Use of an Elastomeric Donor for LIFT of Metal Foils

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The use of laser induced forward transfer (LIFT) techniques for printing materials for sensor and electronics applications is growing as additive manufacturing expands into the fabrication of functional structures. In many LIFT applications, a sacrificial or donor layer is required despite the fact that it must be replenished after being completely vaporized when illuminated with a laser pulse. A better solution would be to employ a reusable donor layer to which the transferable ink or metal foil is attached and then released by a laser pulse but without the donor undergoing damage, therefore allowing repeated use for subsequent transfers. In this work, we describe the use of an elastomeric donor layer based on poly(dimethylsiloxane) or PDMS for LIFT with UV ($\lambda = 355$ nm) laser pulses. Metal foils of varying size and thickness were attached to PDMS release layers initially spin-coated onto glass substrates and then printed onto silicon substrates by LIFT. A parametric study involving both the laser pulse intensity and the gap between the donor substrate and receiving substrate was conducted to determine placement accuracy as a function of laser fluence and gap distance. The effect of these two parameters, fluence and gap is discussed for the transfers of 25 and 50 µm thick copper foils, together with the applications of this technique for the printing of more complex foil shapes of metals and other materials.

1. Introduction

Laser-induced Forward Transfer or LIFT is capable of achieving high speed/throughput, high resolution patterns of a wide range of materials over many types of substrates for applications in flexible-hybrid electronics [1]. For example, by using LIFT of high viscosity Ag nanopastes, it is possible to print 3D microstructures with high aspect ratio micro pillars for vertical device connections and flip-chip applications [2]. Similarly, LIFT can be used to print freestanding, solid metal films/foils across contact pads to make planar circuit interconnects [3]. Both of these approaches require the use of a sacrificial donor layer which needs to be replenished after use. To date, most types of sacrificial layers that hold in place the part or device to be transferred are designed to completely vaporize when illuminated with a laser pulse [4].

This sacrificial donor layer, known as the dynamic release layer or DRL, is first deposited onto the donor substrate. The desired material to be transferred is then subsequently applied to the DRL surface. The DRL, is responsible for absorbing most of the laser intensity, and provides upon vaporization, the forward energy required to propel a fraction of the donor material towards the receiving substrate. A wide range of materials has been used as DRLs for LIFT of weakly- or non-absorbing liquid donor films, from gelatin [5] to metals like titanium [6]. During LIFT with a DRL, some residue from the donor layer is also transferred onto the receiving substrate, [7] and given that it constitutes a minute fraction of the printed material, in most instances it can be ignored. When that is not an option, triazene polymers (TP) comprised of aryl-triazene chromophores can be used since they fully decompose into volatile by-products when illuminated with several common laser wavelengths used for LIFT such as Nd:YAG 266 and 355 nm, and XeCl 308 nm [4].

1.1 LIFT of Intact Structures with Donor Layers

The use of sacrificial donor layers in LIFT has enabled the transfer of entire structures without damage from the laser pulse enabling the placement of microfabricated parts and even entire devices over a determined location on a surface. The first reports of LIFT of prefabricated MEMS, or microelectromechanical systems, structures using LIFT with a sacrificial polyimide DRL were made by Holmes in 2002 [8]. Since then, the use of DRL-enabled LIFT has been applied to the contact-less transfer of very small and thin semiconductor devices, which are easily damaged when mounted using existing pick-and-place tools [9]. The laser transfer of a wide variety of components such as surface mount devices and semiconductor integrated circuits ranging in sizes from 0.1 to over 6 mm^2 in area has been demonstrated using this lase-and-place technique [10]. Other groups have shown the viability of this technique in the placement of individual devices with precision and speeds competitive with well-established pick-and-place tools [11].

1.2 Reusable Donor Layers in LIFT

Despite these successes, the use of LIFT with a sacrificial DRL requires the preparation of a new donor layer after the laser transfers have been completed. A simpler and more practical approach would be to use a reusable donor layer to which the structures to be transferred are attached. This would allow repeated transfers with the same donor layer in a process analogous to mechanical stamping. John Rogers and his group at the University of Illinois at Urbana-Champaign have demonstrated laser-based printing of small devices using PDMS as a viscoelastic stamp. The laser-assisted delamination process that propels the device or structures forward off of the PDMS donor layer is driven by a difference in thermo-mechanical response between the device and the PDMS [12]. This process enables the non-contact printing of various materials and devices.

In this work, we present results of LIFT of copper metal foils using such a reusable donor layer. The reusable layer was made from UV transparent poly(dimethylsiloxane), also known as PDMS, spin-coated onto a quartz wafer. PDMS is a type of polymeric organosilicon which is optically clear and chemically inert with viscoelastic properties determined by its preparation conditions. We find that for a wide range of laser pulse energies at or slightly above the transfer threshold, the PDMS donor surface exhibit minimal signs of visible damage and the same region in the donor layer can be reused more than once. We studied the effect of laser fluence and donor-to-receiving substrate gap distance in the transfers of 25 and 50 µm thick copper foils ranging in size from ~2,500 to ~20,000 µm^2. We conclude by discussing the advantages and limitations of reusable donor layers for LIFT and their scalability and versatility across a wide range of printing processes based on LIFT.

2. Experimental Details

A schematic of the LIFT setup used for these tests is shown in Figure 1. Further details of the LIFT apparatus and setup used for this work can be found in previous publications [13,14]. The donor substrate ("ribbon") consisted of a 2" dia. x 1.8 mm thick quartz disk coated with PDMS. First, the quartz wafer was cleaned in Nochromix solution for 5 min. A solution of PDMS was prepared by mixing Dow Sylgard 184 Base with its curing agent in a 10:1 ratio and then mixing for 1 minute at 2000 rpm in a planetary centrifugal mixer. The PDMS solution was then spin-coated onto the substrates and baked at 125°C for 4 min which resulted in 30 to 60 µm thick PDMS layers (depending on spin-coater settings). This PDMS mixture was used to coat both the quartz donor and the Au coated silicon receiving substrates. The latter PDMS layer was used to facilitate the adhesion of the Cu foils to the receiving substrate and prevent their delamination during characterization.
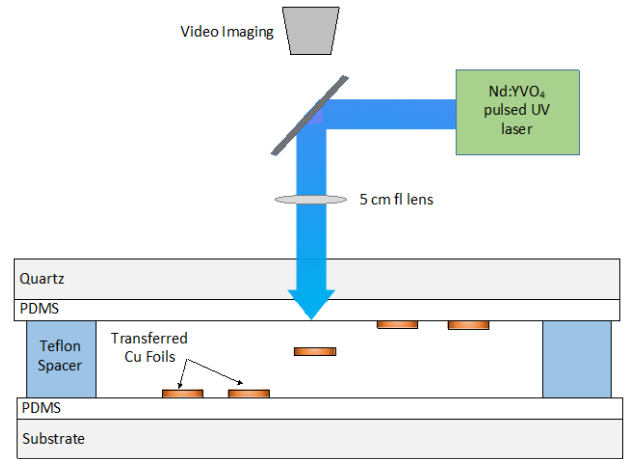

Fig. 1 Schematic (not to scale) of the laser transfer of Cu foils using a PDMS donor layer.

A frequency-tripled Nd:YVO$_4$ pulsed laser (JDSU Q301-HD, $\lambda = 355$ nm, 30 ns FWHM) was used to fabricate the Cu foils from 25 and 50 µm thick oxygen-free high conductivity copper foils (99.95% Cu from Shop-Aid Inc.) on the ribbon and subsequently to laser transfer them onto the receiving substrates. To ensure stable pulse energies, the laser was operated at a constant repetition rate (10 kHz for LIFT, 30 kHz for patterning Cu fliers) while an acousto-optic modulator (AOM) selected individual pulses and their timing. A galvanometric scan head with a 10 cm F-Theta objective (Scanlab, HurrySCAN 10) was used to fabricate the Cu fliers and a 5 cm fl lens was used for the laser transfers. The 5 cm fl lens was in-line with a CCD camera which provided a view of the ribbon and substrate as seen by the laser beam.

To fabricate the Cu foils on the surface of the PDMS ribbon, two different methods were used. The first method used laser micromachining to pattern the Cu foils directly on the PDMS surface. However, some damage to the Cu surface was observed, in addition to damage to the underlying PDMS. To ensure that the PDMS ribbons were reusable, a different fabrication method was investigated to minimize damage to both the Cu foil and the PDMS layer on the ribbon. This method involved laser patterning a protective coating, and then exposing the Cu surface for subsequent etching.

The Cu foil structures were made by first pressing a 25 µm thick Cu sheet onto the PDMS. In order to protect the Cu surface from the etchant, a protective polymer coating (Microposit, FSC-L) was spin-coated at 3000 rpm for 30 sec onto the ribbon and Cu sheet, then baked at 60° C for 1 min. Square annuli of ~ 200-300 µm (inner) and 600-700 µm (outer) were then laser-patterned (laser fluence ~4 J/cm^2) into the sheet. After laser patterning, the entire donor assembly was placed in a ferric chloride etching solution heated to 80°C for ~ 1 min, then rinsed in DI water and IPA. The remainder of the Cu sheet was peeled off the ribbon, leaving behind the desired patterned Cu foil structures. By using this etching technique, both the PDMS and Cu foil surfaces remain undamaged. Figure 2 shows optical micrographs of (a) sample array of Cu foils on the PDMS donor layer after etching and (b) detail of a single 140 µm x 140 µm foil showing the striations in the Cu surface resulting from the rolling process used in the manufacture of these foils.
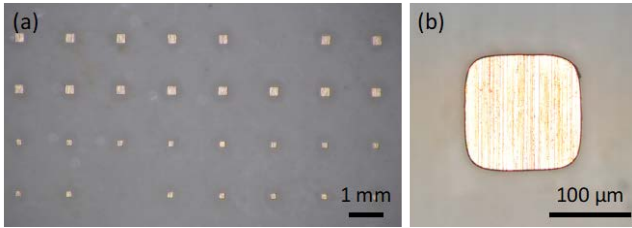
Fig. 2 Optical micrographs of Cu foils attached to the PDMS donor layer after etching and ready for laser-transfer. (a) Array showing two sizes of Cu foils and (b) close-up of a single patterned foil.

The receiving substrates used for this work consisted of a 4" Au-coated Si wafer onto which arrays of bullseye patterns were fabricated via conventional photolithographic methods (see Figure 3). The spacing between successive bullseye rings was 50 µm, with a total diameter of 1 mm. The center of the bullseye pattern was aligned with the laser spot, providing the target reference for the position and rotation of the Cu foil after transfer. The Si receiving substrates were cleaned and then coated with a 34 µm thick PDMS layer to facilitate adhesion of the laser-transferred Cu fliers for subsequent characterization of positional accuracy.
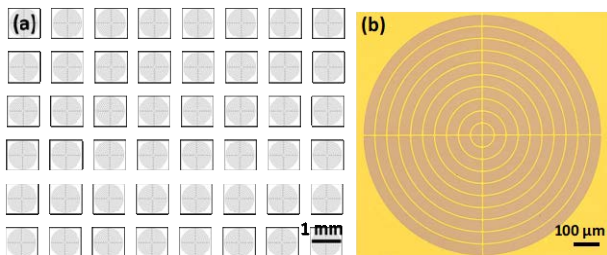

Fig. 3 (a) Bullseye pattern array design used for this work. (b) Optical micrograph of a single Au bullseye pattern on Si.

The receiving substrates were placed on a perforated steel plate over a vacuum chuck mounted on top of computer-controlled x-y translation stages (Aerotech, ALS50100 and ALS50045). Teflon spacer strips placed on the substrate maintained varying gaps (25 µm, 50 µm, 75 µm, 125 µm, 250 µm, or 1 mm) between the ribbon and receiving substrate. After an individual Cu flier on the ribbon was aligned over the bullseye, magnets were placed on the ribbon to ensure the gap distance was uniformly and reproducibly maintained.

For LIFT of the Cu foils, the laser beam divergence was increased using a reversed telescope such that the 5 cm fl lens provided a ~ 324 µm diameter spot on the ribbon. The diameter of the spot was chosen to yield a reasonable operating window for LIFT with the available laser energy. The spot diameter was sufficiently larger than the Cu foil for a more uniform illumination of each individual foil. The beam diameter was fixed for all transfers while the beam energy was varied using the AOM. Maximum energies measured after the 5 cm fl lens were ~ 0.73 mJ (laser at 10 kHz) yielding a fluence near 900 mJ/cm^2. Each Cu foil laser transfer was performed using a single pulse. Series of transfers were conducted in which the laser energy was progressively increased from the energy transfer threshold and the fluence calculated for each transfer. For each series, the gap distance or the foil thickness were varied and the location on the Si receiving

substrate of individual transferred Cu fliers was characterized using a digital microscope (Keyence, VHX-1000).

3. Discussion

The use of a viscoelastic stamp made from a PDMS layer for transferring small structures and devices has been studied extensively by Rogers and his group [15]. Rogers' group then applied these ideas to demonstrate the laser driven stamping of 100 µm x 100 µm x 3 µm thick Si squares, using a process which they named "laser micro transfer printing" [16] that was effectively LIFT based on a PDMS donor layer. For this work, the authors used a cw IR diode laser (808 nm) resulting in unnecessary heating of the PDMS and Si squares. At NRL, we have conducted LIFT tests in which pulsed UV lasers were used to transfer bare die PIN-diodes over a wide range of surfaces using a PDMS donor substrate [17].

For the current work, we investigated how variations in position and alignment of the Cu fliers were influenced by the gap distance between the donor and receiving substrate and also by the laser energy density or fluence of the transferring laser pulses. A sample representative of typical transfers obtained with 140 µm x 140 µm x 25 µm thick copper foils is displayed in Figures 4(a) to (c). In some cases, we also observed flipping of the Cu flier on its side due to uneven release from the PDMS donor layer as illustrated in Figure 4(d).

To simplify the tabulation of positional accuracy of each transfer, the radial displacement was calculated which encompassed displacements both in X and Y from the center of the bullseye at which each Cu foil was aimed during the transfers. Variations in position were relatively insensitive to laser fluence, as demonstrated by the optical micrographs in Figure 5 showing a series of transfers of 140 µm x 140 µm x 25 µm thick copper foils with a 125 µm donor to receiving substrate gap. Overall the total radial displacement of the Cu fliers in these cases was relatively small, i.e. under 20 µm total radial displacement for gaps up to 125 µm.
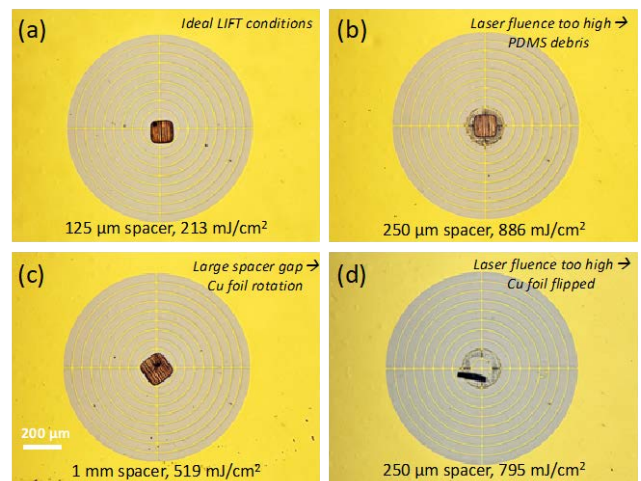

Fig. 4 Sample transfers of 140µm x 140 µm x 25 µm thick Cu fliers under various processing conditions.
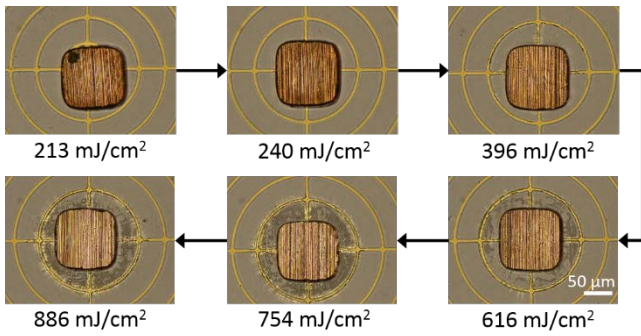
Fig. 5 Transfers of 140 µm x 140 µm x 25 µm thick Cu fliers with a 125 µm gap as a function of laser fluence.

Fig. 6 Transfers of 140 µm x 140 µm x 25 µm thick Cu fliers at a laser fluence of ~350 – 450 mJ/cm^2 as a function of donor to receiving substrate gap.

The variation in positional accuracy was evaluated for a range of fluences from ~250 to ~900 mJ/cm^2 under different donor to receiving substrate gaps. A series of transfers at laser fluences between 350 to 450 mJ/cm^2 for gap distances ranging from 25 µm up to 1 mm are displayed in Figure 6. As expected, the positional accuracy decreases for the larger gaps, i.e. 250 µm and 1 mm. On the other hand, for gaps of 125 µm and below the Cu flier lands with very high position accuracy onto the Si receiving substrate. Similar results were obtained with the tests performed with Cu foils measuring 70 µm x 70 µm x 50 µm thick.

The radial displacement as a function of laser fluence for the series of 140 µm x 140 µm x 25 µm thick Cu foils at four distinct donor-to-receiving substrate gaps (25, 75, 125 and 1,000 µm) is plotted in Figure 7. It should be noted that the graph shows the results for single transfers which explains the lack of error bars for each data point. Despite this lack of statistical data, trends in the radial displacement obtained for each series of transfers at a given gap as a function of the laser fluence become clear. For gap distances up to 125 µm, the radial displacement averages under 10 µm and never exceed 20 µm, which is sufficiently accurate for many applications in microelectronics and micro-assembly. For the largest gap distance plotted in Figure 7, i.e. 1 mm, the radial displacement remained more or less the same (around 10 µm) up to fluences of 500 mJ/cm^2 and then rapidly increased for laser fluences above 600 mJ/cm^2. These results, notwithstanding their preliminary nature, show that in general LIFT with a PDMS elastomeric donor layer is capable of transfers with positional accuracy similar to that obtained with sacrificial DRL's [16].

The rotation of the Cu foils after transfer was also evaluated. The use of rectangular Cu shapes in combination with the bullseye pattern etched in the Au film on the Si receiving substrate enabled the straightforward measurement of the total rotation of each Cu foil after transfer. For transfers across gaps up to 250 µm the rotation of the Cu foils was of less than 10 deg. Only with the 1 mm gap we observed larger rotations up to 60 deg. The radial displacement and rotation observed in these experiments can partially be attributed to the ambient air atmosphere at which the printing was conducted. As the gap increases, air resistance becomes more prominent, causing the radial displacement and rotation to increase.
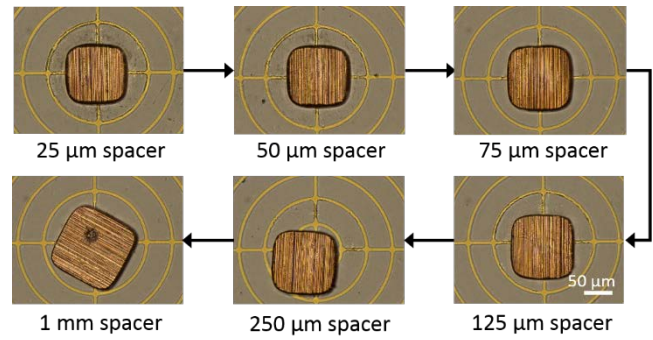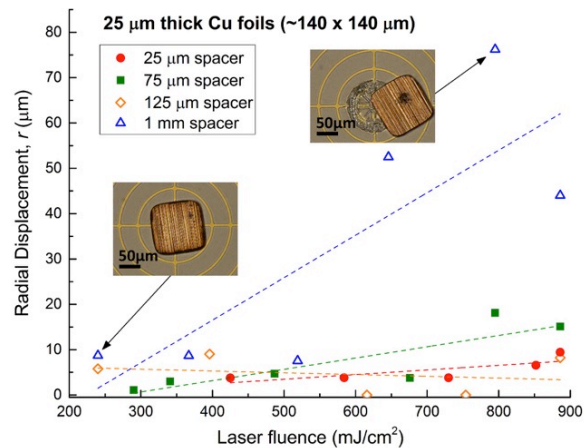

Fig. 7 Radial displacement as a function of laser fluence for transfers of 140µm x 140 µm x 25 µm thick Cu fliers at four different gaps (25, 75, 125 and 1,000 µm). Lines are a guide to the eye. Inserts display micrographs corresponding to two of the points plotted for the 1 mm gap spacer.

Finally, tests to evaluate the ability to reuse the same region on the PDMS layer on the donor substrate for more than a single transfer were conducted. For these tests, individual Cu fliers were manually pressed onto the PDMS layer over the previous laser spot. The results displayed in Figure 8 demonstrate that reuse is indeed possible. The PDMS donor was laser marked to be able to precisely locate the same region for reuse. For this purpose, a cross bar on the PDMS was laser etched prior to these tests and can be seen on the top left corner in the images in Figures 8(a-b) and 8(d-e). The PDMS region thus selected was used first to transfer a rectangular Cu foil, originally attached where the slight discoloration of the PDMS layer is visible in the above images. Two subsequent transfers, this time comprising of two separate ~150 µm dia. Cu disc foils were conducted. Figure 8(a) and Figure 8(b) show the donor region before and after the second time use of this PDMS region, with the resulting transfer shown in Figure 8(c). The entire process was then repeated a third time, with the donor region before and after, plus the resulting transfer shown in Figure 8 (d) through (f) respectively.
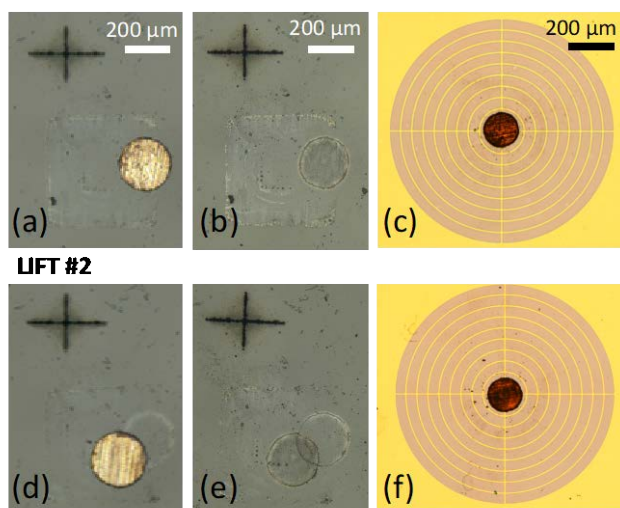
Fig. 8 Demonstration of reusability of the same region of the PDMS donor layer for more than one laser transfer. (a-b) PDMS donor before and after the 2nd use. (c) Resulting 2nd transfer. (d-e) PDMS donor before and after 3rd transfer. (f) Resulting 3rd transfer. The cross bar on the top left corner in (a-b) and (d-e) was laser marked prior to these tests to allow location of the same region in the PDMS donor.

The implementation of a PDMS donor layer with LIFT is compatible with the transfer of more complex shapes of metal foils and other materials including ceramics, composites and multilayer stacks, in addition to functional components or devices such as semiconductor bare die. Furthermore, the ability to reuse the donor layer simplifies the steps required for preparation of the donor substrate and facilitates its implementation in roll-to-roll processes. Roll-to-roll donor preparation can be combined with in-line laser transfer for applications requiring laser printing of large number of devices at high throughput rates. These capabilities help make LIFT more compatible with industrial applications.

4. Summary

The use of a PDMS donor layer for LIFT has been shown for the transfer of Cu foils of various sizes ($\sim$2.5x10^3 to $\sim$2x10^4 μm^2) and two thicknesses (25 and 50 μm). The foils can be laser transferred with positional errors below 10 μm (total radial displacement) when transferred across donor to receiving substrate gaps up to 125 μm. The amount of rotation in the foils during transfer was also evaluated and found to be less than 10 degrees for gaps up to 250 μm.

This work also demonstrated the ability of the PDMS donor ribbon to be used multiple times for LIFT of Cu foils without significant degradation. This feature shows that PDMS can act as a reusable DRL on a donor substrate greatly simplifying the application of DRL for large scale LIFT applications involving the transfer of solid metal foils and other types of solid structures or functional devices.

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