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Effects of bulk charge and momentum relaxation time scales on ac electrospraying

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The behavior at the liquid meniscus under the application of a high voltage ($>4\times10^3 V_{p\text{-}p}$), high frequency ($>10$ kHz) alternating current (ac) signal is shown to depend very sensitively on the liquid conductivity and the applied frequency. For pure ethanol, after resonance effects become unimportant, the meniscus oscillates and then changes to a growing conical mode with increasing frequency. For ionic-liquid doped samples with higher conductivities, however, this transition to an oscillating mode and a conical mode is delayed and additional tip-streaming and elongated-fast dripping modes are seen. A linear proportionality between the transition frequency and the sample conductivity indicates that the critical period is approximately 20 times the bulk charge relaxation time scale; the mode transition takes place only when a specific and universal fraction of charge in the liquid bulk is unrelaxed. This observation for ac cones is in contrast with direct current cones with complete charge relaxation in the bulk. It also suggests that bulk Coulombic forces due to a cumulative net charge in the liquid bulk are responsible for growth of the much more slender ac cones. The vibration frequency of the oscillating mode and the ejection frequency of the elongated-fast dripping mode are found to be related to the momentum diffusion time scale across the drop and not the applied frequency. © *2007 American Institute of Physics*.
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I. INTRODUCTION

Electrospraying with a dc electric field has been the focus of many theoretical and experimental studies and the basic mechanism for it has been elucidated.[1,2] Upon the application of the dc voltage to a capillary-plate system, the bulk liquid within the drop acquires a net charge but rapidly transfers the charges to the interface after a characteristic bulk charge relaxation time. The interfacial charge is especially high at the capillary tip, where the accumulated coions push against the free surface and deform it. When a static balance is reached between the capillary and the Coulombic forces due to the interfacial charge, the liquid meniscus stabilizes into a conical shape.[3] A thin, steady charged jet containing the coions is ejected from the conetip. The jet breaks up into small drops, which undergo further Rayleigh fission to form even smaller drops. This spraying regime with the steady Taylor cone is commonly known as the cone-jet mode. It is the most common dc spray regime and has been extensively studied.[4] Other spraying modes, such as dripping, microdripping, spindle, simple, oscillating, and ramified jets,[5,6] are observed with variations in the applied voltage, liquid properties, ambient permittivity, and spraying geometry to produce different droplet dimension, droplet charge, and spraying patterns.

In contrast, electrospraying under an ac electric field has not been explored fully. Previous studies have been generally limited to either dc superimposed ac fields or low ac frequencies.[7] Dripping at higher frequencies (>30 kHz) and resonance at comparatively low frequencies (<10 kHz) have been reported.[8,9] In a recent work, we have reported the observation of steadily growing conical menisci, with a cone angle much smaller than the Taylor angle, under the application of a high frequency ac field.[10] Such unique ac spray phenomena are counterintuitive and unexplained by extension of the current dc spraying theories. In this article we further explore this frequency regime with variation in liquid properties.

The dominant time scale in this scenario seems to be the semiperiod of the applied field, which ranges from 33 to 2.6 μs for the applied frequency between 15 and 190 kHz. On the other hand, charge relaxation time ($\beta\varepsilon_0/K$), which is the dominant time scale for dc spraying,[11] for ethanol is 3.1 μs, for a conductivity $K\sim0.7\times10^{-6}$ S/cm and dielectric constant $\beta\sim24.5$, and ε_0 is the permittivity of free space. Clearly the half-period is comparable and can even be smaller than the charge relaxation time scale suggesting that, unlike dc cones with complete charge relaxation, ac cones are formed when there is significant charge buildup within the bulk liquid. Hence, it is necessary to understand the interplay and relative importance of the different time scales and to relate some unique features of ac sprays to such interactions. To achieve this end, experiments are carried out by varying the liquid conductivity as well as the frequency and magnitude of the applied field and observing the liquid meniscus in each scenario. In addition to varying the relative magnitude of the time scales to decipher their importance, this also allows us to obtain an all inclusive phase diagram of the spraying behavior depending on the liquid properties and applied field. In this paper, we will attempt to delineate and

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classify all these observed spraying modes. Incidentally, the spraying of pure ethanol reported previously will appear as a subset of this spraying behavior.[10] The information obtained from the occurrence of the different modes and the relevant time scales for them will then help explain the probable charging mechanism for the ac spraying.

II. EXPERIMENTS

The details of the experimental apparatus have been reported previously.[9] Sinusoidal ac signal generated by a wave form generator is amplified by a Powertron rf amplifier and a secondary transformer and applied to a metallic needle containing the liquid to be sprayed. High frequency ac potential up to $7\times10^3 V_{p\text{-}p}$, ranging from 10 to 200 kHz can be generated in this fashion. A piece of copper tape placed at a distance of 5 mm from the metallic needle acts as the ground to complete the circuit. The visualization of the spraying modes is performed via an inverted microscope connected to a high speed camera recording at 5000 frames/s. We use denatured high performance liquid chromatography (HPLC) grade alcohol which contains 90.6% ethanol, with conductivity $K\sim0.7\times10^{-6}$. Varying the conductivity of ethanol proved to be somewhat tricky. Alkali metal salts such as sodium chloride could not be used because of the use of an organic solvent. Hydrochloric acid was able to vary the conductivity; however, the conductivity of the sample was not stable and varied with time, probably due to some kind of denaturing. The problem was finally solved with the use of ionic liquids. Ionic liquids are salts which, in their pure state, are liquids at or near ambient conditions.[12] Due to the tremendous permutations possible, the property of the ionic liquid can be varied by changing the form of the cation and anion present and so it can be tailored to the desired requirements. In addition, the presence of a significant organic component makes it possible to dissolve it in organic solvents. The ionic liquid that we used was butylmethylpyrrolidinium bis(trifluoromethylsulfonyl)imide (Solvent Innovation, 98% purity).

Besides being soluble in ethanol, it is also very stable, which allowed the conductivity of the mixture to remain constant with time, in the absence of any contamination. It also has an extremely high conductivity of 0.274 S/m, which is more than five orders of magnitude larger than that of ethanol. A solution of 20 μl of the ionic liquid with 10 ml of ethanol produced a conductivity of $\sim$220 μS/cm, which was still very high for the experimental requirements. Hence, further dilution was necessary. By varying the concentration of the ionic liquid, a group of solutions with different conductivities was prepared varying between 1 and 10 μS/cm. These conductivity values are quite low and can change within this range easily if any contamination takes place; consequently, experiments were performed directly after making the samples and fresh sets were made for every experimental run. We use a Cole-Parmer conductivity meter 1481-90 for conductivity measurement attached to a cylindrical probe with inner diameter (ID) of 1/8 in. and 6 in. long. The outer diameter (OD) was 7/16 in. It was kept in a constant temperature water bath as we found that the conductivity value was sensitive to temperature.

III. RESULTS AND DISCUSSION

A. Effect of liquid conductivity

For pure ethanol at frequencies above 10 kHz, dripping accelerated by electric field is observed for voltages lower than $4\times10^3 V_{p\text{-}p}$, which is similar to the dripping faucet. At higher voltages ($>6\times10^3 V_{p\text{-}p}$) corona discharge is visible at the needle tip and discharge assisted ejection events take place at the meniscus. Bounded by these two spraying regimes, a steady conical meniscus is observed, which does not alternate with the applied frequency but shows temporal growth. At frequencies between 15 and 30 kHz the meniscus is not perfectly steady and localized waves originate at the meniscus tip, with frequencies of $\sim$500 Hz, which is much lower than the applied frequency. However, the meniscus does not get separated from the needle tip, and ejections, if any, occur from the meniscus tip. We shall henceforth call it the oscillating meniscus mode. With increasing applied frequency the conical meniscus becomes perfectly steady, and so we move from the oscillating mode to the conical mode. This was described in our previous work.[10]

Here, we present the results when the conductivity of the sample is varied. With increasing conductivity the lower frequencies bound for the oscillating meniscus mode and the conical mode both increased and new spraying modes became visible at lower frequencies. Before the onset of the oscillating mode, a tip-streaming mode and a elongated-fast dripping mode appeared. With pure ethanol these modes are difficult to observe since the oscillating mode appears as early as 15 kHz, and below $\sim$10 kHz, resonance effects are dominant because the meniscus dimensions are commensurate with the natural vibrating frequencies of free drops having the same dimensions.[13] Consequently, the frequency window available for the tip-streaming and elongated-fast dripping modes is very small. However, as the conductivity is increased the frequency at which the oscillating and conical meniscus modes appear is also increased and, consequently, there is a larger window which makes the observation of tip-streaming and elongated-fast dripping modes possible.

Figure 1 depicts schematic voltage-frequency phase diagrams for three samples, pure ethanol and ethanol doped with different amounts of ionic liquid resulting in a sample with intermediate conductivity and a sample with higher conductivity. As discussed, additional spraying modes are introduced in the intermediate and high conductivity samples, as compared to the pure ethanol sample. Also, with increasing conductivity, the onset of oscillating and conical meniscus modes is delayed to even higher frequencies, as compared to a sample with slightly lower conductivity, as can be seen by comparing Figs. 1(b) and 1(c).

The actual variation in the onset frequency of the oscillating mode with changing liquid conductivity is shown in Fig. 2. We can see that with increasing conductivity the oscillating mode occurs at higher applied frequencies. The ex-

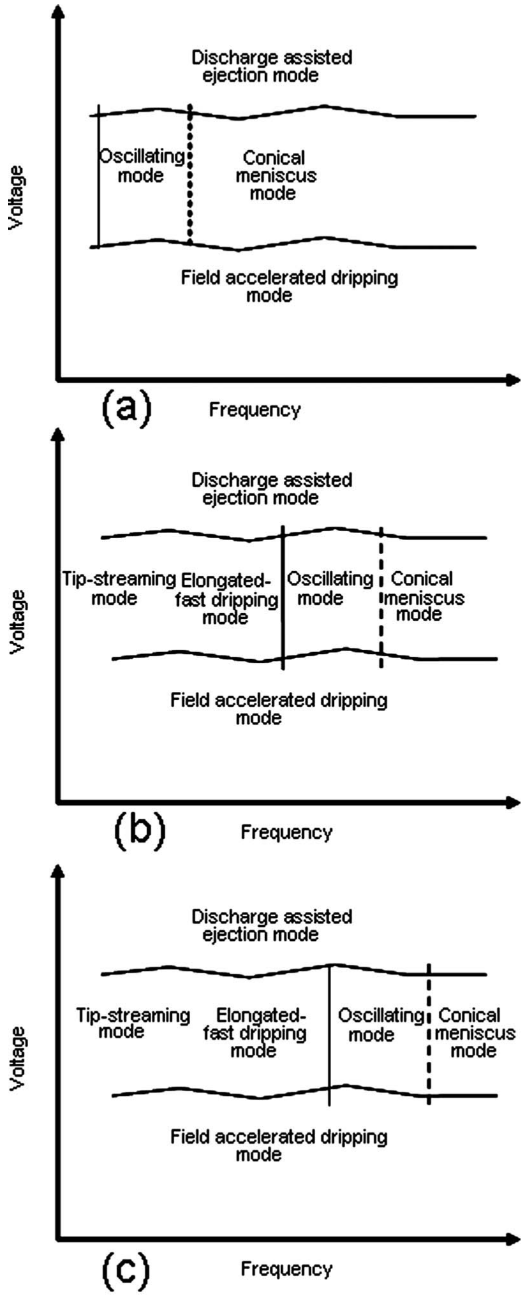

FIG. 1. Schematic phase diagrams for (a) pure ethanol, low conductivity; (b) doped ethanol, intermediate conductivity; and (c) doped ethanol, high conductivity. In the first case, there is only the oscillating mode and the conical meniscus mode. Additional modes are introduced with increasing conductivity. The onset of oscillating and conical meniscus modes are delayed to higher frequencies with increasing conductivities, as can be seen from (b) and (c).

perimental results also point toward a linear proportionality between the transition frequency and the liquid conductivity. The onset frequency of the oscillating mode has been reported since a very distinct change in meniscus behavior is observed at the juncture when the meniscus changes from the elongated-fast dripping mode to the oscillating mode.

B. Different spraying modes

We shall now discuss the different spraying modes observed. We have limited our discussion to the modes occurring between approximately $4\times10^3 V_{p-p}$ and $6\times10^3 V_{p-p}$ since the dripping mode below 4 kV and the discharge mode above 6 kV are driven by phenomena that are independent of liquid conductivity and polarization, consequently, are not relevant here. In fact, we notice from the schematics in Fig. 1 that the change in conductivity does not affect the frequency-voltage window for these modes. The features that we focus on are specifically the meniscus shape, behavior, and ejection rate during each mode and the frequency window in which each of these modes are favored.

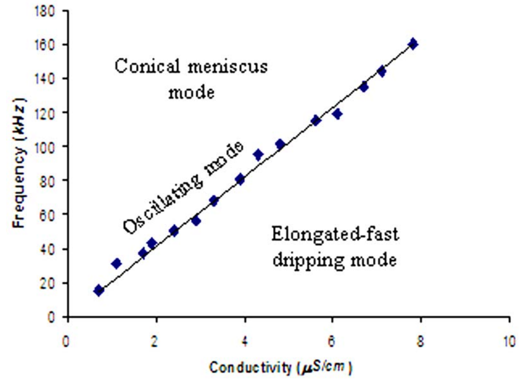

FIG. 2. (Color online) Onset frequency of the oscillating mode as a function of conductivity. With increasing conductivity of the ethanol sample, the onset of oscillating mode is delayed to higher frequencies. The change in the onset frequency is also approximately linear with the sample conductivity.

1. Tip streaming

In the mode classified as tip streaming, small fragments are torn intermittently from the liquid meniscus. The dimension of these fragments is much smaller ($\sim 5\ \mu$m) than the entire liquid meniscus, which has a radial dimension comparable to the diameter of the electrospray needle (300 μm). This mode is more prevalent at lower applied frequencies (<30 kHz) than at higher frequencies. The frequency of ejection events in this mode varies with the applied frequency and initially increases with frequency, until approximately 20 kHz. Beyond this frequency, elongated-fast dripping mode starts appearing along with tip streaming and the behavior at the meniscus tip changes. The tip-streaming mode is shown in Fig. 3. The shapes of the meniscus as well

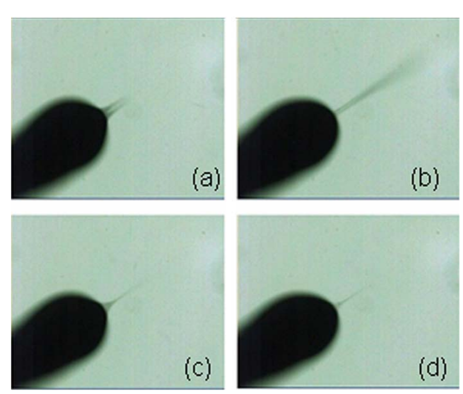

FIG. 3. (Color online) Tip streaming: Successive images at 0, 0.5, 1, and 1.5 ms at an applied frequency of 13 kHz for ethanol. Flow under gravitational head of the liquid in the needle.

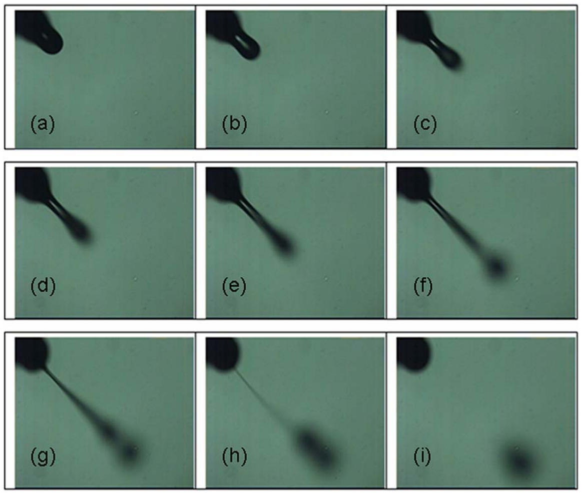

FIG. 4. (Color online) Elongated-fast dripping: Successive images at 0, 2.2, 2.4, 2.6, 2.8, 3.2, 3.6, 3.8, and 4 ms at an applied frequency of 40 kHz for glycerine-ethanol (20%–80%, by volume) mixture. Flow under gravitational head of the liquid in the needle.

as the dimensions of the ejected fragments and the fact that the ejection frequency increases with applied frequency suggest that this mode shares some similarities with dc spraying and particularly the microdripping and cone-jet modes, and is comparable to dc spraying with a varying field.

2. Elongated-fast dripping mode

In the elongated-fast dripping mode liquid accumulates near the needle tip initially and then it is stretched to form a cylindrical jet with velocities as high as 1 m/s. This jet is then detached at the needle tip to form drops of approximately 50–100 μm in dimension. In contrast to the steady jet in dc electrospraying, where a thin (~ 1 μm) but steady jet emanates from the cone tip, this jet is unsteady, retracting and reforming subsequently, and much larger in radial dimension. It still suffers from the capillary or inertial instabilities for jets as it pinches off at the needle to form drops with the size of the needle, as seen in the images of Fig. 4. With the advent of this mode, ejection events decline with frequency and then become approximately constant after $\sim$30 kHz, where the elongated-fast dripping mechanism becomes predominant. The elongated-fast dripping mode is shown in Fig. 4. Unlike the tip-streaming mode, the ejection frequency for this mode does not depend on the applied frequency but varies between 10^2–10^3 Hz.

In Fig. 5 we have shown the variation in ejection frequency for a doped sample, where the oscillating meniscus mode appears at 50 kHz. The initial increase and then decrease of the ejection events point to the initial dominance and then decline of the tip-streaming mode because of the occurrence of elongated-fast dripping. For comparison, the ejection frequency in the discharge assisted ejection mode is also shown as a function of the applied frequency. This mode is a result of corona discharge occurring due to high voltage being applied at the needle tip. Ejection frequency for this mode remains constant throughout, showing that the liquid conductivity has no influence on gas discharges, which is expected.

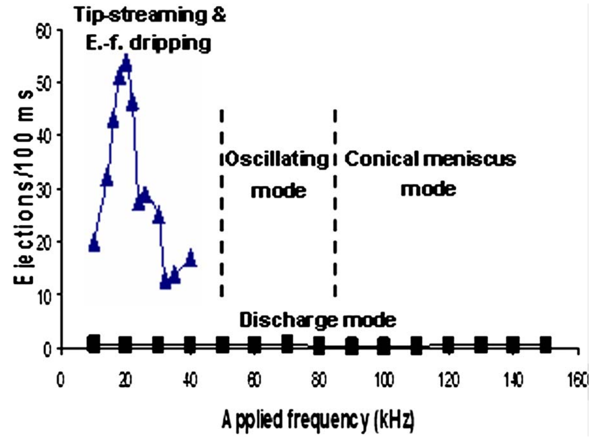

FIG. 5. (Color online) Variation in ejection frequency with applied frequency for a doped sample. The triangles correspond to the tip-streaming and elongated-fast dripping modes, while the squares represent the discharge assisted ejection mode.

3. Oscillating mode

The transition from the elongated-fast dripping mode to the oscillating mode is very sharp. This is because in the elongated-fast dripping mode drop pinch off at the needle tip takes place. In contrast, the oscillating mode seems to be immune to capillary or inertial instabilities and the jet does not pinch off at the needle tip at all (contrast Fig. 6 to Fig. 4). Instead it vibrates and whatever ejection takes place occurs from the tip of the meniscus, when it is at its fully elongated position. The meniscus is similar to a cylinder with a spherical tip. The cylindrical part of the meniscus in the oscillating mode mostly does not pinch off but oscillates with time. The radius of the cylinder consequently oscillates in a single cycle of vibration, attaining its largest value when the length is smallest and its least value when the cylinder is fully elongated. However, the radius can increase over a longer time scale due to accumulation of liquid from the needle. Also, the ejection from the meniscus tip does not take place every

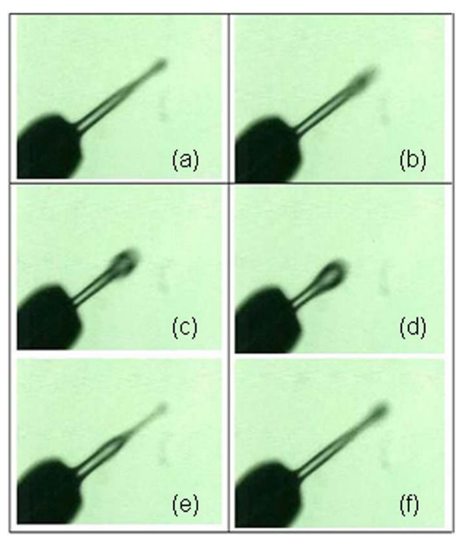

FIG. 6. (Color online) Oscillating meniscus mode: One oscillation of the meniscus, successive images at 0, 0.5, 1, 1.5, 2, and 2.5 ms at an applied frequency of 81 kHz, and for doped ethanol with conductivity 3.9 μS/m. Flow under gravitational head of the liquid in the needle.

TABLE I. Time scales in oscillating meniscus mode: The variation in the oscillation frequency of the jet has been compared with the applied frequency and the inverse momentum diffusion time scale. It can be seen that jet oscillation frequency is strongly correlated to the diffusion time but not the applied frequency.

Diameter (μm)	Applied frequency (kHz)	Oscillation frequency (Hz)	Inverse momentum diffusion time (Hz)
66.7	55	312.5	342
70	81	307.7	310
75	135	295.1	270.2
91.7	55	230.8	180.8
97.1	101	192.8	161.2

period, and the ejection frequency is less than the oscillation frequency. There can be either single or multiple drop formation from a single ejection event. Total meniscus breakup, akin to the elongated-fast dripping mode, is unfavorable in the oscillating mode. However, similar to the elongated-fast dripping mode, the meniscus vibration frequency in the oscillating mode is totally independent of the applied frequency or the liquid conductivity. It depends only on the diameter of the meniscus and decreases with increase in this length scale. Figure 6 shows one cycle of the oscillating meniscus mode at 81 kHz. During the course of an experiment the meniscus diameter generally increases as more and more liquid accumulates there; subsequently, the oscillation frequency shows a gradual decline. This suggests that liquid viscosity is playing an important role in this behavior. In fact, this can be clearly deduced from Table I, where the oscillation frequency and the meniscus diameter have been provided along with the applied electrical frequency and the inverse momentum diffusion time scale. The inverse momentum diffusion time scale is ν/l^2, where ν is the liquid momentum diffusivity (1.52×10^{-6} for ethanol) and l is the drop length scale, taken as the diameter of the liquid cylinder at its most compressed state. The measurement of oscillation frequency has been carried out during intervals when droplet ejection from the meniscus tip has ceased since ejection can vary the oscillation frequency.

As clearly seen in the preceding table, the applied frequency and the oscillation frequency differ by approximately three orders of magnitude. Besides, there is no observable correlation between the two. In contrast, the inverse momentum diffusion time scale is quite close to the observed oscillation frequency and explains the variation of the experimental frequency with the meniscus diameter. The difference between the two might be due to inaccuracies involved in the measurement of the diameter when the comparatively small meniscus is oscillating at high velocities ($\sim$1 m/s). We believe that this time-scale behavior can also explain the observed range for ejection frequencies in the elongated-fast dripping mode.

4. Conical meniscus mode

This is the final spraying mode observed, which occurs after the oscillating meniscus mode with increasing applied

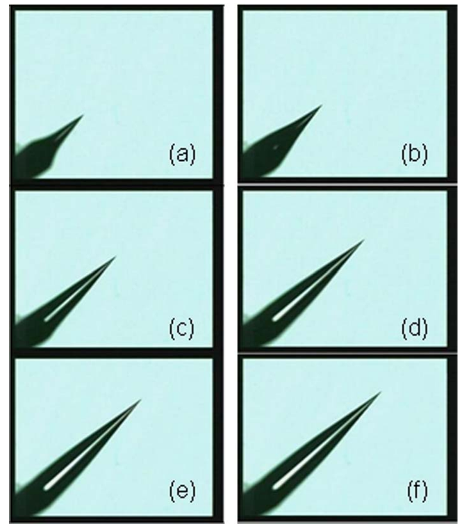

FIG. 7. (Color online) Conical meniscus mode: Successive images at 0, 2, 4, 8, 10, and 12 s for ethanol at 100 kHz. The growth of the ac cone and its slender shape can be clearly observed. Flow under gravitational head of the liquid in the needle.

frequency. The meniscus takes a conical shape with a thin ($\sim$1 μm) jet ejecting from the tip of the conical meniscus, very similar to the dc–cone-jet mode. However, it has two major differences from the dc–cone-jet mode. Firstly, the cone half angle ($\sim9°$) is not the same as the dc case ($\sim47°$) for the same liquid, ethanol. Secondly, the meniscus shows a longitudinal growth with time, over a time scale much longer than the ac period or the charge relaxation time. These features are depicted in Fig. 7. The elongation of the cone continues, with a constant cone angle, until it becomes unstable at a large aspect ratio ($\sim$15:1) and breaks up. Then, the meniscus is reformed and the entire cycle is repeated. This was described in our previous work for pure alcohols. Here, we report the observation of these cones for doped ethanol, whose conductivity is increased. However, we cannot report any upper bound in frequency, if any, for the observation of these conical modes as we cannot generate frequencies beyond 200 kHz. We want to clarify that we find that the cones are perfectly stable but are not perfectly steady, in the sense that the cone tip retains its shape but its length is elongating in time over a period much longer than the ac period.

From the preceding information, we can find several features that can explain the relevant charging time scales for the ac spraying phenomena. Figure 2 shows the linear increase in the frequency for the onset of oscillating mode with the liquid conductivity. For charging of electrolytes, the relevant time scale is the charge relaxation time scale, which describes the gradual discharging of the liquid bulk, until all the residual charge is confined to the surface,

$$\rho = \rho_0 e^{-t/\tau}. \tag{1}$$

Here, ρ_0 is the initial charge density, while ρ is the charge density after time t, and τ is the charge relaxation time ($\beta\varepsilon_0/K$). For our case the relevant time scale is the semiperiod of the applied field; consequently, Eq. (1) can be rewritten as

$$\rho = \rho_0 e^{-K/2\omega\beta\varepsilon_0}. \quad (2)$$

Here, ω is the applied frequency. Consequently, it is clear that with increasing frequency, there is less time available for the bulk to discharge and it retains a higher fraction of the original charge. However, with increasing conductivity the bulk discharges faster and retains a lesser amount of charge. With these arguments and the results of Fig. 2, it is clear that the oscillating meniscus has a greater amount of charge than the meniscus in the elongated-fast dripping mode. More importantly, the linearity of the reported curve demonstrates the existence of a threshold amount of charge for the advent of the oscillating meniscus mode, which is independent of the liquid conductivity or the applied frequency. By fitting the experimental results with a straight line, we find that when the period is approximately 20 times the charge relaxation time, the transition from the elongated-fast dripping mode to the oscillating meniscus takes place. Under such conditions, at every half cycle, approximately 10^{-3}% of the bulk charge is unrelaxed. Hence, we can say that for the oscillating meniscus to appear, 90% of the liquid bulk charge needs to be retained. We also recall that in the oscillating mode there is no pinch off near the needle tip unlike in the elongated-fast dripping mode, where the jet pinches off near the needle tip. Consequently, it can be inferred that this transition occurs at a specific charge fraction since the stabilizing effect of charge against the capillary instability prevents breakup of the liquid cylinder and allows for the transition from the elongated-fast dripping to the oscillating mode. The vibrating menisci in the oscillating mode are the precursor to the steady growing conical menisci, which appear at higher frequencies; it is clear that even more bulk charge needs to be retained for the conical meniscus to appear and grow in length. Evidently, sufficient bulk charge exists with the oscillating mode to suppress the instability for jet break up. However, this bulk charge does not grow in time and hence cannot elongate the jet. The elongation occurs at even higher frequencies when the ac conical shapes appear, suggesting that an ever-increasing bulk charge is responsible for the ac cone formation and its elongation. This is in complete contrast to the classic cone-jet mode in dc spraying, where the charge in the liquid bulk is relaxed because the charge relaxation time scale is the smallest relevant time scale for that process, and the bulk becomes electroneutral.

This clearly points to the distinction between dc and high frequency ac sprayings. However, it does not explain why steadily growing ac cones appear in the first place since the applied high frequency should form either cones every half cycle or no cones at all. The appearance of these cones might be a result of charge entrainment, i.e., ions of specific polarity not totally discharging every period, as a result of which that specific charge progressively builds up and ultimately when it crosses a certain threshold, the meniscus changes to a cone. The amount of charge buildup would be sensitively dependent on the relaxation time scale since discharging of the bulk depends on that. This would explain why cones appear only when the period of the applied field is a specific fraction of the charge relaxation time scale. This charge buildup model can also explain the growth that is observed in high frequency ac cones and not in dc cones. Since there is an entrainment of specific polarity charge every period, there is a temporal growth in the charge density in the bulk, and Coulombic repulsion forces the cones to grow. Ultimately when the aspect ratio of the cone becomes too high, capillary instability eventually destroys the conical meniscus and then the entire process is repeated. For dc cones, there is no charge buildup and, consequently, the conical meniscus does not vary with time. Similarly, the absence of a cone and jet elongation in the elongated-fast dripping and oscillating modes suggests that the bulk charge does not grow in time for these modes. Further experimental verification of this model is being carried out.

In summary, we have reported the observation of different spraying modes for ac spraying and the transition between them and examined the role played by different time scales in the occurrence of these modes. The unique growing slender cones with an ac field are attributed to the accumulation of a net charge due to unrelaxed bulk charges in every half cycle. The vibration and drop ejection frequencies for elongated-fast dripping and oscillating modes are, however, related to the momentum relaxation times within the drop. The transition between the different modes depends on the fraction of the charge relaxed in the liquid. Future work includes the quantification of the growth rate of the conical meniscus with applied frequency and understanding of the charge accumulation behavior, which might elucidate the mechanism responsible for this spraying behavior.

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