# Fluid interfaces with very sharp tips in viscous flow

#### Sylvain Courrech du Pont<sup>a</sup> and Jens Eggers<sup>b</sup>

<sup>a</sup>Laboratoire Matière et Systèmes Complexes, , UMR CNRS 7057, Université Paris Diderot , 10, rue Alice Domon et Léonie Duquet, 75205 Paris cedex 13, France; <sup>b</sup>School of Mathematics, University of Bristol, Fry Building, Woodland Road, Bristol BS8 1UG, United Kingdom

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When a fluid interface is subjected to a strong viscous flow, it tends to develop near-conical ends with pointed tips so sharp, their radius 2 3 of curvature is undetectable. In microfluidic applications, tips can be made to eject fine jets, from which micron-sized drops can be produced. Here we show theoretically that the opening angle of the 5 conical interface varies on a logarithmic scale as function of the dis-6 tance from the tip, owing to non-local coupling between the tip and 7 the external flow. Using this insight we are able to show that the tip 8 curvature grows like the exponential of the square of the strength 9 of the external flow, and to calculate the universal shape of the in-10 terface near the tip. Our experiments confirm the scaling of the tip 11 curvature as well as of the interface's universal shape. Our analytical 12 technique, based on an integral over the surface, may also have far 13 wider applications, for example treating problems with electric fields, 14 such as electrosprays. 15

Free surface flows | singularities | selective withdrawal | microfluidics

In many problems of science and engineering, or in daily life, one is confronted with fluid interfaces subject to strong external flows. For example, consider a bubble rising in a viscous fluid (1) (such as in a shampoo bottle), emulsions of drops or bubbles being stirred (2, 3), a viscous layer being withdrawn from near an interface (4), or two fluids meeting in a microfluidic channel (5).

In Fig. 1 we show three typical situations: at the top, a 8 viscous liquid (light) flows out through a circular hole at the 9 bottom of the picture, deforming the interface between liquid 10 and air - a flow geometry known as selective withdrawal (6-11 8). The interface is focused into a near-conical shape, which 12 ends in a tip so small, it can no longer be resolved by optical 13 means. Therefore, in the inset we show profiles obtained from 14 a numerical simulation: as the tip is plotted with increasing 15 resolution (decreasing a), the observed opening angle increases. 16 This is a reflection of the logarithmic variation of the interface 17 slope, which is the key feature of the solution described below. 18

In the middle we see a drop of liquid, whose viscosity is 19 much smaller than that of the outer liquid, being drawn apart 20 by an extensional flow. Once more, very sharp tips are formed 21 at the ends of the drop, and the ends appear conical. At the 22 bottom, a conical interface is produced when a stream of oil is 23 forced by water in a microfluidic assembly (5, 9, 10). However, 24 now a liquid thread escapes from the tip (a phenomenon 25 called tipstreaming (11), whose subsequent decay is a means 26 of producing micron-sized droplets in a highly reproducible 27 fashion (12) of interest for chemical analysis and in soft matter 28 research. 29

Similar conical structures, known as Taylor cones, appear when a liquid is placed in a strong electric field (13–15). However, the Taylor cone is generally unstable, and a tiny jet is emitted from its tip (11, 16–18), akin to the tipstreaming phenomenon described above. This has led to a vast number of applications in spraying and materials processing (19, 20),



**Fig. 1.** Top: A liquid-air interface is deformed into a sharp tip as the fluid below escapes through a hole at the bottom (8);  $\ell_c$  is the capillary length. The dimensionless source strength (flow rate q) is  $\chi = q\eta/(\ell_c^2\gamma) = 6.31$ , where  $\eta$  the dynamic viscosity, and  $\gamma$  the surface tension. The local capillary number (details below) is  $Ca_{tip} = 1.64$ . The inset shows closeups of the tip found from a numerical simulation of the experiment: as one zooms in, the opening angle increases. Middle: A low-viscosity drop in an extensional flow generated by four rollers (2), with capillary number  $Ca = GR_d\eta/\gamma = 0.7$ , and unperturbed drop radius  $R_d = 0.25$ cm, where *G* is the extension rate of the flow. Bottom: A stream of oil is being forced into a sharp tip by water flowing in the reverse direction at a junction of small channels (12). A jet a few microns thick is ejected from the tip (see inset). The scale bar represents 75  $\mu$ m.

but the fundamentals of how a Taylor cone leads to secondary 36

structures has not been understood properly. 37

Almost a century ago, G.I. Taylor set up a research program 38 to understand the production and stirring of emulsions (2, 21), 39 by considering drops subjected to simple external forcing, 40 as seen in Fig. 1. After stirring, very small drops of about 41 1/100 the size of the original drop can be observed (2), which 42 can be attributed to the ejection of small threads, similar 43 44 to those shown at the bottom of the figure. Taylor later developed a theory for the shape of slender drops of small or 45 negligible viscosity (22), which predicted conical ends with 46 tips of vanishing size, violating the assumption of smoothness, 47 on which continuum theory is based (23). 48

Later refinements of Taylor's theory (24) show that it breaks 49 down near the tips. Other theoretical proposals for drop 50 shapes also exhibit singular tips (24, 25). Our later numerical 51 calculations (26) suggest a finite curvature at the tips, which 52 grows exponentially with the square of the external flow speed. 53 It remains to understand this issue of singular tips theoretically, 54 and to calculate the true shape near the ends. This task has 55 hitherto proven impossible, since it represents a fully non-56 linear, three-dimensional free-surface problem, for which few 57 analytical methods of solution exist. In particular, a slender-58 body or lubrication type approach fails here, since the end is 59 not a slender shape. 60

The axisymmetric geometry we consider is particularly 61 significant, since it represents "optimal" focusing of the flow. 62 In the two-dimensional analogue of the same problem, the 63 interface shape is a cusp (27, 28), whose tip traces out a line 64 65 in three dimensions. Since this involves focusing along a whole line, it is much less efficient than focusing on a single point, 66 as in the present problem. As a result, the curvature only 67 increases exponentially with the flow strength in the quasi 68 two-dimensional case, instead of the exponential of the square. 69 Our analytical calculation of the curvature of an axisymmetric 70 tip establishes theoretically that steady flows with surface 71 tension are always smooth, although rounding may occur on 72 very small scales only. 73

The idea of our analysis is that the region near the tip is 74 on a scale very different from the bulk of the flow. Thus if 75 we introduce the distance  $\zeta = (z - z_{tip})\kappa_m$  from the tip (see 76 Fig. 1, middle), made dimensionless with (twice) the mean 77 curvature of the tip  $\kappa_m$ , the radius h(z) of an axisymmetric 78

## Significance Statement

Turn a shampoo bottle upside down, and the rising bubble develops a very sharp tip at its rear. Similar conical structures have been observed placing a drop in an electrical field. Owing to the significance of such structures in chemical processing or in geophysics, they have been studied for almost a century. Here we present a consistent analysis of the shape near the tip in flow. We show that the size of the tip always remains finite, but decreases so rapidly in size with the flow strength, that it may be considered zero for practical purposes.

S. C. du P. performed selective withdrawal experiments; J. E. developed the theory and wrote the manuscript

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<sup>b</sup> To whom correspondence should be addressed. E-mail: jens.eggers@bris.ac.uk

interface can be written in the form

$$h(z) = \kappa_m^{-1} H(\zeta), \qquad [1] \qquad \text{so}$$

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where  $H(\zeta)$  should be a universal similarity solution, independent of the geometry or of the external flow. In (26) we provided numerical evidence for Eq. (1), comparing  $H(\zeta)$  calculated from a drop in extensional flow, and from a selective withdrawal configuration. Below we calculate the similarity function  $H(\zeta)$  analytically for large  $\zeta$ , which corresponds to the limit of large  $\kappa_m$  at a small but constant distance from the tip. We then use this to calculate the scaling of the tip curvature as function of the local flow strength.

Asymptotic analysis near the tip. To calculate the profile and the flow near the ends, we consider the simplest case of axisymmetric viscous (Stokes) flow, with vanishing viscosity inside. Even if inertia becomes important on a large scale, this will still be a correct description locally, where the local Reynolds number is small, i.e. viscous forces are much stronger than inertial forces. An axisymmetric cross section is a good approximation even if the external flow is not axisymmetric (29, 30), for example in a shear flow. A finite viscosity fluid inside the tip eventually leads to a tipstreaming bifurcation (22, 31), but we will be describing the regime before this occurs, yet tips 100 have become very sharp. 101

Instead of solving the flow equations in the bulk, with 102 boundary conditions applied at the free surface, we use the 103 equivalent boundary integral description (32), which is formu-104 lated on the free surface alone, and which has proven extremely 105 effective addressing free surface problems numerically with 106 very high resolution (26, 32, 33). However, this technique is 107 very unwieldy as an analytical tool, since the surface to be in-108 tegrated over is unknown a priory. Here we break new ground 109 by using the boundary integral technique to find analytical 110 solutions, using the fact that the interface slope is changing 111 very slowly. 112

The integral equation to be solved for the velocity  $\mathbf{v}(\mathbf{x}_1)$ on the surface S (see Fig. 1, middle, more details in *Materials* and Methods) is (32)

$$\frac{\mathbf{v}(\mathbf{x}_1)}{2} = \mathbf{v}^{(ext)}(\mathbf{x}_1) - \int_S \kappa(\mathbf{x}_2) \mathbf{J} \cdot \mathbf{n} \, \mathrm{d}\sigma_2 - \int_S \mathbf{v}(\mathbf{x}_2) \cdot \mathbf{K} \cdot \mathbf{n} \, \mathrm{d}\sigma_2.$$
[2]

Velocities are written in units of the capillary speed  $v_c \equiv \gamma/\eta$ , 117 with  $\gamma$  the surface tension and  $\eta$  the dynamic viscosity. In 118 Eq. (2),  $\mathbf{v}^{(ext)}(\mathbf{x}_1)$  is an externally imposed velocity,  $\kappa$  the 119 mean curvature, and  ${\bf n}$  the outward normal. The kernel  ${\bf J} \cdot {\bf n}$  is 120 the velocity at  $\mathbf{x}_1$ , generated by a point force at  $\mathbf{x}_2$  in Stokes 121 flow, and  $\mathbf{K} \cdot \mathbf{n}$  is the corresponding stress tensor at  $\mathbf{x}_1$ . 122

Since the flow is axisymmetric, one can perform the az-123 imuthal integration explicitly (32), and choose the dimen-124 sionless distance  $\zeta$  from the tip as the integration variable. 125 Transforming to the logarithmic variable  $l = \ln \zeta$ , we obtain 126 from Eq. (2)127

$$\frac{\mathbf{v}(l_1)}{2} = v_{\text{tip}} \mathbf{e}_z + \mathbf{v}^{(J)} - \int_{-\infty}^{\ln(L\kappa_m)} \mathbf{v}(l_2) \zeta_2 \mathbf{k}(l_1, l_2) dl_2, [3] \quad \text{126}$$

$$\chi^{(J)}(l_1) = -\int_{-\infty}^{\ln(L\kappa_m)} \zeta_2(\kappa(l_2)/\kappa_m) \mathbf{j}(l_1, l_2) dl_2,$$
 [4] 129

where L is a characteristic size of the setup, such as a drop 130 size, and  $v_{\rm tip} = v_z^{(ext)}(z_{\rm tip})$  is the external velocity at the tip. 131

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**Fig. 2.** Schematic representation of our treatment of Eq. (6). The self-similar profile  $H(\zeta)$  has a slope H' which changes on a logarithmic scale, see inset. Integrands like  $\zeta_2 k_{zz}$  are peaked at  $l_2 = l_1$ , except  $j_z$ , which decays like  $j_z^e$  for large  $l_2$ .

The kernels **j** and **k** correspond to  $\mathbf{J} \cdot \mathbf{n}$  and  $\mathbf{K} \cdot \mathbf{n}$  in Eq. (2), integrated over  $\theta$ , and written in units of the tip curvature  $\kappa_m$ .

The integrands in Eq. (3) and Eq. (4) are now invariant 135 under a scale transformation: if the surface were a perfect 136 cone of slope s, the integrands would be functions of s and 137  $\Delta = l_2 - l_1$  alone. Thus if all integrals were convergent in the 138 limit  $L\kappa_m \to \infty$ , each term in Eq. (3) would be a function of 139 s alone, and a cone would result as a (similarity) solution to 140 the flow equations. However, as first pointed by Taylor (22), 141 and confirmed by Buckmaster (24), such a conical solution 142 does not exist. Indeed, we will see that the z-component 143 of  $\mathbf{v}^{(J)}$  in Eq. (4) is in fact divergent for  $L\kappa_m \to \infty$ , so L 144 cannot be eliminated from the problem. Instead, we must 145 keep the upper limit in Eq. (4) finite, and cancel the resulting 146 contribution against the external velocity  $v_{tip}$ , introducing 147 an intrinsic coupling between the external flow and the local 148 behavior at the tip. 149

The key idea of our approach is to suppose that the interfacehas the form (cf. Fig. 2):

$$H(\zeta) = \zeta s(\ln \zeta) \equiv \zeta s(l), \qquad [5]$$

where s(l) is a local slope, which varies on a logarithmic scale. We will show below that for large l (far from the tip), which is the asymptotic behavior which interests us,  $s(l) \propto 1/\sqrt{l}$ , i.e. the slope becomes small, as shown in the inset of Fig. 2. In doing so, we are always in the limit  $\zeta \ll L\kappa_m$ , and hence the inner solution near the end applies. If we write Eq. (3) in the form

$$\frac{1}{2} \begin{pmatrix} v_z \\ v_r \end{pmatrix} = \begin{pmatrix} v_{\rm tip} + v_z^{(J)} - K_1 - K_3 \\ v_r^{(J)} - K_2 - K_4 \end{pmatrix}, \quad [6]$$

<sup>161</sup> in evaluating the dominant contribution to each term we can <sup>162</sup> assume the integral to be over a cone of constant slope (since <sup>163</sup> s(l) is varying very slowly), which is a tractable problem.

Our treatment of Eq. (6) is illustrated in Fig. 2: to find  $v_{z/r}$  at a point  $l_1 = \ln(\zeta_1)$ , according to Eq. (3) and Eq. (4), we have to perform an integral over  $l_2$ . However, as shown for the example of  $\zeta_2 k_{zz}$ , integrands are peaked at  $l_2 = l_1$ , and hence to leading order the contribution to  $K_1$  is proportional to  $v_z(l_1)$ , multiplied by the area of the peak. The exception is the integral over  $j_z$ , which decays very slowly for large  $l_2$ . Namely, taking a cone of slope  $s_2$  as the interface, one finds that for  $\Delta \to \infty$  171

$$j_z(l_1, l_2) \to j_z^e \equiv -\frac{s_2^2(1 - s_2^2)}{4\left(1 + s_2^2\right)^{3/2}} \approx -\frac{s_2^2}{4},$$
 [7] 173

where  $j_z^e$  is shown as the red line in the plot of  $j_z$  in Fig. 2. 174 In the same limit of small slopes,  $\zeta_{2\kappa}/\kappa_m \approx 1/s_2$ ; thus the 175 integrand of  $v_z^{(J)}$  in Eq. (4) is  $\zeta_{2}(\kappa/\kappa_m)j_z \approx -s_2/4$ . On 176 the other hand, for  $\Delta \to -\infty$ ,  $j_z(l_1, l_2)$  vanishes rapidly (cf. 177 Fig. 2), and we obtain 178

$$v_z^{(J)} \approx \frac{1}{4} \int_{l_1}^{\ln(L\kappa_m)} s(l_2) dl_2.$$
 [8] 179

Clearly since  $s \propto 1/\sqrt{l_2}$ , this integral makes a contribution  $\propto \sqrt{\ln(L\kappa_m)}$  from its upper limit. Once this has been removed by balancing it against  $v_{\rm tip}$  in Eq. (6), we anticipate that  $v_z \propto \sqrt{l_1}$  from the lower limit. (83)

Now we compute the other integrals in the same spirit as integrals over cones, but using the fact that the remaining kernels are localized about  $\Delta \approx 0$ , and decay for large  $\Delta$ . Thus since  $\mathbf{j}(l_1, l_2) = \mathbf{j}(s_1, \Delta)$  for a conical interface, we can approximate

$$v_r^{(J)} \approx (\zeta_1 \kappa(l_1)/\kappa_m) \int_{-\infty}^{\infty} j_r(s_1, \Delta) d\Delta = 0,$$
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which is confirmed by an expansion of  $j_r(s_1, \Delta)$  for small  $s_1$ . 190

The kernels  $\mathbf{k}(l_1, l_2) \approx \mathbf{k}(s_1, \Delta)$  can be treated in the same way. Beginning with z-component of Eq. (6), 192

$$K_1 \equiv \int_{-\infty}^{L\kappa_m} \zeta_2 k_{zz} v_z(l_2) dl_2 \approx v_z(l_1) \int_{-\infty}^{\infty} \zeta_2 k_{zz}(s_1, \Delta) d\Delta, \qquad \text{193}$$

where we have used, as illustrated in Fig. 2, that  $\zeta_2 k_{zz}$  is strongly peaked, so that  $v_z(l_2)$  varies little over width of the peak. The remaining integral (the area of the peak) can be shown to be 1/2 to leading order in an expansion for small  $s_1$ (see *Materials and Methods*); thus  $K_1 \approx v_z(l_1)/2$ . In the same vein,

$$K_3 \equiv \int_{-\infty}^{L\kappa_m} \zeta_2 k_{zr} v_r(l_2) dl_2 \approx v_r(l_1) \int_{-\infty}^{\infty} \zeta_2 k_{zr}(s_1, \Delta) d\Delta.$$
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Now the integral is  $\approx s_1/2$  to leading order, which becomes 201 small, and  $K_3$  can be neglected compared to  $K_1$ . 202

Coming to the r-component of Eq. (6), to a first approximation we have 204

$$K_4 \equiv \int_{-\infty}^{L\kappa_m} \zeta_2 k_{rr} v_r(l_2) dl_2 \approx v_r(l_1) \int_{-\infty}^{\infty} \zeta_2 k_{rr}(s_1, \Delta) d\Delta. \qquad \text{205}$$

To leading order, the value of the last integral is -1/2, so that  $K_4 \approx -v_r/2$ ; but this cancels  $v_r/2$  on the left, and we have to go to the next order, which yields  $K_4 \approx v_r(l_1)(-1/2 + s_1^2/4)$ . The remaining component is 209

$$K_2 \equiv \int_{-\infty}^{L\kappa_m} \zeta_2 k_{rz} v_z(l_2) dl_2 \approx v_z(l_1) \int_{-\infty}^{\infty} \zeta_2 k_{rz}(s_1, \Delta) d\Delta, \qquad \text{210}$$

but the integral is again zero to leading order, and even the next 211 order vanishes. To capture the leading nonzero contribution, 212 we expand  $v_z(l_2) = v_z(l_1) + v'_z(l_1)\Delta + \dots$ , using that  $k_{rz}$  213

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**Fig. 3.** Comparison between the similarity solution  $H(\zeta)$  as predicted by theory (black solid line), and six different experimental data sets for different values of  $\kappa_m \ell_c$ , rescaled according to Eq. (1) (colored lines);  $\ell_c = \sqrt{\gamma/(\rho g)} = 1.49$ mm is the capillary length. The main panel shows all six experimental data sets on a logarithmic scale, the inset only the first three sets on a linear scale. The dashed line is the asymptotic behavior Eq. (9) with  $l_0 = 2.55$ . For the first three experimental profiles,  $\kappa_m$  has been measured directly, (colored dots in Fig. 4), for the profiles with the highest tip curvatures,  $\kappa_m$  has been calculated from a numerical simulation (colored pluses in Fig. 4).

is peaked around  $\Delta = 0$ . Thus an improved approximation becomes

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$$K_2 \approx v'_z(l_1) \int_{-\infty}^{\infty} \Delta \zeta_2 k_{rz}(s_1, \Delta) d\Delta \approx -v'_z(l_1) \frac{s_1}{2}$$

217 to leading order.

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This completes the necessary calculations of all the integrals. Next we will interpret Eq. (6) as a dynamical system for  $s(l), v_z(l)$  and  $v_r(l)$ .

## Flow equations and scaling. To summarize, Eq. (6) becomes to leading order, putting $l_1 = l \equiv \ln \zeta$ ,

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$$v_z = v_{\rm tip} + \frac{1}{4} \int_l^{\ln(L\kappa_m)} s(l_2) dl_2, \quad 0 = sv'_z/2 - v_r s^2/4.$$
 [9]

The contribution  $v_{\rm tip}$  of the external flow cancels against the 224 contribution from the upper limit of the  $v_z^{(J)}$  integral, as we 225 see below. However, we first calculate s(l) to leading order. 226 To close the system Eq. (9), we use that streamlines must be 227 parallel to the free surface, and thus  $v_r/v_z = H' = s + s' \approx$ 228 s. Differentiating the First of Eq. (9) with respect to l, we 229 find  $v'_z = -s/4$ , and substituting into the second equation 230 gives  $v_r = -1/2$ . This corresponds to the familiar result 231  $(34, 35) - v_c/2$  for the rate of collapse of a cylindrical cavity, 232 remembering that velocities have been made dimensionless 233 with the capillary speed  $v_c$ . It follows that  $v_z = -1/(2s)$  from 234 the kinematic condition, leading to  $s' = -s^3/2$ . Solving, we 235 find the asymptotic solution far from the tip (but still in the 236 self-similar region) to be 237

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$$s = (l - l_0)^{-1/2}, \quad v_r = -\frac{1}{2}, \quad v_z = -\frac{(l - l_0)^{1/2}}{2}, \quad [10]$$

consistent with the previous assumption of small slopes for  $l \to \infty$ . In Fig. 3, Eq. (10) (dashed line) is found to be in excellent agreement with  $H(\zeta)$  as calculated from Eq. (1) (solid line), using a full numerical simulation of a drop in an extensional flow (26); the constant of integration  $l_0$  was adjusted to match with the numerical profile. The major conclusion from Eq. (10) is that the pointed ends seen for example in Fig. 1 only appear conical since *s* is nearly constant over an observable range of scales, but this opening angle will change when the scale of observation is changed, and one zooms into the tip. 249

The theoretical tip similarity solution  $H(\zeta)$  (black solid 250 line) is compared in Fig. 3 to six experimental data sets 251 from a selective withdrawal experiment, described in detail 252 in (8) (colored lines). A container 3 cm wide is half filled 253 with silicone oil of viscosity  $\eta = 30$  Pa s and surface tension 254  $\gamma = 2.13 \times 10^{-2} \text{Nm}^{-1}$ . The liquid is evacuated through a 255 circular sink hole of 1 mm diameter in a solid plate, at a 256 constant flow rate of  $q = 9.97 \times 10^{-9} \text{ m}^3/\text{s}$ , but replenished at 257 a rate which is slightly smaller, so that the distance between 258 the hole and the mean liquid level decreases adiabatically, over 259 a period of several hours. 260

As the liquid-air interface comes closer to the sink hole, it 261 is increasingly deformed. We record the distance  $z_t$  between 262 interface tip and the hole, as well as the shape of the inter-263 face. The experimental cell is lit from behind, so that light is 264 refracted away by the interface, whose cross section appears 265 black (cf. Fig. 1), top. The measured interface shape (colored 266 lines) has been rescaled according to Eq. (1), with tip curva-267 tures given in the figure. The crossover between the similarity 268 solution  $H(\zeta)$  and the outer solution, which is shaped by the 269 sink flow out of the container, takes place at a fixed outer scale 270  $z - z_{\rm tip}$ . Thus to increase the range of the similarity variable  $\zeta$ 271 over which to compare to the experiment, one has to increase 272 the tip curvature. 273

For the first three profiles of Fig. 3, the tip curvature  $\kappa_m$  has 274 been measured directly by interpolating the tip region. The 275 teal curve with the highest curvature shows significant noise, 276 as it has been zoomed in to the limit of our resolution. To 277 be able to compare to theory over a larger range, we included 278 three profiles with a much larger tip curvature, focusing on 279 the far field. The tip sizes of these profiles have become so 280 small, that they are below our optical resolution. Instead, 281 we calculate  $\kappa_m$  from a full numerical simulation of the flow 282 equations, which matches closely the experimental geometry. 283 We can argue on the basis of Fig. 4 below that the numerical 284 estimates of the curvature are very reliable. 285

To compute the tip curvature  $\kappa_m$  analytically, we insert 286 Eq. (10) into the First of Eq. (9) to find to leading order 287

$$-\frac{\sqrt{l}}{2} \approx -\operatorname{Ca_{tip}} + \frac{1}{4} \int_{l}^{\ln(L\kappa_m)} \frac{dl_2}{\sqrt{l_2}},$$
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where the local capillary number  $\operatorname{Ca_{tip}} = -v_{tip}$  is the dimensionless z-velocity at the tip. From this we find  $\operatorname{Ca_{tip}} \approx 290$  $\sqrt{\ln(L\kappa_m)}/2$ , and thus

$$\kappa_m \propto L^{-1} \exp\left(4 \operatorname{Ca}_{\operatorname{tip}}^2\right),$$
 [11] 292

which is the main result of this paper. For Eq. (11) to be valid,  $Ca_{tip}$  needs to be sufficiently large so that there is an appreciable range over which *s* follows the scaling Eq. (10). Owing to the faster-than-exponential growth of  $\kappa_m$ , Eq. (11) shows that while the curvature always remains finite, it can easily reach microscopic dimensions even at moderate values of the capillary number. For example, taking the drop in the



Fig. 4. The dimensionless tip curvature  $\kappa_m \ell_c$ , as function of the square of the local capillary number  $\mathrm{Ca_{tip}},$  based on the  $\mathit{unperturbed}$  velocity at the tip. The solid line is a numerical simulation (details in (26)), parameters the same as Fig. 1 (top). The open orange circles are the experimental measurements, applying no adjustable parameters. The filled circles correspond to the measured curvature of the first three profiles of Fig. 3, the pluses are the curvatures for the last three profiles, based on the numerical simulation. The dashed line has a slope of  $a=4/\ln 10$ , corresponding to the theoretical prediction Eq. (11).

middle of Fig. 1, Eq. (11) would predict a radius of curvature 300 of about  $10^{-89}$  m! This shows that while the tip itself is often 301 too small to observe, what is experimentally relevant is the 302 slow variation of the slope described by our asymptotic theory 303 Eq. (10). 304

In Fig. 4 we test Eq. (11) against experiment (circles) and 305 simulation (solid line). As the capillary number increases, the 306 logarithm of the curvature, plotted as function of the square 307 of the capillary number at the tip, quickly converges toward a 308 straight line. The dashed line has a slope of  $4/\ln 10$ , which 309 is the asymptotic prediction of Eq. (11), and is seen to agree 310 very well with both simulation and experiment. It can be 311 shown (36) that the slight over prediction of the slope can be 312 traced to a slow variation of the *prefactor* in Eq. (11) on the 313 capillary number. 314

The orange circles in Fig.4 come from measurements of 315 the tip curvature in the selective withdrawal geometry for the 316 same parameters as in Fig. 3. The external velocity at the tip 317 was calculated from the flow field of a point source inside a 318 solid wall, evaluated at  $z_t$ . The black solid line is the result of 319 a numerical simulation performed using the boundary integral 320 method (26). The simulation assumes a point source in a solid 321 wall, unbounded in the horizontal direction (instead of a finite 322 experimental cell), an approximation which works very well, 323 except far from the center (26). No adjustable parameters 324 were applied to achieve the near-perfect agreement between 325 simulation and experiment. The colored solid circles are also 326 obtained from direct measurements of the tip curvature, and 327 correspond to the first three profiles of Fig. 3. The pluses have 328 been picked out from the solid line, and mark the curvatures 329 of the last three profiles of Fig. 3. 330

**Conclusions.** In this paper, we have given the first analytical 331 description of axisymmetric interface tips, created by a con-332 verging viscous flow. The shape of the interface near the tip 333 is universal, independent of flow conditions. The tip curva-334 ture always remains finite, but increases dramatically with 335 flow strength. Thus although surface tension always keeps 336

the interface smooth, the continuum hypothesis will in many 337 cases fail in practice. Moreover, it follows that the numerical 338 solution of free-surface problems places extreme demands on 339 the spatial resolution. Our analytical solution can be used to 340 provide effective boundary conditions on intermediate scales, 341 to drastically reduce the necessary numerical effort. Numerical 342 calculations agree well with our experiments, which resolve 343 the tip size down to a few microns. 344

Using the theoretical framework established in this paper, 345 we hope to address the important issue of tipstreaming, used to 346 produce colloidal drops (5, 12), and studied numerically in (37, 12)347 38). The same is true for a much wider class of flows involving 348 electric fields, where the analogous flow is known as the cone-349 jet mode (11, 39). In (12), it was shown that in a suitable 350 microfluidic geometry (see Fig. 1, bottom), the transition to 351 tipstreaming is of second order, i.e. the ejected thread can 352 be arbitrarily thin. Thus we expect the transition between a 353 tipstreaming state and a closed tip to be continuous, which 354 means that previous theories of tipstreaming, based on slender-355 body theory (40, 41), suffer from the same shortcomings as 356 for a tipped state. By adding an inner fluid to our description, 357 we expect to be able to include the thread into our theory, 358 finally being able to address the tipstreaming problem in a 359 consistent fashion. 360

#### Materials and Methods

The idea of the boundary integral Eq. (2), is to use the linearity 362 of the Stokes equation to write the velocity as a superposition of 363 an externally imposed velocity field  $\mathbf{v}^{(ext)}$ , and the velocity  $\mathbf{v}^{(J)}$ 364 produced by surface tension. The latter can be seen as coming 365 from a collection of point forces of strength  $\gamma \kappa \mathbf{n}$ , distributed over 366 the surface S. This makes  $\mathbf{v}^{(J)}$  a superposition of Stokeslets  $\mathbf{J}$ , 367 integrated over the free surface S, with  $\gamma \kappa \mathbf{n}$  as a weight. 368 369

The kernels in the boundary integral description Eq. (2) are (32)

$$J_{ij}(\mathbf{r}) = \frac{1}{8\pi} \left[ \frac{\delta_{ij}}{r} + \frac{r_i r_j}{r^3} \right], \quad K_{ijk}(\mathbf{r}) = -\frac{3}{4\pi} \frac{r_i r_j r_k}{r^5}, \qquad [12] \quad \text{370}$$

where  $\mathbf{r} = \mathbf{x}_1 - \mathbf{x}_2$ , so that  $\mathbf{J} \cdot \mathbf{f}(\mathbf{x}_2)$  is the velocity at  $\mathbf{x}_1$ , generated 371 by a point force  $\mathbf{f}$  at  $\mathbf{x}_2$ , and  $\mathbf{K} \cdot \mathbf{f}(\mathbf{x}_2)$  is the stress at  $\mathbf{x}_1$ . The 372 factor 1/2 on the left-hand side of Eq. (2) and the second integral 373 over the stress  $\mathbf{K}\cdot\mathbf{n}$  corrects for the jump in viscosity between the 374 two phases. 375

Putting  $\mathbf{x}_1 = (y_1, 0, x_1)$ ,  $\mathbf{x}_2 = (y_2 \cos \theta, y_2 \cos \theta, x_2)$ , and  $\overline{\mathbf{n}} =$ 376 (-h'(z), 1), we can perform the integration with respect to the 377 azimuthal angle  $\theta$ : 378

$$\mathbf{j} = y_2 \int_0^{2\pi} \mathbf{J} \cdot \overline{\mathbf{n}} d\theta_2, \qquad [13] \quad \mathbf{37}$$

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and analogously for  $\mathbf{K}$ , yielding the kernels  $\mathbf{j}$  and  $\mathbf{k}$  in Eq. (3) and 380 Eq. (4). They result in well-known expressions (32) involving elliptic 381 integrals in terms of the (logarithmic) coordinates  $l_1, s_1$  and  $l_2, s_2$ . 382

The entries in Eq. (6) are calculated by performing the integrals 383 over  $l_2$  assuming a conical interface with slope  $s_1 = s(l_1)$ . This 384 results in integrals over the kernels, such as  $\zeta_2 k_{zz}(s_1, \Delta)$  for the 385 case of  $K_1$ , but as a function of  $\Delta = l_2 - l_1$  alone. The dominant 386 contribution to the integral comes from a central region of size  $s_1$ . 387 At higher orders, contributions for  $\Delta \geq 1$  also need to be taken into 388 account, but can be disregarded here. 389

To capture contributions at scale s, we put  $\xi = \Delta/s$  and expand the kernel:

$$\zeta_2 k_{zz} = K_1^{(-1)}(\xi) s^{-1} + K_1^{(0)}(\xi) + \dots$$
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Here  $K_1^{(-1)}(\xi)$  is an even function, which is expressible in terms 393 of the elliptic integrals  $E\left(2/\sqrt{4+\xi^2}\right)$  and  $K\left(2/\sqrt{4+\xi^2}\right)$  (42). 394 Thus the integral becomes

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$$\int_{-\infty}^{\infty} \zeta_2 k_{zz} d\Delta = \int_{-\infty}^{\infty} K_1^{(-1)} d\xi + O(s^2) = \frac{1}{2} + O(s^2),$$

 $_{\rm 397}$   $\,$  as shown by an explicit calculation. The remaining integrals to

calculate  $K_2 \dots K_4$  can be evaluated in a similar fashion, expanding the integrand in s at constant  $\xi$ .

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