (TCL) has velocity $u_{cl}$ at time $t$ after a voltage is applied. The driving force acting on the TCL is $(\cos \theta_t - \cos \theta_d)\sigma$, and the frictional force is $-\lambda u_{cl}$. Because inertia is neglected in the overdamped regime, a force balance at the TCL gives $(\cos \theta_t - \cos \theta_d)\sigma = \lambda u_{cl}$, where $\theta_t$ and $\theta_d$, respectively are the contact angles at the equilibrium state and at time $t$. From the high-speed recordings of the TCL motion, we calculate $\lambda$ based on the measured values of $\theta_t$, $\theta_d$ and $u_{cl}$ (Fig. 2, Supplementary Information).

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Author Contributions
T.T. and Q.V. conceived the experiment, Q.V. conducted the experiment, T.T., Q.V. and H.S. analysed the results. All authors reviewed the manuscript.

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