

Fast Solidification of a Gravity-Stretched Liquid Jet

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Spinning is widely used in nature and industry to form thin solid fibers from a liquid. While the fiber radius is readily determined when draw speed is controlled, many situations involve a draw force, making fiber radius difficult to predict. Here, we take advantage of a gravity-stretched jet configuration with tunable light-induced solidification, to elucidate spinning dynamics for a sharply localized liquid-to-solid transition. We show and explain how light intensity and solidification kinetics control the solidification locus, which opens routes for characterizing fast solidification kinetics. We also determine how the fiber radius, the transient regime following light switch-on and jet stability are set by gravity, inertia, and capillarity. These results offer predictive control on the spun solid (fiber versus beads), including for intermittent solidification.

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Spinning is vastly used to produce fibers in textile and building industries [1,2], for biomaterial engineering [3–7], drug delivery [8], particle manufacturing [9–11], or sensing and actuation [12–16]. It is also naturally used by silk-spinning arthropods to nest, displace or catch prey [17,18]. The process involves a feeding fluid, which is drawn while changing rheological properties, because of cooling, evaporation, solvent exchange, concentration, or chemical reaction [19–25]. This coupling between rheology and shape change generally makes spinning intricate, because rheological changes depend on the draw and determine stability, and because the draw is not always set by imposing a speed, which fixes the fiber radius [26–30], but by possibly complex forces—e.g., weight, drag [31], electrostatics [32–37], or centrifugation [38–40]—which depend on solidification. Therefore, important information, such as the locus of solidification, the radius of the spun fiber [41–43], or its uniformity [44,45], remains often unpredictable for force-driven spinning.

Here, we show that spinning complexity is significantly reduced for gravity-driven situations with a sharp change in rheology relevant to in-air microfluidic applications. We use a model gravity-stretched jet configuration with light-induced tunable solidification, providing independent control on the jet speed, solidification delay, capillary effects,

and draw force. This control allows us to address the key facets of the spinning dynamics, for a sharp liquid-to-solid transition, in particular, how the jet dynamics and solidification kinetics set the solidification locus, and how the fiber radius, transient dynamics, and jet stability are set by a subtle balance between gravity, inertia, and capillarity. Our results suggest a route for characterizing fast light-induced reaction kinetics. They also yield a predictive map for the spinning output (fiber versus disjointed beads, stability against global capillary modes), including for intermittent spinning.

Experimental setup—The setup consists in solidifying, rapidly and on demand, a liquid jet stretched by gravity in the air [see Fig. 1(a)]. The jet is formed by forcing a photocurable liquid through a vertical cylindrical nozzle with a constant flow rate. The flow rate Q sets the steady jet velocity at the nozzle outlet $u_0 = Q/(\pi h_0^2) \sim 0.1 - 1$ m/s, with h_0 the nozzle outlet jet radius ($0.35 \text{ mm} \leq h_0 \leq 0.45 \text{ mm}$ due to die swelling). The liquid is a highly reactive resin mixed with a photoinitiator, which is activated by ultraviolet (UV) light absorption (see the Supplemental Material, Sec. 2 [46], for details on the setup). Before illumination, the liquid is Newtonian with a mild viscosity $\eta_0 = 50.3 \text{ mPa s}$ (and a surface tension $\gamma = 44 \text{ mN/m}$, giving a nozzle Weber number $\rho h_0 u_0^2 / \gamma \sim 0.3 - 10$). At a distance $z_{UV} \sim 5 \text{ mm}$ below the nozzle, the jet is exposed from four sides to intense UV illumination obtained with UV LEDs focused over an effective length $\ell_{UV} = 1.93 \text{ mm}$ (see the Supplemental Material, Sec. 2 [46]). The light intensity I is varied between 0.5 and $\sim 6 \text{ kW/m}^2$ (measured with a thermal power sensor; see the Supplemental Material, Sec. 2 [46]), which yields solidification times down to $\sim 10 \text{ ms}$. Light

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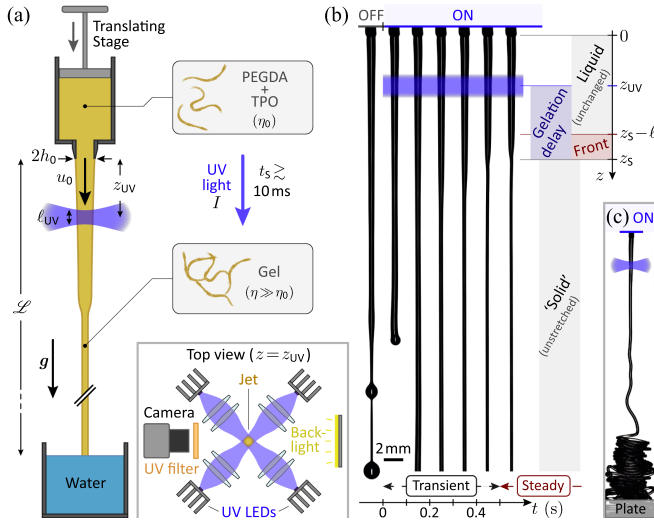


FIG. 1. Setup and phenomenology. (a) Sketch of the setup. (b) Image sequence of the transient and steady solidification upon illumination. Below the light, the jet shows a sharp thinning front ($u_0 = 0.24$ m/s, $I = 1.96$ kW/m², $\mathcal{L} = 385$ mm). (c) Below the front, the jet is solidlike, as revealed by placing a plate in its path.

rise time (~ 1 ms) is shorter than the jet timescale ($h_0/u_0 \gtrsim 1$ ms). The jet is collected in a water bath, at a distance $\mathcal{L} \sim 100$ –400 mm below the nozzle. The obtained fiber radius ranges from ≈ 0.13 to ≈ 0.23 mm.

Jet response to UV illumination—Prior to illumination, the jet is entirely liquid [see Fig. 1(b) and Movie 1 of the Supplemental Material [46]]. It is stretched by gravity and breaks in droplets by capillarity (Plateau-Rayleigh instability). As light is switched on, the jet changes drastically. Within 50 ms, it is interrupted a few millimeters below the light spot, and the lower part is gradually replaced by a solidlike thread, thinning slowly over ~ 0.5 s. This transient regime settles a steady jet, featuring a sharp thinning front, with length $\ell \sim 1$ mm. Above the front ($z < z_S - \ell$), the jet thickness is unchanged relative to the no-light case. Below the front, the jet is much thinner and uniform. This uniformity indicates the jet is solidlike [from the tensile stress due to the jet weight, $\sim \rho g \mathcal{L}$, the observed stretching rate and deformation, $\partial_z u = -(2u/h)\partial_z h \lesssim 10^{-2}$ s⁻¹ and $\Delta h/h \lesssim 10^{-2}$, suggest either a jump in viscosity by over five decades to $\sim \rho g \mathcal{L}/\partial_z u \gtrsim 10^4$ Pa s, or a Young modulus $\sim \rho g \mathcal{L}/(\Delta h/h)$ above 10^5 Pa; see the Supplemental Material, Sec. 3 [46]]. This change is illustrated in Fig. 1(c) (see the Supplemental Material, Movie 2 [46]), by intercepting the jet below the front. The jet does not form a puddle, but coils, like an elastic or highly viscous thread [49–51], confirming it is a solidlike fiber at the bottom of the thinning front ($z = z_S$). Importantly, the jet response involves three timescales: (i) the delay between illumination and solidification [$\sim (z_S - z_{UV})/u \sim 10$ –100 ms] revealed by the distance $z_S - z_{UV}$ from the light spot to the uniform fiber, (ii) the

short solidification duration ($\lesssim \ell/u \sim 1$ ms) reflected by the short length ℓ of the thinning front, and (iii) the long duration of the whole jet transient ($\sim \mathcal{L}/u \sim 0.5$ s), which relates to the collection distance $\mathcal{L}$. These timescales set the thinning front, the regime of solidification, and the settle time of the process. To understand how, we first tackle the locus of solidification and its dependence on illumination.

Locus of solidification—Figure 2(a) (see the Supplemental Material, Movie 3 [46]) illustrates how steady solidification depends on the light intensity I . At low I ($\lesssim 0.5$ kW/m²), the jet is unaffected and keeps breaking in drops. For a larger I (≈ 0.5 –0.6 kW/m²), solidification competes with breakup, and beaded or wavy fibers are formed [11]. At higher I ($\gtrsim 1$ kW/m²), waviness vanishes, and the steady solidification of Fig. 1(b) is recovered. This evolution reflects the dependence of the

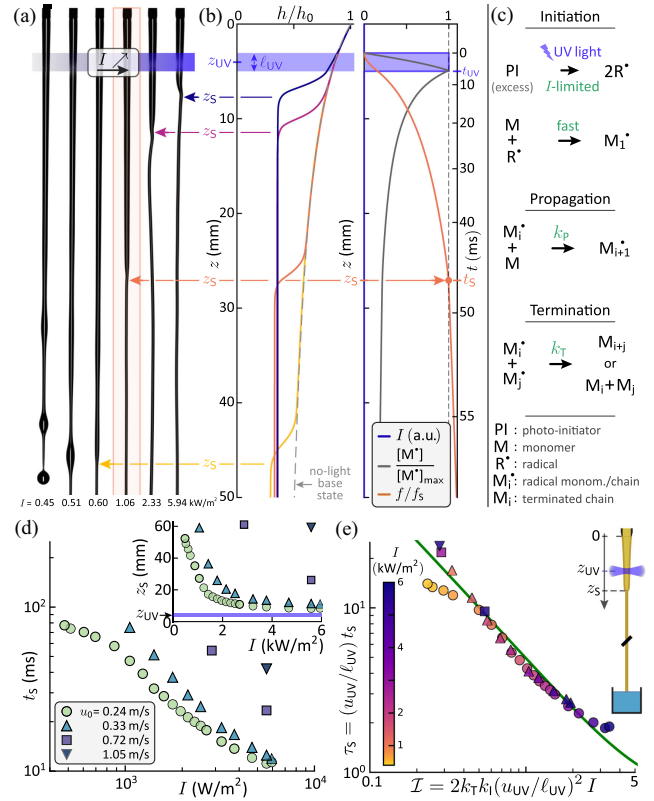


FIG. 2. Locus of solidification. (a) Steady solidification for increasing light intensity I ($u_0 = 0.24$ m/s). (b) Left: normalized jet radius vs distance z to the nozzle. Dashed line: Eq. (1). Right: model light intensity profile (arb. units), radical concentration $[M^*]$, and polymerization fraction $f = 1 - [M]/[M]_0$ vs z , for $I = 1.06$ kW/m². Right axis: travel time from top of light spot ($z_{UV} - \ell_{UV}/2$). Solidification occurs at $f = f_S$ (orange disk). (c) Linear polymerization reaction scheme. (d) Inset: solidification position vs I . Main: solidification time vs I (t_S is the measured travel time $\int h(z)^2/(h_0^2 u_0) dz$ from $z_{UV} - \ell_{UV}/2$ to z_S). (e) Dimensionless solidification time τ_S vs dimensionless intensity I . Solid line: Eq. (3).

solidification delay t_S on I . The higher I , the shorter t_S , hence, the shorter the solidification distance z_S . This is clear from the thickness profiles shown in Fig. 2(b)—which also confirm that the thinning front is sharp and the flow is unaltered down to a short distance ℓ above z_S . This is also shown by Fig. 2(d), which reports the measured solidification locus z_S (inset) and corresponding solidification time t_S (main), for increasing I and nozzle outlet velocity u_0 .

To rationalize these trends, we first model the jet flow. Since the flow above the thinning front ($z < z_S - \ell$) is unchanged by lighting, we describe it like the no-light case, assuming a low viscosity $\sim \eta_0$. For a steady and slender jet ($\partial_z h \ll 1$), mass conservation implies $h^2 u = h_0^2 u_0$, with $h(z)$ the local jet radius and $u(z) = \mathbf{u}(z) \cdot \mathbf{e}_z$ the vertical flow velocity, which is essentially uniform across the jet [52]. Additionally, since the initial velocity is larger than the gravitoviscous scale [$u_0 \gtrsim (\eta_0 g / \rho)^{1/3}$] [26,52–55], viscous stresses above the thinning front can be neglected. Therefore, from momentum conservation, the no-light velocity follows a close to freefall law:

$$u(z)^2 = u_0^2 + 2gz - \frac{2u_0^{3/2}}{We_0} (\sqrt{u(z)} - \sqrt{u_0}), \quad (1)$$

with a small capillary slowdown (last term) involving the nozzle outlet Weber number of the flow $We_0 \equiv \rho u_0^2 h_0 / \gamma$ (see the Supplemental Material, Sec. 4 [46] for derivation). As shown in Fig. 2(b), Eq. (1) captures the evolution of the jet down to the thinning front for the different light intensities.

The next step is to understand the solidification delay. In the present case where the photoinitiator is in excess and the radical-monomer reaction is fast, polymerization is limited by light, and solidification occurs after only a small fraction of monomers ($\sim 1\%$) has reacted. This implies that, although the monomer (PEDGA-700) is bifunctional [56,57], solidification can be modeled as a three-step linear radical-chain polymerization [58,59], such as presented in Fig. 2(c) (see also the Supplemental Material, Sec. 5 [46]). The initiation step is controlled by the absorbed UV light, which activates monomers $\mathbf{M}$ into radical monomers $\mathbf{M}^*$. It only occurs during the transit across the light spot and depends on light intensity I and exposure time $t_{UV} = \ell_{UV} / u_{UV}$, with u_{UV} the jet velocity at the light spot. Radical monomers may grow into radical chains, which increases the polymerization fraction $f \equiv 1 - [\mathbf{M}] / [\mathbf{M}]_0$, or deactivate into terminated chains. The competition between initiation and termination sets the relaxation timescale $t_{IT} = (2k_T k_I I)^{-1/2}$ of the process, given by the termination and initiation constants k_T , k_I (k_I depending on photoinitiator concentration and light absorption coefficient; we assume uniform absorption across the jet and uniform temperature along it and neglect oxygen inhibition, since the absorption length ≈ 0.8 mm is fairly larger than

$h_{UV} \approx 0.3$ mm, the temperature increase due to reaction exothermicity is $< 0.5^\circ\text{C}$, the liquid is initially purged from oxygen, and the atmospheric oxygen penetration depth $\lesssim 10$ μm is much smaller than h_{UV} ; see the Supplemental Material, Sec. 5 [46]). Figure 2(b) illustrates how the radical concentration $[\mathbf{M}^*]$ evolves and the polymerization fraction f increases along the jet as a result of the two timescales (t_{UV} , t_{IT}), until the jet solidifies. To model this, we solve the linear reaction scheme (see the Supplemental Material, Sec. 5 [46]) for the polymerization fraction as

$$f(t) = 1 - \left[\cosh \frac{t_{UV}}{t_{IT}} + \frac{t - t_{UV}}{t_{IT}} \sinh \frac{t_{UV}}{t_{IT}} \right]^{-\frac{k_P}{2k_T}}, \quad (2)$$

with $t = \int_{z_{UV} - \ell_{UV}/2}^z (1/u) dz$ the travel time from the top of the light spot, and we assume that the jet is solidlike when $f(t)$ reaches a fixed solidification threshold f_S , which is specific to the liquid we use [11,59] (though further modulus evolution may occur below). Introducing the natural dimensionless intensity $\mathcal{I} = t_{UV}^2 / t_{IT}^2 = 2k_T k_I (\ell_{UV} / u_{UV})^2 I$, we obtain an expression for the dimensionless solidification time:

$$\tau_S \equiv \frac{t_S}{t_{UV}} = 1 + \frac{1}{\mathcal{I}^{1/2} \tanh \mathcal{I}^{1/2}} \left[\frac{(1 - f_S)^{-\frac{2k_T}{k_P}}}{\cosh \mathcal{I}^{1/2}} - 1 \right], \quad (3)$$

valid for $\tau_S \geq 1$ (see the Supplemental Material, Sec. 5 [46]).

Combining Eqs. (1) and (3), i.e., assuming that the velocity above z_S is unaffected by light (which neglects the small change in the no-light velocity between $z_S - \ell$ and z_S ; see the Supplemental Material, Sec. 6 [46]), we obtain the solidification distance as

$$z_S = z_{UV} - \frac{\ell_{UV}}{2} + \int_0^{t_S} \frac{dt'}{u[Z(t')]}, \quad \begin{cases} \dot{Z} = u(Z), \\ Z(0) = z_{UV} - \frac{\ell_{UV}}{2}. \end{cases} \quad (4)$$

At low I , Eq. (3) gives $t_S \sim (u_{UV} / \ell_{UV}) [1 / k_T k_I I] \propto u_{UV} / I$, and the solidification distance follows $z_S \sim g t_S^2 \propto u_{UV}^2 / I^2$.

As shown in Fig. 2(e), this model matches experiments. Albeit for a slight deviation at low I for $u_0 = 0.24$ m/s (where the breakup length z_B compares with z_S), Eqs. (1) and (3) capture well the evolution of the solidification time across the different light intensity and nozzle velocity investigated, using fixed values for $(1 - f_S)^{-2k_T/k_P} = 6.2$ and $k_T = 2.75 \times 10^6$ L/mol/s. Moreover, the magnitudes of the inferred reaction constants k_T and k_P ($= 3.03 \times 10^4$ L/mol/s, assuming $f_S = 0.01$) agree with those reported in the literature ($k_T = 2.5 \times 10^6$ L/mol/s, $k_P = 3 \times 10^4$ L/mol/s for similar chemicals [60]).

Thinning front and fiber radius—We now turn to the impact of solidification on the jet dynamics, considering first the steady state shown in Fig. 3(a). Immediately above

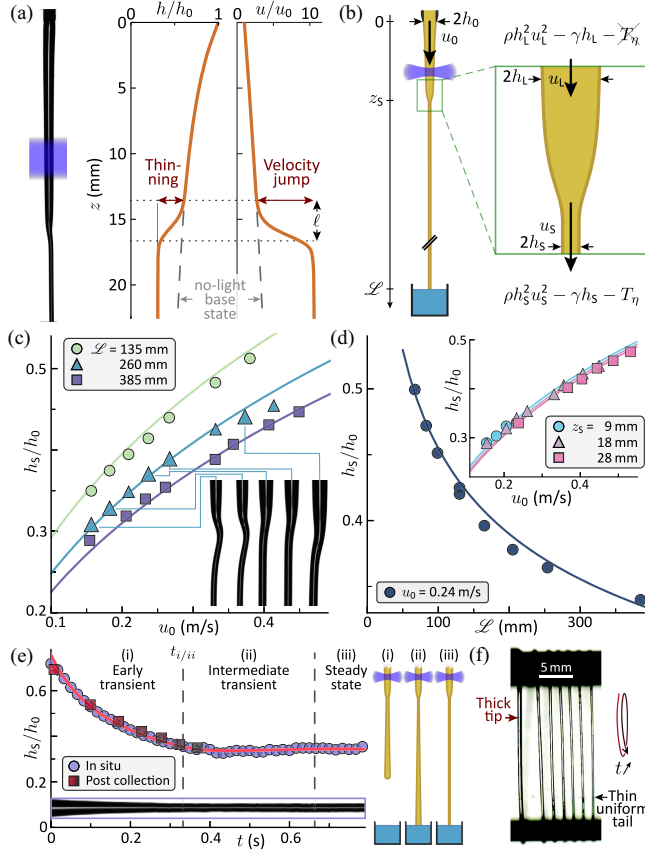


FIG. 3. Thinning front and fiber radius. (a) Image, radius profile, and velocity profile for steady solidification ($u_0 = 0.24$ m/s, $I = 1.24$ kW/m², $\mathcal{L} = 385$ mm). Dashed lines: Eq. (1). (b) Sketch for the momentum balance. (c) Normalized solid radius h_S/h_0 vs u_0 for different collection length $\mathcal{L}$ ($z_S = 18$ mm is kept fixed by tuning I). Inset: close-up of the thinning front. (d) Main: h_S/h_0 vs $\mathcal{L}$ ($u_0 = 0.24$ m/s, $z_S = 18$ mm). Inset: h_S/h_0 vs u_0 for different solidification position z_S ($\mathcal{L} = 385$ mm). (e) Typical transient evolution of h_S/h_0 as light is switched on ($u_0 = 0.24$ m/s, $I = 5.58$ kW/m², $\mathcal{L} = 385$ mm, $z_S = 9$ mm). (i) Early transient phase, before the fiber reaches the bath. (ii) Intermediate transient phase. (iii) Steady state. Circles: *in situ* measurements (see spatiotemporal inset image). Squares: postcollection measurements [see (f)]. Solid line: Eq. (6). (f) Collected fiber (coiled on a cylinder).

the solidification locus z_S , the jet thins down, from the liquid base state radius h_L to the much smaller radius h_S of the solid fiber, over the short length ℓ . Since flow is steady, this sharp thinning implies a steep increase in the jet velocity, akin to a jump [see Fig. 3(a)]. The fiber radius is thus set by the balance between this steep change in momentum flux, $\rho(h_S^2 u_S^2 - h_L^2 u_L^2)$, and the tensile stress difference, $(T_{\eta,S} + \gamma h_S) - (T_{\eta,L} + \gamma h_L)$, between the liquid (top) and solid (bottom) sides of the thinning front, with T_η and γh the viscous-elastic and capillary contribution to the jet tension [scaled by π to keep expressions short; see Fig. 3(b)], respectively. The fiber radius is modeled by noticing three crucial points: (i) the tension $T_{\eta,L}$ on the

liquid side is negligible in practice [since $u_0 \gtrsim (\eta_0 g / \rho)^{1/3}$], (ii) since the fiber velocity is constant and uniform, the tension $T_{\eta,S}$ must carry the fiber weight down to the collecting bath, $\rho g h_S^2 (\mathcal{L} - z_S)$, where the bath carries the rest of the fiber, and (iii) for a large collecting distance $\mathcal{L}$, the proper weight $\sim \rho g h_S^2 \ell$ of the thinning front is negligible relative to the tension $T_{\eta,S}$. Making use of mass conservation [$h_S^2 u_S = h_L^2 u_L = h_0^2 u_0$, and neglecting, for simplicity (see the Supplemental Material, Sec. 7 [46]), the small density change upon solidification], we obtain (see the Supplemental Material, Sec. 7 [46])

$$\frac{u_S}{u_0} - \frac{u_L}{u_0} = \frac{g(\mathcal{L} - z_S)}{u_0^2} \frac{u_0}{u_S} - \frac{1}{We_0} \left[\sqrt{\frac{u_0}{u_L}} - \sqrt{\frac{u_0}{u_S}} \right]. \quad (5)$$

Since the liquid velocity $u_L = u(z_S - \ell)$ is known from the no-light base state [Eq. (1)], Eq. (5) provides a parameter-free expression for the overall thinning ratio $h_S/h_0 = \sqrt{u_0/u_S}$, which is found to match observations quantitatively. As shown in Figs. 3(c) and 3(d), Eqs. (1) and (5) capture the evolution of h_S/h_0 with the initial velocity u_0 . They also recover the direct dependence on the collecting distance $\mathcal{L}$, and consistently predict little dependence on z_S over the threefold increase explored [see Fig. 3(d), inset]. Note that Eq. (5) neglects air drag, which is valid for low drag to fiber weight ratios (here, $\sim \sqrt{\rho_{\text{air}} \eta_{\text{air}} / h_0^2 u_0 \mathcal{L} u_S^2 / \rho g} \sim 10^{-2}$; see the Supplemental Material, Sec. 8 [46]).

The above approach also explains the transient regime following light switch on, when the thickness $h_S(z, t)$ below the front is time dependent and non-uniform. In this case, the momentum balance at the thinning front is still $(u_S - u_L)/u_0 = T_{\eta,S} / \rho h_0^2 u_0^2 - (1/We_0) [\sqrt{u_0/u_L} - \sqrt{u_0/u_S}]$, but the viscous stress is $T_{\eta,S} = m(t)(g - \ddot{u}_S)$, with $m(t) = \int_{z_S}^{\mathcal{L}} \rho h(z)^2 dz$ the time-dependent mass (scaled by π) and $\ddot{u}_S$ the acceleration of the pending fiber. Expressing the fiber mass $m(t)$ from the history of the solidification section $h_S(z_S, t)^2 = h_0^2 u_0 / u_S(t)$ (see the Supplemental Material, Sec. 7 [46]), we obtain

$$\frac{du_S}{dt} = g - \left(u_S - u_L + \frac{u_0}{We_0} \left[\sqrt{\frac{u_0}{u_L}} - \sqrt{\frac{u_0}{u_S}} \right] \right) \frac{1}{t - t'},$$

with $\begin{cases} t' = 0, & \text{for } t < t_{i/ii} \text{ (i: early transient),} \\ \frac{dt'}{dt} = \frac{u_S(t)}{u_S(t')}, & \text{for } t \geq t_{i/ii} \text{ (ii: interm. transient).} \end{cases} \quad (6)$

Equation (6) predicts an early transient stage, with an initially short and thick fiber, which progressively thins down as the drawing weight increases. This early transient lasts until the fiber tip reaches the collecting bath [see Fig. 3(e), inset]. For small capillarity ($We_0 \gg 1$), it follows that $u_S(t) = u_L + gt/2$, i.e., $h_S(t)/h_L = (1 + gt/2u_L)^{-1/2}$,

and $t_{i/ii} = (2u_L/g)\{[1 + g(\mathcal{L} - z_S)/u_L^2]^{1/2} - 1\}$. After this time, the fiber mildly thickens over an intermediate transient converging to the steady state of Eq. (5). As shown in Fig. 3(e), Eq. (6) matches quantitatively the fiber radius evolution observed immediately below the front. It is also validated by postcollection measurements of a fiber thickness profile [Figs. 3(e) and 3(f)]. However, it does not capture interactions between solidification and fast capillary modulations of the jet. In this case, the averaged thinning ratio is close to the steady model [Eq. (5)], but wave crests and necks are not thinned in exact proportion (radius modulations are damped by the front; see the Supplemental Material, Sec. 9 [46]).

Discussion—The analyses above draw a comprehensive picture of gravity-driven spinning for fast solidification of a nearly freefalling jet, in particular, of how the spun radius and its history depend on solidification kinetics, inertia, gravity, and capillarity. Their validation with a light-activated solidifying jet allowing on-demand solidification, with temporal and spatial control, offers several leverages to characterize and tune spinning.

First, the solidification locus reflects the chemistry-specific solidification kinetics, and jet solidification could be used to characterize fast light-activated kinetics within fractions of h/u . This represents $\sim 100 \mu\text{s}$ for our system, and could be reduced below $\sim 0.1 \mu\text{s}$ with ultrafast systems, as employed in microfluidics [61,62] or extreme UV-light sources [63,64]. Alternatively, when kinetics are known, light intensity pilots the solidification locus, which can be used to tune interaction with the jet breakup and control the spinning output. This is shown (for our system kinetics) by the dimensionless collection map of Fig. 4(a) built on Eq. (4). The ratio $Z_{SB} = z_S/z_B$, comparing solidification and breakup distances, indicates whether a uniform solid fiber ($Z_{SB} \ll 1$) or disjointed beads are expected ($Z_{SB} \gg 1$). The ratio $Z_{SC} = z_S/\mathcal{L}$, comparing solidification and collection distances, determines whether the jet or droplets should be liquid when collected ($Z_{SC} \gg 1$) or not ($Z_{SC} \ll 1$). Using Eqs. (1), (3), and (4) for z_S and a simplified model for z_B based on prior works [65–69], (see the Supplemental Material, Sec. 4 [46]), this collection map captures the output for all our experiments (all conducted for $Z_{SC} \ll 1$).

Importantly, the chemistry-independent gravito-capillary-inertial dynamics discussed above are not specific to photopolymerization. They are relevant to all gravity-stretched jets solidifying rapidly enough (this can be validated, e.g., with a core-shell jet solidifying rapidly by a diffusion-limited reaction; see the Supplemental Material, Sec. 10 [46]). Our model is also relevant for intermittent spinning and short fiber production, when unsteadiness yields nonuniform fiber radius. Figure 4(b) demonstrates the effect on short fibers obtained with pulsed illumination. Fibers are thinner at the top than the bottom, and nonuniformity increases for longer fibers, in agreement

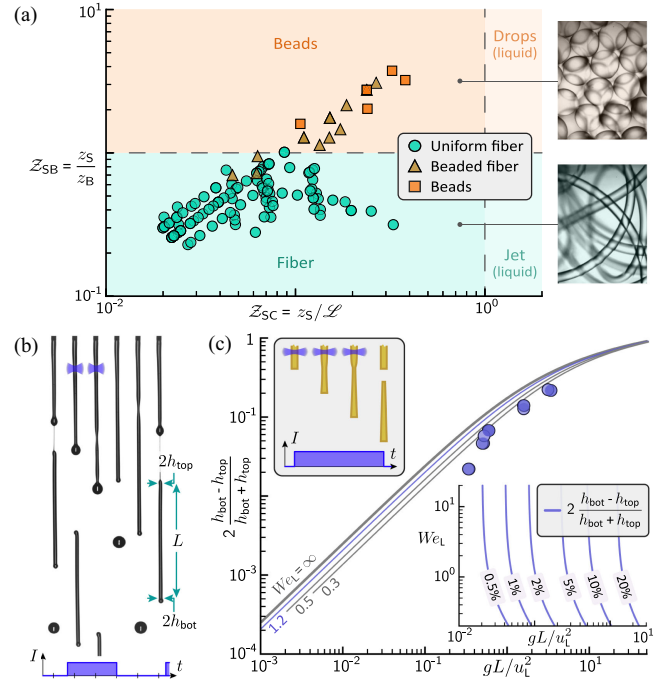


FIG. 4. Steady collection map and short fibers. (a) Solidification to collection distance ratio Z_{SC} vs solidification to breakup distance ratio Z_{SB} . Symbols: experimental output [$\mathcal{L}$ is set, z_S is computed from Eqs. (1), (3), and (4) with the same parameters as in Fig. 2, and z_B is computed from Eq. (S8) calibrated with no-light experiments in the Supplemental Material, Sec. 4 [46]]. (b) Short fibers obtained with periodic lighting (50 ms on, 50 ms off, images are separated by 21.4 ms, $I = 4.68 \text{ kW/m}^2$, $u_L = 0.32 \text{ m/s}$). (c) Fiber radius nonuniformity vs dimensionless fiber length gL/u_L^2 . Symbols: measurements for $We_L = \rho u_L^2 h_L/\gamma = 1.2$ [light frequency of 10, 20, and 50 Hz, 50% duty cycle]. Lines: Eq. (6) (with $h_{bot} = h_L$, $h_{top} = h_S(t)$, $t < t_{i/ii}$, and $\int_0^t u_S(t') dt' = L$) for $We_L = \infty, 1.2, 0.5$ and 0.3 . Bottom inset: nonuniformity map [Eq. (6)].

with our transient model [Eq. (6); see the Fig. 4 caption], which provides a predictive map for fiber tailoring. Analyses also give insights on flow stability. Indeed, jet solidification allows remote perturbations to propagate upward via elastic modes. Most often, these modes do not alter solidification, because they cannot propagate through the jet liquid portion to reach the light spot. However, for low velocity they do, which induces a strong jitter at the light (see the Supplemental Material, Sec. 11 [46]). The force balance behind Eq. (5) explains how capillary and viscous stresses permit propagation, and provides a criterion ($u_L \gtrsim \sqrt{\gamma/\rho h_L}$; see the Supplemental Material, Sec. 11 [46]) for not compromising spinning stability. These points stress that, though fluid mechanics and photopolymerization kinetics are partly decoupled for sharply changing rheology, they nonetheless interfere to set the spun product.

Last, the simple mechanisms highlighted above open questions, in particular, to address situations where drag is

important [31] (see also estimates of the Supplemental Material, Sec. 8 [46]), but not purely viscous [70,71], or the situation when solidification is gradual or uncompleted.

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Data availability—The data that support the findings of this article are openly available at [72].

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