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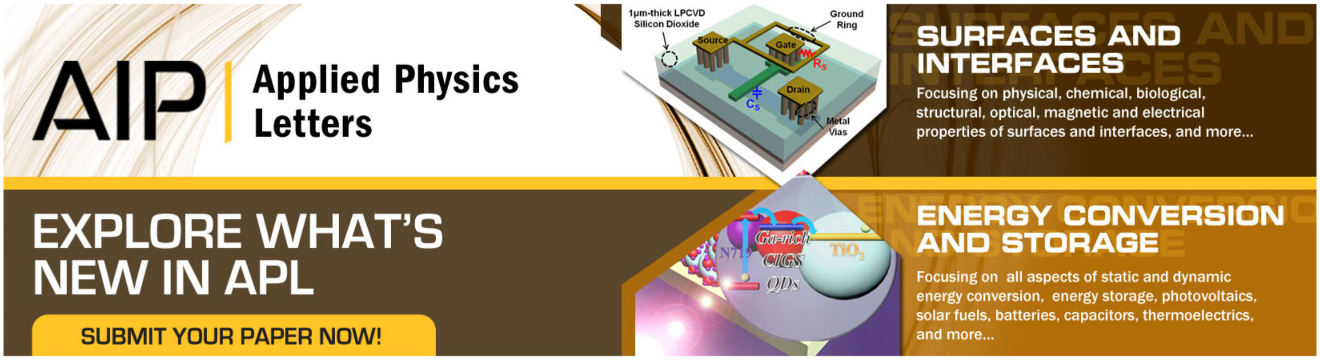

APPLIED PHYSICS LETTERS **91**, 204102 (2007)

Marangoni flow on an inkjet nozzle plate

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In piezo inkjet printing, nozzle failures are often caused by an ink layer on the nozzle plate. It is experimentally shown that the ink layer at the nozzle is formed through streamers of ink, emanating from a central ink band on the nozzle plate. The streamers propagate over a wetting nanofilm of 13 nm thickness, directed toward the actuated nozzles. The motion of the front end of the streamers follows a power law in time with an exponent $\frac{1}{2}$. The observations are consistent with a surface tension gradient driven flow. The origin of the Marangoni flow is an effective lower surfactant concentration of the ink around the nozzle. © *2007 American Institute of Physics*.

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Piezo driven inkjet systems[1] are highly flexible drop-on-demand (DOD) systems with a well-controlled and extremely stable production of monodisperse micron-sized droplets. In piezo inkjet printing, the piezo material is deformed resulting in pressure waves traveling toward the nozzle. At the nozzle end, the acoustic pressure buildup results in a droplet being ejected.[2,3] The fast response of the piezo ceramics allows for a high production rate of typically 30 kHz with a full control over individual droplet formation: the inkjets can be instantaneously switched on and off. A major requirement for today's fast drop-on-demand inkjet printers is the stability of the droplet jetting process. Air entrapment at the nozzle leads to the formation of a small bubble in the nozzle, this bubble travels to the ink channel and grows by rectified diffusion through acoustic forcing.[4] The air entrainment is often directly linked to the presence of an ink layer on the nozzle plate.[4–6] Within the ink layer, small (dust) particles, which are deposited from the ambient air, can be transported toward the jetting nozzle. In Ref. 6, it was shown that a characteristic ink flow pattern occurs on the nozzle plate. The jetting of droplets induces an air flow that drives the ink (with the dust particles) toward the jetting nozzle. Then the particles reach the jetting nozzle, eventually causing air entrapment into the ink channel and nozzle failure. We now have found that an ink flow on the nozzle plate can even emerge when no droplets are jetted, i.e., without the driving air flow. The objective of this letter is to clarify the origin of this nozzle plate flow.

For the experiments side-shooter printheads, developed by Océ Technologies, are employed. The printhead jets droplets out and consists of two arrays of nozzles on a nickel nozzle plate. Each nozzle is connected to an ink channel equipped with a single piezo element. The DOD frequency is set to 30 kHz. More technical details of the employed printhead can be found in Ref. 4. The ink channel is supplied with (transparent) ink from a reservoir. At the operating temperature of 130 °C, the ink (a mixture of multiple organic components in a polymer-based resin) has a surface tension $\sigma = 0.028\ \mathrm{N\,m^{-1}}$ and a contact angle of 15° on the nickel nozzle plate.

To prepare the printhead for operation, the nozzle plate is cleaned using synthetic wipers. It turns out to be impossible to remove all the ink on the nozzle plate. Instead, a thin ink layer remains on the nozzle plate and within minutes the ink accumulates in the middle of the nozzle plate in a stationary central ink band, as shown in Fig. 1(a). The thickness of the central ink band is between 1 and 10 μm, measured by the number of fringes that arise from narrow-band LED illumination ($\lambda = 545$ nm) placed under an angle of 45°. The stationary ink band is the starting condition for the present experiments.

Experiments with different actuation voltages were performed, thereby varying the strength of the meniscus oscillations. Up to a voltage of 45 V the meniscus oscillates, but not strong enough to eject droplets (nonjetting case). Above a driving voltage of 45 V droplets are ejected from the nozzle (jetting case). In both cases ink originating from the central ink band is slowly flowing toward the actuated nozzle (see Fig. 1). First the central band deforms at a timescale of minutes to form a streamer which protrudes in the direction

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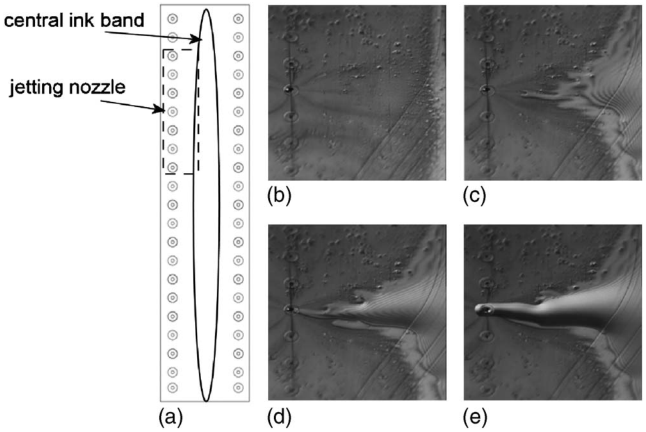

FIG. 1. (Color online) (a) A schematic sketch of the nozzle plate and the central ink band at the initial state. At $t=0$, one nozzle is jetting droplets. The central ink band slowly deforms and a flow develops toward the jetting nozzle. The nozzle is actuated with a trapezoidal pulse (2 μs up, 3 μs flat, and 2 μs down). The actuation frequency is $f_{\mathrm{DOD}}=30$ kHz and the actuation voltage $V_{\mathrm{act}}=70$ V. (b) $t=20$ s after the start of the actuation, (c) $t=390$ s, (d) $t=428$ s, and (e) $t=448$ s.

of the actuated nozzle. Then the streamer rapidly elongates and finally arrives at the nozzle. For the movie of this emanating streamer, we refer to Ref 7.

The motion of the front of the ink streamer is analyzed and its position as a function of time is plotted in Fig. 2. A power law with an exponent 0.46 ± 0.06 is identified.

What are possible driving mechanisms for the ink flow? (i) Nozzle plate vibration: if the nozzle is actuated, the nozzle plate as a whole vibrates due to the moving piezo element. This oscillation could result in a net flow of ink. However, experiments with a sealed but actuated nozzle do *not* exhibit any net ink flow on the nozzle plate, thereby excluding nozzle plate vibrations as an origin of the flow. (ii) Electrostatic effects: the resistivity of the nozzle plate and the other components of the printhead were measured to be 40 Ω, which excludes the presence of significant insulating oxide layers on top of the nozzle plate. Furthermore, the resistivity of the ink is 8×10^3 Ω m. These low values eliminate the possibility of electrostatic effects altogether. (iii) Temperature gradients across the nozzle plate, affecting the ink's surface tension. A temperature increase of 3 ± 1 K from the area around the nozzles to the central ink band area was measured. For typical values of the surface tension temperature coefficient $(d\gamma/dT)/\gamma\approx-10^{-3}$ (Ref. 8), this gives rise to a surface tension variation of at most a few percent. Although this gradient does indeed drive the liquid toward the nozzle, the absolute gradient is insufficient to explain the observations. Furthermore, actuation of the nozzle does not affect the temperature distribution around the nozzle area.

After having excluded possibilities (i)–(iii), we will now give evidence that a surface tension gradient caused by different effective surfactant concentrations and/or selective evaporation is driving the flow. How can such a Marangoni flow develop, as from Figs. 1(a) and 1(b) it seems that the central ink band is physically separated from the meniscus? We employed ellipsometry, allowing for quantitative measurements of film thicknesses much smaller than the wavelength of light, to show that this in fact is not the case. Indeed, we detected a residual wetting ink film with a thickness of 13 ± 1 nm, facilitating the communication between the central ink band and the actuated nozzle. Surface tension gradients can then drive the flow.[9–11] We minimized the surface tension gradient by replacing the ink by silicone oil which is (i) not vulnerable to surfactants and (ii) a pure liquid unlike ink that is a mixture where selective evaporation is possible. Therefore, the surface tension should be constant within the oil film. Indeed, this time, when actuating the nozzles, *no* surface flow or streamers were observed. This finding supports that in the ink case a surface tension gradient is the driving mechanism.

How can the surface tension gradient in the ink layer be built up and sustained? The most likely cause are surfactants. When the meniscus oscillates, its maximum surface area varies between πr^2 and $2\pi r^2$ giving rise to an enhanced average surface area. The oscillation frequency of 30 kHz is too fast for the surfactant concentration at the surface to equilibrate.[8] Hence, the average concentration decreases and the local surface tension increases as compared to the flat configuration and to that of the central ink band. Furthermore, mixing in the nozzle might also reduce the local surfactant concentration. Both effects are more pronounced for larger actuation, in agreement with experiment [see Fig. 2(b)]. Moreover, selective evaporation of one of the components of the complex organic ink could locally modify the surface tension.

We will now derive a simple model accounting for our observations. Fig. 3 depicts a cross section of the nozzle plate with a nozzle to the left and the central ink band to the right. The relevant quantities and parameters are the front

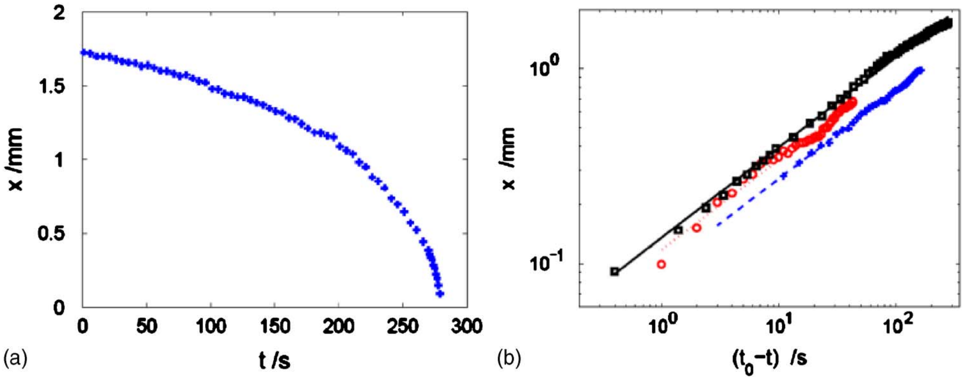

FIG. 2. (a) The position of the ink front x as a function of time when the nozzle is jetting droplets (70 V). The nozzle is located at $x=0$, and t_0 (≈280 s) denotes the time when the front arrives at the meniscus. The front position is defined through a layer thickness of $h=150$ nm. (b) Squares: same as in (a) but on a loglog scale. In addition, we show the front for the nonjetting case (40 V, circles, and 35 V, pluses) and the corresponding fits (solid, dotted, and dashed, respectively). The power law exponents ($\approx0.46\pm0.06$) are comparable, but the prefactor depends on the actuation. It should be noted that the exact value of the power law exponent depends on the choice of t_0, which was determined through a least square fit.

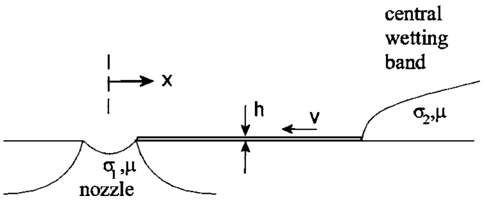

FIG. 3. (Color online) A schematic sketch of the ink on the nozzle plate. The distance of the ink to the nozzle is x. The surface tension at the nozzle is σ_1; at the central ink band it is σ_2. The thickness of the wetting film in between is h.

position of the ink streamer x, its velocity $v=\dot{x}$, the wetting film thickness h, the ink's dynamic viscosity μ, and the surface tension difference $\Delta\sigma=\sigma_1-\sigma_2$ between the ink near the nozzle and near the central ink band. The front velocity of the flow then is [see Ref. 12 or simply from dimensional analysis (apart from the prefactor)]

$$\dot{x} = v = \frac{h\Delta\sigma}{6\mu x}. \tag{1}$$

Note that the velocity of the ink front scales linearly with h for Marangoni flows, in contrast to pressure driven flows, for which the velocity scales with h^2, $v=h^2\Delta P/(12\mu x)$. To achieve the observed velocity of about 1×10^{-5} m/s, a pressure difference of $\Delta P=7\times10^6$ Pa per 1 mm would be required, which is very unrealistic.

Integration of Eq. (1) leads to the position of the moving front as a function of time, similar to[13–16]

$$x(t) = \sqrt{\frac{h\Delta\sigma}{3\mu}}(t_0-t)^{1/2}, \tag{2}$$

where t_0 is the time when the ink front reaches the nozzle. Equation (2) can also be written as $\tilde{x}=\tilde{\tau}^{1/2}/\sqrt{3}$, with $\tilde{x}=x/h$ and $\tilde{\tau}=(t_0-t)/(\mu h/\Delta\sigma)$. The power law exponent $\frac{1}{2}$ is consistent with the experimental observations [Fig. 2(b)]. The prefactor $\sqrt{h\Delta\sigma/(3\mu)}$ in Eq. (2) depends on the surface tension difference $\Delta\sigma$. With $h=13$ nm and $\mu=0.01$ kg m^{-1}s^{-1}, we obtain from the experimentally determined prefactors $\Delta\sigma=0.04$ N m^{-1}, $\Delta\sigma=0.03$ N m^{-1}, and $\Delta\sigma=0.02$ N m^{-1} for the three actuation voltages of 70 V (jetting), 40 V (nonjetting), and 35 V (nonjetting), respectively. The relative increase of the surface tension difference is 50% when going from 35 to 40 V. Taking into account the simplicity of our one-dimensional model, it seems realistic to us that order of magnitude wise these surface tension gradients can build up.

While Marangoni flow seems to be the only explanation for the observations in the nonjetting case, for the jetting case the ejected droplets induce an air flow which contributes also to the ink flow, as shown in Ref. 6. We note, however, that these experiments were performed on preconditioned nozzle plates with a different wetting behavior. This precludes a direct comparison of the results. A full quantitative analysis of the competing shear and Marangoni flows under jetting conditions remains elusive at this stage.

From a practical point of view, the key question is how the nozzle plate flow can be suppressed, in order to prevent nozzle failure.[4] To achieve this goal, it is crucial to have a dry nozzle plate, i.e., suppress the wetting nanofilm by considerably increasing the contact angle between ink and nozzle plate. This can best be achieved by coating the nozzle plate with an anti-wetting coating. In practice, however, the use of such coatings depends on several external constraints, such as its lifetime, chemical reactions with the ink, or its resistance to heat. An alternative would be to use nonwetting inks.

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[7] See EPAPS Document No. E-APPLAB-91-079746 for a movie of the streamer of ink emanating from the central ink band toward the jetting nozzle. This document can be reached through a direct link in the online article's HTML reference section or via the EPAPS homepage (http://www.aip.org/pubservs/epaps.html).
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