Translation–deformation coupling effects on the Rayleigh instability of an electrodynamically levitated charged droplet

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Abstract The breakup pathway of the Rayleigh fission process observed in the past experiments carried out using high-speed imaging of a charged drop levitated in an AC quadrupole trap has shown to exhibit several cycles of shape and center-of-mass oscillations followed by asymmetric breakup by ejecting a jet in the upward direction (i.e., opposite to the direction of gravity). We recently attempted to explain this using boundary integral simulations in the Stokes flow limit, wherein the position of the droplet and the polarity of the end cap electrodes were assigned using physical arguments, and the center-of-mass motion was not estimated consistently invoking quasi-static conditions. In this work, we explain the experimental observation of upward breakup of charged droplets in a quadrupolar field, using numerical calculations based on the boundary element method considering inviscid droplets levitated electrodynamically using quadrupole electric fields. The center-of-mass motion and the end cap are consistently calculated in the numerical scheme. The simulations show that the gravity-induced downward shift in the equilibrium position of the drop in the trap causes significant, large-amplitude shape oscillations superimposed over the center-of-mass oscillations of the drop. An important observation here is that the shape oscillations due to the applied quadrupole fields result in sufficient deformations that act as triggers for the onset of the instability below the Rayleigh limit, thereby admitting a subcritical instability. The center-of-mass oscillations of the droplet within the trap, which follow the applied frequency, are out of phase with the applied AC signal. Thus the combined effect of shape deformations and dynamic position of the drop leads to an asymmetric breakup such that the Rayleigh fission occurs upward via the ejection of a jet at the north pole of the deformed drop.

1 Introduction

An isolated charged droplet is known to become unstable when the repulsive Coulombic force, on account of its charge, overcomes the stabilizing surface tension force of the droplet. This instability is popularly known as the Rayleigh instability of a charged droplet and the critical value of charge at which the instability sets in is given by the expression, 

\[ Q_R = 8\pi \sqrt{\gamma \epsilon_0 a^3} \]

where \( a \) is the radius of the drop, \( \epsilon_0 \) is the permittivity of the external medium (air) and \( \gamma \) represents the interfacial tension \[ 1\]. Thus when the charge on the droplet increases beyond \( Q_R \), it cannot sustain its stable spherical shape and exhibits deformation with time, ultimately leading to its breakup. To demonstrate the Rayleigh instability of a charged droplet and its subsequent deformation pathway with time, experiments were carried out by electrodynamically levitating a micron-sized charged droplet in a quadrupole trap \[ 2,3\]. In these experiments, a droplet is levitated perfectly at the center of the quadrupole field by using superimposed DC voltage on the AC field to balance the gravitational force acting on the droplet. Thus a critically charged droplet exhibits a sequential deformation from the original spherical shape to an elongated prolate spheroid, eventually forming symmetric conical tips from which two jets are ejected out in the opposite directions \[ 3,4\]. This symmetric breakup, observed in experiments \[ 2,3\], is well predicted by numerical calculations \[ 4–7\].

The symmetrical jet ejection from the droplet perfectly levitated at the center of the trap may not correspond to practical situations such as in electrosprays, wherein unbalanced external forces such as gravity or external electric field introduce asymmetry in the drop shape due to different local hydrostatic pressure and asymmetric charge accumulation due to non-uniform fields \[ 8\]. Such broken symmetry can not only impact the pathway of drop deformation and breakup but can also modify the critical limit of charge at which the drop is rendered unstable. Application point of view, asymmetry in drop shape can directly alter the size distri-
culine simulations in the inertial timescales to accurately model a system to study and predict the breakup of the levitated charged droplet.

In experiments, it is observed that, in the absence of DC-biased voltage, a levitated charged droplet undergoes upward asymmetric breakup [18]. Thus detailed experiments were carried out to demonstrate the effect of strength of the applied quadrupole field and conductivity of the droplet on the breakup mechanism of the levitated charged droplet as well as on the characteristics of the jet ejected [18]. This work attempted to validate the experimental observations numerically using simulations based on the boundary element method (BEM) in the Stokes flow limit. Since the drop breakup completes in 1/4th of the AC cycle, the equilibrium position of the droplet in the quadrupole trap (in terms of \( z_{\text{shift}} \), where \( z_{\text{shift}} \) represents the distance of the droplet center from the center of the quadrupole field) was obtained by comparing experimental observation with the solution of the modified Mathieu equation, and the BEM simulations were carried out at this position, with a perturbed drop shape as an initial condition (the perturbation amplitude is obtained by fitting the experimental shape with the Legendre polynomials), after which the drop continuously deforms leading to breakup. The experimental observation of the evolution of aspect ratio of the droplet indicating asymmetric deformation was compared with the simulations to show the effect of \( z_{\text{shift}} \) and that of the quadrupolar field on the upward breakup of the charged droplet [18]. Here the upward breakup refers to the ejection of a single jet from the north pole of the droplet.

Recently, the BEM simulations in Stokes flow limit were extended to unequivocally demonstrate the subcritical asymmetric breakup of a levitated charged droplet [19]. The goal was to explain the physics of the subcritical asymmetric breakup observed in most experiments. Here the BEM simulations were carried out similarly at the shifted position with an initially perturbed shape obtained from the experiments; however, the charge was kept as a fitting parameter in the simulations and was determined through systematic progression–regression methodology [19]. In both these works, the droplet COM position and shape oscillations observed in the experiments could not be simulated since in the Stokes flow limit, quasi-static condition is assumed, and the inertial forces are neglected. However, the comparison of the deformation dynamics showed that the directionality of the asymmetric breakup of the charged droplet is critically dependent on the position of the drop in the trap and the magnitude of perturbation attained by the drop at the end of the oscillation phase [19]. Thus to accurately capture the upward asymmetric breakup of the levitated charged droplet, it is required to consider the translational motion of a droplet in the presence of an applied quadrupole field and its effect on the simultaneous development of the shape deformation that leads to a breakup.

In the present work, we explain the typical experimental observation of upward breakup of a charged droplet levitated electrodynamically in a quadrupolar field, using numerical calculations based on the
potential flow boundary integral formulation. Here the COM motion of the droplet in the quadrupole field is consistently calculated in the numerical scheme along with the simultaneous drop shape evolution leading to breakup. Thus the limitations of our previous works [19], namely ascribing the position of the droplet in the trap with an initially perturbed shape and conducting a quasi-static calculation in the Stokes limit, are overcome, and the effect of translational motion of the droplet on the breakup is demonstrated.

2 Problem formulation

The system considered here is of a liquid droplet levitated in the air, and the simulation setup used in this study is shown in Fig. 1. As the density of air ($\rho_a = 1.2$ kg/m$^3$) is three orders of magnitude lower than the ethylene glycol drop ($\rho_i = 1097$ kg/m$^3$), we have restricted our analysis to a liquid droplet suspended in the dynamically inactive external medium. Thus the boundary integral equation for velocity potential inside the drop is solved to study the oscillation dynamics of the drop. Here we use the integral formulation based on the generalized vortex method, developed by Baker et al. (1982) [20] and applied by Lundgren and Mansour (1988) [21] for an inviscid liquid drop in vacuum or gas with negligible density.

Since the electrical conductivity of ethylene glycol drop used in the experiments is high [19], the relative charge relaxation timescale ($\tau_c = \epsilon_i/\sigma_i$, where $\epsilon_i$ and $\sigma_i$ represent the permittivity and conductivity of the drop, respectively) is much smaller than the capillary timescale ($\tau_c = \sqrt{\rho_i a^3/\gamma}$). For typical experimental parameters, the ratio of two timescales, known as Saville number, $Sa = \tau_c/\tau_c \sim O(10^{-4})$. Thus in this study, the surface charge dynamics are neglected, and the drop is assumed to be a perfectly conducting drop with a net electric charge $Q$ initially distributed uniformly on the drop surface and the drop is suspended in a perfectly dielectric surrounding medium with a permittivity, $\epsilon_e$. The equations can be suitably non-dimensionalized by characteristic scales as; the time by the inertial timescale $\tau_i$; the pressure by $\gamma/a$; while the charge and electric fields are scaled by $\sqrt{\rho_i a^3/\gamma}$, respectively, such that the non-dimensional Rayleigh charge is $Q = 8\pi$.

The non-dimensional electric potential of the drop is denoted by $\phi$ and is assumed to follow, $\nabla^2 \phi = 0$ and the electric field is thus expressed as $E = -\nabla \phi$. The flow is assumed to be incompressible and irrotational inside the drop; thus, the velocity is given by the gradient of the velocity potential $v = \nabla \psi$ such that velocity potential follows the Laplace equation, $\nabla^2 \psi = 0$.

For the case of perfectly conducting charged drop in AC quadrupole electric fields, the integral equation for the electric potential is given by,

$$\phi(r_s) = \phi_\infty (r_s) + \int G(r_s, r) E_n dA(r)$$

where, $G(r_s, r) = 1/4\pi(r - r_s)$ and $r$ and $r_s$ are the position vectors on the surface of the drop while $\phi_\infty$ is the applied electric potential which can be written as,

$$\phi_\infty (\rho, z) = \sqrt{Ca_A \zeta(t)}[(z - z_{\text{shift}})^2 - 0.5 \rho^2]$$

where, $Ca_A = \epsilon_e a^3 A_0/\gamma$ is the non-dimensional intensity of quadrupole field of strength $A_0$ and $\epsilon_e$ is the permittivity of the external medium while $\zeta(t)$ is a time varying function with frequency $\omega = 2\pi f$. Here, $(z - z_{\text{shift}})$ accounts for the shifted position of the droplet from the center of the quadrupole trap. The unknown potential $\phi(r_s)$ is constant on the surface of the droplet and is determined by the condition of conservation of charge,

$$\int E_n(r)dA(r) = Q$$

where $E_n$ is the outward directed normal electric field acting on the drop surface.

According to classical potential flow theory the velocity potential ($\psi$) of an irrotational flow can be expressed as a surface distribution of dipole density per unit area, $\vartheta(r_s)$ at the source point $r_s$ [20]. Following [21], the regularized integral equations for velocity potentials can be written as,

$$\psi(r_s) = \vartheta(r_s) + \int \left[\vartheta(r) - \vartheta(r_s)\right](n \cdot \nabla G(r_s, r)) dA(r)$$

$$A(r_s) = -\int \left[\vartheta(r) - \vartheta(r_s)\right](n \times \nabla_s G(r_s, r)) dA(r)$$
The detailed expansions of \((\mathbf{n} \cdot \nabla G(\mathbf{r}, \mathbf{r}))\) and \((\mathbf{n} \times \nabla_s G(\mathbf{r}, \mathbf{r}))\) are given in our previous work [22]. Once the surface velocity is known, the velocity potential is updated using non-dimensional unsteady Bernoulli equation,

\[
\frac{D\psi}{Dt} = \frac{1}{2} \mathbf{v} \cdot \mathbf{v} - \kappa - Bo z + \frac{1}{2} E_n^2
\]  

(5)

where \(Bo = \rho g a^2/\gamma\) is the gravitational bond number and if \(\mathbf{n}\) is the outward unit normal \(\nabla_s \cdot \mathbf{n}\) gives the curvature of the drop denoted by \(\kappa\). Here, \(\nabla_s = (1 - \mathbf{m}) \cdot \nabla\) denotes the surface gradient operator. It should be noted here that, although the COM motion is considered here with the origin of the coordinate axes at the center of the drop, the added energy term (per unit volume) due to moving frame of reference \((\rho \mathbf{U} \cdot \mathbf{v})\) can be neglected in comparison with the kinetic energy term \(1/2 \rho (\mathbf{v} \cdot \mathbf{v})\), since the COM velocity \(\mathbf{U}\) is much smaller as compared to the surface velocity \(\mathbf{v}\) [23]. This can be seen in the data presented in the appendix for typical parameters. The simulations are carried out in a cylindrical coordinate system by considering azimuthal symmetry across z-axis, and thereby, half contour of the drop is discretized into 150 number of elements. The simulation time step is kept constant at 0.0005, and both spatial and temporal discretization is decided based on the numerical tests carried out for the converged results. It should be noted that in this work, the simulations are carried for typical experimental parameters [19] to elucidate the mechanism of an asymmetric upward breakup which can be extended for different sized droplets levitated at different frequencies of an applied quadrupole field. However, practically when one changes the size of the droplet, the other parameters such as intensity and frequency of the quadrupole field also change according to the Mathieu stability criteria [18]. This gives a very narrow range for the non-dimensional simulation parameters to vary, making it unreasonable to carry out a parametric study for this problem.

3 Results and discussion

In the experiments, a charged droplet is observed to oscillate with the applied frequency before the instability sets in due to a high surface charge near its Rayleigh limit. The positional stability or the dynamical parameters of COM of an un-deformed spherical droplet in the quadrupole field is described by the solution of modified Mathieu equation which can be written in current non-dimensional parameters as,

\[
z''(t) + c_z z'(t) - a_z z(t) \cos(\omega t) + Bo = 0,
\]  

(6)

where, \(a_z = (2QA_0 \tau^2)/m\), \(c_z = \omega \mu_m \alpha_r\) and \(\omega = 2\pi f\) with \(f = \frac{f_e}{c}\) as the non-dimensional applied frequency. Here, \(m\) denotes the mass of the drop and \(\mu_m\) is the viscosity of the external medium (air). Since the BEM simulations presented here are carried out in the potential flow limit, without the inclusion of viscosity, the COM motion of the charged droplet exhibits conserved oscillations consisting of both applied and secular frequencies. In experiments reported in our previous work [19], it is observed that due to the presence of viscosity, natural oscillations of the drop in COM motion damp out quickly, and the drop oscillates with the applied frequency. This observation is also confirmed with the solution of Eq. 6 which indicates that due to the presence of viscous drag (with damping coefficient \(c_z\)), the secular oscillations are damped out, and the droplet steadily oscillates with the applied frequency (with time period \(t_1 = 1/\omega = 40\)) at an equilibrium (Fig. 2) mean position below the center of the trap. We consider the maximum amplitude of COM oscillations as an equilibrium shifted position of the droplet from the center of the quadrupole field and term it as \(z_{shift}\). Such oscillations are not observed if the gravitational force is balanced by some other external force (such as a DC force as used in the experiments [3]). For the typical experimental parameters (borrowed from [19]), the solution of Eq. 6 indicate that the damping of secular oscillations occurs over a non-dimensional timescale, \(t_2 = 1/(c_z \omega) \approx 7000\), to attain steady equilibrium oscillations. The three timescales in the COM oscillations are indicated in Fig. 2. It should be noted here that these results are obtained for linearized Eq. 6, where droplet is moving as a rigid body under the influence of gravity and time varying external force. However, in BEM simulations, the nonlinear coupling between the shape and the center of mass of the droplet is captured. As the inertial timescale is used in BEM simulations to capture the natural oscillations of the drop surface, a large number of calcula-
tion steps are required to obtain the steady-state COM oscillations with the viscous corrections included in the potential flow formalism (as used in the recent work [16]). This results in a tremendous increase in the computational time. To overcome this limitation, the COM oscillations obtained from potential BEM are compared with the solution of the modified Mathieu equation (Eq. 6), and the two solutions are found to overlap with each other (validation can be found in [22]). Taking advantage of this result, to nullify the effect of secular frequency and remove the initial transience in COM oscillations, we obtained the equilibrium position (maximum $z_{\text{shift}}$ value) of the droplet from the solution of Eq. 6. It should be noted that the time-averaged equilibrium position of the droplet, $\langle z_{\text{shift}} \rangle$, can also be obtained from the simple force balance in the $z$ direction. A detailed derivation for the determination of time averaged equilibrium position of the droplet with its corresponding amplitude is discussed in detail in our previous work [19]. The BEM simulations are then carried out with the maximum shifted position, $z_{\text{shift}, t}$, (near the south end-cap) of the droplet as an initial condition. For numerical consistency, instead of shifting the position of the droplet from the center of the coordinate system, the center of the quadrupole field is shifted in the upward direction by $z_{\text{shift}}$. With this strategy, COM oscillations of the droplet at steady state are obtained, which follows applied frequency and exhibits a $\pi$ phase shift with the applied AC cycle (shown in Fig. 3b(i)).

The parameters used in the simulations are borrowed from the experiments with charge as a fitting parameter. The simulations are carried out at various charge values, and the charge at which the droplet loses its stability is considered the critical charge for the given parameters. For typical experimental parameters of $Ca_A = 0.0006$, $f = 0.025$ and $Bo = 0.0038$, the droplet breaks at $Q = 7.7\pi$ in the upward direction, which is 96.25% of the Rayleigh limit. The sequence of drop deformation in one oscillation cycle leading to breakup is presented in Fig. 3a. The figure also shows the temporal variation in the relative position of the droplet, indicating COM oscillations similar to that observed in the experiments [19]. Figure 3b(ii) shows the degree of deformation as a function of non-dimensional time for $Q = 7.7\pi$, indicating that the droplet undergoes few cycles of oscillations, developing finite-amplitude perturbation with time that leads to subcritical instability and eventually breaks in the positive AC cycle at $t = 316$ non-dimensional time. It should be noted that the droplet is at the position near the south end cap (at a position $= -5.2$ in Fig. 3b(i)) at $t = 0$ when the end caps are at a positive potential and is near the center of the quadrupole trap when the end caps are negative due to $\pi$ phase shift. As the droplet undergoes COM and shape oscillations, the amplitude of the degree of deformation is seen to increase. This increase is due to a coupling of charge distribution and the applied quadrupolar AC field. It can be clearly observed in Fig. 3b(ii) that the critical perturbation (amplitude of degree of deformation) is attained at the negative peak of the AC cycle (at about $t = 500$) when the drop position is near the center of the quadrupole field. At this point, the droplet ceases to respond to the applied field, and the further deformation is dominated by charge distribution on the deformed drop. At this point, it appears that the droplet would have undergone a symmetric subcritical, final amplitude instability with this shape perturbation. However, while the deformation continues to increase, the drop starts shifting toward the south end cap (Fig. 3b(i)), in a positive cycle of the AC field. Hence, a positively charged drop near the south end cap experiences higher repulsion at the south pole of the drop from the end cap at a positive potential causing higher charge migration toward the north pole of the already deformed drop, and the drop breaks in the upward direction.

In order to understand the effect of shape mode coupling on the breakup behavior, the drop shape given by, $r_s(\theta, t) = 1 + \alpha_1(t)P_1(\cos \theta)$ is decomposed into its linear modes (defined in terms of Legendre polynomials, $P_l$) and the perturbation coefficients ($\alpha_l(t)$) of various modes are obtained using orthogonality condition of Legendre polynomials [21] which gives,

$$\alpha_l(t) = \left(1 + \frac{1}{2}\right)\int_0^\pi (r_s(\theta, t) - 1)P_l(\cos \theta)d\theta \quad (7)$$

The amplitudes of three linear modes ($P_2$, $P_3$, and $P_4$) as a function of time are shown in Fig. 4a. The amplitude of the second-mode $\alpha_2$ is observed to be significantly greater than $\alpha_3$ and $\alpha_4$. This reconfirms that the $P_2$ mode is dominant in the deformation of a charged drop. Although the amplitude of third-mode ($\alpha_3$) is an order of magnitude smaller than $\alpha_2$, it increases as the drop approaches breakup, causing an asymmetric deformation. These results also validate the late onset of asymmetry observed in experiments and numerical analysis in the Stokes flow limit [19]. The positive value of $\alpha_3$ indicates higher curvature at the north pole and is considered to be responsible for the upward breakup of the drop.

Further, to understand the dynamics of the process, we analyzed the charge density distribution on the drop surface at times that correspond to maximum prolate deformation in the deformation curve. The charge density as a function of arc length is shown in Fig. 4b for various times. Figure 4b indicates that initially, when the droplet is undergoing oscillations with small deformation (arc length ($s$) is almost constant), the charge density at the south pole is marginally higher as compared to the north pole of the drop. This is mainly due to the fact that the positively charged droplet is shifted from the center in the downward direction and thus experiences greater attraction from the south end cap in the negative AC cycle, resulting in greater accumulation of charges at the south pole of the drop. However, as the drop starts deforming (arc length of the drop increases) with time, the charge density distribution becomes symmetric about the equator of the drop with equal charge densities at the north and the south poles. Further, when the drop attains a critical deformation,
Fig. 3  a Evolution of drop shapes in one cycle of oscillations before the breakup indicating COM motion as well as progressive surface deformation leading to upward breakup. Parameters used are: $Ca_{\Lambda} = 0.0006$, $f = 0.025$ and $Bo = 0.0038$ and $Q = 7.7\pi$. b(i) Comparison of COM oscillations obtained from BEM and solution of equation 6, (ii) Degree of deformation (DD) as a function of time with the corresponding applied AC field ($\cos(\omega t)$). Note that the amplitude of applied AC signal is scaled for better visualization. The intersection point of vertical and horizontal dash-dotted black line indicates the critical point. c Time evolution of the total charge on the north ($Q_N$) and south ($Q_S$) hemisphere of the droplet.
Fig. 4  a Temporal evolution of amplitudes of $P_2$, $P_3$ and $P_4$ shape modes obtained by decomposing the drop shapes with charge, $Q = 7.7\pi$. Note that the simulations are initiated with spherical drop shape. b Surface charge density as a function of arc length of the drop for various times when the droplet is near the center of the quadrupole trap (i.e., at the time when DD of the drop is maximum observed in the negative cycle of the applied field).

the shape coupling with charge distribution becomes dominant, and the charge density is found to increase at the north pole (Fig. 4b(a)) due to higher repulsion from the south end cap. This leads to the accumulation of charges in the north pole, which in turn causes the droplet to break in the upward direction. This can also be seen in Fig. 3c, where the time evolution of total charge on the north hemisphere ($Q_N$) is observed to blow up while the total charge in the south hemisphere ($Q_S$) reduces suddenly near the breakup. It is also observed that $Q_N$ is always higher than $Q_S$ due to higher repulsion from the south end cap.

At a lower value of charge, $Q < 7.7\pi (7.65\pi)$, the drop exhibits stable oscillations for a long time without showing any signature of increase in the degree of deformation. (Simulations are carried out till $t = 250$ as shown in Fig. 5a.) This shows that, at $Q = 7.65\pi$, the drop cannot attain sufficient deformation to become Coulombially unstable, and thus it continues to respond to the applied AC field and exhibit stable, sustained oscillations. On the other hand, for $Q > 7.7\pi (7.72\pi)$, the drop breaks in the downward direction (Fig. 5b). This indicates that a critical deformation is achieved at the negative peak potential of end caps, which is sufficient to induce Rayleigh instability in the drop. It should be noted that, as the droplet considered is positively charged, it attains the highest prolate perturbation when the end caps have negative polarity. In this situation, due to the presence of a high surface charge, the south pole of the drop experiences higher attraction, and the droplet breaks in the downward direction before the AC cycle (and thereby the polarity of the end cap) changes (Fig. 5b). The summary of the experimental observations of the asymmetric breakup of a levitated charged droplet [19] is explained using BEM calculations using Fig. 6. Figure 6(a) indicates that at a lower charge, the droplet oscillates without breakup, exhibiting stable shape oscillations responding to the applied AC field. As time progresses, the total charge density on the drop increases due to evaporation, and it undergoes large-amplitude deformations. As soon as the value of charge reaches a critical value (which is less than the Rayleigh limit), an instability sets in, leading to droplet breakup, clearly demonstrating the subcriticality of the instability (Fig. 6(b)). At this stage, if the droplet is positively charged and attains the critical deformation in the positive cycle of the applied AC field, the asymmetry is rendered in such a way that
the droplet breaks in the upward direction. However, if the critical conditions with respect to deformation and charge on the droplet are attained when the droplet is near the end cap whose polarity is still negative, the symmetric subcritical instability is rendered asymmetric with downward direction of breakup (Fig. 6(c)).

4 Conclusions

Detailed numerical simulations that simultaneously describe the electrodynamic levitation, deformation and breakup of a charged drop in a quadrupole field are presented for the first time. The BEM simulations not only capture several cycles of coupled COM motion and shape oscillations of a levitated charged drop before the breakup but also show the simultaneous build-up of critical perturbation that is observed to be responsible for the subcritical asymmetric Rayleigh breakup. This confirms that there exists a strong coupling in the dynamics of COM and the shape of the levitated charged drops. The extended numerical calculations show that the asymmetry in drop shape at the onset of breakup increases with the strength of the applied quadrupole field and the positional shift of droplet from the center of the field. Thus, it can be conjectured that, at the electric field strengths required to levitate the droplet, the external fields only act as initiators of finite-amplitude shape deformations that only influence the Rayleigh breakup process while the induced instability can be seen at much higher values. Thus, it can be inferred from the present study that the symmetrical pathway of the Rayleigh breakup is not universal and perhaps is an exception in realistic situations.

The occurrence of subcriticality of the instability and its dependence on the polarity and strength of the confining electrodes can critically influence the size and charge distribution of droplets thus formed. The relevance of the instability in practical applications stems from several recent advancements in mass spectrometry which remains one of the most popular use of quadrupolar ED traps, wherein Rayleigh breakup is employed. These include investigating chemical reactions in microdroplets in a controlled environment [24], understanding complex chemistry of aerosol particles in the atmosphere [25] or using ED balances [26–28] to measure surface tension, viscosity, diffusivity, and hygroscopicity in aerosol droplets as well as contact free ice-nucleation processes [29,30]. These state-of-the-art advancements in understanding atmospheric physics, apart from continuous use in mass spectrometry as well as the use of electrosprays for the formation of charged droplets, their breakup, and ultimately, their use in the production of nanoparticles using supersaturation and crystallization, has made the understanding of the stability of charged droplets in general and their response in an ED balance in particular, very relevant to advancements in these technologies. Hence, it is necessary to carefully consider the findings presented here in future studies and their outcome in various processes.
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A Appendix

In the potential flow BEM formalism the unsteady Bernoulli equation is solved to obtain the change in velocity potential at each time step due to variation in various energies of the droplet as it undergoes simultaneous surface and COM oscillations. Since in the simulation setup, the center of the coordinate axes lies at the center of the drop (Fig. 1), the reference frame moves with the oscillating droplet. To account for such moving frame of reference, an added energy term is required to be considered [23]. However, in the present simulations, for typical experimental parameters, this added energy term (due to COM motion), $\rho \cdot v \cdot U$ is observed to be small as compared to the kinetic energy ($v \cdot v / 2$) of the drop surface as shown in Fig. 7. Thus the added energy due to moving frame of reference is neglected in the present calculations.

Fig. 7 Comparison of kinetic energy term ($v \cdot v / 2$) with the added energy term $U \cdot v$ due to moving frame of reference

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