Formation, Stability, and Breakup of Nanojets

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Atomistic molecular dynamics simulations reveal the formation of nanojets with velocities up to 400 meters per second, created by pressurized injection of fluid propane through nanoscale convergent gold nozzles with heating or coating of the nozzle exterior surface to prevent formation of thick blocking films. The atomistic description is related to continuum hydrodynamic modeling through the derivation of a stochastic lubrication equation that includes thermally triggered fluctuations whose influence on the dynamical evolution increases as the jet dimensions become smaller. Emergence of double-cone neck shapes is predicted when the jet approaches nanoscale molecular dimensions, deviating from the long-thread universal similarity solution obtained in the absence of such fluctuations.

Liquid jets of macroscopic dimensions and their breakup into drops have been of great scientific interest (1, 2) and have been used in fuel injection (3), cutting and machining (4, 5), spraying and coating, fiber spinning (6), needle and syringe injection, ink-jet printing (7), and micro- and optoelectronic device manufacturing (8). Perturbations at the nozzle opening and surface tension help drive capillary instabilities in jets. Through the application of linear stability analysis to inviscid incompressible fluid columns, Rayleigh concluded that out of all disturbances the fastest growing one has a wavelength of about nine times the column’s radius (9). The accepted mechanism of jet breakup involves flow from regions of the liquid column with smaller radii (necks where the Laplace pressure is larger) to crest regions where the pressure is lower, until pinch-off occurs; extensions of Rayleigh’s original work have shown (10–14) that the effect of viscosity can reduce the breakup rate and increase the drop size.

The highly nonlinear nature of the Navier-Stokes (NS) equations that govern fluid flow prevents reliable analyses of the nonlinear dynamics near breakup with linear stability methods or higher order perturbative treatments and requires the development of a full nonlinear theory. Progress in this direction has been achieved with NS simulations (1) and through analytical formulations based on the “lubrication theory” (1, 15–18). Particularly pertinent to our study are treatments based on the “slenderness approximation” allowing systematic derivation (1) (starting from the NS equation) of viscous one-dimensional (1D) equations that can be solved numerically with relative ease (unlike the original three-dimensional (3D) NS equation). Indeed, such treatments have been used recently with remarkable success in investigations of various liquid structures (1), yielding quantitative correct predictions in the neighborhood of breakup, as well as far from it.

Substantial efforts have been invested in studies addressing the design of nozzles (3, 14, 18, 19) including geometrical factors (such as nozzle shape) and material properties (such as wettability of the nozzle by the fluid) and their effects on the efficiency of conversion from potential to kinetic energy (sometimes termed “pressure” and “flow” energies) as well as on the internal and exit flow (velocity profiles and the magnitude of the disturbances (particularly at the nozzle opening). As the scale of the jets is reduced for use in or fabrication of miniaturized devices, the jet diameter approaches the molecular size, and the commonly used continuum fluid dynamics treatments are of questionable validity (1, 20).

We present the results of large-scale atomistic molecular dynamics (MD) simulations (21–23) aimed at extending our knowledge of jet processes to the nanometer-scale regime (that is, to nanojets, or NJs). Although such NJs are envisaged to substantially impact various technologies [such as nanoscale materials’ machining and patterning, printing of high-density miniaturized circuitry, and transfer of biological materials (24), such as genes, into cells with minimal damage], understanding the conditions for the formation of NJs and their properties is a most challenging fundamental problem. In particular, because of the reduced dimensions, the behavior of such systems involves large spatiotemporal variations and fluctuations of the liquid properties (such as temperature gradients and liquid density changes caused by viscous heating and evaporative cooling) that complicate (and in many cases invalidate) the use of continuum mechanics treatments and require the development of physically reliable and accurate atomistic-scale simulations.

We used a newly developed MD method for atomistic simulations of jet processes to show that NJ formation can indeed be achieved through high-pressure injection and thus overcome large internal viscous energy losses and surface tension effects. We also propose new design principles, such as the use of nonwetting nozzles as well as heating of the nozzle exterior surface, as means for speeding up flow and for enabling NJ formation. Furthermore, aiming at the long-standing goal of relating and integrating atomistic simulations with continuum hydrodynamic modeling, we derive a stochastic modification of the hydrodynamic (deterministic) lubrication equation (LE) for the description of free-surface flow problems and show through comparative MD and hydrodynamic simulations that fluctuation effects increase as the jet radius decreases and that the jet dynamics near breakup is dominated by thermal fluctuations. Our simulations reveal that such fluctuations influence the jet profile near breakup as well as control the breakup length, which we found for NJs to depend only weakly on the strength of perturbations generated in the vicinity of the nozzle just after exit, in contrast to macroscopic jets where the latter processes are the determining factor.

In our MD simulations, we used a common fluid (propane, C₅H₁₂) as a fluid confined and injected into vacuum through a nozzle (25–30). Various configurations of the inlet nozzle (modeled as crystalline metal) were used, including a long tube of constant diameter, a hole in the wall of the container, and a convergent nozzle (31); below, we show results for the convergent nozzle (Fig. 1A).

To determine the conditions for jet formation, we performed a number of simulations for a range of exit orifice diameters D between 2 and 6 nm and nozzle inlet pressures Pᵯ between 250 and 500 MPa (the upper limit was chosen to prevent gasification of the propane fluid), with an overall nozzle temperature Tₖ = 150 K. A steady-state jet (with a velocity of 200 m/s) was produced (32) only in simulations with Pₑ = 500 MPa and with the gold atoms of the outer surface of a 6-nm-diameter nozzle heated to the boiling temperature of propane (230 K) while the temperature of the inside gold atoms was kept at Tᵯ = 150 K. Under these conditions, the thickness of the wetting film on the outer surface of the nozzle (Fig. 1A, time t = 20 ps) was limited to three propane layers. Without heating of the outer surface, a transient initial jet formed, but it was slowed down because of viscous frictional interactions with a thickening wetting film, as well as by surface tension effects, culminating in blocking of the flow. Jet formation was also inhibited for Pₑ = 500 MPa when smaller orifice
diameters were used ($D < 6 \text{ nm}$) because of a strong pressure drop caused by frictional losses inside the nozzle, and also for the 6-nm-diameter orifice with a smaller driving pressure $P_s = 250 \text{ MPa}$. As an alternative to preventing formation of thick wetting blocking films by heating of the outside surfaces of the nozzle, as described above, we found for the 6-nm-diameter nozzle with $P_s = 500 \text{ MPa}$ and $T_b = 150 \text{ K}$ that a jet can be formed (with a higher velocity of 440 m/s) under nonwetting conditions, which we modeled by truncating the interaction potential between the propane molecules and the exterior gold atoms of the nozzle to exclude the attractive interaction between them (in practice, one could use a nonwetting thin coating applied to the exterior of the nozzle).

Atomistic configurations during the initial formation stages of the jet (with heated outside surfaces of the nozzle exit) are shown in Fig. 1A, and the evolution of the jet during the first 2 ns of the simulation is shown in Fig. 1B. The exit of the fluid from the nozzle is accompanied by evaporation of molecules and lateral (radial) spreading resulting in thin-film formation, accompanied by thickening (swelling) of the fluid column (Figs. 1A and 2A); a much reduced swelling of the jet is observed, resulting in breakup events and formation of droplets with a narrow size distribution corresponding to an average intact length of the jet (breakup length) $L_b \approx 170 \text{ nm}$ (we also obtained similar $L_b$ values in simulations with a hotter propane fluid, $T_b = 200$ and 250 K).

In both the wetting (solid lines) and nonwetting (dashed lines) cases, $v_z$ increases in the converging section ($z \leq L_0$) of the nozzle (as expected because of the continuity of the fluid), and the increase of $v_r$ reflects focusing of the flow; however, for the wetting case, near the nozzle exit the attractive interactions of the propane molecules with the nozzle surfaces cause a substantially smaller increase in $v_r$ and a somewhat larger increase in $v_z$, resulting in drag forces on the flowing fluid. Upon exit, the internal pressure and density in the liquid drop precipitously (Fig. 2D). The pressure in the fluid after the nozzle remains constant and near zero, whereas the density shows a slow gradual increase that is related to evaporative cooling of the jet.

Internal frictional processes increase the kinetic temperature of the molecular fluid substantially in the nozzle exit region. In the wetting case, friction occurs even outside the nozzle because of the molecular film adsorbed on the exterior surface of the nozzle, and consequently, the temperature is further increased (Fig. 2E). Immediately past the nozzle (for $z \leq 20 \text{ nm}$), the total internal energy of molecules near the surface of the jet is positive (gray areas in Fig. 2E), leading to a decrease in the temperature due to inefficient evaporative cooling and consequent reduction in the jet radius along the jet axis; the rate of these processes slows for $z \approx 30 \text{ nm}$.

The initial (transient) stage of the evolution of the jet (Fig. 1B, $t < 1 \text{ ns}$) is characterized by the generation of small fast-moving droplets and molecular clusters, portrayed by sawtooth-shaped discontinuities in $L$ at the droplets’ detachment times (Fig. 1B, inset), where $L$ is the intact length of the jet. At steady state ($t \approx 1 \text{ ns}$), the jet instabilities result in the formation of droplets with a narrow size distribution corresponding to an average intact length of the jet (breakup length) $L_b \approx 170 \text{ nm}$ (we also obtained similar $L_b$ values in simulations with a hotter propane fluid, $T_b = 200$ and 250 K); also, a smaller satellite droplet that eventually merged with the preceding larger one occasionally formed (Fig. 1B, $t = 1 \text{ ns}$; Fig. 3C, $t = 1.65 \text{ ns}$). Typically, the neck shape before pinch-off resembles a double cone (see Fig. 3B and the
top configuration in Fig. 3D for the wetting and nonwetting cases, respectively; occasionally, the jet profile deviates from axisymmetry (Fig. 3A), or it may be somewhat elongated, forming a small cluster between the intact part and the main droplet (Fig. 3C). The neck radius decreases steadily to its final value of one or two molecular diameters, and a smooth separation of the few remaining molecules in the double-cone apex completes the pinch-off process.

In the hydrodynamic description (1, 18), the radius of the neck shrinks as breakup of the jet is approached, and the fluid velocity in this region increases strongly (developing a singularity at the pinch-off time), thus localizing the breakup process both spatially and temporally (1, 35). Such "locality" is reflected in the independence of the fluid motion near breakup from the initial conditions and from details of the experimental setup (e.g., the radius of the nozzle); that is, the fluid motion near pinch becomes universal (1). Under these circumstances, the only relevant length and time scales (1, 18, 35) are \( l_{\text{obs}} = \eta / (\rho g) \) and \( t_{\text{obs}} = \eta / (\rho g) \) (expressing the balance between viscous and surface tension forces), respectively, which are determined by the viscosity \( \eta \), the density \( \rho \), and the fluid surface tension \( \gamma \). Furthermore, asymptotic analysis (1) reveals that the fluid motion close to the breakup singularity is a similarity solution of the equations of motion (see below), with the jet profile consisting of long thin threads connected by strongly sloped segments to the rest of the fluid.

In general, the neck shape of macroscopic jets (1, 35) depends on the ratio between the observation scale \( l_{\text{obs}} \) and the intrinsic material-dependent length scale \( l_{\text{c}} \). For \( l_{\text{obs}} / l_{\text{c}} \gg 1 \), the shape of the neck near the pinch point approaches that of a cone connected to a spherical cap (35), whereas for \( l_{\text{obs}} / l_{\text{c}} < 1 \), the aforementioned similarity solution thin-thread profile occurs (1); for macroscopic values of \( l_{\text{obs}} \) [that is, \( l_{\text{obs}} \sim 1 \mu m \) which is the limit for optical detection (20)], the above classes of behavior are associated with fluids whose viscosities are low (such as water) or high (such as glycerol), respectively. For the propane NJ (with \( l_{\text{obs}} \) of the order of the neck radius, that is, a few nanometers), this ratio is \( < 1 \) (even though propane is a low-temperature fluid), and thus, thin-thread formation is expected and indeed confirmed (Fig. 3D, LE simulation) by numerical integration (36) of the LE simulations (1) for the axial velocity \( v(z, t) \) and the radius \( h(z, t) \) of the jet along the \( z \)-axis.

\[
\delta v(z, t) + \frac{v(z, t)\delta v(z, t)}{h(z, t)} = -\frac{\partial_p}{\partial h} \text{[Laplacian]} \\
+ 3\nu \frac{\partial_z^2 h(z, t)}{h(z, t)} h(z, t) (1)
\]

\[
\delta h(z, t) + \frac{v(z, t)\delta h(z, t)}{h(z, t)} = -\frac{\partial_p}{2\nu} h(z, t) (2)
\]

with the Laplace pressure given by

\[
p(z, t) = \gamma k(z, t) = \frac{1}{h(z, t)} \left( \frac{\partial_z^2 h(z, t)}{1 + [\partial_z h(z, t)]^2} \right)
\]

where \( k \) is the surface curvature and \( v = \eta / \rho \) (in Eq. 1) is the kinematic viscosity.

However, the long thread-shaped neck (Fig. 3D, middle), the mean breakup length of \( l_{\text{mb}} \sim 530 \text{ nm} \) (Fig. 3E, bottom), and the steady-state variation with time of the smallest neck radius of the jet (minimal jet radius \( l_{\text{min}} \)) obtained through the LE simulations are in obvious disagreement with the corresponding MD simulation results, which exhibit a double-cone (sometimes called "hourglass") neck shape (Fig. 3D, top), a much smaller mean breakup

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**Fig. 2.** Steady-state properties of the propane NJ formed by injection of propane through a nozzle with a 6-nm diameter. Velocity profiles of the propane fluid are shown for simulations (A) where the propane wets the exterior surface of the nozzle and (B) where the exterior surfaces of the nozzle are nonwetting. At the nozzle exit (\( z = 0 \)), the flow-velocity profile is laminar with a parabolic variation (as shown by the length of the arrows) from the solid gold surface to the middle. Formation of an adsorbed film (with a thickness of three propane layers) in the wetting case causes swelling of the exiting jet with a rapid relaxation to a pluglike flow profile [at \( z = 5 \text{ nm} \) in (A)]. Minimal swelling is found in the nonwetting case (B). The steady-state flow velocities of the NJ are 200 and 440 m/s for the wetting and nonwetting cases, respectively. (C) Time-averaged axial \( v_z \) and radial \( v_r \) velocities of the jet (calculated as an average over the corresponding velocities of the propane molecules in cylindrical slices with a radius of 2 nm and a width of 0.5 nm centered on the jet axis) versus distance along the \( z \)-axis for the wetting (W) (solid lines) and nonwetting (NW) (dashed lines) cases. There is a larger increase in \( v_z \) upon exit and a relaxation to a higher flow velocity for the NW nozzle. (D) Time-averaged variation of the pressure \( P \) and density \( \rho \) of the propane fluid plotted versus \( z \), exhibiting sharp drops in the convergent (\( z \leq 0 \)) part of the nozzle. (E) Time-averaged variations of the propane NJ radius \( R \) (bottom two curves) and molecular kinetic temperature \( T \) (upper curves) for the W (solid lines) and the NW (dashed lines) cases; \( T \) is calculated by subtracting the local flow velocity of the jet from the instantaneous molecular velocities and time-averaging the resultant molecular kinetic energies in slices of the jet along the \( z \)-axis. The emanating jet has a higher temperature for the wetting case with \( T \) going above the boiling point of propane (230 K) right after the exit. With efficient cooling of the emanating "gaseous fluid" through molecular evaporations from the surface of the jet, which is accompanied by gradual shrinking of the jet radius, \( T \) decreases and the propane density in the jet increases [\( \rho \) in (D)], causing condensation of the propane fluid. The gray areas superimposed on the radius plots indicate spatial regions where the total internal energy (potential plus molecular kinetic energy) of the propane molecules is positive, corresponding to regions of the surface layers of the jet where molecular evaporation is most probable.
length $L_n \approx 250$ nm (Fig. 3E, top), and a manifestly less regular variation of $h_{\text{min}}$ that exhibits faster decay near pinch-off (Fig. 3E, bottom). These observations indicate the inapplicability of the deterministic LE continuum treatment for the description of nanoscopic jets and of nanoscale free-surface flow problems in general with a likely source of the difficulty being the neglect of fluctuations ([1, 20, 37, 38]). Indeed, the continuum description of such small systems requires the use of exceedingly small volumes, each containing a very limited number of particles, and consequently, continuum variables associated with such small volume elements, which represent (local) averages over the properties of the microscopic constituents, are expected to exhibit large fluctuations.

To extend the LE continuum approach to the nanoscale regime, we derived a stochastic version of the LE (SLE) by applying the simpleness approach to the NS equation $\rho(\partial_x v + \Sigma_j \partial_j \sigma_{ij}) = \Sigma_j \partial_j p_x$ (here, $i$ and $j$ are coordinate indices $i, j = 1, 2$, and 3), with a Gaussian noise obeying the fluctuation-dissipation theorem (FDT) (39, 40) added to the deterministic stress tensor $\sigma_{ij}$. In the resulting SLE, the dynamics of the jet is described by Eqs. 1 to 3 with a term $-\alpha f/(\pi h^2)$ added to the right-hand side of Eq. 1, where $f$ is a Gaussian white-noise variable obeying the FDT $\langle f(z, t)/f(z', t') \rangle = 6 k_B T \eta h^2 (z, t) g(z - z')(t - t'); k_B$ is the Boltzmann constant. The magnitude of the added stochastic term increases as the lateral dimension of the jet (the radius $h$) decreases.

The results of simulations of the propane NJ using the SLE are in remarkable agreement with the MD simulations, as evident from comparison of the neck shapes (Fig. 3D), the breakup length (Fig. 3E, top), and the evolution of $h_{\text{min}}$ (Fig. 3E, bottom). In the deterministic LE solution, the minimum neck radius decays (close to pinch-off) linearly in time (measured from the pinch-off time $t_0$); that is, $h_{\text{min}} = 0.031 (t - t_0)/a_0$, following the universal law associated with the similarity solution (1), whereas both the SLE and the MD simulations exhibit a less regular pattern of $h_{\text{min}}$ versus $t$ and a faster decay (for results obtained with MD, LE, and SLE comparative simulations pertaining to breakup processes in a liquid bridge at constant temperature, as well as a discussion of the effects due to the disjoining pressure (41), see the supplementary material (42]).

The above results suggest that in NJs the very nature of the dynamical evolution is influenced strongly by hydrodynamic fluctuations, deviating in a substantial way from the behavior predicted through the analysis of the deterministic LE. To further elucidate the role of such fluctuations on the dynamical evolution, we rewrite the force balance equation of the SLE (that is, Eq. 1 with the aforementioned added term) in dimensionless form

$$\partial_t h + v \partial_x h = -\left(\partial_x \phi + 3 \partial_z (h^2 \partial_z h) / h^2 - M_l \sqrt{6/\pi} \partial_z N / h^2\right)$$

with all quantities expressed in units of $L_n$ and $t_n$, $N$ denoting a standard Gaussian (white) stochastic process, and $M_l = l_f / h_n$, where $l_f = \sqrt{h_n T / \gamma}$ is the thermal capillary length. For most materials, $l_f$ is of the order of interatomic distances and is smaller than $L_n$; thus, $M_l < 1$.

As mentioned above, a key concept, which emerged first from experimental (photographic) (35) studies motivated by early theoretical work (43) on the shape of a liquid bridge close to bifurcation (that is, separation of a nascent drop from its parent body), is that of similarity solutions (1, 35) and the associated classification of the neck shapes according to the ratio $l_{\text{obs}}/l_n$ interestingly, $\sqrt{l_f / l_{\text{obs}}} = Oh$, where $Oh$ is the Ohnesorge number occurring often in viscous free-surface flow problems (14, 18).

The appearance of an additional length scale in the SLE simulations (that is, $l_f$ entering Eq. 4 through the number $M_l$) is a direct consequence of the extension to include temperature-dependent stress fluctuations, and its magnitude determines the nature of the jet evolution, including the appearance of solutions other than the universal ones predicted through the deterministic LE (1).

For example, it is known that when the observation scale $l_{\text{obs}}$ (which we take here to be of the order of the neck radius) is in the range $l_{\text{obs}} \gg l_n$, cap-cone neck shapes occur (35), whereas when $l_{\text{obs}}$ is less than $l_n$, but still substantially larger than $l_f$ (that is, $M_l \ll 1$), thread formation will be observed for several decades of reduction in $l_{\text{obs}}$; this is the case
with a further decrease in fluctuations will dominate the breakup process and double-cone neck shapes will appear (Fig. 3D, SLE simulation (44)). For fluids with $M_l > 1$ (e.g., the model propane fluid used in our simulations, where at 210 K, $I_p = 0.56$ nm, $L_p = 2.3$ nm, and $M_l = 0.24$), the range of $I_{obs}$ values corresponding to thread formation is exceedingly narrow, and consequently, a direct transition with decreasing $I_{obs}$ between the cone-cap to the double-cone neck shapes is predicted, as is observed in our MD and SLE simulations. In intermediate cases (that is, $M_l < 1$ but not $M_l < -1$, as in water, where at 300 K, $I_p = 0.24$ nm, $L_p = 13.7$ nm, and $M_l = 0.018$), we expect a tendency for thread formation at $I_{obs} = 10$ nm, but unlike the case of fluids with $M_l < 1$ (e.g., glycerol), this stage is predicted to transform to the double-cone regime early on with a further decrease in $I_{obs}$ (that is, for a neck radius of ~1 nm), without the development of a hierarchy of secondary necks on the threads (as in the glycerol case mentioned above).

References and Notes

2. The review in (1) contains original material as well as a comprehensive discussion and list of references to earlier work on dynamics and breakup processes in liquid jets and other free-surface flow problems.
10. ———, Philos. Mag. 34, 145 (1892).

23. In the early simulations (21), with a relatively small number of Lennard-Jones particles used to investigate stability issues of liquid threads [see also (22)], in addition, the burst of a liquid drop immersed in a second liquid undergoing shear and the establishment of a steady-state flow were considered. Formation, stability, and breakup processes of liquid jets were not studied in (21) and (22).
24. For a review on gene transfer by biolistic processes, see work by P. A. Furth [Mol. Biomol. 7, 139 (1997)].
25. The propane molecules were modeled after (26) and superposed by angle-bending potentials following (27). For the gas, we used a many-body embedded-atom interaction (28). The interaction potentials between the propane and the metal atoms were adopted from (26).
30. To simulate the liquid, we chose $Re = 10^4$, a velocity of propane to the nozzle assembly, we first prepared a 14-nm-diameter, 128-nm-long cylinder of propane, containing ~200,000 molecules that were equilibrated initially at $P_s = 500$ MPa and then frozen instantaneously into an arbitrary liquid configuration. At the start of the simulation, the inlet part of the nozzle assembly was filled with the top part of the frozen propane cylinder, and the dynamics of the propane molecules in the nozzle inlet as well as of the nozzle metal atoms were turned on with Langevin temperature control to the desired temperature $T_D$ above the melting point of propane (Fig. 1A, top left). At the same time, a force $F_p$ (where $A$ is the cross-sectional area of the cylinder and $P_s$ is the chosen driving pressure) was applied at the back of the propane cylinder, and the force acting in the opposite direction from the liquid propane on the frozen cylinder was calculated. The resultant total force was then calculated and used to move the whole frozen propane according to Newton’s equations of motion with an effective mass of 1000 molecules. As the propane cylinder was pushed into the body of the nozzle, the molecules atom by atom entered the nozzle without forming new bonds. When the frozen cylinder was calculated. The resultant total force was then calculated and used to move the whole frozen propane according to Newton’s equations of motion with an effective mass of 1000 molecules. As the propane cylinder was pushed into the body of the nozzle, the molecules atom by atom entered the nozzle without forming new bonds. When the frozen cylinder entered a 0.4-mm-thick zone at the nozzle inlet was also turned on with thermalization to $T_D$. In this way, the propane is forced through the nozzle outlet ($P_o$). To maintain and the preserved pressure at the inlet, new molecules enter the nozzle in a continuous manner. The simulation time zone past the nozzle exit consisted of a rectangular box (vertical ~16 nm by 16 nm by 340 nm [the long dimension in the direction of the propagative jet], and molecules leaving this region were removed from the simulation. With a load-balanced parallelized code (employing an atom decomposition technique), a typical simulation lasted 2 ns, involving the dynamic evolution of ~100,000 propane molecules and 17,000 nozzle metal atoms.
31. The nozzle (Fig. 1A) was modeled as an inlet tube with a 14-nm inner diameter and 4-nm length leading to a 4-nm-long convergent section (with an angle of 135° between the straight part of the inlet tube and the sloping part) and culminating in an orifice of diameter $D$ and a length of 1 nm. The two innermost atomic layers of the metal (gold) surrounding the whole nozzle [including the top layers of the surface of the metal at the outlet] were treated dynamically and were thermally controlled to the desired temperature through Langevin dynamics. The other metal atoms in the metal block were held fixed and the temperature control to the desired temperature $T_D$ above the melting point of propane (Fig. 1A, top left).
32. The time step was adjusted according to the mass of the smallest fluid particles, and two parts of the jet were disconnected whenever their connecting neck became ~2 Å. In the LE and SLE simulations, we used viscosity ($\eta = 0.000011$ Ns/m) and surface tension ($\gamma = 0.0009$ N/m) values that were determined at a temperature of 20 K (i.e., the average molecular kinetic temperature in the jet obtained with the MD simulations) through separate MD simulations of liquid propane, and the velocity and velocity fluctuations obtained from the MD simulation of the jet [evaluated for the nonwetting nozzle case in a thin slice of the jet at $z = 5$ nm, i.e., well beyond the velocity relaxation region (Fig. 28)] were applied as in-flow initial conditions in these hydrodynamics simulations.
33. In (20) [see also (38)], it has been shown that the similarity solution (long threads) is unstable to finite amplitude perturbations, with the critical amplitude going to zero at the singularity, and it was concluded that thermal fluctuations in the liquid can trigger the instabilities. In the numerical simulations in (20), such fluctuations were introduced into the LE simulations in an ad hoc manner.
38. Supplemental material is available at Science Online at www.sciencemag.org/feature/data/1050049.shl.
40. SLE simulations where the inflow initial conditions did not include fluctuations in the velocity yielded very similar results.
41. This research is supported by the U.S. Department of Energy, the Air Force Office of Scientific Research, and the Deutsche Forschungsgemeinschaft (to M.M.). We thank W. D. Lueckel and J. Gao for many useful conversations and R. F. Fox for bringing it to our attention. Simulations were performed on an IBM SP2 computer at the Georgia Tech Center for Computational Materials Science.

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