Digital Microfabrication by Laser Decal Transfer

Alberto PIQUE, Raymond C.Y. AUYEUNG, Heungsoo KIM, Kristin M. METKUS and Scott A. MATHEWS

Naval Research Laboratory, Materials Science and Technology Division, Code 6364,
4555 Overlook Ave. SW, Washington, DC 20375, USA
E-mail: pique@nrl.navy.mil

Laser decal transfer is a novel direct-write technique that allows the digital microfabrication of thin film-like patterns or structures on many substrates without the use of lithography and etch steps. Laser decal transfer is based on the non-phase transforming laser-induced forward transfer of complex suspensions or inks. This type of laser transfer allows the direct printing of materials such as metallic nano-inks from a donor substrate to a receiving substrate while maintaining the size and shape of the area illuminated by the laser transfer pulse. Consequently, this technique does not exhibit the limited resolution, non-uniform thickness, irregular edge features and surrounding debris associated with earlier laser forward transfer techniques. Laser decal transfer can be used to make continuous and uniform metallic lines typically 5 micrometers or less in width, and a few hundred nanometers in thickness. These lines are of similar scale as patterns generated by lithographic techniques. After transfer, the lines are cured either thermally or by a CW laser to become electrically conductive. *In situ* laser curing results in lines with resistivities as low as 3.4 μΩ cm. This novel laser direct-write technique is ideally suited for digital microfabrication applications and can be used for the development, customization, modification, and/or repair of microelectronic circuits.

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1. Introduction

Lithography and etching processes have reigned supreme in the micro and nanofabrication areas since the beginning of the microelectronics industry. There are considerable challenges, however, in adapting lithographic processes to new applications requiring processing on plastic or flexible substrates, production of small batch sizes and customization or prototype redesign. In these cases, the complexity and high capital and operating costs of the equipment involved, and the limited range of materials that can be patterned represent significant shortcomings. Furthermore, the use of lithographic techniques requiring the vacuum deposition of a thin film and its subsequent etching to achieve a desired pattern from a given material is not practical for many applications requiring the modification, and/or repair of existing microelectronic devices or circuits. As a result, there is a pressing need for the development of new microfabrication techniques and approaches that avoid these limitations.

An alternative to lithography is provided by direct-write techniques. Direct-write techniques are digital microfabrication processes that allow the formation of patterns or structures under complete computer control. Examples of direct-write techniques include inkjet, laser chemical vapor deposition or LCVD and laser direct-write or LDW. In general, these non-lithographic techniques allow the deposition of individual 3-dimensional pixels or "voxels" of virtually any type of material at precisely defined locations to generate a given pattern or shape with little or no material waste. For applications requiring the modification or repair of an existing microelectronic circuit or device, direct-write techniques offer the best chance for success. However, most direct-write techniques are not capable of depositing patterns of electronic materials with placement precision under a micron, with thickness of a few hundred nanometers and with feature morphology and size similar to the surrounding thin film structures already present in the device. This has limited their use and implementation outside the laboratory.

In this paper, we describe the laser decal transfer of metallic nanoinks for the direct-write of thin-film-like patterns on various types of substrates. Laser decal transfer is based on the non-phase transforming laser-induced forward-transfer of complex suspensions or inks. This new approach represents a significant advance in LDW processes given the improved spatial resolution, increased thickness uniformity, sharper edge features and minimal surrounding debris as compared to previous laser forward-transfer processes. The laser-transferred material is deposited onto the substrate surface maintaining the size and shape of the transfer laser spot, thus the name "decal transfer". This capability has far greater potential than LCVD or inkjet since it can minimize writing times by varying the shape or profile of the transferred voxel, thus generating the required pattern for an interconnect or repair in considerably fewer steps. Such ability to dynamically control the shape or size of the voxels during the writing process is unique among other digital microfabrication processes and can be used to fabricate many new types of structures.

2. Background

Some direct-write processes, like LCVD, can produce thin-film-like patterns, but are limited by their slow deposition rate, narrow choice of materials and complexity [1]. As a result, LCVD has found limited applications, mainly for the repair of high value-added parts such as high resolution

lithography masks. Simpler additive direct-write processes like inkjet can operate without the need for a vacuum and at room temperature. However, despite inkjet's inherent simplicity, achieving pattern resolutions under 5 micrometers on different types of substrate surfaces is very challenging with inks containing electronic materials such as metals or ceramics. This is due in part to the difficulties in reliably dispensing 2 to 3 μm diameter droplets and the wetting characteristics affecting the contact angle between the dispensed droplets and the targeted surfaces. In these cases, the inadequate consistency of the inkjet droplets and the variable phobicity of the surfaces limit the achievable minimum feature size, surface morphology and edge quality of the printed pattern. As a result, inkjet performs poorly in the fabrication of thin-film-like patterns of electronic materials over existing microelectronic circuits.

Laser direct-write is a general term that encompasses laser-based modification, subtraction and addition processes that can create patterns of materials directly on substrates without the need for lithography or masks [2]. In additive mode, the LDW technique behaves effectively as a "functional materials printer". In this case, laser-forward transfer processes are used for the deposition of metals, oxides, polymers and composites under ambient conditions onto virtually any type of surface. This laser printing process has been used with great success in the fabrication of sensors, microbatteries, interconnects, antennae and solar cells [3-6]. A combination of different LDW techniques can be used for fabricating embedded electronic devices and circuits. For example, devices first embedded in laser micromachined pockets can then be wired to form working circuits by laser printing the metallic interconnects [7,8]. It is also possible to use the LDW technique to transfer entire devices such as semiconductor bare dies or surface mount passive and active components inside a pocket or recess in a substrate, thus performing the same function of pick-and-place machines used in circuit board assembly [9, 10]. No other direct-write technique offers this broad range of capabilities for the rapid prototyping of electronic circuits on a single platform.

Despite these capabilities, there has been limited use of LDW techniques for the repair, modification and customization of integrated circuits, thin film transistor displays and other microelectronic circuits. This is the result of the strict requirements in feature size, layer thickness, cross contamination and material properties associated with these devices. The greatest challenge involves the development of non-contact LDW processes capable of generating patterns of functional materials such as metals or dielectrics with feature sizes of a few microns in width and a few hundred nanometers in thickness. This is what we refer to as the transfer of thin-film-like patterns. Furthermore, these patterns need to be deposited under atmospheric conditions, at room temperature, with good adhesion to various types of organic and inorganic surfaces, with little or no cross-contamination of adjacent areas, and at high writing speeds (up to cm/sec).

The repair of thin film transistor-liquid crystal displays (TFT-LCD) is a relevant example where the above requirements need to be satisfied. As the size of TFT-LCD panels continues to grow, the repair of local circuit defects is of great importance to manufacturers aiming to increase yield while bringing costs down. A particular type of defect that is very hard to repair is a line open defect. The repair of line open defects is far more difficult since it requires the deposition of a conductive pattern on the surface of the panel in order to complete the circuit. Given that conventional semiconductor processes are not suitable for the repair of these kinds of defects, direct-write processes offer the best alternative. Currently, the only direct-write process in use for the repair of line open defects in TFT-LCD panels is LCVD [11]. However, LCVD is far from ideal given its inherent complexity (the deposition must be performed under vacuum), limited choice of materials (each requires an organometallic precursor) and slow deposition rates (typically 10 μm/sec).

The use of LDW techniques based on laser forward-transfer processes offers a better alternative to LCVD for the repair of TFT-LCD circuits if they can be adapted to deposit thin-film-like patterns. In principle, this should be possible with the LDW process known as laser-induced forward-transfer or LIFT [2]. LIFT allows the transfer of patterns with micron resolution and submicron thickness. Unfortunately, LIFT results in either the vaporization or melting of the material being transferred. As a result of these phase transformations, the quality of the material deposited by LIFT is negatively affected by oxidation, decomposition, formation of undesired phases and poor uniformity, particularly at the interface between adjacent voxels.

To avoid these limitations, we investigated a different LIFT process that allowed the printing of solutions or inks (comprising of suspensions of particles of the material of interest) without any phase transformation during the transfer [12]. The laser transfer of these complex multiphase rheological systems has proved extremely successful and has allowed the LDW of many different types of material systems for diverse applications such as microbatteries, photovoltaics and polymer-ceramic membranes [13-16]. Further refinements of this approach resulted in the adaptation of laser printing of rheological systems to the direct-write of thin-film-like patterns. To achieve the required resolution, we began using suspensions of nanoparticles. Early results demonstrated that the combination of laser printing and subsequent laser curing of the transfers could generate high quality transfers with thickness under a micron and conductivities near bulk [17]. However, the transfers still showed poor edge definition, thickness non-uniformities and the presence of debris, which made them unsuitable for the repair of TFT-LCD circuits.

More recently, a joint effort between our group and Photon Dynamics Inc. demonstrated that under the right conditions, laser forward-transfer processes can be used to generate metallic thin film patterns of several micrometers in width and sub-micron thicknesses [18]. By significantly increasing the viscosity of the nanoparticle suspension, we were able to achieve the required thin-film-like patterns with no debris, while maintaining excellent conductivity and adhesion. This novel laser decal transfer technique is promising for the fabrication of thin-film-like patterns given its additive capability under atmospheric conditions, highly precise transfer placement, compatibility with a wide variety of electronic materials and resolutions of the order of a few micrometers. After laser printing, the thin

film patterns are *in-situ* cured using a second laser or in an oven to achieve the desired properties (adhesion, chemical resistance and electrical conductivity.)

3. Experimental

The laser used for the transfers performed in this work was a frequency tripled Nd:YVO_4 laser operating at 355 nm with pulse energies of a few hundred μJ at kHz repetition rates. The laser pulses were controlled in amplitude and time by an acousto-optic modulator (AOM). These laser pulses were directed through an aperture and then through a 20X, 0.4 N.A. objective, forming an image at the focal plane as small as 2 μm (as measured on polyimide). An in-line CCD camera provided direct viewing of the receiving substrate (referred simply as the substrate) and the actual laser transfer from the target substrate or ribbon. Typical laser energies used for laser decal transfer were ~ 2 to 10 nJ (30 ns FWHM) as measured with an Ophir PD10 energy meter after the objective resulting in a fluence of 8 to 40 mJ/cm^2. The substrate was placed on top of a computer-controlled X-Y stage motion control system. Figure 1 shows a schematic of the basic setup.

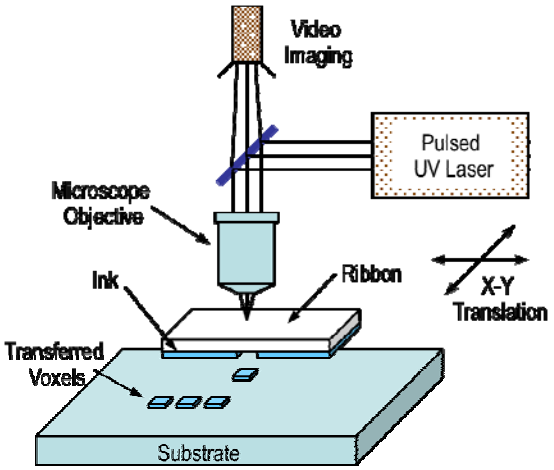

Fig. 1 Schematic diagram showing the basic components of the laser direct-write system used for laser decal transfers.

The ribbon was made from a 50 mm x 75 mm glass microscope slide to which a suspension or ink of silver nanoparticles (particle size: 3 to 7 nm) was applied using doctor blading. The resulting ribbon was then carefully placed with the ink layer side parallel and facing the receiving substrate separated by a 10 to 50 microns adjustable gap. The laser spot was focused onto this ink layer and a series of voxels were laser decal transferred by translating the ribbon to a new area after each laser pulse. Afterwards, the ribbon was removed for visual inspection of the pattern and/or laser curing. The shape of the laser spot was adjusted by using different apertures.

Lines were laser decal transferred by superimposing individual voxels along one direction. Transfers were performed on a variety of surfaces including polyimide, glass and p-type Silicon. The surface of the substrates was not pre-treated by any special techniques other than rinsing with organic solvents (acetone and isopropanol) and dried with nitrogen. Periodic inspection of the transfers using an optical microscope with differential interference contrast (Olympus BX60) was conducted to verify transfer quality and pattern dimensions. After transfer, the samples remained on the stage for laser curing or were placed in a convection oven for 30 min. at 200 °C for thermal curing.

The setup used for *in-situ* laser curing has been described previously [17]. Briefly, single-pass laser curing of as-deposited lines on silicon and glass substrates was performed with a cw green laser operating at 532 nm. This laser beam was first sent through a circular aperture to select the central region of the Gaussian intensity profile and then directed through the same 20X, 0.4 N.A. objective used for the laser decal transfer and focused onto a ~ 20 μm diameter spot as measured on polyimide. Typical laser powers measured after the objective ranged from 30 to 375 mW (higher powers damaged the transfers) resulting in intensities from 9 to 120 kW/cm^2 on the sample. The lines were scanned at different speeds ranging from 25 to 100 μm/s.

3.1 Characterization of laser decal transfers

Optical microscopy was used to characterize the transfers before and after curing and also to characterize the ribbon before and after the transfers. Once cured, the thickness, width and surface morphology of the transfers were determined using contact profilometry (KLA Tencor P-10), atomic force microscopy or AFM (Digital Instruments Dimension 3100) and scanning electron microscopy or SEM (LEO 1550). Elemental analysis using SEM energy dispersive X-ray or SEM-EDX was performed on the same instrument. The adhesion and chemical resistance was evaluated by subjecting the transfers to tape peel tests and immersion in solvents (water and isopropanol) respectively, and measuring any changes in morphology or electrical properties. Characterization of the electrical properties of the lines transferred across Au-pads on glass substrates was performed using standard 4-probe measurement techniques using a Keithley 2400 sourcemeter with 200 μA input current.

4. Results and Discussion

Comparison of the size of the removed material on the ribbon with the resulting transfer demonstrated that the laser decal transfer resulted in virtually a 1-to-1 correspondence in size and shape between the laser spots illuminating the ribbon and the transferred voxels. Figure 2(a) shows an SEM image from a series of silver nanoink voxels laser decal transferred onto a silicon substrate using three different apertures (rectangular, oval and circular, from top to bottom). As the SEM image shows, the resulting decal transfers matched the shape of the aperture imaged at the ribbon with no noticeable distortion. This is very important for repair applications, since specific voxel lengths and forms can be generated with a variable shape aperture, allowing the transfer of a complete repair pattern with one single laser pulse. The SEM image also shows that little or no debris exists between individual voxels. To determine the thickness and morphology of the laser decal transfers, AFM analysis was conducted on selected samples. The AFM image in Figure 2(b) shows two transferred voxels with a thickness of ≈200 nm. In general, the thickness of the transfers depended on the thickness of the ink layer on the ribbon and ranged between 100 and 300 nm. For any given thickness, however, the AFM scans demonstrated the

excellent edge definition and thickness uniformity of the laser decal transfers. Such features are similar to those obtained by lithographically patterning and then etching a vacuum deposited thin film layer of similar thickness. To our knowledge, no other laser forward transfer technique can generate the kind of thin film-like patterns that can be achieved using laser decal transfer.

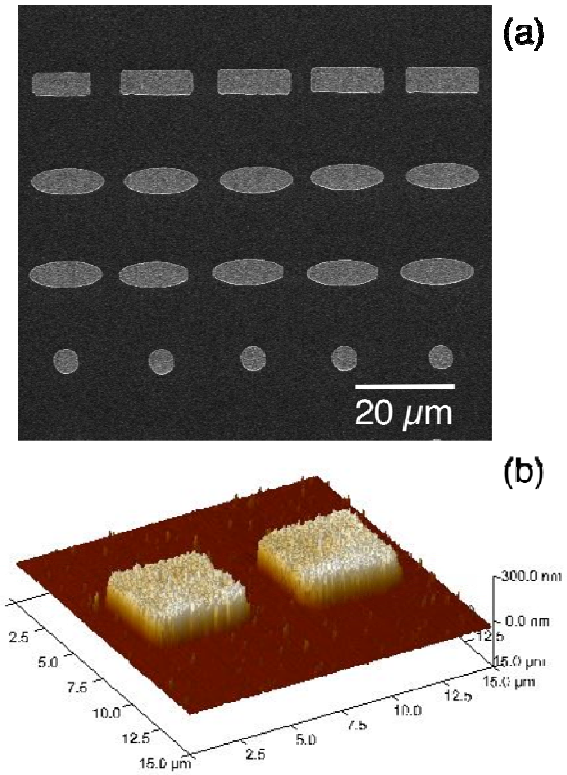

Fig. 2 Examples of laser decal transfer of silver nanoink on a silicon substrate. (a) SEM image showing the transfer of rows of voxels using different apertures. (b) AFM image from two square voxels showing the high thickness uniformity and sharp edge definition of each transfer.

To verify the absence of debris resulting from laser decal transfers, SEM-EDX analysis of the silver (Ag) metal was performed across one of the samples. This sample was oven cured and no further post-processing steps were used prior to the SEM-EDX analysis. Figure 3(a) shows the SEM image from the region on a silicon substrate with an array of laser decal transferred square voxels mapped by SEM-EDX. Figure 3(b) shows the corresponding EDX mapping of the Ag_L line. Note how the Ag element mapping (red dots) exactly follows the area covered by the voxels, while virtually no signal (dark color) is found in the regions in between.

The use of high viscosity nanoparticle suspensions (≈100,000 cP) as inks for the ribbon plays an important role in the ability to perform the decal transfers. In the past, most of the nano-inks used in our transfers were of relatively low viscosity (< 100 cP) and would transfer in the form of droplets as long as the area illuminated by the laser pulse was ≥ 100 μm^2. As the droplets reached the substrate surface, patterns of varying shape and thickness would be generated analogous to those formed by inkjet. Clearly these did not qualify as decal-type transfers. When the area

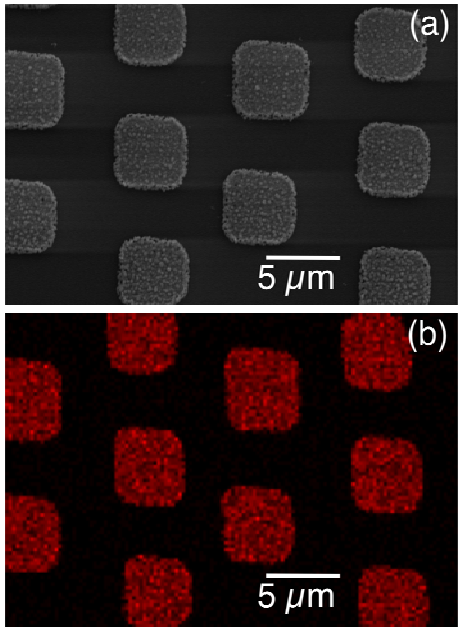

Fig. 3 SEM-EDX analysis of an array of laser decal transfer silver voxels. (a) SEM image and (b) corresponding SEM-EDX element mapping of the Ag_L line. Red indicates the presence of silver while dark color indicates its absence outside the regions covered by the voxels.

illuminated by the laser pulse was further reduced, the release of the ink from the ribbon no longer took place uniformly and the resulting transfer showed a high degree of spattering with a marked increase in surrounding debris. In fact, rather than forming droplets, the transfer broke into a diverging spray. Figure 4(a) shows a SEM image of such a transfer from a ≈10 cP Ag nanoink suspension. This behavior might be caused by the high shear stresses experienced by the nanoink as it is released from the ribbon resulting in a spray-like transfer. By using much higher viscosity nanoparticle suspensions, it is possible to take advantage of shear thickening effects that prevent the breakup of the transfer into a discontinuous ensemble. This is the basis of the laser decal transfer and it is shown in figure 4(b) where the same conditions were employed as in 4(a), except for using a ribbon with an ink of significantly higher viscosity (≈100,000 cP).

In order to characterize the electrical properties of the laser decal transfers, multiple sets of continuous lines across gold pads on glass substrates were prepared. For this particular case, each line was made by laser decal transfer of individual 15 μm x 5 μm voxels, as shown in Figure 5(a). After completing the transfers, the resulting lines were laser cured *in situ* using the 532 nm cw laser. Individual lines were cured at various laser intensities ranging from 9 to 120 kW/cm^2 and their electrical resistances measured using a four-probe setup. For laser powers over 25 kW/cm^2 most lines showed low resistivities with no cracking or delamination and no signs of damage to the underlying glass substrate. For laser powers over 120 kW/cm^2 damage to the line was observed. The lines were cured using laser scanning speeds from 25 to 100 μm/sec. No significant difference in the quality of the lines was observed, indicating that higher curing speeds can be used as long as the

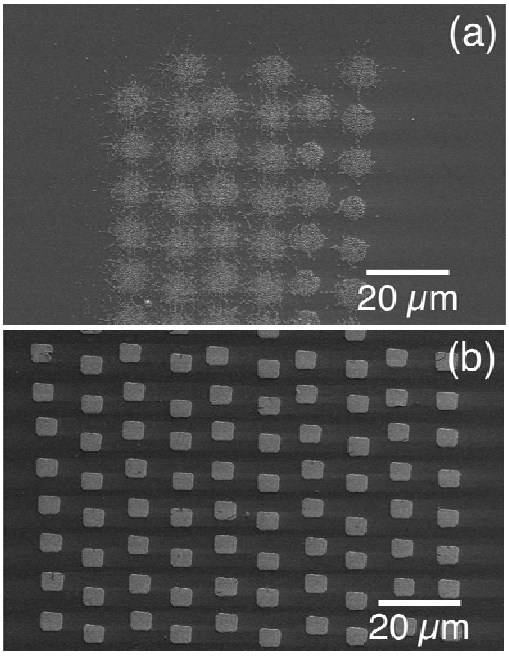

Fig. 4 SEM image showing laser transfers under identical conditions for (a) low viscosity (≈10 cP) silver nanoink and (b) high viscosity (≈100,000 cP) silver nano-particle suspension.

appropriate laser power is available. For those lines cured at laser powers between 25 and 120 kW/cm^2 the measured resistivities were all under 10 μΩ cm. For some samples, resistivities as low as 3.4 μΩ cm were measured, which corresponds to about 2.1 times the resistivity of bulk silver metal (1.6 μΩ cm at room temperature). Figure 5(b) shows a more complex line pattern made from the combination of two different shaped voxels (rectangle and rounded corner) rotated as necessary to form the "s" shaped line. The SEM image on Figure 5(c) shows how the adjacent voxels coalesce once transferred forming uniform and continuous lines on the substrate surface. This merging of individual voxels into one continuous pattern is unique to the non-phase transforming laser transfer of rheological fluids and impossible to achieve with traditional LIFT, which relies on the vaporization or melting of the transferred material. The ability to laser decal transfer complex fluids and suspensions without degrading their properties is crucial for printing highly conductive thin-film-like patterns devoid of discontinuities, interfaces or steps from one voxel to another directly on the surface of an existing circuit.

Since laser decal transfer is able to generate patterns with high edge definition and low debris outside the transferred region, it is also well suited for the deposition of patterns or lines in close proximity or with small gaps. Figure 6 shows an AFM image from a region between two closely spaced lines (≈2 μm) made by laser decal transfer. Note that despite the close gap between the lines, they are well isolated from each other. This is very important for fabricating high density interconnects and electrodes for organic thin film transistors.

The high degree of control in size and shape of the transferred voxels achievable with laser decal transfer can in principle also be used to build 3-dimensional stacked structures. Such capability represents a true digital microfabrication process by which complex geometries can be generated voxel by voxel. An example of an array of

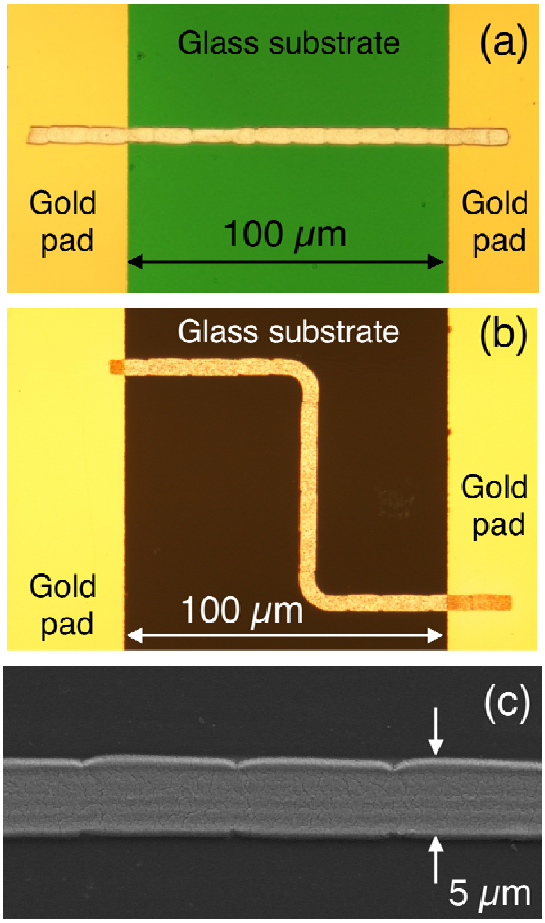

Fig. 5 Optical micrographs showing conductive silver lines made from laser decal transferred individual voxels across gold pads on glass substrates. (a) Line made from 15 μm long x 5 μm wide adjacent voxels; (b) line made by combining the rectangular voxels from (a) with corner shaped ones; (c) SEM image showing a section from the line in (a). Note the continuous pattern across voxels.

stacked voxels is shown in Figure 7 where a combination of alternating voxels and gaps was used to build a two-layered line. The bottom layer, labeled A on Figure 7(a), was cured before the top layer, labeled B, was laser transferred. A glancing angle SEM image of an individual voxel from the top layer across two voxels from the bottom layer is shown in more detail in Figure 7(b). Note that the top

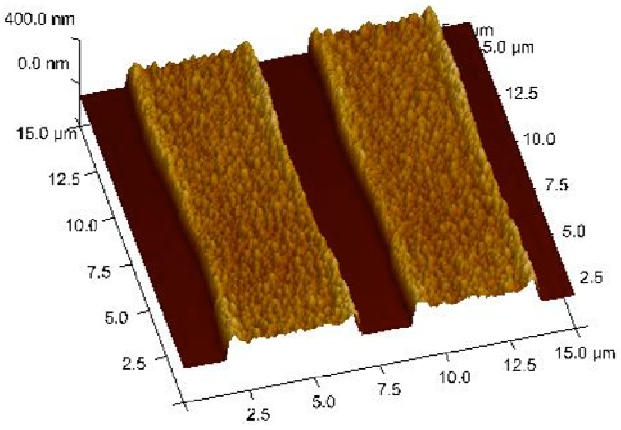

Fig. 6 AFM image showing two 5 μm wide silver lines with a 2 μm gap made by laser decal transfer.

layer voxel actually forms a bridge between the two spaced voxels underneath. This 3-dimensional stack is an example of the type of structures that can be assembled using laser decal transfer. As the AFM image in Figure 7(c) demonstrates, the second layer voxels bridge the gaps with minimum sagging or deformation. Such free-standing structures are impossible to generate with other direct-write techniques without the use of sacrificial layers. This capability is unique and opens the possibility for the digital microfabrication of other types of structures such as MEMS on low temperature substrates, which are impossible to generate by lithographic techniques.

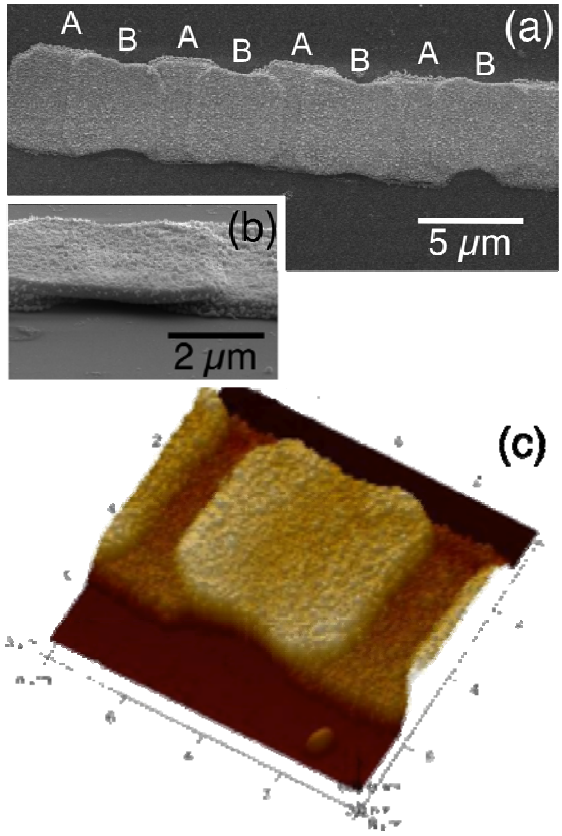

Fig. 7 (a) SEM showing a two-layered line made by laser decal transfers of 5 μm x 5 μm voxels on silicon (A: 1st layer, B: 2nd layer). (b) Glancing angle SEM image showing one of the top layer voxels across the two voxels below. (c) AFM image showing one of the top layer voxels bridging the gap formed by the first layer with minimum sagging and distortion.

5. Summary

This work describes a novel laser transfer process for the direct-write of thin-film-like patterned structures onto various types of substrates. The transfers correspond 1-to-1 to the size and shape of the laser spot imaged on the ribbon, analogous to a decal-like transfer. The resulting transfers are extremely uniform in thickness, show precise edge definition and are free of debris. The transferred material is made of high viscosity (≈ 100,000 cp) suspensions of silver nanoparticles, which once transferred form a continuous pattern that can be cured *in-situ* using a second laser. The resulting laser decal transfers exhibit resistivities 2 to 3 times higher than that of bulk metallic silver and show excellent adhesion to the substrates. Since laser decal transfers allow the printing of different shapes and sizes, any given pattern can be deposited in just a few steps, thus allowing further optimization of the writing time. In fact, laser decal transfers correspond to a form of digital microfabrication processes where the shape and size of each "bit" can be changed at will. The resulting decal transfers are well suited for the repair, modification and customization of microelectronic circuits, such as TFT-FPD's, photovoltaics, integrated circuits and other semiconductor devices, as well as the direct-write of 3-dimensional structures.

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