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Multi-beam interferometric patterning in optically transparent materials

Nicholas Tham^a, Y. J. Kim^a, V. M. Murukeshan^a

^aNanyang Technological University, 50 Nanyang Avenue, Singapore 639798

ABSTRACT

A method to directly pattern nanoscale periodic features within optically transparent solid materials by means of a configurable multi-beam femtosecond laser interference is proposed. While femtosecond micromachining of optically transparent solid materials has been explored in great detail in the recent past, research in direct interference patterning on such materials has not been well established. Therefore, different design considerations such as complete void formation and flexibility of the patterned substrate are investigated as part of this study. The relationship between intensity distributions and actual void formation in optically transparent materials is also investigated to establish critical parameters for pattern formation. It is envisaged that this proposed method and data obtained may enable to explore the untapped domains of developing substrates for solid-state 3D microbattery applications.

Keywords: Multi-beam, femtosecond, interference lithography, optically transparent materials, 3D microbattery

1. INTRODUCTION

Laser interference lithography is a widely recognised tool for researchers to develop periodic patterns quickly and efficiently without the use of masks. Laser interference in a wide variety of configurations and methodologies is used to develop complex patterns on a variety of photoresists and even directly onto substrates, from simple 1D and 2D periodic gratings [1-5] to 3D photonic lattices and lenses [6, 7] and even complex 3D structures like metal crystal structures [8] or all 14 Bravais lattices [9] were shown to be theoretically possible. The flexibility of the multi-beam laser interference system has lent itself to application in a variety of fields, most notably in the highly competitive field of micro-electronics and increasingly nano-electronics [10], where it has been put to use to develop integrated circuits [11] and transistors [12-14]. One particularly intriguing application of interference lithography is in the production of 3D solid-state microbatteries, a higher energy density alternative to current thin film solid-state microbatteries [15] for flexible electronics applications.

A challenge of fabricating 3D solid-state microbatteries, however is developing a method to fully integrate these power sources into specific systems themselves. One method proposed by other researchers would be to fabricate the battery before encasing the battery in a suitable substrate [16]. A potentially simpler and more direct method to directly form the 3D microbattery in the solid substrate itself, by 1D or 2D patterning of high aspect ratio voids [17] or pillars [18] into the substrate before adding battery components, layer by layer to form a surface-conformal 3D solid state microbattery. However, to fully harness the strength of laser interference in forming complex 3D features, the authors of this report propose a novel approach to direct bulk patterning of transparent flexible substrates. One potentially suitable optically transparent flexible electronics substrate is PDMS, or Polydimethylsiloxane, a silicone elastomer that is the subject of a wide range of research investigating applications in flexible electronics [19]. Direct patterning of transparent flexible substrates is only possible with ultrashort pulse lasers such as femtosecond lasers, through non-linear absorption processes [20] at high peak fluences. However, while most of the research is focused on micromachining of optically transparent substrates [20] with a single focused beam, there were not many reported works on direct femtosecond interference patterning in the bulk of optically transparent materials.

In this context, this paper discusses the details of dual beam femtosecond laser interference assisted patterning inside optically transparent bulk materials. Different design considerations such as complete void formation and flexibility of the patterned substrate were investigated as part of this study. The intensity distributions and void formation in optically transparent materials is also investigated.

2. THEORY

A classic horizontal two-beam set-up as schematically in Figure 1 was used in this experiment. For a classic two-beam set-up, the theoretical fringe pitch Λ formed by two beams would be found by the formula

$$\Lambda = \frac{\lambda}{2 \sin \theta} \quad (1)$$

where λ is the wavelength of the incident beam and θ is the angle made by the interfering beams with the optical axis, assuming that both beams are aligned at the same angle. Similarly, in a material with refractive index n , the pitch width in the material [1] would be

$$\Lambda_{\text{material}} = \frac{\lambda}{2 \cdot n \cdot \sin \theta} \quad (2)$$

where n is the refractive index of the material. The relative difference in intensities between the interfering beams is also an important factor in producing interference patterns with sufficient contrast. Contrast V of the two beams can be directly calculated from

$$V = \frac{2\sqrt{I_1 I_2}}{I_1 + I_2} \quad (3)$$

where I_1 and I_2 are the intensities of the two beams respectively. For maximum contrast and clearer patterns, i.e. $V = 1$, the intensities of both beams must therefore be equal. Finally, to characterise the optical transmittance and absorbance α of PDMS at 343 nm, transmittance can be measured and absorbance α calculated from

$$\alpha = \frac{2.3026}{t} * \log_{10} \frac{100\%}{T\%} \quad (4)$$

3. MATERIALS AND METHODOLOGY

PDMS used as test samples were prepared following the procedure detailed next. Sylgard® 184, purchased from Dow Corning Co. was prepared in a 10:1 base: binding agent ratio. The mixture was first thoroughly mixed for 1 to 2 minutes in petri dishes before degassing for 45 minutes. The degassed mixture was then cured in an oven at 80 °C for 2 hours to produce cured PDMS of approximately 1 mm thickness. Sections of PDMS were then cut and mounted into on a sample holder of 12 mm by 25 mm to be exposed.

The optical configuration used for the experiment is shown in Figure 1. A linearly polarised 220 s femtosecond beam of wavelength 343 nm, pulse frequency 500 kHz was split into two using a plate beam-splitter. A path length compensator (UV silica window) was added to the reflected beam path to account for the path length difference introduced by refraction in the plate beam-splitter. Both transmitted and reflected beams were reflected by two mirrors onto a sample (Bulk PDMS is used as the test sample here) plane at an angle of 36.25°. The refractive index of PDMS is approximately 1.455 at 343 nm [21] and therefore the fringe pitch is calculated as 199.30 nm using Equation 2.

PDMS samples were loaded onto the sample stage so that the interfering beams with fluence ranging from 9.55 μJcm^{-2} to 31.80 μJcm^{-2} were incident on it at the desired location. Care has been taken to ensure that fluence range is within the damage threshold such that the patterns are written within the bulk. Prior to patterning, transmitted average power through

the PDMS was measured, which was then divided by the incident power to obtain values for percentage of transmission loss.

Figure 1. Schematic top-down view of two beam set-up. A 343 nm 220 fs laser at 500 kHz pulse frequency was used. A UV silica window was also used as a path length corrector in the reflected arm to account for refraction through the plate beam-splitter. PDMS samples were mounted at the sample stage for sample positioning.

At the sample stage, care was taken not to focus the interfering beams into the PDMS sample as reported previously [22]. The beams were collimated instead for the purpose of large area patterning. Since the diameter of the interfering beams incident onto the sample was relatively large compared to experiments in other groups, where the focused beam waist was in the order of μm , the shape of the beams incident on the surface have to be considered. The area of the interfering beams incident on the sample was calculated by approximating the beam shape falling on the sample.

Before patterning, the fluence of each incident beams was measured by power meter and total required fluence was kept within $\pm 1\%$ between the beams so as to enable maximum contrast. Patterned samples were then evaluated and analysed using a transmission optical microscope (Zeiss Axiotech 100 HD) with 50X and 200X magnifications and a confocal microscope (Keyence VK-X250). Images were taken with a CCD mounted onto the transmission optical microscope (ToupTek ToupCam).

4. RESULTS & DISCUSSION

Transmittance percentage of the PDMS was found to be 92.05% across eight different fluence readings. Absorbance α of PDMS at 343 nm was then calculated from Equation 4 and was found to be 83.38 m^{-1} . Average contrast of the beams was calculated from Equation 3 and was found to be more than 99.99 across all experiments.

At and above a lower minimum fluence for optical damage was found to be $15.90 \mu\text{Jcm}^{-2}$, periodic patterns were observed below the surface layer. The observed regular periodic features formed were oriented parallel to the plane of interference (Figure 2(a)), contrary to the perpendicular orientation of interference fringes and we are currently investigating this further.

The patterned area found to increase from 20.8% to 48.7% of the incident area as the incident fluence increased, in line with trends reported in related literature [23]. At 200X magnification, the pitch of the regular periodic features were measured to be between $10 \mu\text{m}$ to $13 \mu\text{m}$ over the patterned area. The spatial extent and details of these periodic features can be seen in Figure 2. Surface profile measurements at 500X magnification with a confocal microscope revealed the features to have a height variation of less than $0.2 \mu\text{m}$ from peak to trough. While the patterned area increased as incident fluence increased, the pitch spacing of the periodic features did not change as inferred from the comparison made between the 200X magnifications in Figure 2(d) ($15.90 \mu\text{Jcm}^{-2}$) and 2(e) ($31.80 \mu\text{Jcm}^{-2}$).

Figure 2. (a) Schematic figure of periodic patterns observed indicating the wider illuminated area and the smaller patterned area. The incident interfering beams are both contained in the x-z plane. (b) 50X image of patterned area with fluence $15.9 \mu\text{Jcm}^{-2}$. Note the right side of the patterned area which overlapped the cut in the PDMS. (c) 50X image of pattern area with fluence $31.8 \mu\text{Jcm}^{-2}$. (d) 200X image of patterned area with micro-voids visible. (e) 200X image of patterned area with some micro-voids visible. Note the area to the right that cannot be focused properly, providing evidence towards a more complex interference pattern.

In order to investigate the obtained periodic features further, the patterned PDMS samples were sectioned and rotated 90° to observe the depth of the features. The PDMS sample exposed to $31.80 \mu\text{Jcm}^{-2}$ was sectioned lengthwise and breadthwise across the patterned area (Figure 3(a)) and then observed under 50X and 200X magnification. In the breadthwise section (Figure 3(b) & (c)), the surface-level period pattern was found to extend into the bulk of the sample to a depth of $600 \mu\text{m}$. The regular periodicity of the pattern extended throughout the depth of the patterned area until the pattern itself was no

longer observable at about $600\text{ }\mu\text{m}$ in depth. Examining the lengthwise section further revealed the shape and distribution of the periodic features was angled in a V towards the centre of the patterned area (Figure 3(d) & (e)). The measured half-angle of the pattern, 24.1° was similar to the calculated value of 24.0° , which suggests that the angled features were formed parallel to the beam path through the PDMS sample.

Figure 3. (a) Schematic of sectioned samples and direction of observation for each section. Sectioned sample was patterned with $31.80\text{ }\mu\text{Jcm}^{-2}$. (b) 50X image of breadthwise section sample. Sample surface is to the right of the image. (c) 200X image of breadthwise section sample. (d) 50X image of lengthwise section sample. Sample surface is to the right of the image. (e) 200X image of lengthwise section sample.

From the results of the sectioned samples, a clearer picture of the patterned area in three dimensions was obtained, where the periodic features can be concluded to be formed from the interaction of the femtosecond laser beam with the PDMS. The periodicity of the observed features indicated that these features were not formed as a result of PDMS crumpling or wrinkling upon exposure to the beam. This conclusion was further supported by the lack of such periodic features or any evidence of crumpling with single beam illumination. The relationship between the observed periodic features and the interference fringes will be the subject of future research.

As the formation of any fringes or periodic features is dependent on the intensity distribution of the interfering beams within the PDMS sample, any impurities or inhomogeneity within the bulk of the PDMS sample can correspondingly affect fringe formation. As evident from Figure 1(a) and (b), the micro-scale pattern was periodically spaced until the area where the pattern met the observed cut in the sample. Samples must therefore be free from defects and impurities like cuts and dust, as well as crumpling due to improperly cured samples.

Lastly, regularly spaced micro-voids (perpendicular to the fringes) of diameter approximately 2 to 3 μm were observed at the ends of some of the periodic features. These voids could not be observed directly from the sectioned samples and it is expected that in future research clearer samples could better elucidate more information on these micro-voids.

5. CONCLUSION

In this paper, an investigation into direct dual-beam patterning of PDMS, an optically transparent material at 343 nm was conducted, with the aim of developing substrates for 3D micro batteries. Results of these preliminary investigations were presented, and it is inferred that above a fluence threshold of $15.90 \mu\text{Jcm}^{-2}$ collimated beams interfering within the bulk PDMS substrate formed periodic patterns with 10 to 13 μm pitch width parallel to the plane of the interfering beams. Sectioned samples revealed that the periodic features extended into the bulk of the material, and along the plane of interference, were parallel to the beam path through the PDMS sample. These periodic features do share some characteristics of interference fringes such as the consistency of pitch width with increasing beam fluence. These shared characteristics points towards some relationship between these larger periodic fringes and the smaller 200 nm wide theoretical fringes. The regularly spaced 2 μm wide micro-voids formed during interference also seemed to have a relationship with the periodic features observed. The size and complexity of the patterned area increases both with exposure time and incident fluence. More studies will need to be conducted to elucidate the nature and mechanism of these features, and extend this work towards developing high aspect ratio or high areal density features.

Future studies will focus on identifying the parameters to pattern these voids with electrolyte and electrode materials in order to form 3D solid-state micro-batteries directly within the flexible electronics substrate. It is envisaged that the proposed method and the initial data presented provide a platform to explore the untapped domains novel patterning schemes and substrates for solid-state 3D microbattery applications. Further, the proposed interferometric configuration can accommodate multiple beams with which different types of pattern dimension can be achieved, which will be one of the future research directions.

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