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### Conductive silver ink printing through the laser-induced forward transfer technique

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#### A B S T R A C T

Laser induced forward transfer (LIFT) is a technique which allows printing a wide variety of materials. It presents several advantages over inkjet printing, such as a potentially higher resolution, being free from clogging issues, and the possibility to work with a much broader range of viscosities. LIFT appears, therefore, as an interesting alternative in all those fields where miniaturization is a major requirement, as in the microelectronics industry. The fabrication of electronic devices requires the printing of small, narrow and thin conductive lines, and in this work we investigate the printing of continuous lines of conductive silver ink on glass substrates through LIFT. Lines are initially formed through sequentially printing adjacent droplets with different overlaps. We show that above a certain overlap continuous lines can be obtained, but unfortunately they show bulging, a problem which compromises the functionality of the lines. In order to solve the problem, other printing strategies are tested; they consist in printing adjacent droplets in alternate sequences. It is found that the alternate printing of two overlapping sets of droplets with an intermediate drying step allows obtaining functional continuous lines without bulging.

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#### **1. Introduction**

The feasibility of the laser induced forward transfer (LIFT) technique to transfer a large variety of materials for diverse applications has been widely proved. Originally, LIFT was applied to print materials from thin solid films  $[1,2]$ , but some years later it was demonstrated that it was also possible to transfer materials in the liquid phase [\[3,4\].](#page--1-0) In comparison with inkjet printing, LIFT does not present problems of nozzle clogging or head contamination since the transfer process is contact-free. Additionally, LIFT allows working with a wider range of viscosities  $(1-10^3 \text{ mPa s})$ , attaining high degrees of spatial resolution and transferring sensitive materials without significant damage  $[5-8]$ . Thus, LIFT has been used to print materials as diverse as DNA for sensors [\[9\],](#page--1-0) ITO nanoparticles for

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[http://dx.doi.org/10.1016/j.apsusc.2014.12.100](dx.doi.org/10.1016/j.apsusc.2014.12.100) 0169-4332/© 2014 Elsevier B.V. All rights reserved. thin film solar cells [\[10\]](#page--1-0) or organic semiconductors for organic light emitting diodes (OLEDs) [\[11\]](#page--1-0) among others.

Despite that most of the work devoted to the LIFT of liquids has been focused on depositing isolated droplets, the problem of printing continuous lines appears to be especially interesting, since one of the main applications of any printing technique is the realization of interconnects in microelectronic devices. Actually, the feasibility of printing conductive lines through LIFT has already been proved, both from solid metallic films [\[12\]](#page--1-0) and from nanoparticle inks [\[13–17\].](#page--1-0) In spite that the very small line widths achieved through the LIFT of solid metallic films make this option really attractive in applications requiring very high resolutions, there are still some issues like the presence of splatter or the short length of the printed lines which still limit its industrial implementation. The LIFT of nanoparticle inks on the other hand constitutes a more conventional approach, probably closer to the current industrial demands. However, it is not free from problems either: it is in general difficult to find the right process parameters to obtain stable continuous lines free from bulging [\[18–21\].](#page--1-0) This problem is not specific from LIFT, but also common to other even more conventional techniques, such as inkjet printing, for instance. This not









Fig. 1. Sketch of the experimental setup: (a) laser, (b) focusing objective, (c) Ag layer, (d) vacuum chuck, (e) stages, (f) CCD camera, and (g) IR mirror.

only limits the spatial resolution and quality of the printed feature, but also compromises its functionality: the presence of bulges can result in undesired short-circuits between adjacent parallel conductive lines. The objective of this work is, therefore, the development of strategies which allow printing continuous conductive lines through LIFT avoiding the problem of bulge formation. In order to do so, different printing experiments were performed using LIFT to transfer silver conductive inks on glass substrates. Once a successful strategy was found, the bulge-free printed lines were laser cured and their functionality was tested through resistivity measurements.

#### **2. Experimental setup**

The printing system uses an Amplitude Systemes Yb:KYW laser with an output wavelength at 1027 nm and pulse duration of 450 fs. The laser beam is focused through a microscope objective of 50×, with a working distance of 1.3 mm and numerical aperture of 0.55. The laser spot diameter was 12  $\mu$ m and it remained fixed during all the experiments. The objective is mounted on a translation stage which allows changing the focusing conditions on the sample. A CCD camera placed coaxially to the laser beam axis allows controlling the position of the laser beam focus on the sample. The sample is mounted on a vacuum chuck to ensure the steadiness while the printing process is taking place. The vacuum chuck is in turn mounted on a translation stage to move the sample in the XY plane. A scheme of the setup used is shown in Fig. 1.

A commercially available Ag nanoparticle dispersion from Sigma Aldrich® is used in all the LIFT experiments. The particle size is  $\leq$  50 nm and the solid content is around 30–35%. The donor film is prepared by blade coating thin layers of the ink on glass microscope slides, with film thicknesses between 20 and 30 $\mu$ m; such relatively large thicknesses should help to improve the uniformity of the donor film, avoiding its fast drying. The acceptor substrates are also glass slides and they are separated from the donor substrates by a 200  $\rm \mu m$  gap through cover slips used as spacers. Although during LIFT experiments it is usual to use a sacrificial absorbing layer [\[22\],](#page--1-0) in our experiments it is not used because the Ag ink itself absorbs well the laser radiation.

After printing the lines are left to dry and once dried its conductivity is measured. Next, they are laser cured with a CW Nd:YAG laser (Baasel LBI 6000, 1064 nm wavelength, 1.2W output power) operating at a speed of 2 mm/s with an irradiance on the sample of  $12 \text{ kW/cm}^2$ . At the end of the process the conductivity of the lines is measured again and compared with the initial value.

#### **3. Results and discussion**

The printing process begins just after the preparation of the donor film since the drying time affects the donor layer quality in terms of both viscosity and thickness [\[22\].](#page--1-0) Series of equally spaced droplets are printed in order to generate lines. Different drop to drop shifts (measured from center to center) are set with the aim of investigating the influence of this parameter on the morphology ofthe resulting lines, and the total number of droplets is adjusted to always produce 3 mm long lines. The translation stages are moved between consecutive droplets, but these are printed with the stages at rest. The purpose of this procedure, otherwise inefficient from an industrial point of view, is to analyze the bulging problem under the simplest conditions. Issues related with the relative motion between laser beamand sample, and the influence of printing speed on the process are in progress [\[23\].](#page--1-0)

[Fig.](#page--1-0) 2 shows the results of the first experiment, carried out at a laser fluence of 530 mJ/cm<sup>2</sup> following the procedure described above. The shift between adjacent droplets in each line increases from left to right, and consecutive lines have been printed in alternate directions, with the firstline starting in the upper left corner of the figure; since whenever bulging appears in a line it always does it at the beginning of that line (though not exclusively), such alternate printing sequence allows minimizing the chance of coalescence between bulges corresponding to adjacent lines. The average diameter of an isolated droplet is around  $100 \,\mu m$  (two first lines, with no droplet overlap), and all the droplets show a high degree of uniformity and excellent definition which actually constitutes a proof of the success in the preparation of the uniform donor film. When the printing distance between adjacent droplets reaches  $90 \,\mu$ m the droplets start to coalesce, though such coalescence is not enough to result in a continuous line yet. As the printing distance decreases, the frequency of droplet coalescence increases, until a shift of 70  $\mu$ m is reached, for which a continuous line with some scalloping is observed and a significant bulge in its starting point is formed. Further on, the continuous lines become more uniform but they are always accompanied by bulges, especially in the beginning and occasionally in the middle. In all cases the average width of the lines is around 65  $\mu$ m, substantially smaller than isolated droplets. All along the experiment no continuous line free from bulging is ever obtained. As it has already been pointed out in the Introduction, it is not surprising to find bulging in the printed continuous lines. Previous work on conductive ink printing, both through LIFT and through inkjet, has shown the persistent presence of such detrimental effect in continuous lines [\[21,24,25\].](#page--1-0)

Duineveld [\[25\]](#page--1-0) has analyzed the stability of continuous lines deposited by inkjet printing and he has established a dynamic model which provides an explanation for the bulge formation process. In this model, bulge formation is attributed to capillary flow arising from an instability produced inside the liquid due to variations in the contact angle along the line between the newly printed droplets and the previously printed feature. According to this explanation, it is not surprising that bulging appears at random positions along the printed line. However, the model does not account for the systematic formation of the initial bulge.

The question which arises is: is the initial bulge formed during printing of the first droplets, or rather at some later time, once an incipient line has already been printed? In order to find that out, an experiment was carried out where lines with increasing number of droplets were printed. [Fig.](#page--1-0) 3 shows the results of such experiment, with the number of droplets in each line increasing from left to right and with printing direction always from bottom to top

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