Overlapping Limitations for ps-Pulsed LIFT Printing of High Viscosity Metallic Pastes

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Abstract: Laser-induced forward transfer (LIFT) technique has been used for printing a high viscosity (250 Pa s) commercial silver paste with micron-size particles (1–4 µm). Volumetric pixels (voxels) transferred using single ps laser pulses are overlapped in order to obtain continuous metallic lines. However, interference problems between successive voxels is a major issue that must be solved before obtaining lines with good morphologies. The effects of the laser pulse energy, thickness of the donor paste film, and distance between successive voxels on the morphology of single voxels and lines are discussed. Due to the high viscosity of the paste, the void in the donor film after a printing event remains, and it negatively affects the physical transfer mechanism of the next laser pulses. When two laser pulses are fired at a short distance, there is no transfer at all. Only when the pulses are separated by a distance long enough to avoid interference but short enough to allow overlapping (∼100 µm), is it possible to print continuous lines in a single step. Finally, the knowledge obtained has allowed the printing of silver lines at high speeds (up to 60 m/s).

Keywords: laser-induced forward transfer; metallization; Ag paste

1. Introduction

Laser-induced forward transfer (LIFT) is a laser direct-write technique that allows the printing of a number of materials in high resolution patterns [1,2]. LIFT is comprised of a donor transparent substrate, coated with the material to be transferred, and an acceptor substrate separated by a gap distance from the donor. A pulse laser beam is focused in the interface between the donor substrate and the coating material. Part of the coating is vaporized, generating a vapor bubble that expands and pushes the remaining material forward to the acceptor substrate [1]. Thus, LIFT provides a noncontact, nonlithographic, and nozzle-free method for printing complex materials, because the coating material can be solid or liquid in a wide range of viscosities. LIFT can be used for printing two-dimensional (2D) or three-dimensional (3D) complex microstructures [3–6] or for large area printing applications [7]. The possibility of printing conductive metallic materials, either in solid state or in the form of particles dispersed in a fluidic matrix, has straightforward applications in microelectronics or photovoltaics. Different and recent reviews of LIFT can be referred to in [8,9]. LIFT printed metallic lines should be continuous, with widths in the order of microns and aspect ratios as large as possible in order to maximize the electrical conductivity maintaining the nominal width required. Moreover, industrial applications demand a robust process with very high throughputs, in order to compete with standard metallization techniques such as screen printing. Both robustness and process speed are still a challenge when it comes to LIFT printing.

The LIFT technique is quite sensitive to the experimental conditions and adequate process parameterization is needed. Regarding process throughput, lines are usually printed by overlapping volumetric pixels (voxels) of material, each transferred with a single laser pulse. When using direct LIFT
of low viscosity fluids containing metallic nanoparticles, the possible effects of interference between successive laser pulses [10] and the length of time delay needed between pulses should be taken into account proving that a voxel has reached its steady-state shape before the next voxel is transferred. Interference between successive voxels can lead to debris and reduced uniformity of the line, but can also be used for printing lines using multi-jets under certain experimental conditions [11]. Moreover, this effect is more significant in the case of high viscosity metallic pastes [12]. More recently, a similar effect has also been observed in metallic inks with a viscosity one order of magnitude smaller [13].

This work aims to LIFT print continuous metallic lines with high speeds using a commercial, high viscosity, micron-size particles silver paste, and a ps-pulsed laser. In order to achieve this goal, first, the experimental parameters (laser pulse energy, donor film thickness, and gap distance) for printing single voxels are determined. Secondly, the interference between overlapped printed voxels is studied and discussed in terms of the distance of successive laser pulses (pitch), the experimental parameters, and the morphology of the printed line. Lines, 3 cm long, were printed in order to determine the best overlapping conditions to obtain continuous metallic lines with the largest possible height to width ratio. Finally, continuous lines were printed in a single-step process with the highest velocity (60 m/s) reported for printing metallic ink or pastes using LIFT.

2. Materials and Methods

A diode-pumped solid-state ps-pulsed laser (Atlantic 355-60, Ekspla, Vilnius, Lithuania), emitting at 532 nm a maximum power of 38 W at 400 kHz, was used for the LIFT process. Laser pulses with a duration of 13 ps and energies in the range of 5 to 20 µJ were used. Laser pulse energy was measured after the optical path using a thermal sensor with an accuracy of 3%.

In the first part of the work, a computer-controlled X-Y linear translation stage was used for displacing both the donor and the acceptor substrates with respect to the laser beam. When printing lines, the stage moving speed was fixed at 60 mm/s and the laser repetition rate was varied from 400 to 2000 Hz. The pulse-to-pulse distance or pitch distance between single successive laser pulses is directly determined by dividing the stage moving speed by the laser pulse repetition rate. Hence, the pitch distances varied from 30 µm to 150 µm. The sample started moving prior to firing the laser, avoiding any acceleration effect at the beginning or end of the line. The pitch distance and the number of laser pulses were adjusted for printing lines 3 cm long.

In this case, the optical path was comprised of a 5X beam expander (Sill Optics GmbH & Co. KG, Wendelstein, Germany) and a fixed focusing lens (Linos Focus-Ronar, Qioptiq Photonics GmbH & Co. KG, Goettingen, Germany) with a focal length of 58 mm. The laser beam was focused (beam waist \( \omega_0 = 5 \mu m \) and laser spot size of \( \approx 78.5 \mu m^2 \)) on the interface between the transparent donor substrate and the film of the material to be transferred. The Gaussian peak fluences ranged from 12.7 to 51.0 J/cm².

In the final part of the work, the sample position was fixed and the laser beam was scanned along the sample at high speed using a polygon scanner (LSE170, Next Scan Technology, Evergem, Belgium). In this case, the optical focusing system had a focal distance of 190 mm, which led to a beam waist at focus (\( \omega_0 \)) of 12 µm (laser spot size of \( \approx 452 \mu m^2 \) and Gaussian peak fluences from 2.2 to 8.8 J/cm²).

A commercial silver paste (Solamet PV17F, DuPont, Bristol, UK) was used as the donor material. The silver paste shows non-Newtonian thixotropic fluid behavior. Hence, the silver paste was gently stirred for 5 min before using it, to attain the equilibrium viscosity. The equilibrium viscosity was measured with a cone spindle and plate viscometer (HBDV-II+PCP, Brookfield Engineering Laboratories Inc., Stoughton, MA, USA), obtaining a value of 250 ± 30 Pa·s (cone spindle CPE-52, shear rate 5 s⁻¹, 23 °C). The paste was then deposited onto standard microscope glass slides, which acted as donor substrates, using a commercial coater (K101 Control Coater, RK Print Coat Instruments Ltd., Royston, UK). The thickness of the donor layer was measured in each donor sample both before and after the LIFT experiments. A c-Si polished wafer was used as the acceptor substrate. The gap distance between the donor and the acceptor was achieved by sticking commercial polyimide tape (Kapton, DuPont) onto the donor substrate. The morphology of the transferred paste and the holes
left on the donor substrate after the LIFT process was characterized using a confocal microscope (DCM3D, Leica Microsystems, Wetzlar, Germany). More details on the experimental setup can be found elsewhere [12,14–16].

3. Results

As a first step, LIFT of high viscosity silver paste using a ps laser was optimized by characterizing the morphology of printed voxels of the paste using single laser pulses [12,16]. The effect of different experimental parameters, such as a wide range of pulse energies (0.9 to 57.3 µJ), two gap distances (50 and 100 µm), and a fixed donor film thickness (30 ± 5 µm) was studied. Figure 1 shows the morphology of the best voxel obtained and printed using the shorter gap distance (50 µm) and a pulse energy just above transfer threshold (8.3 µJ). These voxels consist of single well-defined dots of paste, without any splashing or disaggregated cluster of paste. The aspect ratio (height divided by width) is also the largest obtained (0.09). These experimental parameters were the starting point for printing lines [12]. Continuous lines or more complex 3D structures can be printed using LIFT by either scanning the laser beam or moving the sample to overlap different voxels. In the first stage of this work, the beam was fixed, and the sample was moved using a motorized X-Y stage.

![Figure 1](image-url) Best voxel printed using a single laser pulse (pulse energy 8.3 µJ, donor film thickness 30 µm, and gap distance 50 µm): (a) microscope image, (b) false color confocal microscope image, and (c) height profile.

3.1. Influence of the Pitch Distance

Figure 2 illustrates the false color height map of both the donor (Figure 2b) and acceptor substrate (Figure 2d) and the corresponding cross-section profile (Figure 2c) of different pulse overlapping distances. Figure 2a shows the scheme of the temporal pulse train of laser pulses, i.e., when and how many laser pulses have been fired. The dash line in Figure 2c indicates the interface between the silver paste and the glass slide.

In the case of lines with a pitch distance of 150 µm and 100 µm, laser pulses cleaned off all the paste from the donor substrate. Due to the high viscosity of the paste, there is no filling of the generated hole in the donor film, even after very long times. The bottom of the hole is the surface of the glass slide and its depth is the real thickness of the paste film at that point. The depth of the remaining holes decreases as the overlapping distance is reduced. In the case of a 60 µm pitch distance, the holes are hardly observed. Around the holes, a bulge of paste can also be seen. These bulges are due to the displacement of silver paste which is not transferred but recoils to the border of the hole. The size of the bulge decreases as the pitch distance decreases.

The morphology of the transferred line is also strongly affected by the pitch distance. When a long pitch (150 µm) is used, there is no overlapping and the voxels are clusters of dispersed droplets instead of a concrete and well-defined dot as in the case of the single pulse experiment. When the pitch distance is reduced to 100 µm, the voxels start to overlap and a paste line, although not yet continuous, is formed. However, decreasing the pitch distance below 100 µm yields a much smaller quantity of transferred paste. At the shortest pitch distance (60 µm), no paste is transferred at all. This result was unexpected as it was assumed that the higher the number of laser pulses given to the donor film (i.e., the higher energy dose), the more paste to be transferred, although the morphology of the line could be
worse. It was observed that successive pulses overlapping in a proximate distance to each other affects the transfer mechanisms in such a way that the transfer is ceased, and hence there is a maximum pitch distance that acts as the transfer threshold for line printing.

![Figure 2](image_url)  
**Figure 2.** False color confocal images of (b) donor film and (d) acceptor substrate of the middle part of a printed line using 8.3 µJ pulse energy, 30 µm donor film thickness, 50 µm gap distance, and different pitch distances: (from left to right) 150 µm, 100 µm, 80 µm, and 60 µm. The laser pulse train (a) and cross-sectional profile (c) of both holes left in the donor and the transferred pastes are also shown. Dash line indicates the position of the glass-paste interface. Laser beam scanning direction is from right to left.

In order to investigate the effect of overlapping on the transfer mechanism, the beginning part of each line was also characterized by confocal microscopy (Figure 3). The same scheme of Figure 2 is used in Figure 3, illustrating the temporal train of laser pulses (Figure 3a), the false color height map of both the donor (Figure 3b) and the acceptor substrate (Figure 3d), and the corresponding cross-section profile (Figure 3c) of different pitch distances. The hole left in the donor substrate and the voxel transferred by the very first laser pulse is approximately the same in every pitch distance, because this first laser pulse in each line found fresh material, not affected by previous pulses. Lines printed with a pitch distance of 150 and 100 µm show the same trend in the beginning, as well as in the middle of the line. However, when a pitch distance of 60 µm is used, both the depth of the holes in the donor and the size of the transferred voxel decrease along the direction of the scanning laser. In the case of the shortest pitch distance, shown in Figure 3 (60 µm), there is only one hole in the donor substrate, followed by a shallow groove, and only the voxel corresponding to the first pulse is transferred.

### 3.2. Influence of the Laser Fluence

According to the results shown above, the best pitch distance is 100 µm, where almost continuous lines are printed, although the morphology of the independent voxels is not good, showing clustering of pastes and some debris. Using a thicker donor film could improve the morphology of the transferred voxel [14]. Therefore, the donor film thickness is increased to 40 ± 5 µm. It was, thus, necessary to re-adjust the laser pulse energy for printing with the new thicker donor layer. Figure 4 shows the false color height map of both the donor (Figure 4b) and the acceptor substrate (Figure 4d) and the corresponding cross-section profile (Figure 4c) of four lines printed using laser pulses separated by the same pitch distance (100 µm) with energies increasing from 5.6 to 11.2 µJ (Figure 4a).
Figure 3. False color confocal images of (b) donor film and (d) acceptor substrate of the beginning of a printed line using 8.3 µJ pulse energy, 30 µm donor film thickness, 50 µm gap distance, and different pitch distances: (from left to right) 150 µm, 100 µm, 80 µm, and 60 µm. The laser pulse train (a) and cross-sectional profile (c) of both holes left in the donor and the transferred pastes are also shown. Dash line indicates the position of the glass-paste interface. Laser beam scanning direction is from right to left.

Figure 4. False color confocal images of (b) donor film and (d) acceptor substrate of the middle part of a printed line using 40 µm donor film thickness, 50 µm gap distance, 100 µm pitch distance, and different laser pulse energies: (from left to right) 5.6, 8.3, 9.7, and 11.2 µJ. The laser pulse train (a) and cross-sectional profile (c) of both holes left in the donor and the transferred pastes are also shown. Dash line indicates the position of the glass-paste interface. Laser beam scanning direction is from right to left.

In the case of the lowest pulse energy, 5.6 µJ, the pulses are separated and neither the holes left in the donor nor the transferred voxels are overlapped. This result is similar to the one previously shown in Figure 2 for 30 µm donor film thickness and 8.3 µJ laser pulse energy. As the energy increases the
size of the hole increases, forming a continuous, wider groove clean of paste. Voxels increase their height but not their width in the transversal direction to the line as the pulse energy increases. The continuity of the line also improves until it is finally possible to transfer a 3 cm long continuous line while using a pulse energy of 11.2 µJ.

Figure 5 shows the cross-section profile of a visually continuous 3 cm line with the largest height to width ratio obtained using a donor film thickness of 40 µm, gap distance of 50 µm, and laser pulse energy of 11.2 µJ. It has a large aspect ratio of 0.25 with a cross-section area of 2425 µm². The width of the line is 134 ± 2 µm and the height of the line is 33 ± 1 µm (each value represents the average of five measurements in different positions with an uncertainty denoting one standard deviation). With such a profile, ps-pulsed LIFT lines can provide good electrical conductivity [17].

4. Discussion

The transfer mechanisms of LIFT are quite different when using Newtonian liquids or low viscosity inks as a donor as compared with using a paste with a high viscosity and comprised of Ag particles with sizes in the order of 1 to 4 µm [14]. In the latter case, it has been shown that the transfer mechanisms depend on the relationship between laser pulse energy, donor film thickness, and gap distances. When using pulse energies that are too high for a determined combination of donor thickness and gap distance, explosive transfer occurs and the paste splashes on the acceptor. If the energy is lower, typically the resulting voxels consist of small clusters of paste. Only when the donor film thickness is similar to the gap with a pulse energy near the transfer threshold is it possible to transfer a well-defined single dot of paste with a large aspect-ratio (concrete-dot transfer). In this situation, the protruding jet touches the acceptor substrate before disaggregation forming a stable paste pillar between the donor and acceptor. When the donor is lifted, the pillar breaks and the printed voxel remains on the surface of the acceptor substrate.

In this work, the best single voxel transfer, i.e., the dot with the largest aspect ratio (0.09), is obtained when the donor film thickness is similar to the gap distance. The voxel consists of a single dot of paste without disaggregation in several clusters. However, the distribution of paste is not homogenous. Thus, the transfer mechanism should be in the limit of the conditions for the formation of a stable pillar. Using higher or lower pulse energies lead to cluster transfer or no transfer, respectively. These conditions have been selected as a starting point for printing lines.
In the case of lines, a strong influence of the separation of the subsequent dots (pitch distance) on the morphology of the line has been shown. It is worth noting that the diameter of the laser focused beam (≈ 10 µm) is one order of magnitude smaller than the diameter of the hole remaining and the diameter of the voxel transferred (both in the order of 100 µm) and smaller than the pitch distance (30 to 150 µm). Therefore, when the term overlapping is used, it refers to the overlapping of the imprint left in the donor (hole or not) or to the overlapping of the printed voxels.

In the case of the longest pitch distance (150 µm), it should be long enough to avoid any interaction between subsequent dots and, then, the voxels should be similar to the static process. However, the voxels consist of several clusters instead of a well-define single dot of paste. In the case of the shortest pitch distances (shorter than 60 µm), the interaction effect is even more acute, i.e., the shorter the pitch distance the less amount of material transferred. The beginning of the line represents that the very first pulse is the same regardless of the pitch distance. These first voxels always have the same cluster structure. Similar overlapping effects have also been described by Sopeña et al. [13] for Ag inks with viscosity one order of magnitude smaller than the Ag paste used in the present work.

This behavior can be explained in terms of the transfer mechanism for high viscosity pastes [14] as follows: The very first pulse found fresh material and its transfer mechanism is that of the static situation, the protruding pillar expands, and a stable pillar is formed. Once the pillar is formed, the next laser pulse is fired. Time between pulses is in the order of several ms, longer than the typical times needed for the displacement of the paste, the formation of the protruding jet, or the formation of the stable pillar (in the order of µs).

When a second laser pulse is fired at a long pitch, but short enough to see overlapping between printed voxels (pitch distances of 150 or 100 µm), the expanding vapor bubble affects the delicate equilibrium of the previous system bubble pillar, breaking the stable pillar, and projecting clusters of paste onto the acceptor.

When the pitch distance is shorter (pitch distances of 30 and 60 µm), the paste wall between the bubble and the previous hole is very thin. The large pressure generated inside the bubble could break that wall, reducing the pressure, and thus reducing the amount of material pushed to the acceptor substrate. Although there is no transfer, there is movement of paste inside the film, leaving a void volume inside the film. The gas bubbles induced by subsequent pulses reduce their pressure in the same way, enlarging the void volume, and forming a draught tunnel along the laser scanning trace in the film, as shown schematically in Figure 6.

![Figure 6. Schematic of the laser pulse overlapping effect with a short pitch distance (30 µm) on the donor substrate in the LIFT process: (a) pulse laser focused onto the glass-paste interface, (b) gas bubble generation and paste movement without transference, and (c) formation of a draught tunnel due to interconnection of the overlapped pulses.](image)

To verify the proposed mechanism, the donor was characterized again by optical microscopy, but in this case through the glass slide side using a long working distance objective. Figure 7 shows images focused on the film-glass interface of the middle part of laser scanning traces left in the donors with pitch distance of 100 µm, 80 µm, 60 µm, and 30 µm. In the case of pitch distance 100 µm (Figure 7a), individual holes are observed. When the pitch distance is reduced to 80 µm (Figure 7b), instead of holes, craters appear near the film-glass interface, i.e., the crater formed on the paste surface and the crater left on the film-glass interface are not connected. And the diameter of these holes is smaller than the ones with pitch distance of 100 µm.
This can be caused by the overlapping of the two successive generated bubbles. When the bubble expands to the adjacent crater, its pressure decreases, and it is not high enough for forward transferring of the material. With pitch distances of 60 µm and 30 µm (Figure 7c,d), the overlapping effect becomes more evident. The shallow groove formed on the film-glass interface implies the overlapping of the bubbles, which finally forms the tunnel in the donor.

Continuous lines were obtained only by increasing the donor film thickness (40 µm), while maintaining the gap distance. In this case, the laser pulse energy must also be increased because the mass of silver paste to be transferred is larger. The voxel morphology using low pulse energies yields similar results as those obtained with a thinner donor film (30 µm), although the transfer mechanism is different. In the thicker donor film case, the holes remaining in the donor are not homogeneous and the pillar is not stable and disaggregates in several clusters. By increasing the laser pulse energy, it is possible to move more material and form a stable pillar. Because the donor film is thicker, the expansion velocity of the jet is smaller than in the case of the thinner donor film. The walls formed between successive bubbles are also thicker, and thus interference between pulses is more unlikely. The pillar remains stable until the donor is lifted. Only at this moment does the pillar break, pulling the material from the donor to the acceptor, leaving a clean groove in the donor film. The printed line is continuous only at the higher pulse energies, which move enough material to homogeneously overlap the voxels.

Previous results have been related to those observed in multipulse LIFT of low viscosity fluids [10]. In that case, there is an effect of fluid movement, filling up the void after the printing event. Therefore, there is a temporal limit for printing successive voxels, i.e., the time needed for the fluid to move [18]. This limit can be overcome by using high repetition lasers [11,19]. If the time between pulses is short enough as compared with the beam displacement, the cavitation bubbles generated in the fluid by successive pulses interact and merge, the transferred jets are linked, and a continuous line can be printed.

When the viscosity increases, the time that the fluid needs to fill the hole increases, the column between the donor and acceptor remains, and a spatial effect arises [13]. In the present work, the viscosity is so high that there is no fluid movement after the laser pulse and the void remains for a very long times, and thus the high viscosity paste LIFT can be understood as a “frozen” state of the low viscosity ink LIFT. The merge of the cavitation bubbles forming a continuous channel (Figure 7) is now undesirable. The limitation is not temporal but spatial. Successive voxels should be printed at a threshold pitch distance, short enough for good overlapping but long enough to avoid interactions between successive voids. This result is positive in the sense that it is possible to escalate the process for printing at a very high speed, if just the laser repetition rate and the scanning speed are increased in the same factor.

5. LIFT Printing of Lines at High Speed

In the previous section, the physical mechanisms that affect the overlapping of single voxels were discussed. To do so, it was necessary to study lines printed at a low speed (60 mm/s). According to that, the best pitch distance between voxels is 100 µm. From this starting point, it is possible to escalate the
printing process to much higher speeds, by simply increasing the laser repetition rate and maintaining the 100 µm pitch.

In this work a high-speed optical polygon scanner (LSE170, Next Scan Technology, Evergem, Belgium) working at 59.8 m/s was used to print long silver lines. The laser repetition rate was adjusted to 598 kHz in order to keep the 100 µm pitch distance. The laser pulse energy was also adjusted because the focusing optics in the polygon scanner is different than that used in previous sections. The gap distance (50 µm) was the same as in the low scanning speed experiments, whereas the donor film was slightly thicker (50 ± 5 µm). Pulse energy was varied from 6.9 to 19.2 µJ. For example, Figure 8 shows microscopic images of lines printed at two energies, (a) 6.9 and (b) 18.1 µJ. The optimum line was transferred using 18.1 µJ. It has the largest aspect ratio (0.13), a width of 155 ± 5 µm and a height of 20 ± 2 µm. Lines deposited with lower pulse energies are comprised of clusters of paste and are not continuous, whereas lines deposited at higher energies are wider, and thus they have a lower aspect ratio.

Figure 8. Microscope images of silver lines printed at 59.8 m/s using 50 µm donor film thickness, 50 µm gap distance, and two different laser pulse energies: (a) 6.9 and (b) 18.1 µJ.

6. Conclusions

In this article, silver lines were printed using high-speed LIFT of high viscosity commercial Ag paste. First, parameterization of LIFT printing of dots of paste using single laser pulses was performed. Voxel morphology was characterized for different process parameters (laser pulse energy, gap distance, and donor film thickness). Secondly, once the best conditions for single voxel printing were determined, the issues regarding overlapping of the voxels were studied and discussed in terms of the physical transfer mechanisms of high viscosity fluids. Due to the rheological properties of the paste, the void created in the donor film by the laser pulse remains after the printing event, affecting the transfer of successive voxels. When two laser pulses are fired at short pitch distances (<60 µm), there is no transfer at all. Only when the pulses are separated a distance long enough to avoid bubble interference but short enough to allow voxel overlapping (≈100 µm), is it possible to print continuous lines, once the experimental parameters are properly adjusted. Using a laser pulse energy of 11 µJ, a gap distance of 50 µm, and donor film thickness of 40 µm, continuous lines with aspect ratios of 0.25 were printed. Finally, the knowledge obtained allowed us to print silver lines at high speeds (up to 60 m/s) combining a polygonal scanner working at 59.8 m/s with a high repetition rate laser (598 kHz). Laser pulse energy was increased due to the different optical system as compared with a standard fix lens. The best lines were printed using 18.1 µJ.

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