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### Full Length Article Laser-induced forward transfer of low viscosity inks

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### ABSTRACT

Laser-induced forward transfer (LIFT) is a laser-based printing technique which has been revealed as an interesting alternative to inkjet printing for the deposition of inks in direct writing applications. The principle of operation of the technique relies on the focusing of a laser beam on a thin film of the ink and the release of a tiny fraction of material through the action of a laser pulse, being usually assumed that such pulse should be very short, of the order of ns and below. However, with the aim of reducing production costs it would be desirable to operate with longer pulses (hundreds of ns and above), so that less expensive lasers could be employed.

In this work we prove that it is feasible to carry out the LIFT of liquids with relatively long laser pulses (a few hundreds of ns). To that aim we have investigated the influence of laser fluence on the printed droplets and identified an evolution of their morphology with that parameter somewhat different from the one characteristic of the LIFT of liquids with much shorter pulses. A further time-resolved imaging study has revealed the onset of up to three different transfer mechanisms which correlate well with the deposition outcomes.

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#### 1. Introduction

Laser-induced forward transfer (LIFT) is a direct-write technique capable of printing solid and liquid materials by means of a focused laser beam [1,2]. In LIFT a film of the material to transfer is spread along a donor substrate which is separated a convenient gap from an acceptor substrate, where the pattern will be printed. Then, through the action of a laser pulse focused on the donor film, a tiny fraction of the donor material is propelled forward and deposited on the acceptor substrate [3]. Through the repetition of this process at different locations of the donor-acceptor system single pixels [3,4], lines [5,6] or complex patterns [7,8] can be transferred.

The feasibility of LIFT to print materials in liquid phase has been extensively proved through the transfer of many kinds of liquids with different viscosities and rheologies, from simple liquids [9–12] to complex suspensions [13–15] and even pastes [16–19]. This constitutes a clear advantage over its main competitor, inkjet printing [20], which presents important restrictions concerning the printable viscosities and rheologies. In LIFT there is practically no limitation in liquid viscosity, being able to easily transfer low and high viscosity liquids (from a few mPa·s to hundreds of Pa·s) [21,22],

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http://dx.doi.org/10.1016/j.apsusc.2016.11.179 0169-4332/© 2016 Elsevier B.V. All rights reserved. and it is free from issues such as clogging so characteristic of inkjet printing.

The LIFT of low viscosity liquids has been widely investigated both through the analysis of the influence of the main process parameters, such as laser fluence or focusing conditions, on the morphology of the printed features [4,23,24] and through the study of the dynamics of liquid transfer using time-resolved imaging [3,15,25–31]. All of these works, though, have been carried out using very short laser pulses, from several femtoseconds to a few tenths of nanoseconds in pulse duration. However, it would be also interesting to explore the possibility of carrying out LIFT with longer pulses, since this would allow working with less expensive lasers, an obviously attractive option from an industrial point of view. In fact, cost reductions between 30 and 50% can be easily achieved in the acquisition of a Nd:YAG laser source, for example, with a pulse duration of 100 ns respect to one with a few ns pulse duration with the same average power. Although transfer with long laser pulses (from  $\sim$ 100 ns up to  $\sim$ 1 ms) has already been proved with solid materials such as tungsten, molybdenum or aluminum [32–34], the LIFT of liquids with long pulses remains unexplored. Since substantial differences have been observed between the transfer of solids and liquids when using short and ultrashort pulses, it would also be interesting to investigate the transfer of liquids with longer pulses. Therefore, in this work we study the LIFT of low viscosity inks with relatively long laser pulses. To that aim we used an industrial marking laser with a pulse duration of

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hundreds of nanoseconds to deposit droplets of a commercial silver ink at varying laser fluences, thus proving the feasibility of the new approach in printing applications. We combined this study with a further analysis of the liquid transfer dynamics through timeresolved imaging in order to carry out a complete characterization of the transfer process which allowed to correlate the observed transfer behavior with the obtained deposits.

#### 2. Experimental

Laser transfer was performed using a Nd:YAG laser (Baasel Lasertech, LBI-6000) operating in the fundamental mode (1064 nm wavelength) with a pulse duration of approximately 120 ns, a laser pulse energy ranging between 0.01 and 0.30 mJ, and a Gaussian intensity distribution. This laser system is furnished with a gal-vanometric mirrors head that allows scanning the beam following predefined patterns. After the mirrors, an f-theta lens with a focal length of 100 mm focuses the laser beam onto the sample; the resulting beam waist diameter in the focal plane is of about 80 µm.

Both donor and acceptor substrates were microscope glass slides (Deltalab,  $76 \times 26 \times 1$  mm), and the chosen transfer material was a commercial silver nanoparticle ink (Sigma Aldrich, ref. 736481) typically used in inkjet printing. Its particle size is smaller than 50 nm and its solid content around 30–35 wt.%, with a density of 1.45 g/cm<sup>3</sup>, a viscosity of 8–10 mPa·s, and a surface tension of 30–50 mN/m. Donor films were obtained by blade coating of 60  $\mu$ L of ink deposited on a clean slide, leading to a thickness of around 30  $\mu$ m. Then the donor was turned up-side-down and placed above the acceptor using spacers 150 and 250  $\mu$ m thick. Finally, the donor and acceptor slides, separated by the spacers, were placed at the focal plane of the laser system for printing.

A time-resolved imaging setup composed of a light source, a CCD camera and a pulse generator was used in the second part of the experiment for the visualization of the liquid transfer dynamics. The light source consisted in a red LED (Thorlabs, 630E) which output was focused by means of two consecutive lenses (focal lengths of 25.4 and 35 mm, respectively) acting as a condenser. The LED pulse (duration of 200 ns) was triggered by the laser pulse at a controlled delay by means of a pulse generator (Stanford Research Systems, DG645). A monochrome CCD camera (Diagnostic Instruments Inc., Insight IN 1800) with and exposure time of 500  $\mu$ s and coupled to a 20× microscope objective with a numerical aperture of 0.42 was used for the images acquisition. Both light source and camera setup were placed along the same optical axis in shadowgraphy configuration at grazing incidence respect to the donor-acceptor system.

#### 3. Results and discussion

The feasibility of long pulse LIFT for low-viscosity inks printing was proved through the deposition of droplets under the systematic variation of the main process parameter, the laser pulse fluence. The process was investigated through the characterization of the morphology of the deposited droplets and the time-resolved imaging study of the transfer dynamics.

#### 3.1. Droplet printing

Arrays of droplets at different laser fluences were printed with a donor-acceptor gap of 150  $\mu$ m by scanning the laser beam along the sample at a constant speed of 800 mm/s and a pulse rate of 1 kHz, conditions which allowed printing non-overlapping droplets. Deviations due to slight variations in the film thickness were mitigated by printing redundant arrays of many droplets.

Fig. 1a-b shows printed droplets and their corresponding dried pixels at fluences ranging from 2.0 to 6.0 J/cm<sup>2</sup>, measured by dividing the pulse energy by the cross sectional area of the beam at the waist. In fact, a wider range of fluences was explored, but no transfer was observed below 2.0 J/cm<sup>2</sup>, which sets the threshold fluence, and 6.0 J/cm<sup>2</sup> is the highest available fluence with our system. The outcome of this experiment already proves the feasibility of LIFT for printing low viscosity inks with long laser pulses, since well-defined droplets are obtained at least for some fluences along the investigated range. It has to be noted too that this fluences are substantially higher than those required for printing similar droplets with shorter pulses [24,28,35,36]. It can also be observed that as the laser fluence increases the radius of the droplets also increases, from 80  $\mu$ m at the lowest fluence (2.0 J/cm<sup>2</sup>) to 250  $\mu$ m at the highest available fluence with our laser system  $(6.0 \text{ J/cm}^2)$ . The minimum droplet obtained here is substantially larger than the smallest feature dimensions commonly reported with shorter laser pulses [4,37]. The minimum value of our experiment does not represent, though, the ultimate resolution of the technique: the laser beam diameter could be considerably reduced through the use of lenses with shorter focal lengths (this is not possible with our marking laser system, but it might be with many other systems). Besides, there are many attractive applications for printing which do not require extremely high resolutions, like in the production of RFID tags, smart labels in the packaging industry, or in the fabrication of sensors.

The droplets in Fig. 1a are quite circular at low fluences, with a rather uniform and well defined border. As the fluence increases the edge becomes less regular and some satellite droplets appear, as well as some splash. At the highest fluences the largest satellites surprisingly disappear and the edge becomes more uniform again, though some very small corona splash remains. This three regime (low, intermediate and high fluences) behavior, wherein some improvement in droplet quality is observed at high fluences, is rather unusual and has not been reported in previous experiments [4,26,38,39]. In general, the quality of the printed feature tends to suddenly decrease at high fluences, leading to irregular and scattered drops, but never to increase again. When comparing wet (Fig. 1a) and dry (Fig. 1b) droplets no apparent change in dimensions is observed. The droplet radius, on the other hand, increases quite linearly with fluence (Fig. 1c). In previous experiments with liquids with a similar viscosity we observed a linear trend for the droplet volume instead [4,35,40]; the evolution of the droplet radius was clearly not linear. This apparent inconsistency should not be so surprising, since the working conditions are significantly different now. First, those former studies dealt with a simple solution of water and glycerol, while in this experiment we are working with a rather different ink, a suspension of silver nanoparticles. Second, the pulse duration regime is also substantially different (a few nanoseconds in the first case, hundreds of nanoseconds in the second). Furthermore, it has to be noted that a linear dependence of radius versus fluence was also observed by Makrygianni et al. using 10 ns pulses and a similar silver nanoparticle ink [24]. This suggests that the difference observed with our previous works could be more related to the composition of the ink than to the laser pulse duration. In fact, the acceptor wettability can strongly influence both the radius and volume of the deposited droplets and their evolution with laser fluence, and this property can be quite different for the water-glycerol solution and the silver ink.

Confocal microscopy was used to measure the topography of the dry pixels and the corresponding profiles are shown in Fig. 2a. It can be observed that their thickness does not follow the same trend as the radii. The average thickness slightly increases from  $0.4 \,\mu\text{m}$  at 2.0 J/cm<sup>2</sup> to around 0.6  $\mu\text{m}$  at 2.8 J/cm<sup>2</sup> to slowly decrease again to around 0.4  $\mu\text{m}$  at 4.6 J/cm<sup>2</sup>, from which fluence it suffers a dra-

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