Dynamic spatial pulse shaping via a digital micromirror device for patterned laser-induced forward transfer of solid polymer films

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Abstract: We present laser-induced forward transfer of solid-phase polymer films, shaped using a Digital Micromirror Device (DMD) as a variable illumination mask. Femtosecond laser pulses with a fluence of 200-380 mJ/cm^2 at a wavelength of 800 nm from a Ti:sapphire amplifier were used to reproducibly transfer thin films of poly(methyl methacrylate) as small as ~30 µm by ~30 µm with thickness ~1.3 µm. This first demonstration of DMD-based solid-phase LIFT shows minimum feature sizes of ~10µm.

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1. Introduction

In recent years, there has been much commercial interest and technological advances in direct-writing for material deposition, processes that enable the controlled addition of material onto a substrate. Whilst there are a wide range of deposition techniques, a subset of methods for micron to mm printed devices uses a focused laser beam to interact with a surface, causing a localised reaction that results in the addition of material.

In these methods, a user controls the position of a laser spot, enabling a great deal of flexibility with regards to the fabricated structure, as the path of the laser focus with respect to the surface of the sample can be programmed to produce almost any desired pattern. However, this processing method can be relatively slow due to the need to scan a single laser beam over a sample. An alternative method, the use of a fixed illumination mask, can be considerably faster as no laser-scanning may be required, and hence can be ideal for the manufacturing of a large number of identical devices. However, this approach can be a costly technique for rapid prototyping, where the printed device may need several design updates. The use of a *variable* illumination mask however retains the advantage of both approaches, namely the flexibility that allows rapid prototyping but also the speed advantage of using an illumination mask.

In this paper we demonstrate the use of a spatial light modulator, namely a Digital Micromirror Device (DMD), as a variable illumination mask for femtosecond laser pulses, for the shaped deposition of polymer films with features as small as 10 μm, via the technique of laser-induced forward transfer (LIFT) [1].

1.1 Laser-induced forward transfer

LIFT is an additive laser direct-write technique in which laser pulses are used to transfer solid, liquid or paste *donor* materials from a *carrier* substrate to a *receiver* substrate, as shown schematically in Fig. 1. A single laser pulse is focused or imaged at the interface

between the donor and the carrier, such that a small volume of the donor, or an optional dynamic release layer (DRL) at this interface, absorbs a large fraction of the laser pulse energy and undergoes an explosive phase transformation, propelling the remaining donor material, referred to as the *flyer*, towards the receiver substrate. As donor and receiver substrates are prepared independently, LIFT enables the deposition of materials on substrates that may be difficult via other fabrication methods. Multilayer donor materials have allowed the production of complex structures via LIFT, including metamaterials [2,3] and light-emitting diodes [4,5].
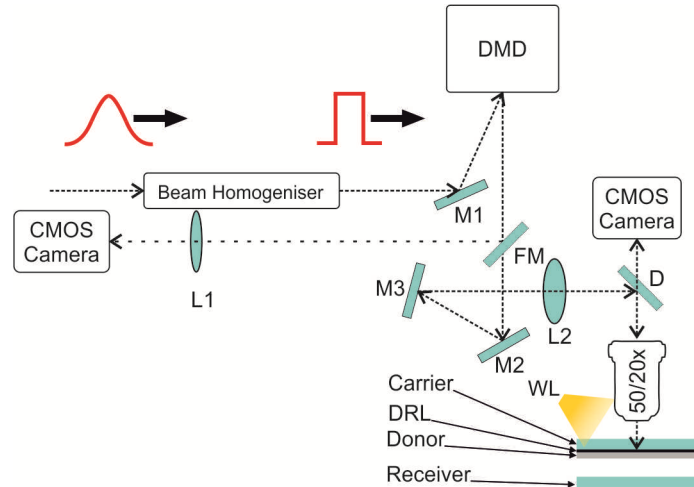

Fig. 1. Schematic of the experimental setup.

In general, the use of a DRL [6] leads to a more confined absorption of the incident pulse energy and hence can result in a lower fluence required for flyer propulsion, as well as less damage to the donor and resultant flyer. DRLs of metals [6,7], polymers [8,9], or more complex materials [5] have been successfully demonstrated. Due to the high speed of the flyer (shadowgraph measurements have shown speeds varying from 34 ms^{-1} [10] to ~2 kms^{-1} [11], where flyer velocities with and without DRLs have been recorded at similar values [12]) and subsequent rapid deceleration when it collides with the receiver substrate, the flyer can experience significant forces that can result in damage. However, it has been shown [13] that use of an additional 'soft' compliant receiver (such as polydimethylsiloxane (PDMS), deposited onto the receiver) can reduce the forces and improve the integrity of the deposited flyer.

Most frequently, donor thicknesses of a few hundred nm to a few μm are used, though this depends on material properties of the chosen donor. Lateral dimensions of the flyer range from tens of nm [14] to a few mm [15]. Pulsed lasers, usually ranging from nanosecond to femtosecond pulse lengths, can be used for LIFT, with typical energy densities of a few tens to hundreds of mJ/cm^2.

Until recently, the majority of LIFT research has used focused or imaged laser pulses that had a Gaussian or 'top-hat' spatial intensity profile, with circular or rectangular beam-shaping (with extension to "smart beam shaping" [16], or the use of custom masks [5]). Whilst individual deposits can be overlapped [17], this approach cannot be easily extended to produce less simple 2D structures. In order to build up more complex structures, the use of a variable illumination mask (or spatial light modulator, such as a DMD), has recently been explored [18].

DMDs can offer a significant speed advantage (30 kHz repetition rate devices are currently possible [19]) over liquid-crystal spatial light modulators, and hence can enable dramatically faster direct-writing. This paper describes results from use of a DMD, acting as an intensity spatial light modulator, for the shaped LIFT of polymer films with minimum

lateral dimensions ~30 μm, and builds upon recent results by Piqué et al. [18,20], which show DMD-assisted LIFT (referred to as Laser Decal Transfer in their work) of a paste-like donor material that produces solid structures after post-processing. In our work, we demonstrate for the first time, the use of a variable illumination mask for the LIFT of intact solid phase materials, which do not require any post-processing step.

4. Experimental setup

The DMD used in this work was a Texas Instruments DLP3000 [21], which consisted of an array of 608 by 684 individually controllable, ~7.6 μm wide mirrors, arranged in a diagonal-square lattice. The centre-to-centre distance between adjacent mirrors in the horizontal and vertical planes was 10.8 μm. Here, the mirrors are described as being either 'on' or 'off', which refers to their angular orientation of either $+ 12°$ or $-12°$ respectively, relative to the plane of the DMD surface.

Due to the periodic structure of the DMD, when working with a spatially coherent light source, as was done in this work, multiple diffraction peaks were produced, where the output angles of these peaks can be determined from the grating equation for non-normal incidence, $\sin(\theta_i) - \sin(\theta_m) = m\lambda/d$, where θ_i and θ_m are the incident and diffracted angles, d is the mirror spacing, λ is the laser wavelength and m is the order of diffraction. In this case, the angular orientation of the mirrors resulted in the mirror array acting as a blazed grating, which was taken advantage of in this work in order to increase the percentage of diffracted light present in a single diffraction order.

A Ti:sapphire amplifier was used to produce ultrashort (150 fs, 800 nm wavelength) laser pulses with a maximum energy of 1 mJ at a 1 kHz repetition rate. Although LIFT is performed using pulsed lasers, the choice of this femtosecond source was for its 800 nm wavelength, rather than its pulse-width; temporal broadening caused by any optical elements in the setup was not important. This wavelength was well reflected by the DMD; a glass layer suspended in front of the DMD mirrors, which contains the microelectromechanical system (MEMS) devices in an inert atmosphere, has ~90% transmission at a 30° angle of incidence [21], while the mirrors themselves have a reflectance of ~88% at 800 nm [22]. The pulses were spatially homogenized (using an AdlOptica 'π-shaper' model 6_6) to produce a 'top-hat' spatial intensity distribution. The homogenized pulses were incident onto the DMD, where they underwent diffraction into multiple orders. By choosing an incident angle of ~24° (taking advantage of the blaze angle), the m = 5 diffraction order (at an optimal output angle of 0°) contained ~30% of the incident pulse energy. A collimating lens captured the chosen order, which was then imaged using a 50x or 20x infinity-corrected objective onto the DRL, as shown in Fig. 1, to replicate the binary pattern loaded on the DMD as a spatial intensity profile at the DRL. A diagnostic beam line, consisting of a flip mirror (FM), lens (L1) and a CMOS camera, enabled the surface of the DMD to be imaged for alignment purposes. The beam path used for LIFT consisted of two mirrors (M2 and M3), a collimation lens (L2), a dichroic mirror (D) and the imaging objective (50x or 20x). A white light source (WL) allowed viewing of the sample in real-time as it was imaged back through the objective and dichroic onto a second CMOS camera. The sample was loaded on a 3-axis stage, which had a positional accuracy of ~1 μm and 50 mm total travel in each direction. By sending monochrome bitmap images via computer to the DMD, all mirrors were switched to their intended on/off positions.

The donor used throughout the experiments was poly(methyl methacrylate) (PMMA, purchased from MicroChem), and was prepared on the carrier substrate via spin coating, resulting in a 1.3 μm thick layer. As PMMA is transparent at 800nm, a DRL was required.

The carrier substrate (a glass slide, Electroverre extra-white) was coated with either a 30 nm gold or 50 nm carbon film via sputtering or evaporation respectively, both acting as DRLs. Although triazene polymers have been used previously as DRLs [9], they were not used here due to concerns over polymer orthogonality. The receiver had a ~10 μm thick spin-coated PDMS layer, acting as a compliant receiver. While the compliant receiver here was

used primarily to preserve the integrity of the flyer, PDMS-coated substrates are widely used in microfluidics, contact-lithography and MEMS devices [23–27], and polymer-coated receivers have even been used in the manufacture of electronic devices [4]. The donor and compliant receiver were placed in contact during the LIFT process in order to reduce the possible rotation of the flyer during transfer. Previous work [10] has shown instability even in approximately rotationally symmetric flyers – the highly asymmetric shaped flyers demonstrated here would likely be released in a non-uniform fashion, leading to an increased chance of damage. Placing the donor and receiver in contact presented no restrictions in the work here – both being chemically stable polymers in the solid phase. Contact may be unfeasible for future work where the donor and receiver would chemically bond to each other, or would be altered by the other's presence in some way.

5. Experimental results

Projected spatial intensity patterns (a selection of alphabetic letters) ranging in complexity and size were trialled for a range of laser fluences, in order to determine the optimal conditions for the LIFT of the chosen samples. Scanning electron microscope (SEM) images of individual deposited structures are shown in Fig. 2, with insets displaying the DMD patterns as loaded (and hence the projected spatial intensity profile) when the laser pulse was incident on the DMD surface. Figure 2 shows a comparison of the deposited structures using (a-d) a 50x objective and fluence of 300 mJ/cm^2, and (e-h) a 20x objective and fluence of 270 mJ/cm^2. For the 50x objective, the size scales in the letter 'B' and 'S' appear to be at the limit of what is possible for this particular material, as the features were not reproduced correctly. For the 20x objective, the size scales were larger and both the letter 'B' and 'S' were reproduced accurately. While many letters were reproduced successfully using the 20x objective, with maximum lateral dimensions of ~70 μm, the quality of some, such as 'S', remained low until they were produced at lateral dimensions of ~100 μm (note the smaller scale bar in Fig. 2 (h)). At this size, the entire alphabet was deposited accurately. In general, for successful LIFT, the region on the donor that is illuminated by the laser pulse must break free from the surrounding donor material, via the explosive phase transformation caused by absorption of the incident laser pulse. There is therefore a limit to the minimum width of a feature that can be successfully removed from the donor via the LIFT process, and is a value determined predominantly by the material properties and dimensions of the donor, and the presence of a DRL. This was an evident concern in the LIFT of solid donor material when compared to the LIFT of pastes by Piqué et al. [20], which could much more easily break free from the surrounding donor film. This limit is observed in Fig. 2, where the features in (a-d) are too small to be removed in the same shape as the projected intensity pattern, whilst the features in (e-h) are almost perfectly replicated. Evidently in (a-d), the deposits' minimum feature sizes are larger than they would have been, had the deposit been a perfectly minified version of the projected intensity patterns (also shown as the insets).

Measurements showed that when using a 50x objective to produce deposits of size ~30 μm, although the overall size of ~30 μm corresponded to the minified image projected from the DMD, minimum feature widths in each deposit were ~10 μm. Compared to the imaged DMD masks, there was an excess of ~3 μm in feature width either side of each edge in the deposits. Similarly, for the deposits of size ~70 μm in (e-g) and ~100 μm in both (h) as well as Fig. 4, the excess of ~3 μm persisted, meaning a lower relative discrepancy between deposit feature width and intensity pattern width with increasing overall deposit size. We attribute these discrepancies between the projected intensity pattern and the shape of the deposits in (e-h), in particular the fact that the deposits have curved (rather than square) corners in (b-d, f-h), to the difficulty in removing a section of the donor that had such sharp features, and hence the typically chamfered results on transferred deposits.
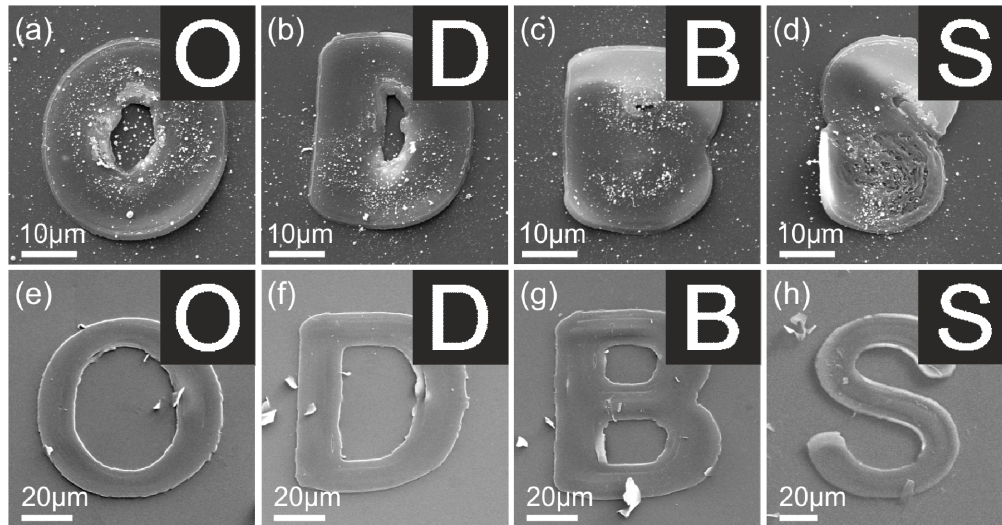
Fig. 2. SEM images of 1.3 µm thick polymer structures deposited via LIFT onto a PDMS-coated glass slide, with the inset images showing the projected spatial intensity patterns, for (a-d) a fluence of 300 mJ/cm² using a 50x objective and a 30 nm Au DRL and (e-h) a fluence of 270 mJ/cm² and a 20x objective with a 50 nm carbon DRL. Note the different scale bars on the figures.

Although the results shown in Fig. 2(a)-2(d) corresponded to the use of a 30 nm gold DRL and (e-h) to a 50 nm carbon DRL, in general the particular characteristics of these DRLs at these thicknesses were not found to affect the quality of the deposit, but rather only the amount of fine debris present after the LIFT process, which generally resided on top of the deposit (visible in (a-d) but not in (e-h)). After initial experiments using a gold DRL that produced deposits as in (a-d), carbon was used exclusively in the interest of reducing fine debris while optimising magnification scale towards that used for the results in (e-f). In order to test the adhesive qualities of the deposits, a sample was left in a sonication bath filled with distilled water for 30 minutes, and out of a total of approximately 300 deposits, all of the deposited structures remained in place.

Figure 3 demonstrates the effect of a variation in pulse energy on the deposited polymer structures, over a 25% range in laser fluences, with values reducing from 300 mJ/cm² (left) to 235 mJ/cm² (right) in steps of ~17 mJ/cm², for a series of letters. The shapes appear to be deposited similarly across this range. Successful deposition of shaped polymer structures was also achieved outside this range. In general, fluences from 200 mJ/cm² to 380 mJ/cm² were found to yield high-fidelity deposits for all letters (and arrays of squares, as in Fig. 4), hence demonstrating robustness to energy fluctuations and reproducibility across an extremely wide variability of shapes. As each deposit only requires a single laser pulse, large areas of sample coverage can be achieved via rapidly LIFT-printing multiple deposits by taking advantage of the high refresh rate of the DMD (hence allowing each laser pulse to be used to deposit a different shape) when using a high-repetition rate laser. For instance, given that 100 µm by 100 µm deposits can be made with a single pulse, at the 1 kHz repetition rate of the laser used, an area of 0.1 cm² could be covered per second, with potentially 1000 unique deposits in that area.

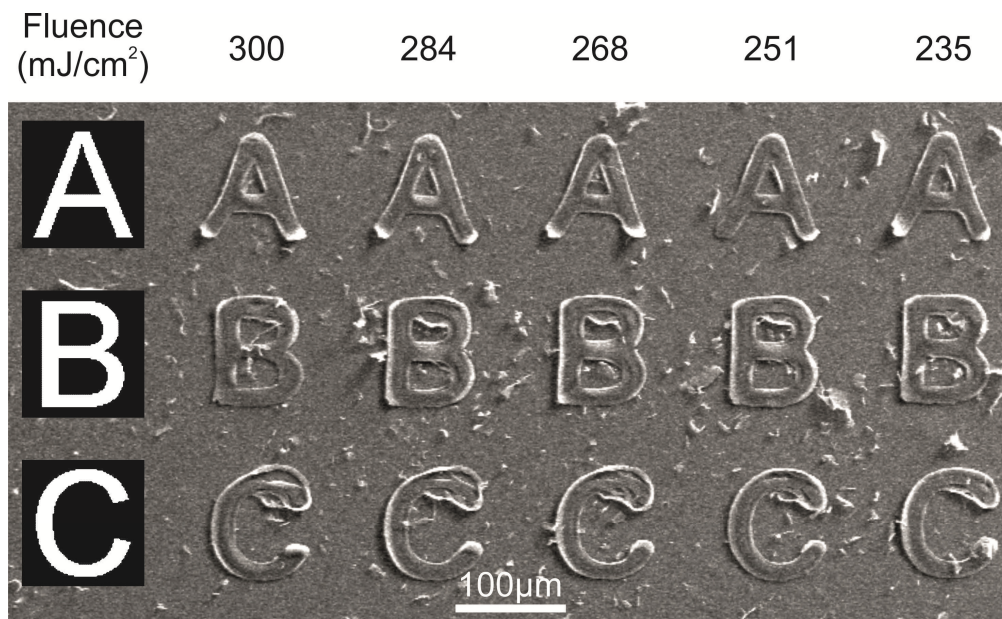Fluence (mJ/cm^2)   300   284   268   251   235

Fig. 3. SEM image of an array of different shaped polymer deposits using a 50 nm thick carbon DRL and 20x objective, for a range of laser fluences reducing from 300 mJ/cm^2 to 235 mJ/cm^2. The insets show the projected intensity profiles.

In order to show another facet of the applicability of a DMD as a variable illumination mask for LIFT, multiple unconnected structures were deposited simultaneously using a single pulse. This approach is demonstrated in Fig. 4, where SEM images show a two by two array of square deposits, for two different projected intensity patterns, namely (a) confined and (b) sparse, for the same conditions as in Fig. 3, save for the use of different fluences. In solid-phase LIFT of PMMA, additional donor material around the edge of the intended flyer can be ejected as the flyer shears free from the donor. In Fig. 4(a), the minimal separation between squares is reflected in the lower quality of the deposited structure, where additional and unintended donor material was ejected from the donor, resulting in the square deposits being larger than the corresponding intensity profile on the donor, which is shown in Fig. 4 as dashed yellow lines.
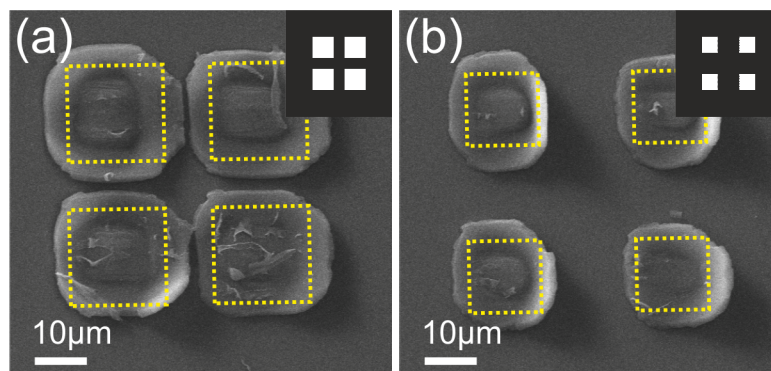

Fig. 4. SEM images of arrays of two by two squares that were deposited simultaneously via the LIFT technique, with squares of different sizes and separations displayed on the DMD. Donor, receiver, objective and DRL are as in Fig. 3. Produced at fluences (a) 250 mJ/cm^2 and (b) 270 mJ/cm^2. The inset shows the projected intensity pattern, where the correctly scaled outside edges of the pattern are also displayed as yellow dashed lines.

In agreement with results found in [28,29], the smaller squares in Fig. 4(b) required a higher fluence than those in (a), to achieve transfer. For any LIFT transfer, energy will be required to shear the perimeter of the flyer, to release at the interface between carrier and donor, and to propel the flyer forward. Of these energy costs, it is important to compare that of shearing at the perimeter to releasing at the interface. As the region of contact at the interface is exactly the single-side surface area of the flyer itself, one would expect this release energy to scale linearly with area, and so would have no effect on the required fluence (energy per unit area). The energy required to shear at the perimeter will increase linearly with perimeter length, which will depend on the particular geometry of the flyer, but typically will increase with flyer area. However, the ratio of perimeter length to area increases as flyer area decreases, meaning that for smaller flyers a greater proportion of incident energy is required to shear at the perimeter. In order to account for the constant fluence needed to release at the interface, and an increased share of total incident energy to shear at the perimeter, smaller flyers require higher fluences to transfer.

6. Conclusion

In conclusion, we have presented the use of a DMD, acting as a spatial light modulator, for laser-induced forward transfer of shaped 2D deposits from a solid thin polymer (PMMA) film, with lateral dimensions as small as ~30 μm by 30 μm, and minimum features of 10 μm. This first demonstration of DMD-assisted LIFT of solid, intact donors without the need for additional post-processing for the placement of structures with controllable shape and position with high repeatability and adhesion provides added versatility to the LIFT process.

Previous related work has included the LIFT of pastes via DMD [20], which are post-processed to form solids on the receiving substrate, and the shaped LIFT of solids via exposure masks [5,16], where deposit sizes are on the order of ~500 μm lateral dimensions.

While the fidelity of shaped deposits achieved with pastes has been impressive, not all materials are amenable to preparation in paste-form, and receiver substrates may not withstand necessary post-processing. Our work with DMD-assisted solid-phase LIFT retains a key advantage of the LIFT process, namely that preparation conditions of the donor and receiver substrates are truly separate. Further, the mechanical and chemical stability of solids increases the lifespan and ease of manipulation of donor substrates in comparison to pastes and liquids. We have extended the LIFT process to enable, to our knowledge, the only viable method of printing solid deposits that are shaped differently shot-to-shot, without any post-processing requirements, on a size scale of tens of microns. This initial proof-of-principle study will lead to further work in the field towards the shaped LIFT of a wider range of solid donor materials onto other receiver substrates.

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