



# Laser patterning of hierarchical structures on metal cylinders for UV-NIL replication

Pierre Lorenz<sup>a,\*</sup>, Lukas Bayer<sup>a</sup>, Theodoros Tachtsidis<sup>b</sup>, Klaus Zimmer<sup>a</sup>

<sup>a</sup>Leibniz Institute of Surface Engineering (IOM), Permoserstr. 15, 04318 Leipzig, Germany <sup>b</sup>Nanotypos, P.O.Box. 60756, 57001 Thermi, Greece

#### **Abstract**

Highly functionalised surfaces are requested by various applications for water repellent, self-cleaning, antifouling and friction reduction purpose. These applications require regularreplication such as UV-NIL (ultraviolet nanoimprint lithography) to allow micron pattern transfer into functional materials and realise a high throughput, low-cost processing techniques. Therefore, large area and seamless (360°) laser patterning of nickel sleeves was performed using ps – laser radiation with wavelengths of 1064 nm and 355 nm producing structures with sizes of  $\geq$  20  $\mu$ m and  $\geq$  5  $\mu$ m, respectively. Further, the intended hierarchical patterns were written by overlaying of array patterns that were ablated with IR- and UV-laser pulses. A high lateral precision of better than 10  $\mu$ m was achieved on sleeves for large structured areas (A  $\geq$  66 cm²). In addition, beam splitting phase masks for 355 nm were used to reduce the fabrication time. The fabricated structures on sleeves were replicated into silicone for imaging and moulded into ORMOCER®-based photoresist film on polymer foil in a UV-NIL roll to roll process. With this approach complex  $\mu$ m-scale patterns with an aspect ratio up to 1 were achieved.

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Keywords: UV-NIL; ps-laser laser patterning; hierachical structures; metal cylinder; nickel

# 1. Introduction

Ultrashort laser pulses allow the well-defined pattering of flat and curved metal surfaces like metal sleeves [1-3] by direct writing without excessive heat impact. Usually, laser ablation processes are utilised for texturing of surfaces with micron-sized features [4]. In addition to determined surface texturing by laser ablation, the laser-solid interaction can result in the formation of periodic surface structures called LIPSS (laser-induced periodic surface structures) [5]. Micro- and nanostructures can influence the macroscopic properties of surfaces like water contact angle or friction but also LIPSS can contribute to functionalisation [6]. Highly functionalised surfaces, however, require the combination of specific chemical, physical and morphological features and properties to fulfil the characteristics of the particular application. While replicating the laser produced micron structures e.g. by a R2R UV-NIL (roll-to-roll ultraviolet nanoimprint lithography) process onto a foil [7] the laser fabricated morphology is transferred inversely into a functional material resulting in a surface featuring characteristic morphological and chemical properties. In this study, hierarchical structures were fabricated by ps-laser irradiation of nickel sleeves in a two-step process with two wavelengths. The different sized patterns were overlayed with high precision to realise the hierarchical structures that were applied for replication by a R2R UV-NIL process.

# 2. Experimental set-up

The fast, precise and large area fabrication of periodic pattern structures for hierarchic micrometre structures is a challenge for the laser methods. Nickel sleeve surfaces were patterned by a Nd:YVO<sub>4</sub> ps-laser with a pulse duration of  $\Delta t_p = 12$  ps as well as a wavelength of  $\lambda = 1064$  nm (fundamental) and  $\lambda = 355$  nm (third harmonic), respectively. The pulse repetition rate was set to  $f_{rep} = 100$  kHz. Within the optimisation experiments the laser

<sup>\*</sup> Corresponding author. Tel.: +49-341-235-3291 fax: +49-341-235-2584. E-mail address: pierre.lorenz@iom-leipzig.de

energy  $E_L$ , the number of laser pulses N and scanning speed  $v_s$  were varied. The laser is installed in a 3D Micromac microSTRUCT vario workstation that features two beam paths (fundamental + THG) with optics for shaping, scanning and focusing the beam. Therefore, micromachining of hierarchical structures with high overlay accuracy is enabled by precise moving the rotation unit to both paths (see Fig. 1). The installed intelliScan scanners were equipped with f-theta lenses of a focal length of 165 mm and 65.5 mm for  $\lambda = 1064$  nm and 355 nm, respectively. The experiments were performed on nickel sleeves (thin metal cylinders). The sleeves were fixed on a pressure cylinder (SWG, Frankenberg) of the rotation unit installed into the machine (see Fig. 1).

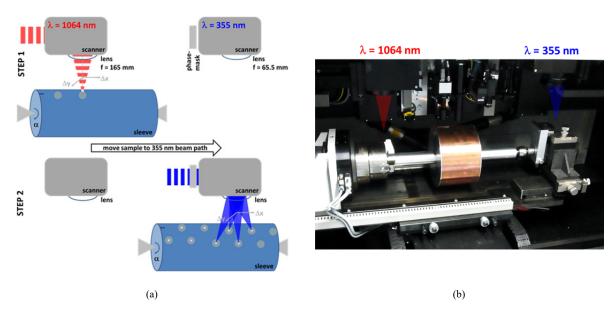


Fig. 1.(a) Schematic illustration of the laser process steps for hierarchical pattern writing on sleeves by combining 1064 nm single spot and 355 nm multi spot processing.

(b) Photographic image of the used laser-system (for a better contrast in the image a copper sleeve was installed)

The laser workstation allows the sleeve movement from the 1064 nm to the 355 nm scan field with a precision of some microns, which enables writing of pattern with a well-defined overlap. The experiments were performed with two different beam set-ups: (i) 1064 nm and 355 nm single beams and (ii) 1064 nm single beam and 355 nm multi-beam approaches to reduce fabrication time. The beam splitting was performed by pure phase mask patterns (periods  $p=150~\mu m-550~\mu m$  were tested) that were produced in fused silica by photolithography and ion beam etching. The pattern period defines the diffraction angles and therefore the lateral distance of the laser spots but the intensity of the diffracted beams can be adjusted independently by the etching depth h in the fused silica. For  $h=\lambda/(2(n_{opt}-1)\sim372~nm$ , the zero order beam are suppressed and the four first order beams hold the most laser power; the intensity of the third order beam is almost one order of magnitude smaller by this spots. Here, the geometry of the binary phase mask was designed to suppress of the even diffraction orders. At this approach for hierarchical pattern writing the distance of the spots achieved by beam splitting must be equal the period of the hierarchical structures. The structures written into nickel surfaces were analysed by optical as well as scanning electron microscopy (SEM) either direct at the sample or by means of moulding them into silicone. The laser patterned metal sleeves were cleaned and passivated by a chemical process prior utilisation for R2R UV NIL to allow a gentle demoulding.

#### 3. Results and discussion

# 3.1 Single beam structuring

The first experiments were performed with a single 1064 nm and a single 355 nm laser spot. First of all, the overlap was tested on a flat nickel sample. An exemplary result is shown in Fig. 2 (a). The optimised laser system allows the well-defined overlap of both structures with a lateral precision of  $\sim 1~\mu m$  for the overlapping of a single dot produced by 1064 nm with 4 dots produced by 355 nm (fabrication time a few second mainly defined by the transfer time of the sample between both beam path). Based on the flat surface experience, curved surfaces were irradiated. Partial sectors of the sleeve surface with a sector size of  $A = 459~mm^2$  ( $\Delta x = 9~mm$ ,  $\Delta y = 51~mm$  ( $\Delta \alpha = 45^\circ$ )) was irradiated by a 1064 nm single beam and 355 nm single beam (fabrication time per sector  $\sim 1~h$ ). In Fig. 2 (b) a photo of the irradiated sleeve, as well as a silicone moulding of a typical test structure, is shown and in Fig. 2 (c) the optical analysis of the test structure on the curved nickel surface is summarised. The experiments show that both laser structured pattern can be overlayed with a high lateral

precision at flat samples. However, the lateral precision at the sleeve is with  $\leq 10~\mu m$  (see Fig. 2 (c)) distinct worse in comparison to the tests on plane nickel samples. An influence of the sleeve holding unit on the overlay precision can be excluded. The overlapping test on the nickel sleeves similar to the flat sample test results in a lateral precision  $\sim 1~\mu m$ , too. The lateral precision is most likely mainly defined by thermal effects like pointing stability of the laser ( $\leq 50~\mu rad/K$ ) and thermal shifts of the scanning system. A reduction lateral overlap deviation down to 5  $\mu m$  over a large area has been achieved by applying a well-chosen thermal management procedure.

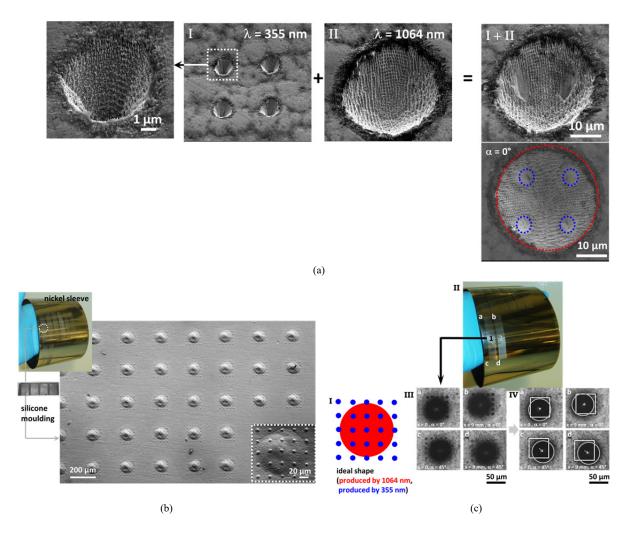


Fig. 2. (a) SEM image of overlapping test of 355 nm and 1064 nm produced on a flat nickel sample (SEM measured at  $45^{\circ}$  and in image (right, bottom) at  $0^{\circ}$ )

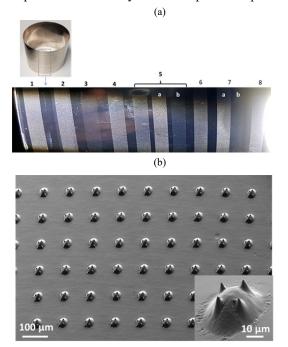
(b) Photo image of laser structured nickel sleeve as well as their silicone moulding of hierarchic structures produced by an overlapping of a 1064 nm and 355nm single beam and SEM image of silicone moulding.

(c) Test of the overlap of a 1064 nm pattern structure with 355 nm pattern structures. I: Ideal overly of the hierarchical pattern structure, II: Optical image of produced pattern structures on the nickel sleeve, III: Microscopic images of the laser-produced structures on the nickel sleeve at different position, IV: Analysing of the mismatch of the 355 nm pattern structure to 1064 nm structures based the microscopic image at different position (big holes:  $\lambda = 1064$  nm, N = 315,  $E_L \sim 1.2~\mu J$ ; small holes:  $\lambda = 355$  nm, N = 1329,  $E_L \sim 0.08~\mu J$ )

#### 3.2 multi beam structuring

Furthermore, the 355 nm beam was beam split by a phase mask ( $p = 450 \mu m$ , measured depth h = 397 nm) into four main spots to reduce the fabrication time for the production of seamless structures (see Fig. 3 (a)). This phase mask together with the optics causes a distance of the four dominant spots (first diffraction order beams) of ~96  $\mu m$  where the orientation of the diffracted beams was set to be parallel to the x- and y-direction of the laser system. The 1064 nm single spot and the 355 nm multi-spot was moved over sample surface the by a combination of scanner and stage movements to produced periodic hierarchic structures over a large area. An exemplary result of structures with beam splitting is shown in Fig. 3 (a) and (b). After optimisation the overlapping process the fabrication time was improved due to the beam splitting of the 355 nm beam and the sleeve was seamlessly structured (see Fig. 3 (c)). Seamless structures with an area up to 66 cm² were produced in

an interruption-free laser irradiation process. Such hierarchic surface structures on sleeves have been replicated onto ORMOCER®-based photoresist on polymer foils by UV-NIL in the R2R-UV-NIL machine of Nanotypos. The anti-adhesion functionalised sleeve surface sticking of the cured polymer resist at the sleeve can be avoided and an easy demoulding achieved. Various patterns types (dot arrays hierarchical pattern, and LIPSS) were replicated successfully with web speeds of up to 5 m/min.



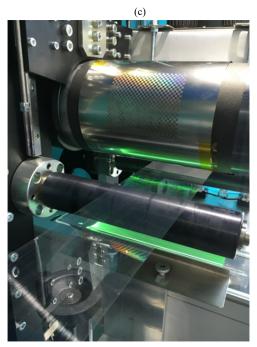


Fig. 3. (a) Optical images of different seamless laser structures on a Ni-sleeve produced by the overlapping of a 1064 nm single spot with a four times split 355 nm beam.

(b) SEM image of silicone moulding of hierarchical structures (moulded from block 6, see (a)) (big holes:  $\lambda=1064$  nm, N=653,  $E_L\sim0.12~\mu J$ ; small holes:  $\lambda=355$  nm (with phase mask), N=2013, energy of the primary beam  $E_{L,0}\sim0.46~\mu J$ )

(c) Optical image during R2R moulding of a laser structured nickel sleeve.

### 4. Conclusion

The high-precision overlapping of structures produced with a 355 nm and 1064 nm with a lateral precision  $\leq 10~\mu m$  (large areas) and  $\sim 1~\mu m$  (dot-dot overlay) allows the fabrication of well-defined hierarchical microstructures on flat and curved nickel surfaces over a large area. Furthermore, the usage of beam splitting allows the reduction of the fabrication time. However, the time and thermal stability of the used system limits the lateral precision of the structuring process. The in-situ measurements and readjustment of the beam position during the laser structuring process might be necessary to improve the lateral precision.

#### Acknowledgements

Research get funding from the European Union (FP7/2007-2013) under APPOLO project (No. 609355).

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