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Non-contact and Breakup of Oppositely Charged Droplets

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Abstract – In this paper, a detailed experimental study on the non-contact and breakup process of oppositely charged drops for different conductivity liquids has been reported. Under different applied voltages, the breakup morphology of oppositely charged droplets with various ionic concentrations has been accurately captured, and the image processing technology was used to analyze the effect of several parameters on the breakup process. The breakup structures based on ionic concentrations are measured to build up their relationship for studying the dynamic behaviors of oppositely charged droplets. For poor or non conductivity liquid, no breakup behaviors are obtained regardless of how high voltage is applied. However, various breakup structures are detected as the applied voltage increases for high conductivity liquid. The behavior of rebound with breakup is a form of Coulomb fission which means the surface charge on droplet tip reaches to Rayleigh limit prior to air discharge. A fitting curve is given to demarcate the behavior which breakup occurs; droplet with high conductivity has the maximum change of breakup volume which indicates liquids in high conductivity are more sensitive to the change of electric field strength.

Keywords**:** Charged Droplet, Conductivity, Coulomb fission, Breakup volume, High speed Microscopy.

1. Introduction

Charged droplet and its dynamic behaviors induced by electric fields have been extensively studied for more than one century years; some technology on charged droplets such as electrostatic spray has been widely applied to spray coating, micro/nanometer thin film, air purification，micro combustion, space micro propulsion, mass spectrometry etc. There exist important phenomena of droplet coalescence and separation in practical elecrtospray and the basic mechanisms about the dynamic behavior of adjacent droplets are only partially understood. Based on the minimum surface energy, undoubted coalescence occur once adjacent droplets contact. However, when oppositely charged droplets are electrically driven to get close to each other, special electrohydrodynamic behaviors appear rather than simple coalescence which are of interest to numerous scholars, and some unsolved mechanisms together with interesting new phenomena have been reported very recently.

There is a new finding that two oppositely charged droplets bounce off each other and do not coalesce if their charge exceeds a threshold value (Ristenpart W D et al, 2009; Mugele F, 2009;), which considers the coalescence-recoil transition is a consequence of the drop geometry. These investigations show that under the influence of an electric field of sufficient strength, two oppositely charged water droplets recoil after contact rather than coalesce, depending on the curvature and local capillary pressure at the liquid bridge between two adjacent droplets. A surface energy model (volume-constrained area minimization) is proposed to describe the transition between coalescence and recoil (Brid J C et al, 2009). According to mean curvature flow, a new model is proposed to explain the remarkable phenomenon of bouncing and coalescence of charged droplets, and one advantage of this approach compared to the existing ones is that strong assumptions on the precise shape of the bridge between the touching fluid droplets is no more needed (Helmensdorfer S, 2014).

By contrast, stable emulsions, which consist of oppositely charged droplets dispersed in a fluid have been reported (Liu T T et al, 2012), non-coalescence of oppositely charged droplets in pH-sensitive emulsions is successful prepared and shows that electrostatic interactions between droplets do not determine their stability and reveal the unique pH-dependent properties of emulsions stabilized. The bounce off after contact behavior of oppositely charged droplets has been verified in great detail. However, the dynamic behavior of breakup prior to contact for oppositely charged droplet has been few reported, other behaviors like breakup after contact and bounce off before contact are also lack of understanding and have no reasonable explanations. Therefore, some new ideas needed to reveal the bounce off and breakup mechanisms of oppositely charged droplets.

The subject of the present work mainly concerns the dynamic behaviors of the non-contact and breakup of oppositely charged droplet under the external electric field. From the perspective of ionic migration, some new explanations to bounce off after contact behavior are conducted and based on Rayleigh limit, liquids with the property of poor and perfect conductivities are used to analyze the breakup prior to contact behavior in great detail. A fitting curve is given to demarcate the behavior which breakup occurs, and the parameters of breakup volume and elongation speed are defined to describe the breakup intensity. Our study aims to provide some new understanding of electrically driven behaviors of oppositely charged droplet.

2. Experiments

The experimental setup for studying the breakup behavior of oppositely charged droplet is described in detail in Fig. 1. The main body of the oppositely charged droplet collision device is made of organic glass. The lower droplet positioned on the super-hydrophobic substrate is strict symmetry with the upper droplet suspended in the cooper wire. With the help of the micrometer, the upper droplet could slowly get close to the lower one; the experiments presented in the immediately following are carried out by use of a high voltage supply, which could make these two droplets get opposite charge with its negative and positive connected to the cooper wire and the super-hydrophobic substrate respectively. Depending on the high-speed camera (MotionProTMX4puls) and a microscopic zoom lens (NAVITAR12X), the dynamic behaviors of the non-contact and breakup of oppositely charged droplets with different conductivity are visualized and recorded with a frequency of 15000 frames per second. LED cold light source is placed closely ahead of high speed camera and rightly faces the capillary so that clearer images can be captured in a very short exposure time.

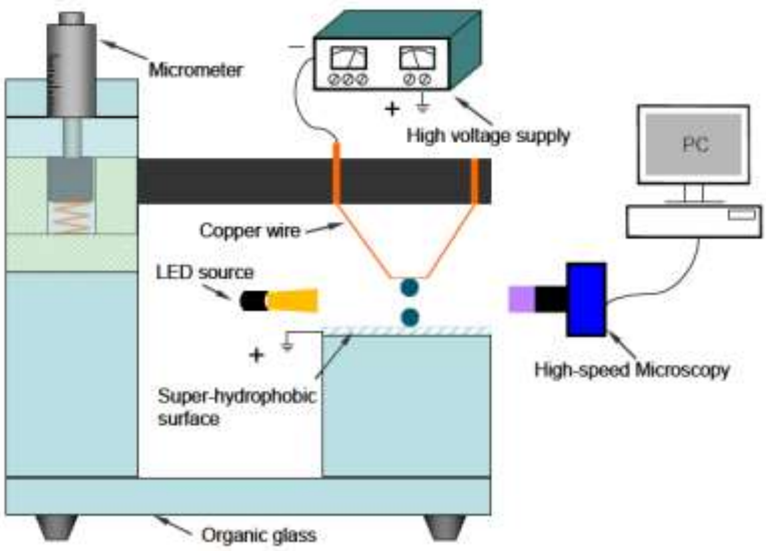

Fig. 1. Details setup of oppositely charged droplet.

The hydrochloric acid solutions with various electrical conductivities were carried out by adding distilled water into hydrochloric acid solution of 6mol/L to change the concentration. The electric conductivities of these solutions have been measured by a potentiometer and given in Table 1. All experiments are carried out in ambient air with temperature of 25±0.5℃.

Table. 1.The electrical conductivities of the solutions used in the experiment.

Liquids	Hydrochloric acid solution							
Mass fraction	0	0.5%	2%	3%	4%	5%	7.5%	20%
Conductivity （μs/cm）	1.3	77	169	285	360	420	571	820

3. Results and Discussion

3.1 Charge Distributions on Electrified Liquid Droplet

When a motionless droplet carries net electric charge, the charges reside on the exterior surface due to Coulomb repulsion (Gomez A, 1994). Theoretically, the charge density on the droplet surface is uniform and droplet stability depends on the relationship between the fluid static pressure differences P_s caused by surface tension and expansion force P_e caused by coulomb repulsion, as shown in fig. 2. A primary theoretical analysis of the breakup condition for a charged droplet shows that Coulomb fission should occur when the total charge on the droplet surface reaches a limiting value which named the Rayleigh limit, given by

$$Q = 8\pi\sqrt{\varepsilon\sigma}\,r^{3/2} \tag{1}$$

Where *e* is the permittivity of the medium surrounding the droplet, s is the droplet surface tension and *r* is the droplet diameter. (Rayleigh, 1882). *Q* is a critical value which makes *e s P* = *P* and the droplet will finally breakup due to the expansion force exceeds the fluid static pressure differences once *q* >*Q*. It is not difficult today to understand the Rayleigh limit expression by analyzing the balance of the electrical energy $\frac{Q^2}{8\pi\varepsilon r}$ caused by electric charge and surface energy 4p*r* 2s maintained by surface tension(Wang J F, 2002). The surface energy maintains the surface stability and prevents the deformation of the motionless droplet; on the contrary, the electrical energy enhances the surface instability of the droplet. Based on the virtual work principle, the difference of total energy W to the radius *r* is

$$F_r\big| = \frac{\partial W}{\partial r} = 8\pi\sigma r - \frac{Q^2}{8\pi\varepsilon r^2} \tag{2}$$

The Rayleigh limit could also be obtained*Q* = 8p es *r*3/ 2 when *r F* =0, which means the virtual work did by the electrostatic force reaches to the virtual work did by the inherent surface tension at this time. Usually the surface charge density is used to describe the charge condition for a droplet of certain size and the charge density limit based on Rayleigh limit is

$$\phi = \frac{Q}{4\pi r^2} = 2\sqrt{\frac{\varepsilon\sigma}{r}} \tag{3}$$

This equation indicates breakup will occur once the surface charge density reaches to a threshold value.

However, these theoretical analysis about droplet instability are based on a hypothesis that the net charge distributed on the droplet surface is uniform, a spherical droplet with its diameter of D won't breakup until the whole surface charge density reach to a certain level according to Rayleigh limit. Actually, the surface charge density of charged droplet is nonuniform under the effect of external electric field, and the electric charge will migrate along the direction of the electric field. As a result, there exists a region where the surface charge density is higher than any other part of the droplet, as shown in fig. 3. If the electric field strength is sufficiently high, the whole charge distribution on the droplet surface is extremely uneven which makes the charge accumulation in a region first reaches to its Rayleigh limit and the droplet will breakup partially. However, the whole droplet hasn't reached to Rayleigh limit at this moment because the inherent charge on the whole surface is unchanged, which means the Coulomb fission would happen before the electric charge on the whole droplet reaches to Rayleigh limit, in other words, a droplet would breakup at a certain level of Rayleigh limit, here we called sub-Rayleigh limit (Huo Y P, 2012). As a result, droplet breakup criterion can't be simply evaluated by Rayleigh limit due to the nonuniform charge density on droplet surface. However, look from the local scope, the partial region of the droplet which leads to the partial breakup has a uniformly higher charge density, and Coulomb fission happens as a result of Rayleigh limit in this area. For droplet breakup behavior, it is the breakup region that first reaches to Rayleigh limit rather than the whole droplet reaches to Rayleigh limit.

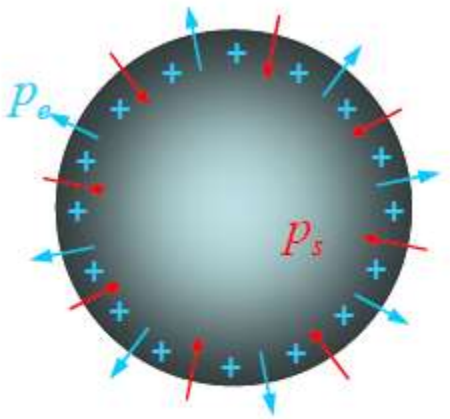

Fig. 2. Theoretical charge distributions of charged droplet

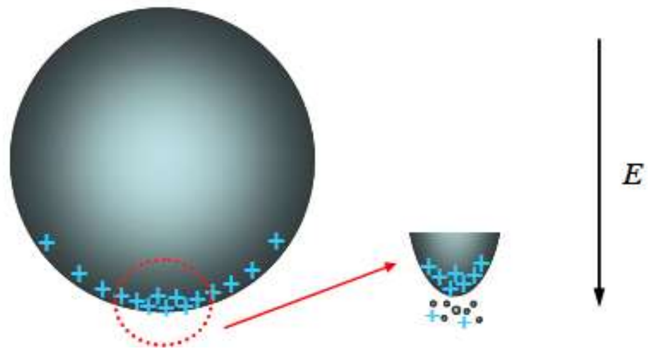

Fig. 3. Actual charge distributions under external electric field

3.2 Charge Distributions on Electrified Liquid Droplet

Theoretically, when two oppositely charged droplets get close to each other, coalescence will finally happen because of the inter-attraction. On the contrary, it is interesting that five dynamic behaviors which include coalescence after contact, bounce off after contact, bounce off prior to contact, breakup after contact and break up prior to contact are observed for oppositely charged droplets. Based on Rayleigh limit, these special phenomena could get reasonable explanation by analyzing charge distributions on electrified liquid droplet (Shrimpon J S, 2005). As we know, ionic migration will occur along the direction of the electric force under the effect of external electric field, and the oppositely electric charge will mainly accumulate at their two tips. The electric field strength around the droplet tip will be continuously enhanced when two droplet tips get close to each other with the effect of the electric force, as a result, the charge density at droplet tip will be gradually increased in the process of moving toward each other.

For charged droplet of prefect conductivity, charge accumulation at the tip could reach to a sufficiently high level under the external electric field due to the high ionic concentration within the droplet, and Coulomb fission will occur once the electric charge density at droplet tip approach to Rayleigh limit. Fig.5 shows the dynamic morphology of oppositely charged droplet with whose conductivity is 820μm/cm. Two new dynamic behaviors named bounce off after contact and breakup prior to contact are observed with the increase of applied voltage. The behavior of bounce off after contact occurs when the applied voltage increase to 1.0kV and this dynamic change results from the charge migration which can get a reasonable excuse by the exciting research on chemical physics (Impey R W, 1983; Koneshan S, 1998). When the liquid bridge formed, the negative ions would run from negative electrode to positive one and at the same time positive ions in the liquid would move to the negative side. One tip will accumulate a certain amount of charge come from another tip and there appear transitory Coulomb repulsion between these two droplet tips. If the initial charge density at droplet tip reaches to a certain level, the transitory Coulomb repulsion caused by more ionic migration will be bigger enough and plays a very important role in the bouncing process. Here the bounce off after contact behavior should not be simply judged by droplet geometry, which just analyzes the relationship between the curvature and local capillary pressure at the liquid bridge without taking the transitory Coulomb repulsion into account. However, this dynamic behavior won't happen at a very low conductivity level because the number of ionic migration is so few that transitory Coulomb repulsion has little effect on the bounce off behavior. The behavior of breakup prior contact occurs when the applied voltage increases to 1.2kV, and this phenomenon can only appear after the droplet conductivity reaches to a certain degree. In the process of getting close to each other, the increase of electric field strength around droplet tips makes the more and more charge accumulation at droplet tips, and Coulomb fission finally occurs when the charge density at droplet tip reaches to Rayleigh limit. What's more, the behavior of breakup before contact means the surface charge accumulates towards the droplet tip reaches to Rayleigh limit prior to air discharge. Otherwise, bounce off will occur because of the decrease of the electric field strength and no Coulomb fission is observed. With the increase of applied voltage, the ionic migration towards droplet tip is accelerated and the number of ionic accumulation increases, the region of approaching to Rayleigh limit enlarges which lead to larger breakup volume, as a consequence, More intense breakup appear when the applied voltage increases ,as shown in Fig.5 (4).

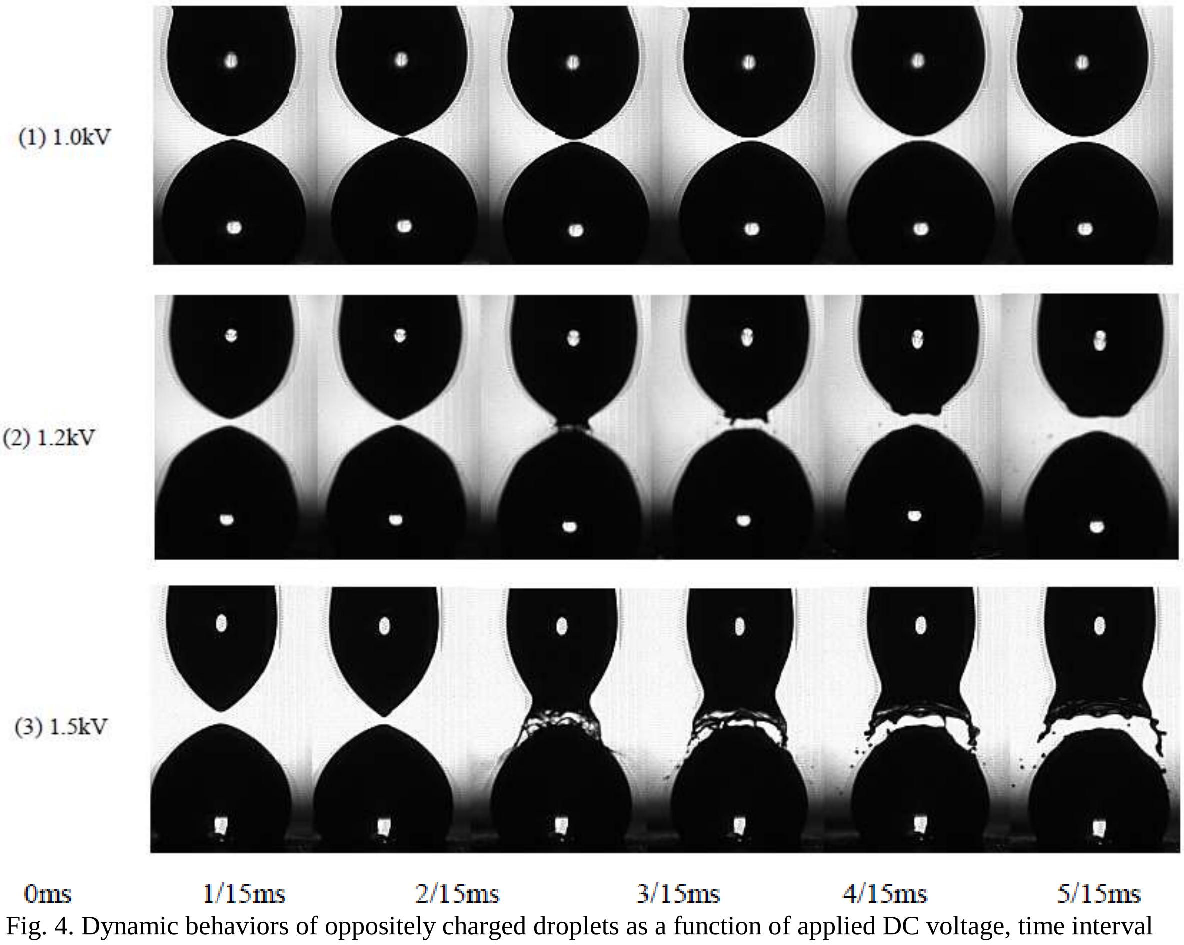

Fig. 4. Dynamic behaviors of oppositely charged droplets as a function of applied DC voltage, time interval is 1/15ms, liquid conductivity is 820µs/cm.

3.3 Conductivity Effect on Breakup Behaviour

As a form of Coulomb fission based on Rayleigh limit theory, breakup prior to contact is a special dynamic behavior of oppositely charged droplet which can be observed only when the liquid conductivity reach to a certain level. In the following experiment, some parameters which include critical field strength, breakup volume and elongation speed are used to evaluate the influence of conductivity on the breakup characteristic of adjacent droplets.

As we known, Rayleigh limit needed for breakup behavior appear at droplet tip, and the charge density which leads to Rayleigh limit directly relative to the relationship between conductivity and applied voltage. Fig.6 gives the critical applied voltage for the breakup of oppositely charged droplet with the change of liquid conductivity. With the help of origin8.0, $y = ax^b$ is chose to give nonlinear Curve Fit based on the experimental data, and the fitting accuracy with $a \approx 11.3$, $b \approx -2$ approaches to 99 percent compared to origin data. This curve indicates a minimum applied voltage for different conductivity liquids which first leads to Coulomb fission and as a boundary line, it also plays a key role which distinguishes the dynamic behaviors of bounce off from breakup before contact when a combination of conductivity and applied voltage is given in the current experiment. For liquid in high conductivity, low applied voltage needed for the occurrence of the breakup behavior, on the contrary, very high applied voltage needed for the low conductivity droplet to breakup. In addition, no breakup behavior occurs for distilled water with whose conductivity is 1.3 μ s/cm in the current experiment condition. It means that the property of conductivity plays a very important role in promoting the breakup of oppositely charged droplets.

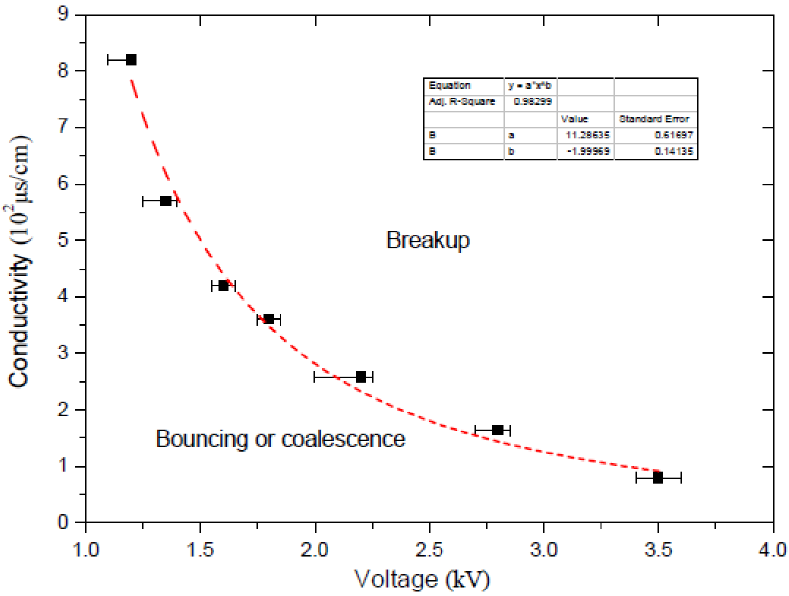

Fig. 5. Critical field strengths for the breakup of oppositely charged droplets under different conductivities.

In order to describe the breakup strength, a special parameter V^b named breakup volume is defined. We calculate the volume change of droplet tip in 2/15 ms from the very beginning of breakup as the value of breakup volume, which can be obtained by measuring the bottom radius and the height of the circular cone. Therefore, direct results can be given out to reflect the break strength. Fig.7 shows the breakup volume as a function of applied voltage under different liquid conductivities. As the applied voltage increases, the breakup volumes for droplets of different conductivities all enlarge once Coulomb fission occurs. This demonstrates the increase of electric field strength accelerates the ionic migration towards droplet tip and leads to the increase of the number of ionic accumulation on droplet tip, therefore, more liquid volumes at the tip approach to Rayleigh limit and more powerful breakup finally occurs. Although the droplets with different conductivities have the same tendency above, the growth rates of their breakup volumes are much differences, which indicate droplet with high conductivity has the maximum change of breakup volume with applied voltage increase. In other word, high conductivity liquids are more sensitive to the change of electric field strength which are caused by the larger number of ionic migration.

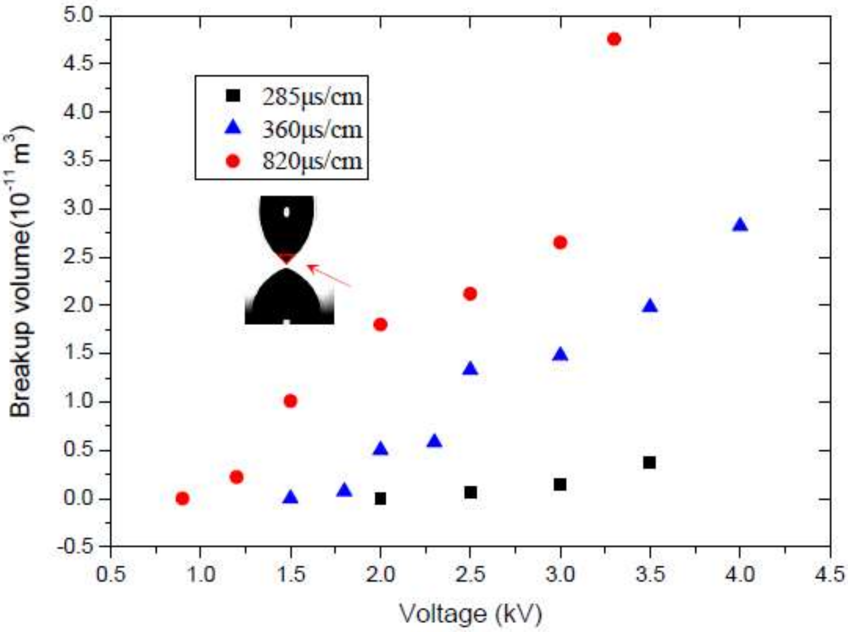

Fig. 6. Breakup volumes as a function of applied DC voltage. Liquid: Hydrochloric Acid

4. Conclusion

In the present study, an experimental investigation is conducted to study the non-contact and breakup behaviors of oppositely charged droplets. Based on Rayleigh limit, some new explanations of electrically driven droplets are conducted in great detail. For charged droplets of prefect conductivity, two new dynamic behaviors named bounce off after contact and breakup before contact are observed with the increase of applied voltage. There appears transitory Coulomb repulsion between the tips of oppositely charged droplet which plays a very important role in the bouncing process. The behavior of breakup before contact can only appear when the droplet conductivity reaches to a certain degree, and the occurrence of Coulomb fission means the surface charge accumulates towards the droplet tip reaches to Rayleigh limit prior to air discharge.

A fitting curve is given to demarcate the behavior which breakup occurs. For liquid in high conductivity, low applied voltage needed for the occurrence of the breakup behavior, on the contrary, very high applied voltage needed for the low conductivity droplet to breakup. The property of conductivity plays a very important role in promoting the breakup of oppositely charged droplets. Breakup volume is defined to describe the breakup strength of the upper droplet. As the applied voltage increases, the breakup volumes for droplets of different conductivities all enlarge once Coulomb fission occurs; droplet with high conductivity has the maximum change of breakup volume which means liquids in high conductivity are more sensitive to the change of electric field strength caused by the larger number of ionic migration.

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