Surface oscillations and breakup characteristics of a charged droplet in the presence of various time varying functions

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Abstract

A charged droplet can be levitated in the air when suspended in a trap subjected to quadrupole potential. The applied potential is typically sinusoidal. Such a charged drop undergoes surface oscillations subsequently building enough charge density to undergo Rayleigh breakup. In this work, we investigate high-speed imaging of large amplitude surface oscillations of a levitated sub-Rayleigh charged drop and corresponding Rayleigh breakup, by applying non-sinusoidal waveform. The droplet oscillation behaviour and Rayleigh breakup (recorded at 100k frames per second) are investigated for different applied waveforms such as sine, square and ramp at different imposed frequencies but a fixed voltage. The surface oscillates in sphere-prolate-sphere-oblate (SPSO) mode and sometimes in the sphere-prolate-sphere (SPS) modes depending upon the complex interplay of forces such as charge (Q), applied field (Λ) and shift of the droplet from the geometric center of the trap (Zshift). The Fast Fourier Transformation (FFT) analysis shows that the droplet oscillates with the imposed frequency irrespective of the type of waveform. While in the sinusoidal case, the nonlinearities are strong, in the square and ramp potentials, there is an admittance of all the frequencies of the applied voltage. Further, the droplet breakup characteristics do not change in the presence of various waveforms. The experimental observations are validated with a potential theory with viscous correction and found to be in a fair agreement.
I. INTRODUCTION

The understanding of how inherently charged droplets oscillate, deform and break is important in many technological applications such as inkjet printing [1], fuel injection [2], crop spraying [3] and electrospray atomization for ion mass spectroscopy of biomolecules [4]. The oscillating droplets or bubbles have been considered a novel experimental platform for understanding a wide variety of natural, analytical and biological applications, for example, interfacial reactions and rheology, thin film, biosensors, biophysical simulations etc.

The experimental techniques involving oscillating droplet/bubble have several advantages such as high rate of surface reaction and larger interfacial activity due high surface area-to-volume ratio, reduced consumption of expensive chemicals, and precise environmental control due to miniaturized droplet platform. The controlled oscillations of a surface can be considered as a basis for many analytical studies. For example, the sinusoidal oscillations in the presence of constant amplitude and imposed frequency of an applied force can be used to measure the surface dilational modulus of a molecular (surfactants or macromolecules) monolayer by estimating the time varying interfacial tension.

Several methods have been used to examine droplet oscillations such as pendent drop [5, 6], sessile drops [7, 8], acoustically levitated drop [9, 10], electromagnetically levitation of drop [11] and electrodynamic levitation [12] of a drop. For understanding the oscillations of a small sized (∼ 10-300 µm) charged droplet in electrodynamic levitation is considered to be one of the most efficient techniques of droplet levitation. Since the droplet size is small the gravity plays a negligible role in the steady state droplet deformation and the droplet remains nearly spherical during oscillations.

The capillary oscillations of an electro-dynamically levitated charged droplet can be considered as an idealised system because a charged droplet is levitated in a contact free environment with no wall effect. Thereby, suitable modelling and development of analytical theory for capillary oscillations of a charged droplet in a quadrupole trap can be helpful in making this system as a very reliable tool for characterizing the fluid properties such as surface tension, viscosity etc.

Towards this, many analytical studies on the droplet oscillations have been reported [13-16]. Recently Singh et al. [13] reported theoretical analysis of surface oscillations of a charged liquid droplet levitated in an electrodynamic balance in the small electro capillary
limit. The oscillations were found to be governed by potential flow description with viscous corrections. The effect of viscosity was taken into consideration by including the viscosity in the normal stress balance condition for potential flow limit. The analytical model was validated through experiments involving the recording of surface oscillations in the presence of sinusoidal applied waveform at a very high frame rate \(\sim 1\) hundred thousand fps. An interesting question which can be asked here is: what will be the effect of non-sinusoidal waveform on the behaviour of charged droplet oscillations.

Droplet levitation using sinusoidal applied electric potential is fairly well established. The mechanism at play is the same as that in the classical Mathieu equation to describe dynamics under oscillatory forcing. A time average net ponderomotive force acting towards the centre of the trap overcomes destabilizing forces such as gravity and charge repulsion. It is then worthwhile to explore the robustness of the mechanism, by subjecting it to non-sinusoidal applied potentials. It is known that sinusoidal potentials of frequency \(\omega\) can lead to responses in the deformation of the droplet in \(\omega\) or \(2\omega\) which can be attributed to the quadrupole potential acting on the net charge or the square of the Maxwell stress tensor of the applied potential, respectively. It is known that non-sinusoidal potentials such as square and a ramp potential consist of several harmonics (odd or even) of the applied frequency. It is then interesting to see the signature of these harmonics in the deformation pattern. This can then establish the deformation response to arbitrary potentials. Undertaking a systematic investigation of this issue forms another motivation for the present study.

In the present manuscript, we have examined the surface oscillation characteristics in the presence of various forcing waveform such as sine, square, and ramp. Unlike Singh et al. [13] the surface oscillations characteristics are examined by Fast Fourier transform (FFT) analysis of surface dynamics. The experimental observations are also compared with the viscous potential flow theory.

II. MATERIALS AND METHOD

The experiments presented in this work involve levitation of charged ethylene glycol (EG) droplets. A small amount of NaCl was added to increase the electrical conductivity \((\sigma)\) of the droplet and the conductivity was measured using a conductivity meter (Hanna instruments). The conductivity range was \(\sim 50-80 \mu S/cm\). The viscosity \((\mu_i)\) of ethylene glycol, measured
using Ostwalds viscometer, was 0.011 Ns/m². The surface tension (\(\gamma\)) of the droplet was measured using the pendant drop (DIGIDROP, model DS) method as well as the spinning drop (dataphysics, SVT 20) apparatus. The value of \(\gamma\) was obtained as 0.047N/m. The experiments were carried out at normal atmospheric conditions (1 atm pressure and 25\(^0\) C temperatures).

The experimental set-up consists of an end-cap electrode (separation distance, \(z_0=6\)mm) and a ring electrode (diameter, \(r_0=6\)mm) to realise a quadrupole trap such that the electrical potential is given as \(\phi = \Lambda_0(r^2 - 2z^2)\), where, \(\Lambda_0 = \frac{\phi_0}{r_0^2 + 2z_0^2}\) and \(\phi_0=11kV_{pp}\) was the applied potential. The details of the setup can be found in our previous paper (Singh et al. [17]).

A function generator (33220A Function /Arbitrary Waveform Generator, 20 MHz), used to apply the potential of the desired waveform, was connected to a high-voltage amplifier source (Trek, model 5/80, high-voltage power amplifier). The applied peak to peak AC potential in our experiments was 11kV\(_{pp}\) with frequency varying from 0.1kHz to 0.5kHz. In a typical experiment, charged ethylene glycol droplets were generated by an electro-spray (in dripping mode) setup, realised by applying high DC voltage to a stainless steel needle tip. The charged droplet was injected between the electrodes and was suspended using the quadrupolar AC electric field. The field was applied between the end caps and the ring electrodes of the trap where the end caps were connected to the live power supply and the ring was kept grounded. The levitated single droplet was observed using a high-speed CMOS camera (Phantom V 12, Vision Research, USA), which was connected with a stereo zoom microscope (SMZ1000, Nikon Instruments Inc.). The camera was kept inclined at 30\(^0\)–40\(^0\) for visualization of the phenomenon. Nikon halogen light (150 W) was used as a light source.

III. RESULTS AND DISCUSSION

A. Surface oscillations of a charged drop:

In typical levitation experiments the gravitational force associated with the mass of the drop is balanced by imposing an additional DC bias voltage, superimposed on the AC voltage to compensate the weight of the droplet (see ~ ref. Duft et al. [18, 19]). Since the gravity is present in most of the practical applications such as electrospray no additional DC voltage was applied to balance the weight of the droplet in ED levitation. It was observed that in
the presence of gravity, the droplet was levitated at an off-centred location in the downward z-direction (at a distance $z_{shift}$) irrespective of the applied waveform. An off-centred droplet experiences a local uniform field ($E = 4\Lambda_{0}z_{shift}$) along with quadrupole field. The presence of $z_{shift}$ modifies the stress distribution over the surface of the droplet and affects the oscillatory response. It was observed in the experiments that the droplet surface oscillated in the sphere prolate sphere oblate (SPSO) mode. However, the magnitude of the prolate deformation was higher than that of the oblate deformation. This experimental observation is different from the experiments of Duft *et al.* [19] wherein they observed symmetric SPSO mode and reported an equal magnitude of the prolate and oblate deformations in each oscillation cycle. In some experiments moreover, it was observed that the droplet exhibited sphere-prolatesphere (SPS) mode of oscillations. This oscillation behaviour can be attributed to a complex interplay between the electrical parameters such as $Q$, $\Lambda$ and $E$ as well as fluid properties such as viscosity, surface tension and density of the levitated droplet, and is discussed later.

B. Deformation under sinusoidal and other time periodic waveform

When a charged droplet is experimentally levitated in a quadrupole AC electric field with sine waveform as an applied signal the droplet surface oscillates with the applied frequency. However, the surface oscillations can admit higher harmonics due to non linear interaction of charge on the droplet with applied quadrupole field and uniform field. In order to characterize the surface oscillation behaviour, the recorded video is processed using ImageJ software and the surface characteristics are obtained by tracking the change in the outline of the droplet in each frame of the video. The accuracy of the analysis and noise in the data depend upon the quality of the images in each of the frames and the method of thresholding in ImageJ software. The present experimental analysis has an error of around $\sim 10$-$15\%$. The droplet is levitated at a constant AC voltage i.e. $11kV_{pp}$ and different types of waveforms are applied to the trap. The oscillation characteristics are examined by evaluating the rate of change of the Taylor deformation parameter ($DD=\frac{L-B}{L+B}$, where, $L$ is the major axis, $B$ is the minor axis and the $L$ & $B$ were obtained by tracking the boundary of the droplet using ImageJ software) with respect to time. The fundamental frequency and other harmonic frequencies are identified by performing the FFT analysis using the software OriginLab. The software automatically chooses a sample interval $\Delta t$. The triangular type of window is used
to suppress frequency leakage and mean square amplitude method is used for power density normalization. The FFT analysis gives the amplitude of each frequency present in the data. Although the $DD$ data contains around 10-15% error due to the blurriness of the image, the FFT data is very accurate. Hence in the present manuscript, the FFT analysis is used for surface characterization and theoretical and experimental comparison.

Singh et al. [13] reported that a droplet surface oscillates only with the fundamental frequency that is the applied frequency of the electric potential. However, their DD vs time data as seen in experiments (figure 7 in Singh et al. [13] and figure 1b in the present manuscript) indicates the presence of higher harmonic frequencies along with the fundamental frequency. Hence to identify the harmonic frequencies, a sinusoidal signal was applied between the endcap electrode and ring electrode to levitate a sub-Rayleigh charged droplet. The typical applied AC voltage was $11kV_{pp}$ and the corresponding applied frequency is $255Hz$. The applied sinusoidal signal data was acquired with the help of an oscilloscope (Tektronix) and is plotted in figure 1a. The surface oscillations of a charged droplet for the applied sinusoidal signal is plotted in terms of DD vs time as shown in figure 1b. It can be observed from figure 1b that the primary oscillation frequency of the droplet surface is the fundamental applied frequency. However an unmistakable noise is observed at the peaks of the deformation cycle. This noise is an indication of the presence of higher harmonic frequencies. Hence to confirm the presence of harmonic frequencies an FFT analysis is done for both applied waveform and the surface oscillation data. The FFT of the applied signal (Figure 1c) shows that there exists only one peak at $255Hz$ which is the fundamental frequency of the applied signal. The FFT of surface oscillation on the other hand (figure 1d) shows the existence of higher harmonics. The fundamental frequency $f_1=255Hz$, the second harmonic frequency ($f_2$) at $2f_1$ and the third harmonic frequency ($f_3$) at $3f_1$ are clearly seen with diminishing magnitude. While the fundamental and second harmonics frequencies can be attributed to the effect of $\Lambda$ on $Q$ and the $\Lambda^2$ term due to quadrupolar dependence of Maxwell stress on electric potential, the third harmonic could be due to the quadrupole field ($\sim\omega$) acting on the charge induced on the deformed sphere ($\sim 2\omega$) due to the quadrupole field itself. To re-confirm the presence of second and third harmonic frequencies the droplet is levitated at a different frequency, $150Hz$, and the FFT analysis is done for both applied signal and the surface oscillation data. It can be seen from figure 1e that while the applied signal has only the fundamental frequency ($150Hz$) the deformation admits harmonic frequencies.
FIG. 1: Droplet oscillation characteristics in the presence of a sine waveform; a) rate of change of applied voltage for sinusoidal waveform, b) surface oscillation of a droplet levitated in an ED balance c) FFT analysis of sinusoidal applied signal used for levitating the droplet d) FFT analysis of the DD vs time data e) FFT analysis of sinusoidal applied signal at 150Hz f) FFT analysis of DD vs time data for 150 Hz applied frequency g) FFT analysis of DD vs time data for 115 Hz applied frequency.

Hence the non-linearities in the droplet oscillations due to large deformation can lead to the admittance of the $3^{rd}$ harmonic frequency. To confirm the results obtained at a lower applied frequency ($150Hz$) the droplet is levitated at a still lower frequency of $115Hz$ and corresponding FFT analysis of surface oscillations is shown in figure 1g. It can be observed
FIG. 2: Droplet oscillation characteristics in the presence of a square waveform; a) rate of change of voltage for non-sinusoidal square waveform at 220Hz b) the surface oscillations of a droplet levitated in an ED balance in presence of square waveform c) FFT of square waveform applied signal for levitating the droplet d) FFT of degree of deformation vs time data for 220 Hz applied frequency.

that the droplet surface oscillates with several harmonic frequencies along with fundamental frequency (115Hz). Its higher magnitude at 150 Hz as compared to that at 225 Hz is in agreement with the large deformation seen at the lower frequency.

The levitation of a charged droplet in sinusoidal applied potential raises the question of the necessity of non-sinusoidal waveform for levitation. Therefore two types of non-sinusoidal waveform, namely square waveform and ramp waveform, are applied for examining the controlled oscillation characteristics. To examine the case of square waveform a charged droplet is levitated at 11kv_{pp} applied voltage and 220 Hz imposed frequency. The characteristics of an applied signal are shown in figure 2a. The noise can be observed at the peaks of the
applied the signal since the square waveform consists of odd order harmonic frequencies of the fundamental sine frequency. The surface oscillations in the form of DD vs time for the droplet levitated in the presence of square wave is shown in figure 2b. The figure DD vs time data is smoothened by 5 point averaging method to reduce the noise. The noise in the deformation data is a consequence of the noise in the applied signal and the noise associated with image processing. For the preliminary confirmation of surface oscillations of a levitated charged droplet in the presence of a square waveform, a square wave function of the same amplitude and frequency is fitted to the DD vs time data using the software OriginLab. It can be observed that the surface oscillation characteristics of a droplet follow the applied waveform and the droplet oscillates with fundamental applied frequency. Further confirmation of droplet oscillation behaviour is done by performing the FFT analysis of the applied waveform and the surface oscillation data. The FFT analysis of the applied signal is shown in figure 2c and it is observed that the applied signal has several harmonic frequencies in odd multiples of the fundamental frequency ($f_1 = 220\, \text{Hz}$). This response is expected because of the characteristic behaviour of the square waveform. The magnitude of the fundamental frequency is observed to be high as compared to the harmonic frequencies. The corresponding FFT analysis of surface oscillation behaviour is shown in figure 2d and it is found that the FFT analysis of the surface oscillation is similar to that of the applied signal. Although there are only two harmonic frequencies present in the experimental surface oscillation data, the order of occurrence of these harmonic frequencies is similar to the applied signal. Presence of only two harmonic frequencies is because of the lower magnitude of the other harmonic frequencies which may be considered as a noise in the DD data. Unlike the sinusoidal waveform, the droplet surface oscillates with applied frequency in the presence of square waveform and the $2^{nd}$ harmonic is also significantly marked. The dominant $3^{rd}$ harmonic in surface oscillations here is really the response of the droplet to the presence of $3^{rd}$ harmonic in the applied voltage.

Similar to the square waveform another non-sinusoidal waveform, a ramp waveform, is applied to levitate a charged droplet in an ED balance. The voltage applied across the ring and end cap electrode is $11kV_{pp}$ at $205\, \text{Hz}$. The rate of change of applied voltage with time for ramp waveform is shown in Figure 3a. The surface oscillations in the form of DD vs time are shown in figure 3b. To confirm the oscillations follow applied waveform a ramp waveform of equal magnitude and frequency is fitted onto the DD vs time data using the
FIG. 3: Droplet oscillation characteristics in the presence of a ramp waveform; a) rate of change of voltage for non-sinusoidal square waveform at 220Hz b) the surface oscillations of a droplet levitated in an ED balance in presence of square waveform c) FFT of square waveform applied signal for levitating the droplet d) FFT of degree of deformation vs time data for 220 Hz applied frequency.

software OriginLab and a good fit is observed. Figure 3c shows the FFT analysis of the applied ramp waveform. It can be observed that there exist integer harmonic frequencies in the applied signal since the ramp waveform is theoretically an infinite series of sine integer harmonic frequencies [21]. Although the magnitude of the fundamental frequency is greater than that of higher harmonics, the higher harmonics do show significant contribution to the ramp signal. This is evident in figure 3d where the FFT of the deformation shows a significant presence of the fundamental and all higher harmonics. These results conclusively prove that an unmistakable signature of the applied waveform is seen on the deformation dynamics.
C. Theoretical validation of droplet surface oscillation:

The surface oscillation dynamics of a charged droplet levitated in an ED balance is governed by both electrostatics and hydrodynamics. Singh et al. [13] conducted a theoretical analysis of surface oscillations of a levitated ethylene glycol charged droplet in the presence of a sinusoidal applied voltage. In the present work, we extend the theoretical analysis for the case of non-sinusoidal waveforms and compare it with the experimental observations. The details of the model are omitted here, only boundary conditions and governing equations for the surface dynamics are shown for completeness. Since the droplet conductivity is high (>30 μS/cm) and the surrounding medium (air) is a dielectric the droplet is considered as a perfect conductor drop. Thus the non-dimensional equations and the boundary conditions governing the electrostatics of the droplet surface dynamics are:

\[
\nabla^2 \phi = 0, \quad 0 \leq r \leq r_s(\theta, t) \tag{1}
\]

\[
\phi \to \phi_\infty, \quad r \to \infty \tag{2}
\]

\[
\mathbf{n} \cdot \nabla \phi = -q(\theta, t), \quad r = r_s(\theta, t) \tag{3}
\]

\[
t \cdot \nabla \phi = 0, \quad r = r_s(\theta, t) \tag{4}
\]

where, the droplet surface is defined with a small amplitude of perturbation around a spherical shape \(r_s(\theta, t)\) as,

\[
r_s(\theta, t) = [1 + \sum_l \delta\alpha_l(t) P_l\cos(\theta)] \tag{5}
\]

where, \(\delta\) is a small perturbation parameter to measure the oscillation amplitude. The \(l = 1\) mode contributes to the centre of mass translation of the drop and the other modes such as \(l = 2, 3, 4\) indicate the deformation of the drop shape from a sphere.

Typically, an ethylene glycol droplet of diameter 100-250μm with viscosity \(\mu_i = 0.016\)Ns/m^2 and density \(\rho_i = 1097\)Kg/m^3 is levitated in the electrodynamic balance to study the oscillations characteristics of the drop surface in the presence of non-sinusoidal waveform. The surface tension \((\gamma)\) of the droplet is 0.047N/m^2. The typical range of Ohnesorge number \((Oh = \frac{\mu_i}{\sqrt{\gamma\rho_i}})\), which relates the viscous and surface tension forces, for our experimental parameters is about 0.15-0.35. Although the \(Oh < 1\), that indicates that for the given experimental conditions inertial effects can be dominant, the contribution of viscous terms cannot
be neglected. Thus for theoretical analysis of the oscillation behaviour of the drop, viscosity is included through the normal stress balance condition in the potential flow analysis. Also, known as viscous potential flow theory and is governed by the following non-dimensional equations:

\[
\nabla \cdot \mathbf{v}_{i,e} = 0 \quad (6)
\]

\[
\frac{\beta}{Oh^2} \left( \frac{\partial v_{i,e}}{\partial t} + v_{i,e} \cdot \nabla v_{i,e} \right) = -\nabla p_{i,e} + \lambda_{i,e} \nabla^2 v_{i,e} \quad (7)
\]

Where, subscript \( i \) and \( e \) refer to drop and the outside medium respectively, \( \beta = \rho_e / \rho_i \), \( \rho_e \) is the density of the outside fluid and \( \rho_i \) is the density of the drop, \( \lambda = \mu_e / \mu_i \), \( \mu_e \) is the viscosity of the outside medium and \( \mu_i \) is the viscosity of the drop. In the potential flow limit \((Oh \ll 1)\), the dynamics of the irrotational and incompressible fluid having velocity potential \( \psi \) and the drop shape \( r_s(\theta, t) \) is given by the following non dimensional equations,

\[
\nabla^2 \psi_i = 0, \quad 0 \leq r \leq r_s(\theta, t) \quad (8)
\]

\[
\nabla^2 \psi_e = 0, \quad r \geq r_s(\theta, t) \quad (9)
\]

and the pressure is given by the Bernoulli equation,

\[
\Delta p + \left( \frac{\partial \psi_i}{\partial t} + \frac{1}{2} \left( \frac{\partial \psi_i^2}{\partial r} + \frac{1}{r} \frac{\partial \psi_i}{\partial \theta} \right) \right) - \beta \left( \frac{\partial \psi_e}{\partial t} + \frac{1}{2} \left( \frac{\partial \psi_e^2}{\partial r} + \frac{1}{r} \frac{\partial \psi_e}{\partial \theta} \right) \right) = 0 \quad (10)
\]

The boundary conditions are continuity of normal velocity

\[
\frac{\partial \psi_i}{\partial r} = \frac{\partial \psi_e}{\partial r} \quad (11)
\]

and the normal stress balance.

\[
\Delta p - 2(Oh \frac{\partial v_{r,i}}{\partial r} + \lambda_e Oh \frac{\partial v_{r,e}}{\partial r}) = \kappa - \frac{1}{32} \left( 8X + 12 \sqrt{Ca} \zeta \cos(\theta) + 5 \sqrt{CaQ} \zeta (3 \cos(2\theta) + 1) \right)^2 \quad (12)
\]

at the interface \( r = r_s(\theta, t) \). The equation \( 12 \) is the non dimensional stress balance equation and \( \kappa \) is the surface curvature and defined as:

\[
\kappa = 2 + \delta \sum_{l=2,3,4} \left( l(l + 1) - 2 \right) \alpha_l P_l(\cos \theta) \quad (13)
\]
The length is scaled by unperturbed droplet radius \( R = D_p/2 \), velocity by \( \sqrt{\gamma/\rho R} \), time by \( \sqrt{\rho R^3/\gamma} \), \( \zeta \) is a time periodic function which depends on the type of applied waveform e.g. \( \zeta = \sin \omega t \) for the sinusoidal applied waveform. \( X \) is the fissility which is the ratio of the actual charge on the droplet to its Rayleigh charge \( Q_R = \sqrt{64\pi^2\epsilon_a R^3\gamma} \), where \( \epsilon_a \) is the electrical permittivity of the surrounding fluid. \( Ca_Q = (R^3\epsilon_a\Lambda_0^2/\gamma) \) is the capillary number based on the strength of the applied quadrupole field and \( Ca = (R\epsilon_a E^2/\gamma) \) is the electrical capillary number based on the uniform field, where \( E = 4\Lambda_0 z_{shift} \). These equations are solved by the expansion of the velocity and electric potentials into spherical harmonics to yield:

\[
\alpha''_1(t) + \frac{1}{2\beta+1} \left( 12\text{Oh} \alpha'_1(t) - 12(\sqrt{Ca}\sqrt{Ca_Q} \zeta^2 + X\sqrt{Ca} \zeta) \right) = 0 \quad (14)
\]

\[
\alpha''_2(t) + \frac{6}{3\beta+2} \left( 2(\lambda + 4)\text{Oh} \alpha'_2(t) + 4\alpha_2(t) - 3Ca \zeta^2 - 10X\sqrt{Ca_Q} \zeta - \frac{25Ca_Q \zeta^2}{7} \right) = 0 \quad (15)
\]

\[
\alpha''_3(t) + \frac{1}{4\beta+3} \left( 24(2\lambda + 5)\text{Oh} \alpha'_3(t) + 120\alpha_3(t) - 108\sqrt{Ca}\sqrt{Ca_Q} \zeta^2 \right) = 0 \quad (16)
\]

\[
\alpha''_4(t) + \frac{1}{5\beta+4} \left( 840(\lambda + 2)\text{Oh} \alpha'_4(t) + 2520\alpha_4(t) - 900Ca_Q \zeta^2 \right) = 0 \quad (17)
\]

Equation (14) is the based on the \( P_1 \) Legendre mode and primarily contributes in the translational motion of the droplet. The equation (15) is based on the \( P_2 \) Legendre mode and similarly, equation (16) and (17) are obtained as coefficients of the \( P_3 \) and \( P_4 \) Legendre modes. Since the focus of the study is the surface dynamics, the centre of mass motion, which is the translation motion of the drop, is neglected. Hence only equation (15), (16), (17) plays an important role in the surface oscillation dynamics. From equation (15) it can be observed that for a highly charged drop the value of \( X\sqrt{Ca_Q} \) term dominates over other terms and the droplet oscillates with the fundamental frequency. On the other hand for mildly or uncharged droplets levitated at the centre of the trap the term \( Ca_Q \zeta^2 \) dominates over other terms, and the droplet oscillates with twice the frequency of fundamental applied frequency. Hence, a lower oblate deformation as compared to the corresponding prolate deformation during the negative and positive cycle of end cap potential in the SPSO mode of surface oscillation of a positively charged drop levitated in a quadrupole field depends upon the relative magnitude of \( Ca\zeta^2 \), \( X\sqrt{Ca_Q} \zeta \), \( Ca_Q \zeta^2 \). The equation (16) shows that at
very high value of $z_{shift}$ from the centre of the trap, the droplet shape can become highly asymmetric. The equation 17 show that at high value of applied field the droplet oscillates with twice the imposed frequency. The relative magnitudes of the shape coefficients $\alpha_2$, $\alpha_3$, $\alpha_4$ as determined by equations 15, 16 and 17 finally determines the characteristics of the droplet oscillations. For our experimental parameters $\alpha_2$ found to dominate the all droplet shape. Thus, the dynamics of a charged droplet oscillation in the presence of sinusoidal and non-sinusoidal waveform is explained using equation 15.

![FFT of theoretically obtained and experimentally observed surface dynamics for a sinusoidal waveform.](image)

**FIG. 4:** FFT of theoretically obtained and experimentally observed surface dynamics for a sinusoidal waveform. The typical parameters used for theory is given in the inset of the figure. The magnitude of theoretical FFT is scaled by the factor of 10.

Since the detailed experimental validation of surface oscillations in terms of degree of deformation with time for a sine waveform was reported by us recently[13], in this study, we have experimentally and theoretically validated the frequency response of surface dynamics in term of FFT analysis of surface oscillations, as shown in figure 4. In order to obtain the theoretical oscillations characteristics for applied waveform, equation 15 is solved for the experimental parameters. Since the oscillation are recorded before the droplet breakup, the charge is considered as sub-Rayleigh ($X=0.9$) and the $z_{shift}$ of the droplet is kept as a fitting
FIG. 5: FFT of theoretically obtained and experimentally observed surface dynamics for a square waveform. The typical parameters used for theory is given in the inset of the figure.

The magnitude of theoretical FFT is scaled by the factor of 5.

Parameter. The $DD$ value from the theory is obtained by using the following expression,

$$D = \frac{3\alpha_2}{4} + \frac{\alpha_3}{2} + \frac{5\alpha_4}{16}$$  \hspace{1cm} (18)

Similar to the experimental analysis, the embedded harmonic and higher harmonics are resolved by performing the FFT analysis of theoretically obtained surface oscillation data with the help of the software OriginLab. It can be observed from figure 4 that there exists harmonic frequencies (in experiments) for fundamental applied frequency ($f_1 = 260$ Hz).

The first two harmonics are expected because of significant magnitude of coefficient of $\zeta^2 = \cos(\omega t)^2$ due to off-centre position of the droplet, in addition to $\zeta = \cos(\omega t)$ due to charge on the droplet (term $X\sqrt{C_aQ}$ in equation 15). The resultant droplet oscillation dynamics contains both the frequencies. Similar observations are made in the theoretical oscillations, as shown in figure 4. However, it can be also observed in the figure 4 that the experimental FFT exhibit $3f_1$ and $4f_1$. The presence of these higher order harmonic frequencies suggests non-linear interaction between the charges on the deformed droplet and the quadrupole field acting on these charges. Since the theory presented here is based on linear order approximation it captures only the lower order frequencies i.e. $\omega$ and $2\omega$. 

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FIG. 6: FFT of theoretically obtained and experimentally observed surface dynamics for a ramp waveform. The typical parameters used for theory is given in the inset of the figure. The magnitude of theoretical FFT is scaled by the factor of 5.

The analysis is then extended to surface dynamics for square and ramp waveforms. In the case of a square waveform, the time varying function $\zeta$ can be given as:

$$\zeta = 2 \tan^{-1} \left( \frac{2\pi ft}{\delta} \right), \quad (19)$$

where, $\delta (= 0.01)$ is a small parameter and used for smoothing the square waveform. If the value of $\delta$ is high the curve will be smooth at the corners. Figure 5 shows the FFT analysis of theoretical and experimental surface oscillations. It can be observed that fundamental frequency and harmonic frequencies occur at the same order. Along with harmonic frequencies, other inter-harmonic frequencies can be observed in figure 5. The presence of inter-harmonic frequencies shows the complex coupling between various terms discussed in the previous section. Additionally, it is also observed that if the magnitude of $Ca_A$ and $Ca_E$ terms is higher the FFT of theoretical surface oscillations contain even multipliers (i.e. $2f_1$, $4f_1$, $6f_1$ etc.) of the fundamental frequencies. Similar to the case of a square waveform the case of a ramp waveform is examined theoretically for $\zeta$, given as:

$$\zeta = \left( \frac{t - \tau t_c \left( \frac{e}{\pi} \right)}{\frac{\pi}{2}} \right) - \frac{\tau}{2}, \quad (20)$$
Where, $\tau = 1/f$, $f$ is the applied frequency, $t_c$ is a defined function which accept only integer part. Since all the equations are solved in the Mathematica (version 10) the function can be defined as, $t_c[t]=\text{IntegerPart}[t]$. The theoretical validation of surface oscillations in the presence of ramp waveform is done by performing the FFT analysis of theoretical surface oscillations, as shown in figure 6. Unlike the case of square waveform, no inter-harmonic frequencies are observed in case of ramp waveform. This is because, while in the case of a square waveform, the harmonic frequencies occur in the multiple of an odd number of the fundamental frequency, the interactions of various terms can generate even order inter-harmonic frequencies along with with odd order harmonic frequencies. On the other hand, in the case of ramp waveform, all integer harmonic frequencies are present in the applied signal itself. Like the previous case the theoretically obtained FFT, as shown in figure 6, is in fair agreement with the experimental observations.

D. Droplet breakup characteristics

![Image of breakup of levitated charged droplet in the presence of different waveform]

FIG. 7: Breakup of levitated charged droplet in the presence of different waveform a) sine waveform b) square waveform c) ramp waveform.

To the best of our knowledge, there exist only two experimental evidence of breakup of
charged droplet levitated in electrodynamic balance\cite{18, 22}. In both the studies the droplet is levitated in the presence of an applied potential having sine waveform. In this work, for the first time, we report the droplet breakup in the presence of non-sinusoidal waveform. The charged droplet is levitated in the presence of sine, square and ramp waveform in the quadrupole trap as discussed earlier. Unlike the surface oscillations of a sub-Rayleigh charged drop discussed in this work, for the breakup studies, we let the droplet evaporate and build a Rayleigh critical charge at which droplet undergoes breakup. The droplet breakup is recorded at 1 hundred thousand fps. The experimental observations of the droplet breakup in the presence of various waveform is shown in figure 7, where it can be observed that droplet breaks in the upward direction and in an asymmetric manner. Since the droplet is levitated in the presence of AC field without any superimposed DC voltage to balance the mass of the drop, the droplet levitates away from the geometric centre of the trap. At this off-centred location, it experiences asymmetric electrical stress which causes an asymmetric breakup. It is also observed that the droplet breaks in the upward direction due to the high initial $P_2$ perturbation and higher curvature (+ve $P_3$) in the upward direction. The magnitude of asymmetry and droplet DD depends upon the $z_{shift}$ and $\Lambda$. As reported by Singh \textit{et al.} \cite{22}, the droplet breaks with more asymmetry at a higher value of $z_{shift}$ even for a fixed value of $\Lambda$. The thickness of the jet depends on the droplet diameter (or $z_{shift}$) and the applied voltage. The detailed explanations of such droplet breakup characteristics are discussed by Singh \textit{et al.} \cite{22}. The important part to note here is that there is no significant difference in the breakup mode in the presence of different waveforms.

IV. CONCLUSIONS:

The surface oscillation characteristics are investigated by performing FFT analysis and it is found that the FFT is an appropriate characterizing tool for identification of the presence of different frequencies in the droplet deformation data. The surface oscillation behaviour in the presence of sinusoidal and non-sinusoidal waveform is successfully demonstrated and found that droplet oscillates with the driving field frequency. In the case of a sine waveform, the droplet oscillation is found to admit greater nonlinearity at lower applied frequency. The surface oscillation is compared with the visco-potential flow theory and a fair agreement is found. The droplet breakup characteristics in the presence of sinusoidal and non-sinusoidal
waveform are shown and it is observed that the application of a different waveform does not alter the breakup characteristics. The extension of linear stability analysis to higher-order analysis can make the system suitable for a contact-free and accurate surface tension measurement device. The work demonstrates that the mechanism of droplet oscillations and breakup is robust and is admitted as long as there is a time periodic driving potential applied to the trap.

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