Laser-induced Forward Transfer (LIFT) of Congruent Voxels

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Abstract

Laser-induced forward transfer (LIFT) of functional materials offers unique advantages and capabilities for the rapid prototyping of electronic, optical and sensor elements. The use of LIFT for printing high viscosity metallic nano-inks and nano-pastes can be optimized for the transfer of voxels congruent with the shape of the laser pulse forming thin film-like structures non-lithographically. These processes are capable of printing patterns with excellent lateral resolution and thickness uniformity typically found in 3-dimensional stacked assemblies, MEMS-like structures and free-standing interconnects. However, in order to achieve congruent voxel transfer with LIFT, the particle size and viscosity of the ink or paste suspensions must be adjusted to minimize variations due to wetting and drying effects. When LIFT is carried out with high-viscosity nano-suspensions, the printed voxel size and shape become controllable parameters, allowing the printing of thin-film like structures whose shape is determined by the spatial distribution of the laser pulse. The result is a new level of parallelization beyond current serial direct-write processes whereby the geometry of each printed voxel can be optimized according to the pattern design. This work shows how LIFT of congruent voxels can be applied to the fabrication of 2D and 3D microstructures by adjusting the viscosity of the nano-suspension and laser transfer parameters.

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1. Introduction

Direct-write processes are evolving into viable techniques for printing functional materials over diverse types of surfaces for the digital fabrication of microelectronic devices. These simple, relatively fast, low cost and environmentally friendly alternatives to traditional photolithographic processes have the potential of completely changing the way microelectronics are presently fabricated. In general, direct-write processes rely on the dispensing, transfer or printing of discrete voxels at predetermined locations on a substrate. These voxels serve as the building blocks for the lines, sheets and 3D structures required to fabricate a device. Among these voxel-depositing direct-write processes, inkjet printing is one of the most widely used given its ability for non-contact printing of a wide range of functional fluids and inks [1]. Another direct-write process that has demonstrated great flexibility and attracted much attention is laser-induced forward transfer or LIFT. LIFT is a non-contact, nozzle-free additive process in which a controlled amount of material is transferred from a donor film (or ribbon) to a receiving substrate upon interaction with a laser pulse [2].

Since the initial reports of laser transferred copper metal patterns by Bohandy, et al. almost 30 years ago [3], the use and development of LIFT has increased steadily. These simple yet powerful techniques employ a pulsed laser to locally transfer material from a source film onto a receiving substrate in close proximity or in contact with the film, thus achieving the direct-write of patterns independent of the type of surface. The source is typically a coated laser-transparent substrate, referred to as the ribbon, source, or donor substrate. Laser pulses propagate through the transparent donor carrier and are absorbed by the film. Above an incident laser energy threshold, material is ejected from the film and propelled toward the acceptor or receiving substrate. Translation of the donor and receiving substrates, or scanning and modulation of the laser beam, result in complex pattern formation in three dimensions with speeds typically limited by the laser repetition rate. Commercially available, computer-controlled translation stages and/or galvanometric scanning mirrors enable rapid motion with sufficient precision to

generate high-resolution patterns from the individually written voxels that result from the laser transfer process.

These laser transfer techniques are capable of generating high-resolution patterns without the need for lithographic processes afterwards [4]. To better understand the applicability and potential new uses of laser direct-write and associated laser transfer techniques for printing electronics, it is useful to compare/contrast them with other well-established digital microfabrication processes such as ink-jet. As with ink-jet, laser transfer techniques are capable of precisely depositing or direct writing many types of functional materials (or their precursors) over virtually any type of surface or substrate in a conformal fashion. Unlike ink-jet, laser transfer techniques are not constrained to deliver the material through a nozzle (thus impervious to clogging problems) and can deposit fluid materials ranging from very low viscosity inks to high viscosity pastes (making them immune to ink-surface wetting issues), as well as solids and even entire devices [5]. Furthermore, laser transfer techniques offer the added benefit of laser processing, such as micromachining, for material removal or trimming (not possible with ink-jet alone) and laser materials modification, for in-situ annealing, curing or sintering, all carried out using the same apparatus. What makes LIFT unique is its ability to laser transfer such a wide range of materials with relatively high resolution without requiring complex or time consuming steps. Furthermore, the laser transfer process does not have a deleterious effect on the properties of the voxels of material undergoing transfer.

When LIFT is used to transfer high-viscosity pastes, it is possible for the printed voxel to match congruently the illuminating laser pulse, a process referred to as laser decal transfer or LDT [6]. LDT offers a unique approach to direct-writing techniques in which voxel shape and size become controllable parameters, allowing the non-lithographic generation of thin film-like structures. The ability to adjust or entirely change the size and shape of a voxel according to the desired final pattern allows for the formation of complex structures in one single transfer step, reducing

the processing time as well as avoiding problems related to the merging of multiple voxels, which is prevalent with inkjet [7]. In this work we describe the various approaches undertaken to achieve congruent transfer of two different types of silver nano-suspensions comprising of a nanoink or a nanopaste, respectively. Examples of voxels laser transferred under similar conditions (laser spot size, shape and pulse energy) for various types of nano-suspensions are presented and the characteristics of the resulting patterns discussed. We demonstrate that the laser transfer of congruent voxels can be achieved with different types of suspensions by adjusting the properties of the nano-suspensions and the transfer gap. We conclude by showing examples of structures generated by the congruent transfer and discuss its applications in printed electronics.

2. Experimental

The experimental details for LIFT of high-viscosity nanopaste have been described elsewhere [8]. In brief, a frequency-tripled Nd:YVO₄ (JDSU, Q301-HD) pulsed laser (λ = 355 nm, 30 ns FWHM) operating at 30 kHz was employed for the congruent transfer process. The laser pulses were controlled in amplitude and time via an acousto-optic modulator (AOM 35085-3-350, NEOS Technologies). The laser beam was shaped through an aperture, which was then imaged through 10X or 20X microscope objectives to create a uniform beam profile onto the donor substrate or ribbon. A CCD camera positioned to view along the same optical axis as the focusing objective provided a real-time plan view of the transfer process, allowing alignment of the transfer pattern on the receiving substrate. The laser fluence was varied from 30 to 150 mJ/cm² at the donor layer interface with the donor substrate. UV transparent glass plates (50 mm x 75 mm) served as donor substrates, while (100) Si wafers were used as receiving substrates. A schematic showing the basic components of the LIFT setup used for this work is shown in Figure 1.

Two types of silver nano-suspensions, a silver nanoink (CCI-300, nominal viscosity 0.01 Pa·s, 20 wt% solids loading, Cabot Corp.) and a silver nanopaste (NPS, nominal viscosity <100 Pa·s, 80 wt% solids loading, Harima Chemicals, Inc.) were used for the

laser transfer experiments. The dynamic viscosity of the silver nano-suspensions as received and after solvent removal (both inks and pastes) was measured with a VROC microfluidic chip sensor (Rheosense, Inc.). By using different channel widths, this sensor can measure viscosities of both Newtonian and non-Newtonian fluids from $<0.01 \text{ Pa}\cdot\text{s}$ to $>500 \text{ Pa}\cdot\text{s}$. The silver nanoink was applied onto glass plates via spin coating at various speeds (1000 – 3000 rpm) in order to form coatings with thicknesses from 0.9 to 2 μm , as determined by confocal microscopy (Olympus LEXT OLS3000) and then heated in a hot plate at 100°C for 1-2 minutes to remove excess solvent and increase its viscosity to $\sim 0.4 \text{ Pa}\cdot\text{s}$. The silver nanopaste was doctor-bladed into 1 - 25 μm deep wells previously etched into a different set of glass plates and then dried for one minute at room temperature using N_2 flowing along the surface of the plate to improve uniformity of the paste layer inside the well and raise its viscosity over $100 \text{ Pa}\cdot\text{s}$. Once prepared, both types of donor substrates with silver nano-suspensions were stored in dry boxes purged with dry nitrogen to avoid moisture absorption and used within 24 hours of preparation. Figure 2 illustrates schematically the steps taken to prepare the donor substrates for the two types of nano-suspensions used for this work.

Each type of donor substrate was then placed, coating side down, over the receiving substrate and secured by a vacuum chuck mounted on top of a computer controlled X-Y stage pair. Arrays of voxels of different shapes were laser transferred as a function of laser fluence or donor-to-receiver gap distance. The actual thickness of the nanopaste layer inside the wells was determined from the voids left in the ink layer after transfer using confocal microscopy. Once printed, the transferred voxels were cured in an oven at 100°C for 1 hour for confocal microscopy and scanning electron microscopy (SEM) (JEOL, JSM-7001F) analysis. The samples were further cured in an oven at 250°C for 1 hour for electrical characterization using four-point Kelvin probes connected to a Keithley 2400 sourcemeter. The height and width of the structures were measured with a KLA Tencor P-16 profiler.

3. Results and Discussion

The transfer of low viscosity fluids using LIFT has been studied extensively by various groups[9, 10, 11] and is characterized by the formation of a bubble which upon collapse gives rise to a jet that breaks up into droplets carrying the fluid across the gap towards the receiving substrate. The formation of a jet and subsequent droplets implies that the shape of the laser pulse impinging on the donor layer is lost due to surface tension forces present in the fluid. Figures 3(a) and (b) show the LIFT of voxels using a circular and square laser spot with a low-viscosity ink. Figure 3(b) illustrates this loss of spatial features by revealing the washed-out shapes of the incident square laser spot obtained after the transfer of the as-received CCI-300 silver nanoink (viscosity ~ 0.01 Pa·s) spin coated onto a donor substrate. Note how each voxel collapses into a drop-like feature with the original shape of the laser pulse barely recognizable. If higher viscosity nano-suspensions are used instead, it is possible to achieve congruent voxel transfers as shown in Figs. 3(c) and 3(d). In this case, a donor substrate comprising a $2\text{ }\mu\text{m}$ thick NPS silver nanopaste layer dried with flowing nitrogen gas (viscosity ~ 100 Pa·s), was used with the same apertures as before and this time the printed voxels accurately reflected both the size and shape from the laser spatial profile generated with each aperture.

As the above example demonstrates, it is possible to increase the viscosity of the nano-suspensions by carefully removing volatile components of the ink through heating or drying while avoiding excessive changes that result in consolidation of the solids in suspension and premature curing. For the CCI-300 silver nanoink this was accomplished by gentle heating in a hotplate at $100\text{ }^{\circ}\text{C}$ for a few minutes. In order to quantify the loss of volatile solvents from the ink during this step, the weight change of the nano-suspension in the donor substrate was measured before and after heating. These measurements revealed a fast drop in weight during the first minute during heating and a reduced rate of weight loss afterwards, indicative of the removal of highly volatile organic components from the ink. Because the composition of the CCI-300 silver nanoink is proprietary, we cannot ascertain the exact nature of the volatile organic removed by this curing process, but we were able to establish that once the nanoink underwent a 55 wt% loss, its viscosity

increased to ~ 0.4 Pa·s, without damaging the nano-suspension. Higher weight losses resulted in nanoink films on the donor substrate that were no longer stable but instead had been cured into silver metallic layers. The time it took to achieve this weight loss varied as a function of the initial thickness of the nanoink layer, which depended on the rotation speed during spin coating as shown in Figure 4. However, for all these thicknesses, a 55 wt% change resulted in coatings with the silver nano-suspension still intact as determined by their non-metallic, uncured nature.

The donor substrates with the CCI-300 nanoink which underwent a 55 wt% reduction were used for transfers using a range of fluences to determine the optimum laser transfer conditions. The laser fluence was incrementally adjusted from the transfer threshold energy until the transfer was far from optimal (20 to 75 mJ/cm² for this work.) A graphical summary of the results obtained for donor to receiving substrate gap distance of 5 μ m is shown in Figure 5. As the images of the transferred voxels reveal, congruent transfers are possible only for a very narrow parameter space just above the transfer threshold of 24 mJ/cm² for this donor substrate, with individual voxels shown in Figures 5(e) and (f). In fact, the majority of the transfers with the thermally-treated CCI-300 nanoink are far from congruent and show clear signs of interaction between the transferred voxel and the laser pulse as revealed by the square void left in their middle. The influence of the gap between the donor substrate and the receiving substrate on the voxels using low viscosity nano-ink was also studied. It is clear that the shape of voxels is fairly sensitive to the gap separation and that the square-like voxels were achieved with the smaller gaps. As expected, when the gap separation increases higher fluences are required to produce the square-shape of voxels. At larger donor-to-receiving substrate gaps, the square shape of the voxels is lost completely for all fluences making congruent transfers impossible to achieve. This is illustrated by the confocal microscope images of transfers shown in Figures 6(a-d).

The previous results indicate that congruent transfers require higher viscosities than those achievable with the use of nanoinks. By using nanopastes designed for

screen-printing rather than nanoinks tailored for inkjet, higher viscosities can be obtained. For example, silver nanopastes like NPS (manufactured by Harima Chemicals) can be used for congruent transfers as previously reported by various groups [12, 13]. Because of its high viscosity, the as-received paste cannot be spin coated onto the donor substrate and is instead doctor bladed over a well on the donor substrate to form a film of uniform thickness. Still, the viscosity of the NPS nanopaste needs to be raised over 100 Pa·s to achieve congruent laser transfer [14]. Given the very high solids content of the NPS paste (80 wt%) heating would result in damage of the coating surrounding the nanoparticles that prevents them from reacting with each other [15]. Instead, removal of excess solvent was accomplished by flowing nitrogen gas over the surface of the paste in the wells of the donor substrate. Upon reaching lower volatiles content, donor substrates with silver nanopaste layers ranging in thickness from less than a micron to several microns were evaluated for congruent transfers. The results for four different silver nanopaste thicknesses (0.8, 2.2, 2.8 and 3.8 μm) at three different donor-to-receiving substrate gaps (5, 25 and 75 μm) are shown in Figures 6 (e-h). In this case, a 50 μm x 50 μm square laser spot was used and the laser fluence corresponding to the threshold for transfers for the four different nanopaste layers on the donor substrates was recorded. As expected, the threshold for transfer increases with nanopaste thickness, however, the integrity of the transferred voxels depends both on their thickness and the gap distance between the donor and receiving substrate. This can be attributed to the fact that upon release from the donor substrate, the voxel must travel across the gap before it lands on the receiving substrate and in the process be subject to drag forces at atmospheric conditions. Thus, the 0.8 μm thick voxels break apart for the 25 and 75 μm gap transfers. The thicker voxels (> 2 μm) fare better, although as the gap increases the accuracy of their placement onto the donor substrate worsens. Overall, the thicker the voxel, the less susceptible it is to breakage and drifting ~~it is~~ during transfer independent of the gap distance. These results agree with reports by other groups whereby LIFT is conducted under reduced atmospheric pressure to minimize fracture and shifting of the voxel during transfer [16].

The successful transfer of congruent voxels also depends on the laser fluence applied to the nanopaste coating on the donor substrate. As the SEM images in Figure 7 illustrate, the overall size of the voxels generated under congruent transfer is not as sensitive to the applied laser fluence as in the case of LIFT with low viscosity nano-inks, see Figure 5. However, as the fluence further exceeds the transfer threshold for a particular silver nanopaste layer in the donor substrate, fracturing and deformation of the voxels becomes an issue as shown in Figure 7 for a series of $25\text{ }\mu\text{m} \times 25\text{ }\mu\text{m} \times 2.2\text{ }\mu\text{m}$ voxels transferred at progressively higher fluences. However, as these results show, the range of fluences over which congruent transfers can be achieved with no change in the size of the voxel is wider than for transfers of low viscosity nano-inks (Figure 5). In this case, the shape of the voxel remains uniform, but its size increases with fluence as previously reported [17].

Despite these difficulties, the capability to perform congruent laser transfer across a gap is highly relevant for many applications in printing electronics. First, the ability to generate congruent voxels by LIFT of high viscosity nanopastes makes possible the printing of entire patterns with one single laser pulse rather than having to assemble them from individual voxels. This is demonstrated by the printing of entire devices, as shown by the array of double split-ring resonators in Figure 8(a), where each voxel comprises a pair of resonators. Stacking congruent voxels over each other by adjusting the gap between the donor and receiving substrates allows the printing of 3D structures with high aspect ratios, which are useful for fabricating vertical interconnects as shown in Figure 8(b). Finally, by combining the ability to vary the shape of each voxel with their precise placement, more complex 3D microstructures become feasible, as shown in Figure 8(c). Here a series of interconnects built above a gap were fabricated directly with congruent LIFT without the need for sacrificial support layers. The free-standing rectangular pads on top of the square posts survived the transfer and curing process intact due to the high viscosity of the treated NPS silver nanopaste and spanned the gap without

shorting to the central conductive gold trace in this particular configuration. Electrically, these planar interconnects performed well as shown by the measured I-V plot in figure 8(d), with a resistance of $2.0\ \Omega$, corresponding to a resistivity of $25\ \mu\Omega\cdot\text{cm}$ (about 15 times higher than the conductivity of bulk silver, $1.6\ \mu\Omega\cdot\text{cm}$). Such printable planar interconnects are essential for connecting discrete devices in circuits fabricated on thin substrates for applications requiring embedded components or for electronics on flexible substrates.

4. Summary

The advantages of using LIFT to print patterns of high-viscosity silver nano-suspensions with high congruence with respect to the spatial profile of the incident laser beam have been demonstrated. By first investigating the transfer process with a low-viscosity nanoink by varying parameters such as laser fluence and ink viscosity, it was clearly shown that the quality of the transferred voxels reached a limit for low-viscosity inks and that nano-suspensions with higher ink viscosities, as those found in nanopastes, is required for congruent transfers. We have shown that the laser fluence window for congruent transfers with low viscosity nanoinks is very narrow and can only be achieved at donor-to-receiving gaps of $5\ \mu\text{m}$ or less, making such transfers impractical for most applications. In addition, the size of the transferred voxels increases with laser fluence for nanoink transfers, while remaining almost constant for transfers with donor layers comprised of silver nanopaste. This fact is demonstrated by the transfers of high-viscosity voxels, which remain congruent with the laser beam shape across donor-receiver gaps of up to $75\ \mu\text{m}$. The ability to use LIFT to print congruent voxels is important for applications in embedded and flexible electronics which call for non-lithographic processing of thin-film-like patterns and/or free-standing structures. This feature is shown clearly in applications such as the printing of split-ring resonators, planar interconnects and other 3D constructs. Such 3-D structures would be difficult if not impossible to achieve with techniques that only work with low-viscosity inks such as inkjet.

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Figure captions:

Figure 1. Schematic illustrating the main elements in a system for laser-induced forward transfer or LIFT.

Figure 2. Schematic illustrating the two processes employed to prepare the donor substrates coated with (a-c) CCI-300 silver nanoink and (d-f) NPS silver nanopaste.

Figure 3. SEM images of transferred voxels with two different viscosities onto Si receiving substrates for different aperture shapes: (a,b) low viscosity silver nanoink (Cabot CCI-300) and (c,d) high viscosity silver nanopaste (Harima NPS). The apertures (top-left) and their corresponding laser beam profiles (top-right) are shown above each column of arrays.

Figure 4. Plot of weight change of spin coated CCI-300 silver nanoink on donor substrate as a function of curing time on a hot-plate at 100 °C for four separate nanoink thicknesses produced at four different spin coating speeds (between 1000 and 3000 rpm). The dotted line shows a 55 wt% reduction, which was found to work best.

Figure 5. (a) Confocal micrographs of voxel arrays with the donor to receiving substrate at a 5 μm gap and sequentially transferred at laser fluences starting at threshold (24 mJ/cm²) and increasing. (b – f) Higher magnification confocal micrographs showing the resulting voxels at various selected fluences.

Figure 6. (a-d) Confocal micrographs of voxel arrays of $50\text{ }\mu\text{m} \times 50\text{ }\mu\text{m}$ printed using low viscosity NP ink with various ink thicknesses of (a) 0.9, (b) 1.2, (c) 1.7 and (d) 2.0 μm , plotted as a function of transfer distance (5 – 75 μm). (e-h) SEM images of arrays of $50\text{ }\mu\text{m} \times 50\text{ }\mu\text{m}$ printed using high viscosity NP paste with various paste thicknesses of (a) 0.8, (b) 2.2, (c) 2.8 and (d) 3.8 μm , plotted as a function of transfer distance (5 – 75 μm). Each array was printed at the laser threshold energy for each thickness.

Figure 7. SEM images of $25\text{ }\mu\text{m} \times 25\text{ }\mu\text{m} \times 2.2\text{ }\mu\text{m}$ congruent voxels printed by LIFT displayed as a function of laser fluence (40 – 72 mJ/cm^2).

Figure 8. SEM images of congruent voxels prepared by LIFT: (a) double split-ring resonators, (b) High aspect ratio 3D structures for vertical interconnects, (c) Free-standing planar interconnects. (d) I-V curve measured across the interconnects in (c).

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