



## Experimental analysis of the evolution of an electrified drop following high voltage switching

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### ABSTRACT

We report experimental results on the electrohydrodynamic behavior of drops following high voltage switching. A pendant drop was placed between two parallel electrodes. Then, a DC high voltage power supply was switched on. The voltage difference between the two electrodes reached the prescribed value through an exponential relaxation process with a characteristic time of the order of some tenths of a second. The drop's free surface deformation was precisely measured as a function of time. The drop's evolution can be regarded as a quasi-equilibrium process for subcritical voltages. The instantaneous free surface deformation perfectly matched the equilibrium solution for a voltage which increased over time. The rate at which the electric charge migrated to the free surface was measured precisely by fitting the equilibrium solution to the experimental free surface deformation. For supercritical voltages, the drop's evolution consisted of two stages: a quasi-equilibrium process until the voltage difference reached its critical value, and a dynamical regime driven by the surface tension force and leading to the drop's breakage.

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### 1. Introduction

Understanding the effects associated with the action of electric fields on capillary systems is of great importance in a number of applications including inkjet printing, spring coating, electrowetting, mass spectrometry, fabrication of microspheres of biological materials, direct handling of living cells, etc. One such application is electro spraying [1], which constitutes a very useful atomization technique to produce a variety of capillary shapes. Many electrohydrodynamic phenomena arising in electro spraying have been studied both theoretically and experimentally over the last decades. The so-called steady cone-jet mode of electro spraying has attracted much attention due to its capability of steadily producing drops with diameters ranging from hundreds of microns down to a few nanometers [2–4]. Drops can also be produced in transient electro spraying modes from the breakage of liquid ligaments periodically formed and emitted [2,5]. In this case, the liquid ligament forms and collapses on time scales of the order of or less than milliseconds, depending on the meniscus size and the applied electric voltage. Transient regimes in electro spraying have been studied experimentally in the past decade [5,6] because

of their importance for drop-on-demand atomization techniques. However, technical difficulties associated with the experiments hinder one from getting information with high enough spatial and temporal resolutions to describe the formation and collapse of the liquid ligaments. That information would be useful not only for gaining insight into the physical mechanisms governing the transient regimes, but also as a reference against which to validate novel simulation techniques.

Electrified drops are the precursors of the elongated liquid menisci from whose tips jets are emitted in electro spraying. This fact is one of the practical reasons for studying the dynamic behavior of electrified drops. The action of electric fields on drops gives rise to a variety of interesting phenomena, such as drop deformation and rotation, breakup and coalescence, and liquid ejection, among others. The static deformation of floating, pendant, and sessile drops exposed to electric forces has been studied over the last decades [7–12]. There is also a considerable body of literature dealing with the dynamic response of drops to oscillatory electric fields (see, e.g., Refs. [13,14] and references therein). The dynamic deformation of a pendant drop due to a step change in the electric field magnitude is closely related to drop-on-demand electro spraying atomization techniques, transient regimes [5,6], and, to a certain extent, the establishment of the steady cone-jet mode [15,6]. For relatively low values of the applied voltage, the drop deforms following an extensional/oscillatory motion to reach a more elongated shape [7,16,11]. If the electric field magnitude exceeds a certain critical value, the pendant drop ejects a liquid

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ligament from its tip [17]. This ejection can be sustained over time if liquid is injected at an appropriate flow rate, which yields the steady cone-jet mode [2–4]. The dynamic deformation of both subcritical and supercritical highly conducting drops due to a step change in the electric field magnitude has been examined numerically by Notz and Basaran [16].

Low-conductivity liquids are commonly used in electrohydrodynamic applications. In these applications, the appearance of shear stresses at the liquid free surface feeds complex flow patterns including recirculation cells [18,19]. The analysis of the dynamic response of low-conductivity liquids to the action of electric forces is important at both fundamental and practical levels. The response of a low-conductivity drop to a step change in the electric field magnitude has been studied both numerically and experimentally by [20]. They examined in detail the drop-jet transitional region and the emitted jet for supercritical electric fields. The numerical results for the subcritical regime showed an exponential decay to the steady drop shape.

A pendant drop located between two electrodes undergoes the following three processes when the voltage supply is switched on. Firstly, a voltage drop is established between the two parallel electrodes. Secondly, electric charges are transferred to the free surface and accumulate there. Lastly, the liquid flows until a new equilibrium shape is reached. The electric time characterizing the second process is expected to be much shorter than any characteristic hydrodynamic time, even for very small liquid electric conductivities. Thus, the migration of free electric charges to the free surface can be regarded as instantaneous. The electric charges accumulated over the free surface screen the external electric field. The electric field inside the droplet vanishes, and hence the liquid polarization forces can be neglected for any time. The liquid flow is originated by the appearance of electric stresses at the free surface. The sum of the electric and hydrostatic pressures is balanced by the capillary stress (viscous stresses normal to the free surface can be generally neglected). The electric charge density and the electric stress increase at the apex. Therefore, the hydrostatic pressure decreases in that region, and thus the liquid is sucked towards the drop tip. This liquid motion stretches the drop, increasing the free surface curvature at the apex. Consequently, a restoring capillary force appears in that region. If the electric stresses are not high enough to overcome the surface tension force, then the drop reaches an equilibrium state. Otherwise, a liquid ligament is ejected from the droplet tip.

If the prescribed voltage difference between the electrodes (i.e., the value selected in the voltage power supply) were established much faster than the hydrodynamic processes, the drop's evolution towards the equilibrium state would be governed by the liquid dynamic properties. In this case, the time scale on which the drop reaches its new equilibrium shape can be determined by establishing a balance between the viscous stress  $\mu \partial^2 w / \partial z^2 \sim \mu v / R^2$  ( $\mu$  is the liquid viscosity, and  $v$  and  $R$  are the characteristic axial velocity and length, respectively) and the axial inertia term  $\rho dw / dt \sim \rho v / t_c$  ( $\rho$  is the liquid density and  $t_c$  is the characteristic evolution time), which leads to  $t_c \sim t_\mu \equiv \rho R^2 / \mu$ . In principle, the new equilibrium shape could be reached through either an overdamped extensional deformation or the damping of free surface oscillations, depending on the relative magnitude of the viscous  $t_\mu$  and capillary  $t_\gamma \equiv (\rho R^3 / \gamma)^{1/2}$  ( $\gamma$  is the surface tension) times. This latter characteristic time determines the oscillation frequency. This scenario can be completely different if no special precaution is taken to ensure a sufficiently fast voltage increase. If the time scale characterizing the relaxation of the voltage to the prescribed value were much larger than the capillary time  $t_\gamma$ , then the drop's evolution is expected to correspond to a succession of equilibrium states given by the instantaneous value of the voltage difference between the electrodes.

It must be noted that several studies have considered both the equilibrium shape and stability of drops under the action of electric fields [7–11]. The perfect conducting [7,10] and dielectric [8,9,11] limits were analyzed both theoretically and experimentally. However, the dynamical process leading to the drop's static deformation was not considered in any of those studies. Reznik et al. [20] did examine the response of a low-conductivity drop to a variation of the electric field. The subcritical regime was analyzed only numerically, and assuming both a step change in the field magnitude and negligible inertia. The evolution of drops subject to an instantaneous step change in the field strength was also studied numerically by Notz and Basaran [16]. In this case, the opposite limit (perfectly conducting and inviscid liquids) was considered.

In this paper, we analyze experimentally the electrohydrodynamic behavior of low-conductivity drops following high voltage switching. The voltage difference between the two electrodes reached the prescribed value through an exponential relaxation process with a characteristic time of the order of some tenths of a second. For subcritical voltages, the electrohydrodynamic mechanisms governing the drop's deformation were much faster than that relaxation process, and hence the drop adopted the equilibrium shapes corresponding to the instantaneous values of the voltage difference. The drop's evolution was essentially independent of its volume, the liquid dynamic properties, and the prescribed voltage. We measured precisely the rate at which the electric charge moved to the free surface by fitting the equilibrium solution to the experimental free surface deformation. We shall also show that these results can be extended to the first part of the evolution of supercritical drops, where the surface tension restores the equilibrium "instantaneously" by balancing the electrical stresses. The subsequent breakage process is, however, driven by the surface tension force, and the equilibrium description is no longer valid.

The paper is organized as follows. The experimental method is described in Section 2. We developed a numerical procedure to calculate the equilibrium solution as a function of the applied voltage. That numerical procedure is presented in Section 3. In Section 4, we present and discuss our results. Some concluding remarks are presented in Section 5.

## 2. Experimental method

### 2.1. Experimental setup

The experimental configuration studied in this work (Fig. 1) consisted of two parallel circular electrodes of radius  $R_0 = 5$  cm made of stainless steel. A copper disk of radius  $R = 2$  mm and width  $h = 0.7$  mm was attached to the upper electrode. Both the upper electrode and the disk had a circular orifice of radius  $r = 0.1$  mm at their centers. The disk was separated from the lower electrode by a distance  $H = 10$  mm. A drop of volume  $\mathcal{V}$ , density  $\rho$ , viscosity  $\mu$ , surface tension  $\gamma$ , electrical conductivity  $K$ , and relative permittivity  $\epsilon$  hung from the disk. The triple contact line was anchored to the disk edge in the course of the experiment. Table 1 shows the nominal values of  $\rho$ ,  $\mu$ ,  $\gamma$ , and  $\epsilon$  for the liquids used in the experiments. The table also shows the liquid's electrical conductivity  $K$ , which was measured by applying a voltage difference between the ends of a cylindrical borosilicate capillary full of the working liquid, and then measuring the resulting electric current.

Fig. 2 shows the apparatus used in the experiments. The experimental cell (A) that contained the two electrodes and the supporting disk was situated horizontally on a goniometer (B). An electric potential  $V$  was applied to the lower electrode with a standard DC high voltage power supply (BERTAN 205B-10R) (C), while the upper electrode was connected to ground. The electrical potential  $V$  reached its prescribed value following an exponential relaxation process with a characteristic time of the order of some

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