Aerodynamically-assisted electrified microscopic jets

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We introduce a way to produce steady micro/nano-liquid jets via electrohydrodynamic fields together with co-flowing gas streams. We study the dripping-jetting transition of this configuration theoretically through a global stability analysis as a function of the governing parameters involved. A balance between the local radial acceleration to the surface tension gradient, the mass conservation and the energy balance equations enable us to derive two coupled scaling laws that predict both the minimum jet diameter and its maximum velocity. The theoretical prediction provides a single curve that describes not only the numerical computations but also experimental data from the literature for cone-jets. Additionally, we performed a set of experiments to verify what parameters influence the jet length. We adopt a very recent model for capillary jet length to our configuration by combining electrohydrodynamic effects with the gas flow through an equivalent liquid pressure. Due to the diameters below 1 micrometer and high speeds attainable in excess of 100 m/s, this concept has the potential to be utilized for structural biology analyses with X-ray free-electron lasers at megahertz repetition rates as well as other applications.

The ubiquitous study of liquid jets and drops during the last decades [1] has accompanied their increasing use in applications within analytical chemistry [2], [3] or material science [4] among others. In this sense, a prominent example is Serial Femtosecond Crystallography (SFX) [5], with the use of flow focusing (FF) [6], [7], [8] and electrospray (ES) [9], [10] for carrying biological samples to the region where they interact with the focused X-ray pulse [11], [12]. With the introduction of X-ray free-electron laser facilities that produce pulses at megahertz rates, even smaller and faster liquids are required [13], [14]. The interaction with each X-pulse is quite violent and influences more than just the intersecting volume of the X-ray beam with the jet. The jets need to run fast enough to introduce fresh sample on each pulse, taking into account the explosive interaction with the X-rays [15].

It is well known that smaller nozzles in flow focusing or electrospray geometry lead to tinier jets. However, miniaturization also reduces the robustness of such systems (e.g. increasing the likelihood of clogging). Thus, it is desirable to work at optimum conditions for both maximum speed \( v_j \) and minimum diameter \( D_j \) given a certain nozzle size. This entails the maximization of the driving pressure \( \Delta P \) and the minimization of the liquid flow rate \( Q_l \) for a steady operation, as can be noted from the established scaling laws for capillary cone-jets as a function of the flow properties and governing parameters (e.g. surface tension \( \sigma \), viscosity \( \mu_t \), density \( \rho_t \), electrical conductivity \( K \), permittivity \( \varepsilon \), gas pressure \( \Delta P_g \) and electrical pressure \( \Delta P_{ES} \): \( v_j \approx (2\Delta P/\mu_t)^{1/2}, D_j \approx (8\rho_t Q_l^2/\pi^2 \Delta P)^{1/4} \), with \( \Delta P = \Delta P_{FF} \approx \Delta P_g \) and \( \Delta P = \Delta P_{ES} \approx (\sigma^2 K^2 \rho_t \varepsilon^2)^{1/3} \) for flow focusing and electrospray under the dominance of inertia and electrostatic suction, respectively [16], [17]. But any accurate modeling of the jet behavior must contend with instability mechanisms within the fluid domain that may prevent the ideal steady jetting and imposes some additional constraints to the problem. So, for a given Weber number \( We_l^* \) within the range \( 1 \leq We_l = \rho_t v_j^2 D_j/2 \sigma \leq 20 \), the maximum jet speed is limited by the minimum liquid flow rate \( Q_{min} \). Consequently, we can transform this optimization problem into that of finding the dripping-jetting transition, which has been recently linked to the global stability analysis of focused liquid jets [18], [19].
We propose here a method to generate liquid jets using electrohydrodynamic forces together with co-flowing gas streams producing shear axial forces on the jet (Fig. 1). Specifically, we fluid with a flow \( Q_\ell \) from a capillary with an inner diameter \( D_i \). The liquid stream is forced to discharge into a low density ambient gas with an electrical permittivity close to vacuum \( \epsilon_o \). The main driver is an imposed electrical potential \( \Delta V \) between the liquid capillary and a grounded electrode placed at a distance \( H \) downstream from the nozzle end. Additionally, a mass flow rate \( Q_{mg} \) of a dielectric gas with density \( \rho_g \) and viscosity \( \mu_g \) is forced to flow co-axially through an annulus nozzle (Fig. 1) with a hydraulic diameter \( D_j^{(h)} \) that leads to a gas Reynolds number \( Re_g = Q_{mg} / \mu_g D_j^{(h)} \), with a characteristic velocity \( v_g \sim Q_{mg} / \mu_g D_j^{(h)/2} \). Then a steady liquid jet with a diameter \( D_j \) and an intact length \( L_j \) is issued. Each flow has a boundary layer of thickness \( \delta_g = (\mu_g D_j^{(h)}) / (\rho_g v_s) \) and \( \delta_l = (\mu_l D_j / (\rho_l v_s))^{1/2} \) for the gas and liquid, respectively, with a characteristic velocity \( v_s \). The electrical charge density in the bulk, is approximately zero owing to the electrical relaxation time of the charges \( t_e \sim \epsilon / K \), which is much smaller than the characteristic hydrodynamic time \( t_h \sim L_j D_j^2 / Q_\ell \) along the cone-jet transition length \( L_t = \epsilon_o \Delta V^2 / \sigma \). At the vicinity of this region, shear electric stresses \( E_s \) are comparable to the normal electrical stress on the cone \( E_n \sim (\sigma / \epsilon_o L_t)^{1/2} \), the voltage mainly decays as \( \Delta V \sim E_n L_t \) and the electrical current transported by the jet turns out \( I \sim K D_j^2 \) [20], [21]. For conditions close to the dripping-jetting instability, the section at the stagnation point has a characteristic diameter \( D_o \sim D_j \) and is located within the electrical cone-jet transition region. Between that stagnation section and where the jet emerges from the hydrodynamic transition region at a distance \( l \) downstream, mass continuity dictates that the liquid flow rate through the liquid boundary layer of thickness \( \delta_l \) is equal to to the liquid flow rate through the jet, leading to \( v_s D_j \delta_l \sim v_j D_j^2 \). Here, we postulate that the local radial acceleration of perturbations grows downstream through the liquid boundary layer until it becomes eventually of the same order of magnitude than the surface tension gradient at \( l \) downstream, which demands

\[
\rho_l \frac{v_j \delta_l}{l e_o} \sim \frac{\sigma}{l D_j} \tag{1}
\]

This temporal amplification would play the role of triggering the underlying global instability mechanisms that lead to the dripping-jetting transition within the liquid domain. Since the characteristic time of surface wave package \( t_o \) is scaled at first order as the capillary time \( t_o \sim (\rho D_j^2 / \sigma)^{1/2} \), the algebraic manipulation of the equation (1) and the mass balance equations enable us to derive the first-order estimates for \( v_s \sim \sigma^2 / (\mu_l D_j v_j^2) \) and \( D_j \sim \sigma / (\rho_l v_j^2) \). Making the later dimensionless with the intrinsic viscous-capillary length of the liquid \( l_\mu = \mu_l^2 / \rho_l \sigma \) and the capillary velocity \( v_\mu = \sigma / (\mu_l \epsilon) \), we obtain the following relation

\[
\left( \frac{v_j}{v_\mu} \right)^2 = c_d \frac{l_\mu}{D_j} \tag{2}
\]

where \( c_d \) is a constant to be determined later. An energy balance closes the scaling analysis. Specifically, two power sources exist in the problem, the electrical part given by \( I \cdot \Delta V \sim \sigma K D_j^3 / \epsilon_o \) and the contribution from the kinetic energy of the gas flow \( O(\rho_g v_j^2) \) carried by the boundary layer, whose associated flow rate \( O(v_s D_j \delta_l) \) leads to a total power as \( O(\rho_g v_j^2 D_j^3) \). These energy sources are converted by the jet in the form of kinetic energy \( O(\rho_l v_j^2 D_j^3) \). Hence, the power balance evaluated at the dripping-jetting transition is found to be

\[
c_j \hat{v}_j^3 - c_g (\rho_\ell \Upsilon_g) v_j^2 \hat{v}_j = c_e \alpha \tag{3}
\]

where we include the following dimensionless numbers \( \rho = \rho_g / \rho_\ell \), \( \mu = \mu_g / \mu_\ell \), \( \Upsilon_g = D_j^{(h)} / D_i \), \( \hat{v}_j = v_j / v_c \), \( \hat{v}_g = v_g / v_c \), \( \alpha = (\rho_\ell D_j^3 K^2 / (8 \pi \sigma))^{1/2} \) and \( v_c = (2 \sigma / \rho_\ell D_i)^{1/2} \). The dimensionless electrical term \( c_e \alpha \) is balanced to the dimensionless kinetic energy of the jet \( c_j \hat{v}_j^3 \) minus the contribution of the dimensionless kinetic energy from...
where the wrapping gaseous boundary layer \( c_d (\rho \mu \bar{Y}_g)^{1/2} \bar{c}_g^2 \). The coefficients \( c_c, c_g \) and \( c_d \) are dimensionless numbers that should be found within the explored parametric domain, as it will be shown later by the global collapse of the numerical data cloud around the equation (3). In order to validate the coupled scaling relations derived at the dripping-jetting transition, we will conduct a comprehensive numerical study where both base flows and their global stability are evaluated (Fig. 2). Given that it is possible to define a steady surface charge density \( \sigma_c \) at the liquid interface, one reads the dimensionless axisymmetric Navier-Stokes equations for the pressure \( p^{(j)}(r; z; t) \) and velocity \( \mathbf{v}^{(j)}(r; z; t) = (u^{(j)}(r; z; t))^t \mathbf{e}_r + (w^{(j)}(r; z; t))^t \mathbf{e}_z \), where \( r \) and \( z \) are the radial and axial coordinates, respectively. Likewise, the superscripts \( j = \ell \) and \( g \) refer to the liquid and gas phases, respectively, and the subscripts \( t, r \) and \( z \) stand for the corresponding partial derivatives for the variables involved.

So, one reads

\[
\nabla \cdot \mathbf{v}^{(j)} = 0,
\]

\[
\frac{\partial \mathbf{v}^{(j)}}{\partial t} + \rho^{(j)} (\mathbf{v}^{(j)} \cdot \nabla) \mathbf{v}^{(j)} = -\nabla p^{(j)} + \mu^{(j)} C \nabla^2 \mathbf{v}^{(j)},
\]

where \( C = (2 \mu^2 / \rho \sigma D_t)^{1/2} \) is the Ohnesorge number of the liquid, \( t \) is the time and \( \delta_{ij} \) is the Kronecker delta. As the Bond number \( Bo = \rho g D_t^2 / 2 \sigma \ll 1 \) and the Froude number \( Fr = v^2 / (g D_t)^{1/2} \gg 1 \), we can neglect gravitational effects along the fluid domain. Additionally, the electrical potential has to fulfill the Laplace equation

\[
\nabla^2 \phi^{(j)} = 0
\]

Given a potential solution, the electric field at both sides of the jet surface is

\[
E^{(j)}_k = \frac{F^{(j)}_{x_k, n} \phi^{(j)} - F^{(1 - \delta_k, n)}_{x_j} \phi^{(j)}}{(1 + F^2_j)^{1/2}},
\]

where the subscripts \( k = n \) and \( s \) denote the normal and shear directions, respectively. Besides, by the imposition of the kinematic compatibility and the balance of tangential and normal stresses on the interface, one reads

\[
F_t + F_z w^{(f)} - u^{(f)} = F_t + F_z w^{(g)} - u^{(g)} = 0
\]

\[
\sum_j \beta^{(j)} \mu^{(j)} C [(1 - F^2_z)(u^{(j)}_t + w^{(j)}_z) + 2F_z (u^{(j)}_r - w^{(j)}_z)] - \frac{1}{2} \sigma_c \mathbf{E}_c^{(j)} = 0
\]

\[
\sum_j \beta^{(j)} \mu^{(j)} C [u^{(j)}_t - w^{(j)}_z] + \beta - 1] E^{(j)}_c^2 / (1 + F^2_j)^{3/2} = 0
\]

where \( \chi = 2L_t / D_t \) is the electric Bond number and \( \beta = \epsilon / \epsilon_0 \). Considering both the jump condition for the normal electric field and the charge conservation at the
free surface, we obtain
\[
\begin{align*}
\sigma_e &= E^{(s)} - \beta E^{(t)} \\
\frac{\partial \sigma_e}{\partial t} + F_z u^{(t)} + w^{(t)} - \sigma_{ez} - \frac{\sigma_e}{1 + F_z^2}[u^{(t)} + F_z w^{(t)} - F_z(z u^{(t)} + w^{(t)})] &= \alpha E^{(t)},
\end{align*}
\]
(11)

At the inlet section \(z=0\), we set a flat profile for the gaseous stream \(U_g = 4\alpha_g Re_g/\rho_g \pi D_g^{(h)}\) and a Hagen-Poiseuille parabolic profile \(w^{(t)} = 8Q_i/(\pi D_g^2 v_e)(1 - r^2)\) for the liquid phase. Previous flow profiles evolve along the domain downstream, drawing their respective boundary layers. Besides, the non-slip boundary condition is imposed at the solid walls and by simplification of the electric problem \(\phi^t = \phi^o = \chi^{1/2}\). We prescribe the standard regularity conditions \(\phi_r^t = w^{(t)} = r^{(t)} = 0\) at the symmetry \(z\)-axis, and outflow conditions \(u^{(t)} = u^{(t)} = F_z = 0\text{ at } z = L_z\). As boundary conditions for the electrical potential at \(r = L_r\), we assume an approximated analytical solution [25] successfully used in pure electro-spray simulations [23],[17]

\[
\phi_1(r', z') = -\chi^{1/2}K_o \ln \left\{ \frac{\left[ r'^2 + (1-z')^2 \right]^{1/2} + (1+z') \right\} \ln (8H/D) \right\},
\]
(13)
together with a logarithmic decreasing of the voltage in the radial direction at \( z = 0 \) and \( D_i/2 < r < L_r \).

The fluid domain is radially discretized via Chebyshev spectral collocation technique with 12 and 50 points for the liquid and gas domains, respectively, and through a fourth-order finite differences scheme for the longitudinal coordinate with 751 points. Once the numerical model is established, it is possible to obtain both the non-linear response and the eigenvalue problem to evaluate the global stability analysis of a basic flow with the same equation framework [26]. This allows us to capture the critical points of the dripping-jetting transition instead of classical spatio-temporal stability analysis that overestimates \(Q_{min}\) [27]. In order to calculate the linear global modes, we can assume a temporal dependence of a certain hydrodynamical variable \(U(r, z; t) = U_o(r, t) + \eta \delta U(r, z)\text{ (}\eta \ll 1\text{)\), where } U_o(r, t) \text{ and } \delta U(r, z) \text{ are the steady flow and the eigenmode, respectively. This yields a complex eigen-frequency spectrum } \omega = \omega_i + i\omega_p \text{ and their corresponding eigenmodes } \delta U \text{ as a function of the governing parameters. If the highest imaginary frequency is positive, then the system turns out asymptotically unstable [28]. An accurate validation demands an exhaustive exploration of the dimensionless parameters involved in the problem, namely } C \in [0.01234, 3.3242], \chi \in [0, 12], \alpha \in [13, 124479], \beta \in [10, 50], \mu \in [0.00016, 0.01625], \rho \in [0.00104167, 0.0015625], Re_g \in [0, 150], U_o \in [0.0015, 0.02]. \text{ From such an exploration we determined that both scaling relations collapse the numerical points with a remarkable agreement (Fig. 3), leading to the constants } c_d = 11.4, c_e = 2.58, c_j = 0.52 \text{ and } c_d = 24.8. \text{ Interestingly, we also find a generality for both flow focusing and electrospray, comparing our prediction to several dripping-jetting experiments from the literature, finding an excellent degree of matching. These results may suggest a common origin for the dripping-jetting instability of cone-jet configurations. We now focus on conditions and parameters relevant for SFX experiments. In particular, we use a liquid (12\% water, 88\% glycerol doped with KCl) with } \rho_t = 1050 \text{ kg/m}^3, \mu_t = 120 \text{ mPa-s, } \sigma = 0.065 \text{ N/m, } K = 0.1 \text{ S/m and a flow rate ranging from 0.1 to 5 } \mu\text{L/min carefully fed from a syringe pump (CETONI, neMESYS 600). The voltage is applied onto the liquid through a metal wire connected upstream to a high voltage power supply (FUG HCP 35-20000) and the grounded electrode is placed at } H = 1.9 \text{ mm. The gas flow is pure nitrogen } N_2, \text{ which is controlled via a pressure regulator upstream (SMC, IR2000-F02) and measured by a flow meter (Bronkhorst, EL-FLOW). A high-vacuum discharge ambient gas is kept at a pressure of about } 10^{-3} \text{ Pa using vacuum pumps (Pfeiffer Vacuum). The nozzle fabrication by means of a laser lithography nano 3D printer (Nanoscribe GmbH, Photonic Professional GT) [30] allowed us to accurately explore the influence of the nozzle geometry in terms of } H_p = H_p/D_i \text{ and } \chi_p \text{ for } D_i = 40 \text{ \mu m (Fig. 4). A parameter island for steady jetting was found, with stable points for } We_t \approx 45. \text{ This entails a potential increase of 50 \% over the maximum jet speed of both flow fo-}

FIG. 4. Gañán-Calvo et al. jet length scaling law (dashed line) and their supporting experimental data (grey) [29] together with experimental realizations for this aero-electro configuration (color). In the embedded figure, the experimental points collected are plotted in terms of the gaseous Reynolds number and the dimensionless voltage. Mark color: \( We_t = 6.24 \) (black), 10.80 (yellow), 15.28 (blue), 19.72 (green), 44.11. Mark shape: \{ \( H_p, \chi_p \} = \{(0.326, 1.245) \} \text{ (circle), \{ 0.7, 1.98 \} \} \text{ (triangle), \{ 0, 0.847 \} \text{ (square)} \{ 0.1, 7.79 \} \} \text{ (diamond).}
We limit m/s, mg/min, $^{\hat{29}}$ and expressed as recently derived for steady jetting of mechanical means parameters. In this regard, we use a jet length scaling a description of the jet length in terms of the governing plays a major role for SFX application. This motivates $Q = 300$ nL/min, $Q_s = 50$ nL/min, $\Delta V = 2400$ V, $Q_{mg} = 1.86$ mg/min, $H_p = 0.7$, $Y_g = 1.96$, $D_j \approx 260$ nm and $v_j \approx 110$ m/s.

Upscaling and pure electrospray at their non-axisymmetric limit $W_{e_j} \sim 20$. Apart from jet steadiness, jet length also plays a major role for SFX application. This motivates a description of the jet length in terms of the governing parameters. In this regard, we use a jet length scaling recently derived for steady jetting of mechanical means $^{[29]}$ and expressed as

$$\frac{L_j}{d_o} = k_3^{1/2}W_{e_j}^2 \left( \frac{d_o}{D_j} \right) \left( 1 + k_4 \frac{C_{a_e}}{W_{e_j}^{-2}L_j/d_o} \right)^{1/2}$$

where $d_o = \sigma/\Delta P$, $C_{a_e} = \mu v_j/\sigma$, $k_3 \approx 163$ is an universal factor and $k_4$ is a constant that depends on the type of ejector. Here, we take the same value than that for gas focused jets $k_4 = 0.175$. To adapt properly that model to our system, we assume a negligible influence of $\Delta V$ at first-order, given its narrow range at steady jetting (Fig. 4). Thus, we use an equivalent liquid pressure $\Delta P \sim \Delta P_{es} + \psi \Delta P_g$ including both electrical $\Delta P_{es}$ and gas pressure $\Delta P_g$, according to classical gas discharge through apertures. Despite that most of our experimental realizations properly fit the adapted scaling law for $\psi = 25$, the upper limit found for flow focused jets is exceeded (Fig. 4). Additionally, given a $\Delta P_g$, the nozzle geometry does not show any significant influence over the jet length.

Lastly, we show the possibility of delivering two miscible liquids with a double configuration. The inner liquid (s) can be steadily and coaxially issued through an outer liquid (c) at liquid flow rates even smaller than what the physical limit dictates for the same liquid (s) in a single configuration. For instance, we generate steady jets for $Q_{c}/Q_s \in (1.6)$, delivering pure water through the liquid previously mentioned. Interestingly, we found conditions for steady jets with $D_j \approx 260$ nm and $v_j \approx 110$ m/s, values obtained by using our model with a total flow rate $Q_t = Q_c + Q_s$ of a liquid equivalent to the resulting mixture at the ratio $Q_c/Q_s = 6$ (Fig. 5).

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