



Single step high-speed printing of continuous silver lines by laser-induced forward transfer



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ABSTRACT

The development of high-speed ink printing process by Laser-Induced Forward Transfer (LIFT) is of great interest for the printing community. To address the problems and the limitations of this process that have been previously identified, we have performed an experimental study on laser micro-printing of silver nanoparticle inks by LIFT and demonstrated for the first time the printing of continuous conductive lines in a single pass at velocities of 17 m/s using a 1 MHz repetition rate laser. We investigated the printing process by means of a time-resolved imaging technique to visualize the ejection dynamics of single and adjacent jets. The control of the donor film properties is of prime importance to achieve single step printing of continuous lines at high velocities. We use a 30 ps pulse duration laser with a wavelength of 343 nm and a repetition rate from 0.2 to 1 MHz. A galvanometric mirror head controls the distance between two consecutive jets by scanning the focused beam along an ink-coated donor substrate at different velocities. Droplets and lines of silver inks are laser-printed on glass and PET flexible substrates and we characterized their morphological quality by atomic force microscope (AFM) and optical microscope.

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1. Introduction

The Laser-Induced Forward Transfer (LIFT) is a printing technique introduced by Bohandy et al. almost three decades ago [1] which has been used to transfer a large variety of materials in solid [2–4], paste [5] or liquid [6,7] state from a transparent donor substrate to a receiver substrate placed nearby. The use of LIFT is widespread and appears very attractive to print microelectronic devices especially on flexible substrates such as conductive lines [8–11], microelectromechanical systems (MEMS) [12,13], or organic light emitting diodes (OLEDs) [14–16] and organic thin film transistors (OTFTs) [17,18].

Time-resolved imaging experiments have been used to study the ejection dynamics of solid [19,20], powder [21] and liquid materials [22–24]. For the ejection in liquid phase, which is the purpose of this study, the dynamics of formation and expansion of the liquid jet induced by a single laser pulse has been extensively studied by means of time-resolved shadowgraphy [25–27]. The absorption of the laser energy, either by the liquid itself or by a thin absorbing layer deposited between the substrate and the liquid film,

generates the formation of a cavitation bubble which expands away from the surface and propels the ink, forming a jet [25]. The liquid displaced by the jet is collected on a receiver substrate and forms a single droplet. A continuous line can thus be printed by the overlapping of adjacent droplets. The dimensions and the quality of the line depend on the liquid viscosity, the thickness of the film on the donor substrate, the laser fluence, the distance between the donor and receiver substrates [28,29] and the presence or absence of an absorbing layer [9].

In order to print continuous lines at high velocity, we combine a very high repetition rate pulsed laser (0.2–1 MHz) with a galvanometric mirror scanner system that can scan the beam over a large surface at high speed (max. 17 m/s). This allows generating adjacent jets close to each other with a short delay time between them and that can lead to strong jet–jet interactions. Recently, our group has demonstrated the printing of micrometer conductive lines at velocities up to 4 m/s in multi-passes configuration with a picosecond laser operating at 500 kHz [30]. However, to develop a fast and reliable LIFT printing process suitable for industrial applications [31], such as flexible electronic, some improvements are required and especially a single pass approach must be demonstrated. Previous studies performed on high-speed laser printing by time-resolved shadowgraphy [32,33] showed that the strong interactions between successive jets strongly modify the ejection

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dynamics of the jets compared to the well-known mechanisms of a single jet generation. These studies demonstrated that the expansion of the second jet generated few μs after and next to the previous jet is perturbed by the interactions between two adjacent cavitation bubbles. Moreover, if the generation of the second bubble occurs on a non-flat surface, due to the perturbations induced by the previous laser irradiation, the second jet is tilted and can interact with the previous and/or the following jet. These effects are the main limitations for printing continuous lines in a single step at high velocity [34].

In this work, we present a time-resolved imaging study of the sequential multi-jets ejection as a function of the silver ink donor film properties (thickness) and irradiation parameters (laser frequency and scan velocity). The dynamics of the liquid transfer of the silver ink (20% silver content) at high velocity shows that for some specific conditions all the successive jets are linked together and the stability of this multi-jet structure is maintained. In these conditions, we printed 30 μm wide silver lines at velocities up to 17 m/s in a single pass.

2. Experimental setup

Printing experiments have been performed with a frequency-tripled fiber laser (Hegoa, EOLITE Systems; wavelength = 343 nm; pulse duration ~ 30 ps; max. repetition rate = 1 MHz). The delay between consecutive laser pulses (≥ 1 μs) and the total number of pulses are adjustable. A wave plate and a polarizing beam-splitter cube control the pulse energy. The laser beam is scanned by a galvanometric mirror and focused by an F-theta telecentric objective (focal length = 160 mm). The diameter of the Gaussian beam is ~ 34 μm (at $1/e^2$) and have been determined with the method proposed by Liu et al. [35], based on the curve representing the measurements of the diameter of the ablated holes in an Aluminum layer deposited on a Kapton substrate as a function of the natural logarithm of the pulse energy.

The SunTronic silver nanoparticles ink used in this work was supplied by Sun Chemical with a silver nanoparticles content of 20 wt% (size = 60–150 nm; density = 1.22 g/mL; viscosity = 10–13 mPa s; main solvents: ethanediol, ethanol, glycerin, 2-isopropoxyethanol). The donor substrates were prepared by spin-coating the ink onto quartz substrates with different speed and time parameters in order to create donors with different homogeneous film thicknesses. The thickness of a fresh ink layer is measured by weighting the sample before and after the spin-coating process.

The shadowgraphy system consists in (1) a NANOLITE KL-M flash lamp (High-Speed Photo-Systeme) as illumination device, (2) two quartz lenses aligned to collect the light from the flash lamp and focus it below the donor substrate, on the trajectory of the laser beam and perpendicularly to it, and (3) a fast QICAM 12-bits camera equipped with a 12 \times zoom lens system. The duration of the flash (~ 16 ns) determines the temporal resolution of the entire acquisition system. A calibration is carried out with a stage micrometer in order to determine the scale of the images. A computer controls the laser firing and the scanning and imaging systems. We set the delay between the emission of the laser pulses and the triggering of the flash lamp with a digital delay generator (DG535, Stanford Research Systems).

All the experiments are done at room temperature and ambient pressure 10 min after the preparation of the donor substrate. Shadowgraphy experiments are conducted with and without receiver substrate. Two liquid jets seem to evolve in opposite directions in the time-resolved images (see for instance Fig. 1), but only the jet propagating downward must be considered, since the upward-propagating one corresponds to the reflection on the ink surface,

which is represented by a white dashed line in every figure. The volume of the cured deposited droplets and lines is measured with an Atomic Force Microscope.

3. Results and discussion

3.1. Dynamic of a single jet formation

Previous studies have shown that the formation of the jet induced by the laser irradiation of a film of silver nanoparticle inks is correlated to the dynamics of the expansion of the cavitation bubble [22,33]. This dynamics is strongly linked to the film thickness, and it appeared interesting to investigate the influence of this parameter on the formation of the jet. As the interaction between the successive bubbles is the limiting process for printing at high velocity, this study on the bubble dynamics is particularly relevant to understand the phenomena observed in the multi-jets experiments.

Fig. 1 shows shadowgraphy images taken at several delays after the irradiation with a single laser pulse for three different donor film thicknesses: (a) 1.5 μm (1 h after the spin-coating ink during 30 s at 2900 rpm), (b) 2.6 μm (30 s at 2600 rpm) and (c) 4.9 μm (30 s at 1600 rpm), without receiver substrate. The laser fluences are different, (a) 35 mJ/cm^2 and (b)–(c) 64 mJ/cm^2 , because the threshold to generate an expanding jet is higher when the donor film thickness increases and we always selected a laser fluence slightly above the threshold to guarantee the generation of stable jets. It is important to note that the general trends of the jet dynamic are similar for the different irradiations series and the different fluences. The images show both the evolution of the cavitation bubble and the formation of the jet.

A detailed description of the temporal evolution of the cavitation bubble and the formation of the jet has been reported by Biver et al. [33] for donor film thickness of 1.5 μm (a). Consequently, we will only point out and discuss the main differences in the jet dynamics induced by the variation of the ink thickness. The first difference is the dimension of the bubble diameter (delay < 1 μs). Whereas the maximum diameter achieved during the expansion phase is 24 μm at 1.5 μm donor thickness (a), it is two times bigger, 48 μm and 50 μm , at 2.6 μm (b) and 4.9 μm (c), respectively. Also, the jet width during the stabilization step is higher when the donor film thickness increases: ~ 3 μm (a, 600 ns), ~ 4.5 μm (b, 1.3 μs), and ~ 6.5 μm (c, 2 μs). The dynamics of the bubble expansion is slower when the donor thickness increases. Indeed, the maximum diameter is observed at 200 ns (a), 400 ns (b) and 700 ns (c) after the laser irradiation. These evolutions are not surprising because the irradiation of a thicker film will lead to the motion of a larger amount of liquid (wider jet) and that requires a bigger amount of energy (higher fluence and larger bubble diameter). In conclusion, although the dynamics of the cavitation bubble and the jet are quite similar for different donor film thicknesses, the variations in time for the expansion, collapse, first rebound and stabilization of the bubble, as well as the different bubble and jet dimensions will induce different interaction mechanisms in multi-jet process for similar irradiation conditions (galvanometric mirror velocity and laser frequency).

3.2. Multi-jet and droplet printing

A theoretical description and the first multi-jet results have been presented in previous works [32–34]. The results point out that the slope of the surface around the irradiation region varies during a few microseconds after the irradiation. This variation can modify the orientation of the following jet which is generated from a tilted surface and propagates with an angle relative to the vertical direction. In order to perform the second irradiation in a liquid volume

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