Singularity during the Onset of an Electrohydrodynamic Spout

Lene Oddershede* and Sidney R. Nagel

The James Franck Institute, University of Chicago, Chicago, Illinois 60637

(Received 24 January 2000)

When one applies a sufficiently large electrical field normal to the surface of a conducting liquid the fluid rises in a spout to form a jet leaving the surface. Using high-speed photography, we have studied the development of this electrohydrodynamical spout and found that its curvature and height can be scaled with respect to a critical time indicating the presence of a critical point in the dynamics underlying the instability.

PACS numbers: 47.20.Ma, 68.10.–m

In some situations, an initially smooth fluid interface spontaneously develops singularities in its shape when perturbed by a uniform force. A commonly observed example is a pendant water drop which, under the influence of gravity, can become unstable; as the drop falls it breaks up into two or more droplets. At the point of breakup where the neck has become infinitely thin, the curvature, and possibly the flow velocities as well, must diverge [1,2]. The system has been forced by the dynamics to go through a critical point at the instant of snapoff. Around this point, the properties of the interface and the flow can be understood in terms of a similarity solution which allows a simplified description of the dynamics [3,4]. Of course, not all cases with smooth initial conditions and smooth forcing are compelled to progress through such a singular point. However, if such a situation does prevail, a deeper understanding of the dynamics may be obtained. In this paper we report the appearance of such a singularity in the dynamics in another well-studied problem, namely, the ejection of a jet of fluid due to the presence of a large electric field normal to the liquid surface.

When an electric field is applied in a direction normal to the surface of a conducting fluid, that surface will distort and create a hump so as to take advantage of the electric field energy. For a sufficiently strong field, such a hump becomes unstable and forms a spout emitting charged droplets. As the field is increased further, the spout stabilizes, emitting liquid at a constant rate. This phenomenon is well known, although not very well understood, and is used in controlled industrial applications where the production of a charged jet or a monodisperse spray is desired as, for instance, in the injection of fuel for combustion [5,6], in the creation of sprays for painting applications, and in the production of ionized beams for mass spectrometry of biomolecules [7]. The phenomenon also arises in an uncontrolled manner when "water trees" penetrate high-voltage cables causing power blackouts [8,9] or when lightning bolts hit the surface of a lake. The discovery we report here of a singularity in the dynamics may help identify the correct balance of forces in the production of the spray.

The first photos of such phenomena were taken in 1917 by Zeleny [10]. Taylor made further investigations of the phenomena, using both oil-water and air-water interfaces, and noted that, neglecting viscous stresses, the steady-state structure should be conical with a constant semiangle of 49.3°, the *Taylor angle* [11]. He was able to observe this angle in a cell with specially machined boundary conditions. In addition, Taylor considered the stability of an interface in the presence of an electric field [12] and the jet emitted from the cone [13]. Investigations [14–17] of this electric-field-induced instability have focused on understanding features such as the diameter of, and the current carried by, the jet [18,19], the length of the jet before breaking into a spray, the angle of the spray, and the size and charge distribution of droplets emitted [20–27]. No theory yet describes all the important aspects of this phenomenon. Much of this theory [14,21,22,25,28] is based on the assumption that the inherent *steady-state* shape of the jet far from the ejected spout is a cone, as described by Taylor. In this paper, we show that, for one geometry, during the formation of the spout the instability progresses to a nonconical shape.

In the design of our apparatus we have focused on studying the formation of a spout and not on maintaining a stable spout, which has been the goal in most other investigations [21,23]. A transparent container holds a conducting fluid (water) covered by a second immiscible liquid of insulating silicon oil. The upper fluid, silicon oil, was used because the instability occurs at about 5×10^5 V/m which is greater than the breakdown voltage of air (2×10^5 V/m). Silicon oil has a high breakdown potential and has a dielectric constant of 2.5, so the voltage needed to create the instability is reduced from that in air. The silicon oil used in the experiments is from Fischer Scientific #S159-500 and has a kinematic viscosity of 50×10^{-6} m^2/s, a surface tension against air of 0.0208 N/m, and a density of 0.963 g/ml. In order to control the conductance of the liquid, NaCl was added to the distilled water; the concentration of NaCl in the water was $[\mathrm{NaCl}] = 8 \times 10^{-2} M$ corresponding to an equivalent conductance of approximately 10^{-2} m^2 S mol^{-1} l.

 0031-9007/00/85(6)/1234(4)$15.00

In order to fix the position of the spout, a cylindrical electrode (with lower end rounded into a radius of 0.32 cm) was used as the upper electrode and immersed in the oil. A conducting mesh immersed in the water was the lower electrode. In order to apply the voltage instantaneously, a capacitor with a capacitance about 3 orders of magnitude larger than the capacitance of the electrodes in the liquid was placed in parallel with the experiment. When a high voltage switch was activated, charge flowed from the storage capacitor to the experimental electrodes and within less than a microsecond created the desired voltage difference between the surface of the water and the upper electrode. With respect to the time for the formation of a spout (typically a few tenths of a second) it was possible to build up the voltage difference instantaneously. We used a "Hycam" high-speed film movie camera capable of taking several thousand frames a second. The precise timing between frames was obtained from a light that marked the border of the film at a constant frequency. The film frames were digitized and analyzed by image analysis software develped for this purpose.

We have analyzed the data from two experiments which have values of the initial electric field that differ approximately by a factor of 2. For the first experiment, the voltage applied instantaneously was 10.5 kV and the distance between the surface of the water and the upper electrode was 1.5 cm. For the second experiment, the applied voltage was 16 kV and the distance to the upper electrode was 1.2 cm. Twelve snapshots taken from the first experiment are shown in Fig. 1. The first few frames show the initially flat interface growing into a hump. The top of the hump becomes increasingly sharp as the spout grows. Between photos 6 and 7, the top of the spout forms a cusp. Thereafter a jet is emitted. In the tenth photo, the spray has reached the upper electrode and current flows in a jagged line. In the last two images, the spray is repelled from that electrode. An analysis of the shape of the entire spout will be given elsewhere [29].

As the spout develops, the shape near its top appears somewhat conical, as in the sixth image in Fig. 1. At this point the semiangle of the cone is 44° (less than 49.3°, the "Taylor angle" [11]). de la Mora has observed semiangles between 32° and 46° [22] and our observations fit into this interval. In the literature the cone has often been denoted as the final shape [11,14,21,22,25] or has been considered to be the steady-state shape starting from which one can add an additional component to form a jet [29]. The conical region that we see does not extend all the way to the apex. Near the peak, the radius of curvature is definitely nonzero so that although *away* from the apex the shape may appear conical, asymptotically close to the peak the shape is not conical. As we show below, the approximate conical shape is observed only as a transition during the evolution towards a nonconical shape where the tip curvature does appear to diverge.

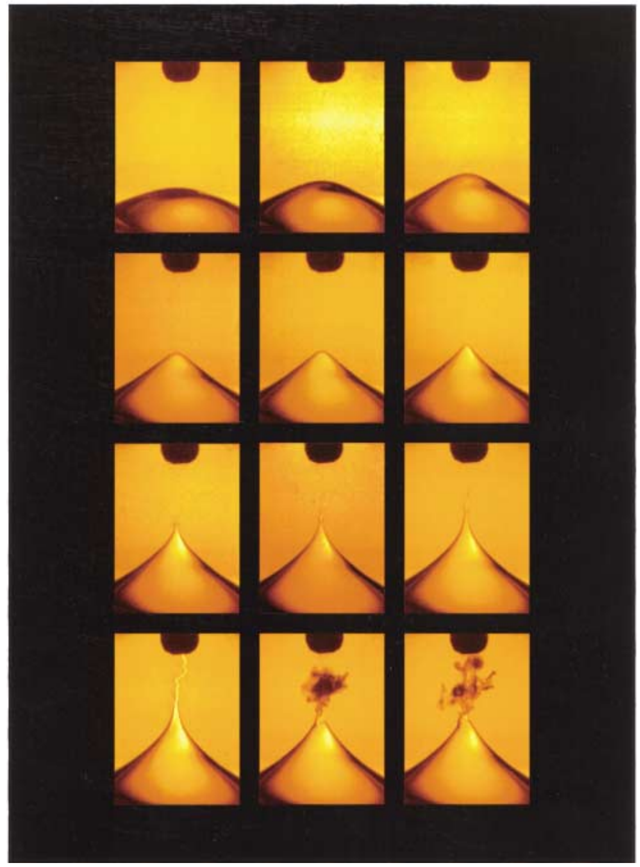

FIG. 1 (color). Photographs from a high-speed movie of experiment I showing the time development of an electrohydrodynamic spout in an experiment where an electrical field was suddenly applied across an oil-water interface. To give the scale of the photographs, the width of the upper electrode which is visible in all photos is 6.4 mm. The first six frames (counting from the upper left corner) show how the initially flat interface grows into a hump whose apex curvature increases with time. Between the sixth and seventh frames the curvature of the tip diverges. Thereafter, the instability continues to develop, emitting a jet as can be seen in frames 7, 8, and 9. In frame 10 the spray has reached the upper electrode and current flows in the jagged, "lightning strike," line seen in the photograph. In frames 11 and 12 the charged spray is repelled from the upper electrode towards the body of the spout. The photographs are taken at the following times from the critical point t^*: Photos No. 1: 0.068 s, No. 2: 0.039 s, No. 3: 0.020 s, No. 4: 0.0058 s, No. 5: 0.0029 s, No. 6: 0.0001 s, No. 7: −0.0028 s, No. 8: −0.0056 s, No. 9: −0.0085 s, No. 10: −0.0098 s, No. 11: −0.011 s, No. 12: −0.012 s.

The height and curvature of the spout can be determined from the high-speed movie as a function of time. Before a critical time t_c, the curvature at the top of the spout is well defined; at that time, the rounded top turns into a cusp and begins to emit droplets. Using the edge-detected interface, the tip has been fit to a Gaussian function centered along the symmetry axis: $\mathrm{fit}(x) = a\exp(-x^2 b)$. $\kappa = -2ab$ is the curvature of the top of the spout. The parameters a and b result from fitting the function to the shape at each frame. For fitting, we chose a narrow region near the tip and checked that κ did not vary with length of the interval used. The inset in Fig. 2 demonstrates the fit of the above Gaussian function to the edge-detected apex of the instability for the frame closest to, but less than, t_c for experiment II. Other functions besides a Gaussian give similar results for κ.

Testing for power-law behavior in both experiments, we plot on double logarithmic axes the curvature κ versus $t^* \equiv t_c - t$, that is, the time t measured from a critical time t_c. This is shown in Fig. 2. The data of experiment I, which is analyzed for scaling and is shown in Figs. 2 and 3, correspond to the stages of the instability represented in images 1–6 in Fig. 1, that is, before the tip forms a cusp. For each experiment, we varied t_c, constrained to be within one movie frame after the last data point used, in order to find the value for which the data were straightest on this plot with the same slope for both sets of data. The curvatures, shown in Fig. 2, are consistent with scaling behavior over 2.5 decades of time t^*:

$$\kappa \propto (t^*)^{-\gamma} \tag{1}$$

for $t < t_c$ with scaling exponent $\gamma = 0.80 \pm 0.05$.

A plot of apex height h versus time is shown in Fig. 3. For each experiment, $\log h^*$ is plotted versus $\log t^*$ for $t < t_c$ where $h^* \equiv h_c - h$. The exact height h_c of the apex at t_c must lie between the heights measured in the two frames of the movie adjacent to t_c which was determined from Fig. 2. Within this strong constraint, h_c is chosen to make the data lie on the best straight line. In Fig. 3, the data from both experiments again exhibit scaling over 2.5 decades of time:

$$h^* \propto (t^*)^{\beta} \tag{2}$$

for times $t < t_c$ with scaling exponent $\beta = 0.42 \pm 0.04$. As was the case for the curvature, the same scaling exponent is consistent with both sets of experimental data. This suggests that the observed scaling relations are independent of the size of the applied electrical field.

For our experiments, the observed scaling exponents are related by

$$\gamma = (1.91 \pm 0.22)\beta \,. \tag{3}$$

One can write a scaling relation for the curvature and height close to the critical time t_c

$$\kappa \propto (h_c - h)^{-1.91 \pm 0.22}. \tag{4}$$

It follows that, close to t_c, the shape of the instability is not conical (i.e., linear around the apex) but is rather approximately a square-root cusp.

Using high-speed photography, we have followed the development in our two runs of an electrohydrodynamical spout revealing the shape of the instability during its onset. Initially, the instability is a "hump" on the interface. At a critical time t_c, the top develops an approximately square-root cusp and starts to emit charged drops. In the existing literature the shape of the instability at its final form has been considered a "cone" with a specified angle [11,14,21,22,25]. This is not consistent with our findings for the shape of a viscous fluid during the *dynamical* transition to the jet geometry. It is interesting to note that recent studies have shown that sinusoidal shaking of a liquid also yields a critical point during the ejection of droplets from the surface [30]. Also the collapse of an axisymmetric film bridge under its own surface tension has been simulated by

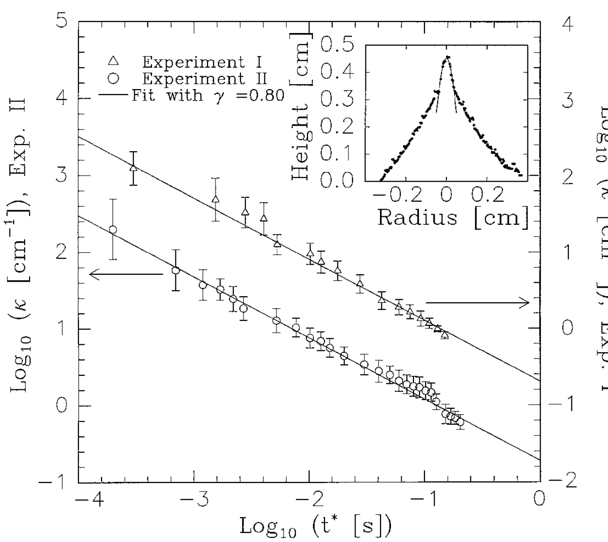

FIG. 2. Double logarithmic plot of κ, the tip curvature, as a function of t^*, the time measured with respect to the critical time: $t^* \equiv t - t_c$. The solid lines are fits to Eq. (1) with the same scaling exponent $\gamma = 0.80$ for both data sets. The initial electrical field in experiment II (left axis) was roughly twice that in experiment I (right axis). The two axes are displaced by 1 decade for clarity so that the data from the two experiments do not overlap. The inset shows the Gaussian fit to the edge-detected frame (corresponding to the first point from the left in the data of experiment II) closest in time to t_c.

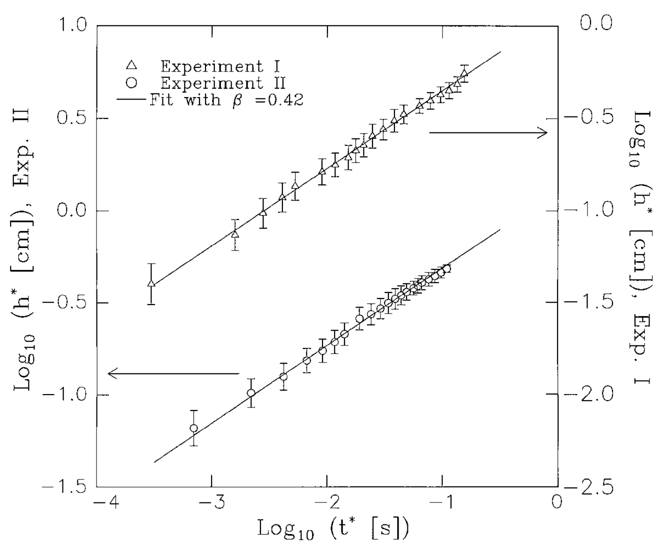

FIG. 3. Double logarithmic plot of the distance to the critical height: $h^* \equiv h_c - h$ versus time measured with respect to the critical time t^*. The solid lines are fits to Eq. (2) with the same scaling exponent $\beta = 0.42$ for both data sets.

Chen and Steen [31] who found scaling in the formation of a cusp in a region prior to the final pinchoff event which had similar exponents to those we have found. We note, however, that this system was inviscid and had an annular three-dimensional geometry as compared to our system which has viscous stresses in a cylindrical two-dimensional geometry.

Both the height and the curvature data exhibit scaling approaching a time t_c at which point the apex of the instability develops a cusp. In addition, the exponents are insensitive to variations in the size of the initial electrical field. This suggests the existence of a critical point at t_c in the dynamics describing the onset of the spout. Although there is no general treatment of this type of problem, other systems with critical behavior in the dynamics have been successfully treated in terms of obtaining the correct force balance around the singularity and devising an appropriate similarity solution for the flow [4,30–34]. Such an approach may be useful for studying the electrohydrodynamical spout and may provide further insight into the formation of the technologically important droplets emitted from this sharp apex.

We thank Itai Cohen and Dan Mueth for technical assistance. We are also grateful to them and to Meredith Betterton, Jakob Bohr, Michael Brenner, Daniel Fisher, Raymond Goldstein, Leo Kadanoff, Howard Stone, and Shankar Venkataramani for stimulating discussions and suggestions. This work was supported by the NSF. L. O. thanks the Danish Research Council for support.

*Permanent address: The Niels Bohr Institute, Blegdamsvej 17, 2100 Copenhagen Ø, Denmark.

[1] D. H. Peregrine, G. Shoker, and A. Symon, J. Fluid Mech. **212**, 25 (1990).
[2] M. P. Brenner, J. Eggers, K. Joseph, S. R. Nagel, and X. D. Shi, Phys. Fluids **9**, 1573 (1997).
[3] J. Keller and M. Miksis, SIAM J. Appl. Math. **43**, 268 (1983).
[4] J. Eggers, Rev. Mod. Phys. **69**, 865 (1997).
[5] G. Chen and A. Gomez, in *Proceedings of the 24th International Combustion Symposium* (The Combustion Institute, Pittsburgh, PA, 1992).
[6] A. R. Jones and K. C. Thong, J. Phys. D **4**, 1159 (1971).
[7] J. B. Fenn, M. Mann, C. K. Meng, S. F. Wong, and C. M. Whitehouse, Science **246**, 64 (1989).
[8] E. W. Bungay and C. McAllister, *Electric Cables Handbook* (Blackwell Science, Oxford, 1990), 2nd ed.
[9] E. Ildstad and S. T. Hagen, in *Proceedings of Electrical Insulation, Conference Record of the 1992 IEEE International Symposium* (IEEE, New York, 1992), pp. 135–139.
[10] J. Zeleny, Phys. Rev. **10**, 1 (1917).
[11] G. I. Taylor, Proc. R. Soc. London A **280**, 383 (1964).
[12] G. I. Taylor and A. D. McEwan, J. Fluid Mech. **22**, 1 (1965).
[13] G. I. Taylor, Proc. R. Soc. London A **313**, 453 (1969).
[14] R. P. A. Hartman, J. C. M. Marijnissen, and B. Scarlett, J. Aerosol. Sci. **28**, S527 (1997).
[15] M. Cloupeau and B. Prunet-Foch, J. Aerosol Sci. **25**, 1021 (1994).
[16] A. J. Mestel, J. Aerosol. Sci. **25**, 1037 (1994).
[17] N. V. Krasnov and S. I. Shevchenko, Rev. Sci. Instrum. **66**, 3623 (1995).
[18] R. J. Turnbull, IEEE Trans. Ind. Appl. **28**, 1432 (1992).
[19] A. J. Mestel, J. Fluid Mech. **274**, 93 (1994).
[20] S. K. Aslanov, A. I. Stroutchayev, and K. Kopyt, J. Aerosol Sci. **28**, S505 (1997).
[21] J. Fernandez de la Mora and I. G. Loscertales, J. Fluid Mech. **260**, 155 (1994).
[22] J. Fernandez de la Mora, J. Fluid Mech. **243**, 561 (1992).
[23] J. Fernandez de la Mora, J. Navascues, F. Fernandez, and J. Rosell-Llompart, J. Aerosol Sci. **21**, Suppl. 1, S673 (1990).
[24] J. Fernandez de la Mora, J. Colloid Interface Sci. **178**, 209 (1996).
[25] C. Pantano, A. M. Ganan-Calvo, and A. Barrero, J. Aerosol Sci. **25**, 1065 (1994).
[26] G. M. H. Meesters, P. H. W. Vercoulen, J. C. M. Marijnissen, and B. Scarlett, J. Aerosol Sci. **21**, Suppl. 1, S669 (1990).
[27] A. Gomez and K. Tang, Phys. Fluids **6**, 404 (1994).
[28] A. M. Gañán-Calvo, Phys. Rev. Lett. **79**, 217 (1997).
[29] L. Oddershede, I. Cohen, and S. R. Nagel (to be published).
[30] B. W. Zeff, B. Kleber, J. Fineberg, and D. P. Lathrop, Nature (London) **403**, 401 (2000).
[31] Y.-J. Chen and P. H. Steen, J. Fluid Mech. **341**, 245 (1997).
[32] X. D. Shi, M. P. Brenner, and S. R. Nagel, Science **265**, 219 (1994).
[33] P. Constantin *et al.,* Phys. Rev. E **47**, 4169 (1993).
[34] R. E. Goldstein, A. J. Pesci, and M. J. Shelley, Phys. Rev. Lett. **70**, 3043 (1993).