Splitting of a Liquid Jet

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We demonstrate that a flowing liquid jet can be controllably split into two separate subfilaments through the application of a sufficiently strong tangential stress to the surface of the jet. In contrast, normal stresses can never split a liquid jet. We apply these results to observations of uncontrolled splitting of jets in electric fields. The experimental realization of controllable jet splitting would provide an entirely novel route for producing small polymeric fibers.

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The fragmentation of liquid jets into droplets has been the subject of intense investigation [1–3] and is at the heart of many technological processes, ranging from ink jet printing [4,5] to microfabrication. Most efforts for controlling droplet breakup have focused on the Rayleigh instability [6]. On the other hand, it has long been known that the Rayleigh instability can be suppressed by either viscoelasticity or by a convective flow [1]. In this situation, a completely different mode for jet breakup might be possible, the splitting of a jet into two separate filaments. In this Letter, we carry out a theoretical analysis of the conditions for splitting a jet. If jet splitting were controllable, it would provide an entirely novel route for producing small fibers.

The possibility of jet splitting is not mere theoretical fantasy: in uncontrolled situations, jet splitting has been observed when fluid jets interact with electric fields. Old experiments [7–9] demonstrate that when large axial electric fields are applied to a liquid jet, both bending of the center line of the jet and (more rarely) splitting of the jet into two filaments is observed. More recently, there has been much effort aimed at understanding electrospinning, a materials process in which submicron fibers are produced by an electrically forced viscoelastic jet. Although in electrospinning the dominant mechanism for thinning the jet involves bending [10,11], splitting events have been carefully documented [12,13].

But what is the mechanism of the splitting? What types of forces are needed to split a jet? Can splitting of a jet be controllably produced in the laboratory? To answer these questions, we have carried out a theoretical analysis of the splitting process. The scenario is outlined in Fig. 1. A jet of liquid with density ρ, viscosity μ, and surface tension γ emanates from a nozzle of radius a with volumetric flow rate Q. At some distance downstream from the nozzle a localized external force is applied which attempts to stretch out and break the cross section of the jet.

We work in the limit that the jet cross section changes slowly as it moves downstream from the nozzle. In this limit, we show jet splitting can occur if and only if there are sufficiently large tangential stresses acting perpendicular to the axis of the jet. Without tangential stresses, splitting the jet into two pieces in a finite time is impossible. We discuss the implications of this result for both the controlled splitting of a jet and for the splitting mechanism in electrospinning experiments.

We begin with the Navier Stokes equations for a fluid moving out of a circular nozzle. As in Fig. 1, there is some distance downstream where a forcing element attempts to split the jet. We denote the velocities in the cross section of the jet by $u_{||}$, and the axial velocity by u_z, and assume that the splitting event is steady in the laboratory frame. In the limit that the in-plane forces $\mathbf{f}$ are sufficiently large that $u_{||} \gg u_z$, the equation of motion is

$$\rho(u_z \partial_z u_{||} + u_{||} \cdot \nabla_{||} u_{||}) = -\nabla_{||} p + \mu \nabla_{||}^2 u_{||} + \mathbf{f}, \quad (1)$$

with $\nabla_{||} \cdot u_{||} = 0$. Here we have also assumed that the radius of the jet is much smaller than the length scale of the deformation in the axial direction, so $\partial_z \ll \nabla_{||}$. Under these assumptions, the axial velocity $u_z \approx Q/(\pi a^2)$ is approximately constant. Hence we can make the substitu-

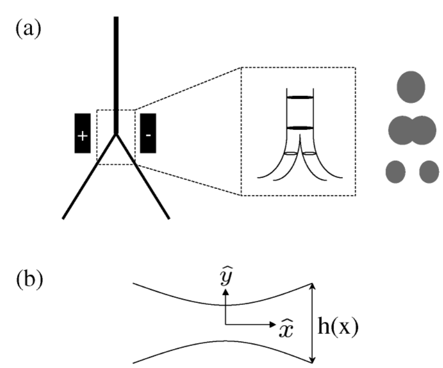

FIG. 1. (a) Schematic of the proposed experiment. A liquid jet passes through a forcing element (e.g., an electric capacitor) that stretches out the cross section of the jet. The stretching splits the jets into two subfilaments. Near the splitting event the cross section of the jet stretches from a circle to break into two separate pieces. (b) The dynamics of the splitting is described by deriving equations for evolution of the thickness $h(x, t)$ of the cross section.

tion $t = z/u_z$, so that $\partial_t = u_z \partial_z$. The equations are then the two-dimensional forced Navier Stokes equation, representing the fluid flow in a cross section of the jet as it advects away from the nozzle.

This derivation demonstrates that under our assumptions, the jet splitting is equivalent to the breaking of a two-dimensional droplet. To determine whether jet splitting is possible, we make the further assumption [14,15] that near a putative splitting event, the jet cross section becomes long and narrow with thickness $h(x, t)$. If the x direction denotes the long axis along the jet cross section, then we assume that variations in the y direction are large compared to variations in the x direction [see Fig. 1(b)]. The validity of this assumption can be checked *a posteriori* once the nature of the solution near a splitting event is known. We enforce boundary conditions on the free surfaces for the jump in normal stress $\gamma\kappa - n(x)$ and tangential stress $\tau(x)$, where κ is the mean curvature and $n(x)$, $\tau(x)$ are externally imposed normal and shear stresses. The resulting equations for the thickness of the neck $h(x, t)$ and axial velocity $v(x, t)$ are [14]

$$\rho(\partial_t v + v v') = p' + 4\mu \frac{(h v')'}{h} + \frac{\tau(x)}{h} \tag{2}$$

$$\partial_t h + (h v)' = 0, \tag{3}$$

where $p = -\gamma h'' + n(x)$, where primes denote differentiation with respect to x.

We now use these equations to understand whether a jet can be split into two filaments. A splitting event corresponds to the vanishing of the cross sectional thickness $h(x, t)$ in finite time. First we consider the case of an applied tangential stress τ and a vanishing normal stress $n(x) = 0$. The jet cross section is taken to be initially circular, and we impose a tangential stress that acts symmetrically around the center of the cross section $\tau(x) = \tau_0 x$. For small τ_0, surface tension balances tangential stress and the jet cross section evolves to a noncircular steady state. Our numerical simulations indicate that when $\tau_0 > 0.39\ \gamma/a^2$ no such steady state exists and the jet cross section splits into two pieces.

Figure 2 shows the time evolution for the time evolution of the thickness h and the velocity v. The equations are simulated using a second order implicit finite difference scheme. The inset shows that the minimum thickness vanishes in finite time t^*, obeying the law $h_{\min} \sim (t^* - t)^2$ while the maximum velocity obeys $v_{\max} \sim (t^* - t)^{-1}$. Tangential stresses can split a jet.

We now construct an approximate analytical description of this splitting event. The solution is comprised of three separate regions: (1) the *lamellar region* is characterized by a flat h profile and a linear velocity field. This region extends roughly for $|x| \leq 0.8$. (2) The *outer region* occurs for $|x| > 0.8$ and is characterized by an essentially nearly time independent h and v. (3) The jump region connects the lamellar and outer regions, and it contains a sharp jump in the velocity field.

The dominant forces in *the lamellar region* are fluid inertia and the applied tangential stress. Motivated by the numerical solution, we assume that the length scale in this region is constant and thus use the ansatz $h(x) = f(x) \times (t^* - t)^p$ and $v(x) = (t^* - t)^q g(x)$. Equation (3) then implies that $q = -1$, and Eq. (2) implies that $p = 2$, in agreement with our numerical results. This dominant balance is consistent since the surface tension and viscous forces that we have neglected are asymptotically smaller $[O((t^* - t)^{-1})]$ than the forces we have kept $[O((t^* - t)^{-2})]$. Without surface tension and viscosity, Eqs. (2) and (3) have the exact solution $g = 2x$ and $f = \tau_0/(6\rho)$. Figure 3(a) shows the lamellar region converges quantitatively onto this solution: we plot $h(x, t) v(x, t)^2/x^2$ for several times before the splitting event. The solution converges to the theoretical value $2/3(\tau_0/\rho)$ predicted by the exact solution over the entire lamellar region.

The *jump region* connects the lamellar solution constructed above to the essentially time independent outer region. The characteristic feature of this region is the strong growth of the maximum fluid velocity given by $v_{\max} \sim a(t^* - t)^{-1}$. Figure 2(b) shows that in the jump

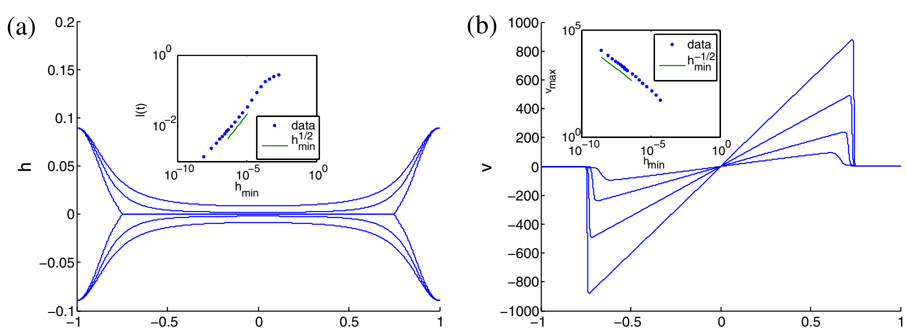

FIG. 2 (color online). Time evolution of the thickness $h(x, t)$ (a) and velocity $v(x, t)$ (b) of the jet cross section, under a tangential stress $\tau_0 = \gamma/a^2$ and $\mu = 0.27\sqrt{\gamma a \rho}$. The inset of (a) shows the minimum thickness vanishes as $h_{\min} \sim (t^* - t)^2$, while the inset of (b) shows the maximum velocity diverges as $v_{\max} \sim (t^* - t)^{-1}$.

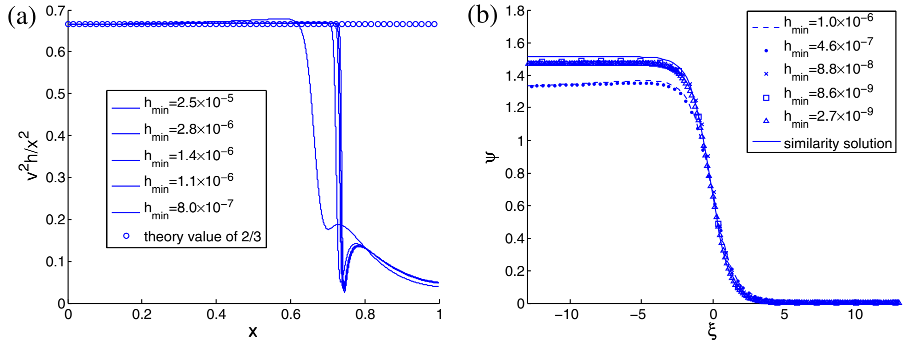

FIG. 3 (color online). (a) Collapse of curves in the lamellar region. The dotted line corresponds to the value of $\frac{2}{3}$. The combination $\frac{v(x,t)^2 h(x,t)}{x^2}$ collapses to the dotted line near the origin. (b) Collapse of the velocity profile in the jump region onto the similarity solution. $\xi = (x - x_0)/\ell(t)x_0$ is the position where the velocity is half its maximum.

region this maximum fluid velocity slows down abruptly over a length scale ℓ. We measured this length scale from our simulations and found that it obeys $\ell(t) = \mu/(\rho a) \times (t^* - t)$. This therefore motivates the ansatz

$$h_{\text{jump}} = \frac{\tau_0}{\rho}(t^* - t)^2 \phi(\xi) \tag{4}$$

$$v_{\text{jump}} = \frac{a}{(t^* - t)} \psi(\xi), \tag{5}$$

where ξ is the similarity variable $(x - x_0)/\ell(t)$ and x_0 denotes the position of the jump region. Using this ansatz in Eq. (3) implies that the mass current is to leading order independent of ξ, so that $\phi\psi = J$. Equation (2) has a dominant balance between the nonlinear fluid inertial term $(\rho v v')$ and the viscous term $(4\mu/h(hv')')$. Therefore, we obtain the following relationship between ψ and ϕ:

$$\psi\psi' = 4\frac{(\phi\psi')'}{\phi}. \tag{6}$$

Using $\phi = J/\psi$, this equation can be integrated directly so that

$$\phi = \frac{J}{C}\left(1 + A \exp\frac{C\xi}{4}\right), \tag{7}$$

as well as $\psi = J/\phi$. Here C, J, A are parameters determined by matching the jump solution to the lamellar and outer solutions, respectively. The matching to the lamellar region requires that (1) the lamellar thickness matches the thickness at the leftmost edge of the jump region $\gamma/(6\rho) \times (t^* - t)^2 = \gamma/\rho(t^* - t)^2\phi(\xi \to -\infty) = \gamma/\rho(t^* - t)^2 J/C$ and (2) the velocity at the edge of the lamellar region matches the corresponding jump velocity $2x_0/(t^* - t) = a/(t^* - t)\psi(\xi \to -\infty) = Ca/(t^* - t)$. The first condition implies $J/C = 1/6$, while the second condition implies $C = 2x_0/a$, implying $J = x_0/(3a)$. Figure 3(a) tests this theory by comparing the velocity field in simulations rescaled in similarity variables: as the splitting event is approached the numerical solutions converge to the similarity solution. The collapse of the (rescaled) $h(x, t)$ yields similar agreement.

Thus, we have demonstrated that a finite tangential stress, which is large enough to overcome surface tension, splits a jet with the thickness shrinking to zero according to the law

$$h_{\min} = \frac{\tau_0}{6\rho a}(t^* - t)^2. \tag{8}$$

We have also examined whether a jet can split with only an applied normal stress [$n(x) = kx^2/2$, $\tau = 0$ in Eq. (2)]. Here there is a critical k above which the normal stress dominates surface tension so there is no steady state solution. In this regime, our numerical simulations indicate that the jet cross section again develops a solution with three regions (lamellar, jump, and outer) as above; however, the minimum thickness of the lamellar region decreases *exponentially* in time, and hence finite time splitting does not occur.

An analysis of this solution demonstrates that now the lamellar region is characterized by the balance between the viscous stress and the applied normal stress, whereas in the jump region the dominant balance is between surface tension and viscous stress. The most important point of this solution is that the exponential decay arises from the balance in the jump region, and is therefore independent of the particular form of the applied normal stress. Therefore, we conjecture that the absence of finite time splitting occurs for any choice of $n(x)$. As above, one can construct similarity solutions which quantitatively capture the numerical behavior [16]; however, given that the splitting event occurs in infinite time, this solution is not of interest and we do not pursue it further here.

We have therefore demonstrated that jet splitting is possible if and only if a sufficiently large tangential stress is applied to cross section of the jet. Normal stresses acting alone cannot split a jet. For jet splitting to occur, (i) the magnitude of the applied tangential stress τ_0 must be large enough to overcome surface tension $\tau_0 > 0.39\gamma/a^2$ and (ii) the tangential stress must act long enough for splitting

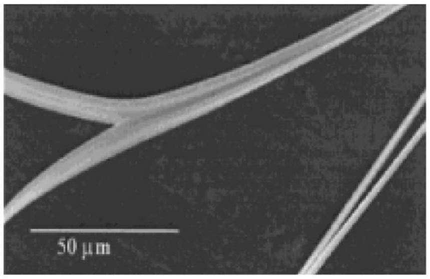

FIG. 4. Scanning electron micrograph of a splitting event from an electrospinning event [13]. The jet radius is originally of order 5 μm, much smaller than the characteristic length scale of the splitting along the fiber. The splitting occurs in a lamellar region as expected from the analysis herein.

to occur. The splitting law [Eq. (8)] implies that the splitting time is approximately $T_{\rm split} \approx \sqrt{6a\rho/\tau_0}$.

We now analyze whether previous observations of uncontrolled splitting under electric fields [9,12,13] are consistent with the mechanism outlined here. We consider a liquid jet with dielectric constant ϵ and electrical conductivity K entering a capacitor where there is an electric field perpendicular to the flow direction (Fig. 1). Initially there are electric fields inside and outside the jet, causing both normal and tangential electrical stresses [17]. Tangential stresses result from the interaction of the surface charge density on the jet σ with component of the electric field that is tangential to the surface E_t, so that $\tau(x) = \sigma(x)E_t(x)$. Surface charge builds up on the jet cross section due to Ohmic currents forced by the electric field inside the jet. After a time of order ϵ/K, the surface charge is sufficient to completely screen out the electric field, so the current stops.

At this point, the tangential electric field, and hence the tangential stress, vanishes. Hence there is a strong constraint for splitting a jet with a time independent electric field: the splitting time must be smaller than the electrical relaxation time $T_{\rm split} < \epsilon/K$, or

$$\sqrt{\frac{6a\rho}{\epsilon E_\infty^2}} < \frac{\epsilon}{K}, \tag{9}$$

where E_∞ is the applied field. For distilled water the electrical relaxation time is of order 1 μsec. Splitting a jet in such a short time requires that the applied electric field $E_\infty \sim 5000$ kV/cm for a jet with radius $a = 1$ mm. A laboratory field is of order several kV/cm and therefore cannot split a jet unless the jet radius is smaller than $\sim$1 μm.

These constraints are extremely prohibitive for steady splitting. On the other hand, a critical assumption of the analysis is violated for uncontrolled splitting events with electric fields: these jets split while their center line is oscillating at a frequency $\omega \sim 10^5$ sec^{-1}. Hence the electric field direction in the jet varies nearly as quickly as the electrical relaxation time. This implies that there will be a residual tangential field (and hence tangential stress) on the surface of the jet, rendering splitting to be theoretically possible. Indeed, the fiber shapes produced by jet splitting have a strong qualitative resemblance to the solutions described herein. Figure 4 shows the solidified remnant of an electrospun fiber that has split into two pieces. The fiber broke in the center of a flat lamellar region.

The controlled splitting of a jet into two subfilaments requires the maintenance of a steady tangential stress over the splitting time scale $T_{\rm split}$. We are aware of three potential mechanisms for creating such a steady tangential stress. These include (i) directly applying an alternating electric field, and hence mimicking the whipping jet of electrospinning, (ii) surface tension gradients could be applied to the surface of the jet, perhaps using lasers and light sensitive surfactants, and finally, (iii) a steady tangential stress can be produced from an electric field as long as an external conducting fluid is present [18,19]. An experimental demonstration of controlled jet splitting is a fascinating challenge for future investigation.

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