



Lasers in Manufacturing Conference 2021

Fabrication of complex periodic patterns on a metallic drum for high throughput roll-to-roll processing

Bogdan Voisiat^{a,*}, Max Menzel^a, Wei Wang^a, Yangxi Fu^a, Marcos Soldera^a, Andrés Fabián Lasagni^{a,b}

^aInstitut für Fertigungstechnik, Technische Universität Dresden, George-Bähr-Str. 3c, 01069 Dresden, Germany

^bFraunhofer Institute for Material and Beam Technology IWS, Winterbergstraße 28, 01277 Dresden, Germany

Abstract

In this study, the development of complex periodic structures on massive metal drums by means of direct laser interference patterning (DLIP) is demonstrated. The DLIP technology allows the formation of high-resolution periodical structures (even with sub-micrometer resolution) at high fabrication speeds on large surface areas. These advantages drastically reduce the patterning costs of the drums that are broadly used in roll-to-roll processing. We demonstrate the ability to control individually each laser spot (e.g. period) to form complex periodical patterns to be used as decorative elements exhibiting structural colors. These patterns are then replicated on a polymer foil by an industrial hot-embossing roll-to-roll process at speeds up to 50 m/min. This process brings the industrial fabrication of such patterns to the next level in terms of throughput and is thus suitable for mass production.

Keywords: Direct Laser Interference Patterning; roll-to-roll; structural coloration; diffraction gratings

1. Introduction

Surface functionalization by engraving a controlled microtopography has become a convenient path for many industrial sectors to improve their products. A process capable of producing micro and nanostructures on thermoplastics in a fast and cost efficient way is roll-to-roll hot embossing, whereby a running polymeric foil is heated above the glass transition temperature and pressed between two heated drums containing the desired surface texture (Ahn & Guo, 2009). Surface patterning of cylindrical tools at high throughputs with a resolution down to the sub-micron scale poses many technological challenges, especially when a high degree of flexibility is required for achieving arbitrary textures on large areas. For instance, electron beam lithography

* Corresponding author. Tel.: +49 351 463 - 34976; fax: +49 351 463-37755 .
E-mail address: bogdan.voisiat@tu-dresden.de

(EBL) or laser interference lithography (LIL) have been used to pattern cylindrical parts with a resolution far below 1 μm , but they require long processing times (up to several days for areas over 0.5 m^2) as well as photoresist deposition and development steps (Taniguchi & Aratani, 2009). Recently, a novel method termed nanocoining was employed to pattern micro and nanostructures on a Ni coated tube, whereby a focused ion beam (FIB) structured diamond die mechanically indents the rotating cylindrical mold (Cates et al., 2021). Although this method features a relatively high throughput of 10 cm^2/min , arbitrary textures can not be easily fabricated and the diamond patterning by FIB is a complex and time-consuming process. A very promising technology that circumvents these issues is Direct Laser Interference Patterning (DLIP), which relies on the interference of multiple beams to form periodical patterns with a single processing step (Rosenkranz et al., 2016; Voisiat et al., 2011). Using a ps-laser source with a relatively low output power of 10 W, Ni sleeves were microstructured at a remarkable throughput of 57 cm^2/min by Rank et al., 2019. Furthermore, with this method it is possible to structure arbitrary areas and to modify the laser parameters, like pulse-to-pulse overlap or spatial period, on-the-fly, for producing customized decorative elements, such as logos.

In this study, further possibilities of the DLIP method are explored to produce complex microstructures over large areas on Ni and Cr cylindrical workpieces. In this way, cylindrical masters with individualized designs for roll-to-roll hot embossing can be fabricated with high precision and throughput.

2. Materials and Methods

2.1. Structuring of cylindrical tools by Direct Laser Interference Patterning

An in-house DLIP (developed by Fraunhofer IWS, TU Dresden) setup equipped with a ps-pulsed solid state laser (Edgewave PX200, Germany) with an output power of 10 W was used to pattern the cylinders. The laser source has a fundamental wavelength of 1064 nm, but also the second (532 nm) and third harmonic (355 nm) can be employed. The pulsed laser radiation has a fixed pulse duration of 10 ps while the repetition rate can be adjusted up to 30 kHz. Two optical heads were used in this study that were designed for the IR (1064 nm) and visible (532 nm) wavelengths. The design principle of the DLIP heads is shown schematically in Figure 1 and it consists in a diffractive optical element (DOE) that splits the primary laser beam into two sub-beams (for the IR head) which are parallelized by prism and then focused on the sample by a lens. In the case of the head designed for 532 nm radiation the selected DOE splits the beam into four sub-beams. The cylindrical parts were mounted on a rotary axis system whose motion can be controlled with a computer. Varying the distance between the prism and DOE the overlapping beams angle can be adjusted allowing a fine-tuning of the spatial period of the interference pattern. Further details can be found elsewhere (Lang et al., 2017; Rank et al., 2017; Soldera et al., 2020).

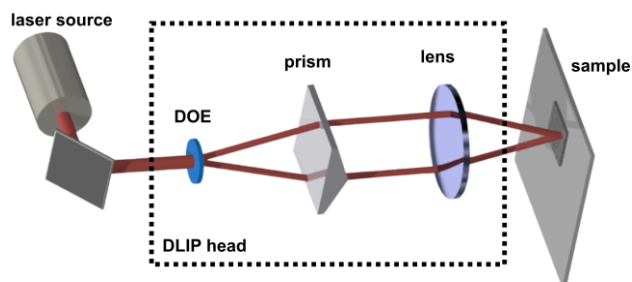


Fig. 1. Operating principle of DLIP head used in this work. The laser source consisted on a ps-pulsed solid-state laser, with 10 W laser power, operating at 532 nm and 1064 nm wavelengths.

2.2. Topography characterization

A confocal microscope (Sensofar S neox, Sensofar S.A., Barcelona, Spain) was used at a magnification of 150× to measure the surface topography of the structured samples, providing a lateral and vertical resolution of 140 nm and 2 nm, respectively. The obtained topographical data were then analyzed with the SensoMap software (SensoMap “Premium” Version 7, Sensofar S.A., Barcelona, Spain). Due to the large size of the treated drums (228 mm in diameter and 762 mm in length), its direct characterization using the above mentioned confocal microscope was not possible. Therefore, a fast curing replication compound (101 RF, Microset Products Ltd., Leicestershire, UK) was applied on the structured cylinders to indirectly measure the surface topography. The compound deposition procedure was performed manually by droplet dispensing the compound onto the structured surface. Afterward, the imprint was peeled of the sample creating an inverse replica of the texture.

3. Results and Discussion

3.1. Line-like textures

Periodic grooves with different geometric dimensions were fabricated on a cylindrical nickel sleeve (300 mm in length, 300 mm in diameter and 200 μm of thickness) using IR radiation at a repetition rate of 10 kHz and a fluence of 0.96 J/cm². The spatial period was varied in the range 1.7 – 5.9 μm , while the pulse-to-pulse feed was swept between 2 and 20 μm resulting in different structure heights and aspect ratios. Figures 2 a) and b) show photographs of the resulting structured sleeve, where each colored ring with a width of 15 mm represents a different set of DLIP parameters.

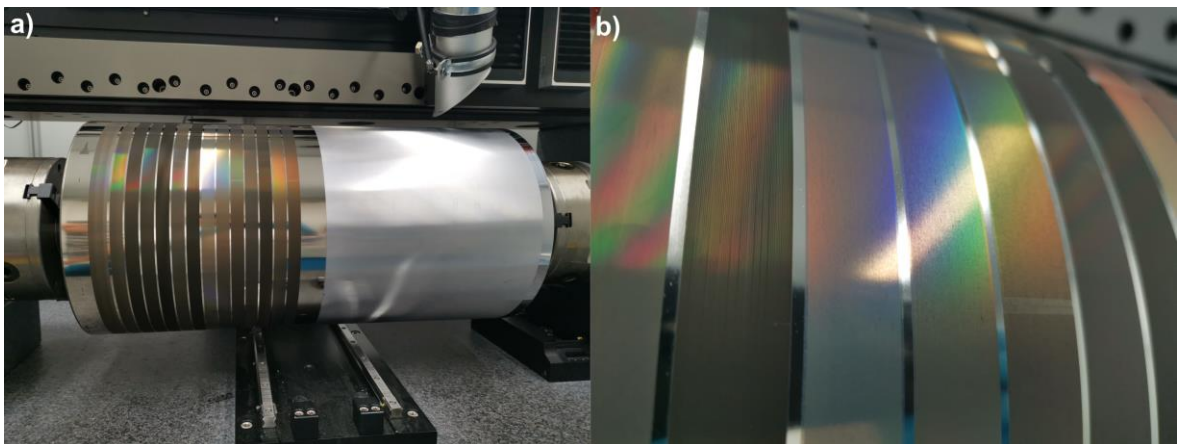


Fig. 2. a) and b) Photographs with different magnifications of a Ni sleeve structured with 15 different set of parameters (see Table 1 for details).

Table 1 lists the used DLIP parameters as well as the resulting structure heights measured with confocal microscopy and calculated aspect ratios. According to the sample labeling of Table 1, the numbering of the samples in the sleeve shown in Figure 2 starts from the left to the right. The topography of three selected samples are shown in Figure 3, together with the corresponding extracted profiles.

Table 1. DLIP parameters and topography results of the structured sleeve shown in Fig. 2. Each structured stripe has a width of 15 mm.

Number	Period (μm)	Pulse-to-pulse feed (μm)	Hatch distance (μm)	Structure height (μm)	Aspect ratio
1	1.25	20	125	0.32	0.26
2	1.47	20	125	0.35	0.24
3	1.71	10	100	0.82	0.48
4	2.1	10	100	1.31	0.62
5	2.1	5	100	2.11	1.00
6	3.5	20	100	0.83	0.23
7	3.5	5	125	2.29	0.65
8	4.7	10	100	1.52	0.32
9	4.7	5	100	2.45	0.52
10	4.7	2	100	4.05	0.86
11	5.9	20	100	0.51	0.086
12	5.9	10	100	1.02	0.17
13	5.9	5	100	2.21	0.37
14	5.9	2	100	4.21	0.71

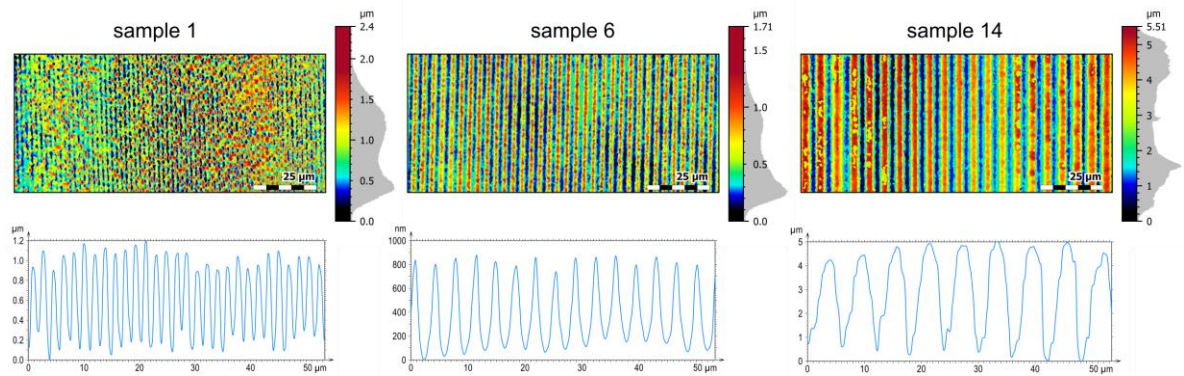


Fig. 3. Topography images taken by confocal microscopy of selected samples structured on a Ni sleeve (see Table 1 and text for details).

3.2. Cross-like textures

A square array of micropillars, or cross-like texture, can be fabricated by two subsequent DLIP patterning steps, in which two-beams are overlapped in each step. First, an underlying line-like texture is patterned on the sample, while in the second step a 90° rotation between the sample and the plane of incidence of the beams is applied followed by the structuring process (Bieda et al., 2015). In order to structure large cylindrical workpieces with such cross-like texture, a DLIP head equipped with a DOE that splits the incoming laser beam into four sub-beams was used. In the first step, a line-like texture was engraved on the sample with the grooves aligned perpendicular to the cylinder axis by allowing only two sub-beams to overlap on the surface, as shown schematically in Figure 4a. In the second step, the optical head was translated to the initial position and the blocked sub-beams are swapped, implying an effective rotation of the overlapping beams on the surface by 90° (Figure 4b). In this way, the patterning of grooves parallel to the cylinder axis was enabled. Following this procedure, a massive steel drum coated with a chromium layer with a length of 762 mm and diameter of 228.6 mm was structured with green radiation (532 nm). For the first DLIP step, repetitive grooves with a spatial period of $1.2 \mu\text{m}$ were patterned at a fluence of 2 J/cm^2 , a repetition rate of 10 kHz and a pulse-to-pulse feed

of $25\ \mu\text{m}$. Afterwards, the interfering pattern was rotated by 90° as described above and the drum was re-irradiated at a fluence of $1.7\ \text{J}/\text{cm}^2$, a repetition rate of $1\ \text{kHz}$ and pulse-to-pulse feed of $50\ \mu\text{m}$. In this second step, the accumulated fluence was significantly lower than in the first one, because the patterned chromium surface after the first DLIP process absorbs the laser light more efficiently. The resulting structured area on the cylinder was $1790\ \text{cm}^2$. The negative of the cross-like texture is shown in the topographical image of Figure 5 taken by confocal microscopy on the replicated imprint (see section 2.2 for details). From this measurement, it can be inferred that the texture on the cylinder resembles a square array of micropillars with a spatial period of $1.3\ \mu\text{m}$ and a structure height of $130\ \text{nm}$, which results in an aspect ratio of 0.1 .

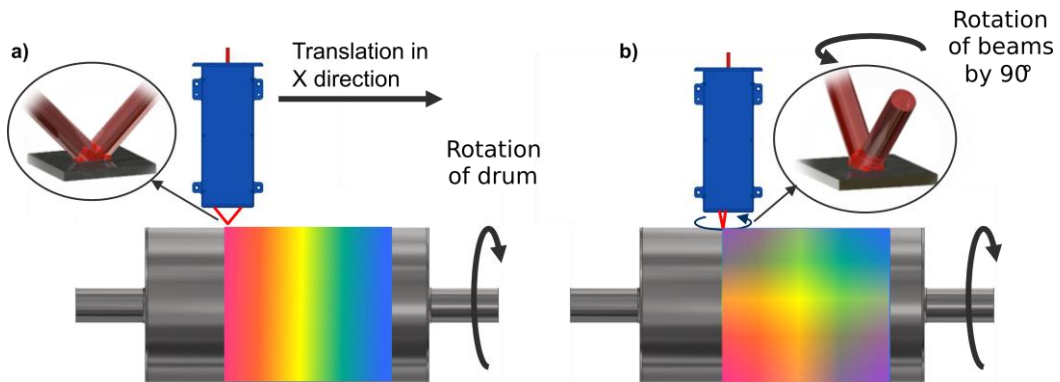


Fig. 4. Schematical representation of the procedure to fabricate cross-like textures on a drum. a) In the first step a line-like texture is patterned, whereas b) in the second step the process is repeated after a 90° degree rotation of the overlapping beams.

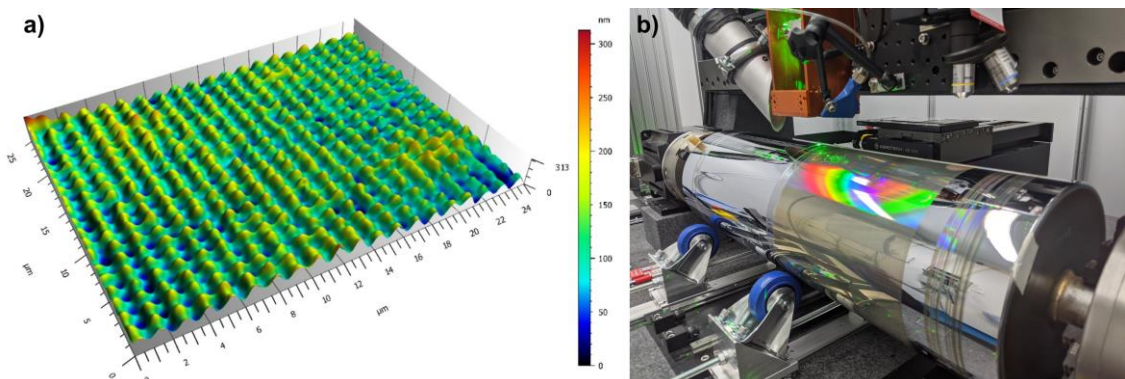


Fig. 5. a) Inverted topography of a cross-like texture structured on a chromium-coated steel drum. b) Large-area patterned drum.

3.3. Dot-like textures

Arrays of periodic microholes, or dot-like textures, with a square symmetry can be patterned by overlapping four sub-beams. To that end, a steel drum coated with a chromium layer with a length of $762\ \text{mm}$ and diameter of $228.6\ \text{mm}$ was structured by four-beams DLIP using laser pulses at a wavelength of $532\ \text{nm}$, a repetition rate of $5\ \text{kHz}$, a pulse-to-pulse feed of $80\ \mu\text{m}$ and a fluence of $1.5\ \text{J}/\text{cm}^2$. The patterned area totalized $1790\ \text{cm}^2$. The confocal image of Figure 6 was taken on the replica of the dot-like texture. The replica features a

square-array of microdomes, implying that the texture on the drum should be a hole-like array characterized by a spatial period of $1.75 \mu\text{m}$ and a structure height of 120 nm .

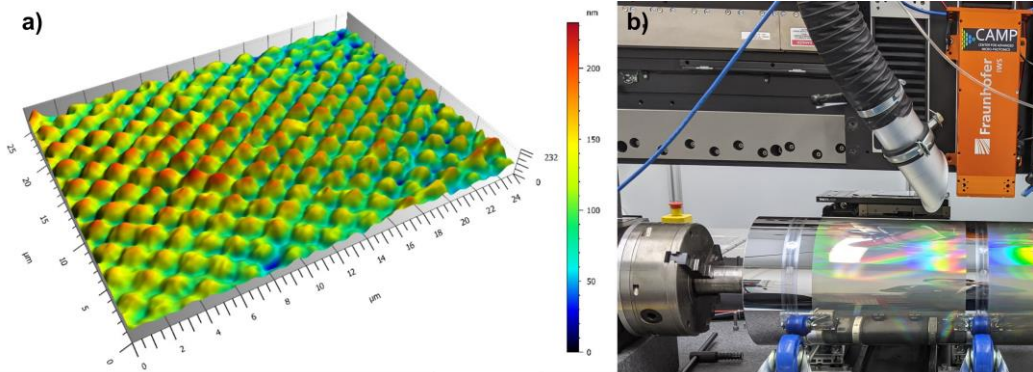


Fig. 6. a) Inverted topography of a dot-like texture structured on a chromium-coated steel drum. b) Large-area patterned drum.

3.4. Perspectives for engineered holographic motives on cylindrical tools

Interaction between incoming white light and periodic microtextures patterned on a surface can induce the appearance of diffraction modes reflected off the surface. Depending on the incidence angle, spatial period and texture shape, specific wavelengths can be directed to an observer, generating an apparent coloration on the surface (Kinoshita et al., 2008; Storm et al., 2019). This holographic effect can be exploited for producing logos, motives or anti-counterfeiting features on a cylindrical mold for roll-to-roll hot embossing. The flexibility of the DLIP method is a key feature for fast processing of individualized textured areas with such holographic appearance on both flat and cylindrical surfaces.

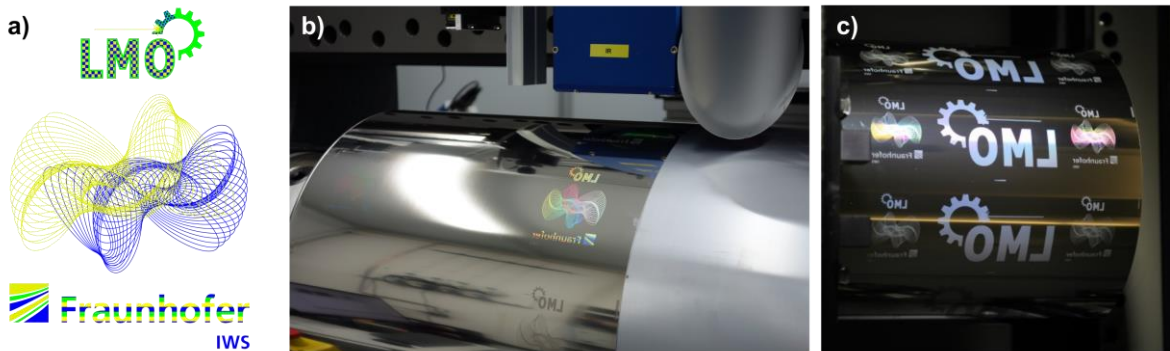


Fig. 7. a) Logos designed with three different colors that were converted to three different spatial periods. b) and c) Photographs of a sleeve structured with logos. The coloration of the engraved logos can be attributed to the different patterned spatial periods.

For instance, the utilized laser system coupled to the two-beam DLIP (1064 nm) head can deliver a spot size of $80 \mu\text{m}$ on the sample surface enabling a maximum resolution of 300 DPI. As each pixel produced on the surface can have a particular spatial period and structure height, the possibilities for patterning customized decorative features are endless. As an example, the logos in Figure 7a were engraved on a Ni sleeve (300 mm

in length, 300 mm in diameter and 200 μm thick) where each of the three colors in the bitmap image was associated to a different line-like spatial period, i.e. 1.8 μm , 1.9 μm or 2.0 μm . In this way, under a given observation angle the patterned image reflects mostly three wavelengths linked to each spatial period, as observed in the photographs of Figures 7b and c. Note that the logos patterned on the sleeve are mirrored, so that the imprinted images on a polymer after roll-to-roll processing are displayed correctly.

4. Conclusions

In this work, it is demonstrated that DLIP is a versatile tool for patterning customized structures and motives on cylindrical workpieces over large areas. Line-, cross- and dot-like textures were produced on Ni sleeves and massive steel drums coated with a chromium layer. It was also shown, that the ability to change the spatial period and the structure aspect ratio during the laser processing enables a high degree of flexibility, which can be exploited for patterning logos, decorative motives or anti-counterfeiting features. These advances pave the way for rapid prototyping of cylindrical masters with arbitrary shapes and microtextures for roll-to-roll hot embossing processes.

Acknowledgements

This work was partially supported by the Federal Ministry for Economic Affairs and Energy (Grant No. ZF4821601LP9). M.S. acknowledges the support of the Alexander von Humboldt Foundation. Part of this work was also founded by the European Regional Development Fund (ERDF) and co-financed under taxation on the basis of the budget adopted by the members of the Saxon State Parliament.

References

- Ahn, S. H., & Guo, L. J., 2009. Large-Area Roll-to-Roll and Roll-to-Plate Nanoimprint Lithography: A Step toward High-Throughput Application of Continuous Nanoimprinting, *ACS Nano* 3(8), p. 2304–2310.
- Bieda, M., Schmädicke, C., Roch, T., & Lasagni, A., 2015. Ultra-Low Friction on 100Cr6-Steel Surfaces After Direct Laser Interference Patterning, *Advanced Engineering Materials* 17(1), p.102–108.
- Cates, N., Einck, V. J., Micklow, L., Morère, J., Okoroanyanwu, U., Watkins, J. J., & Furst, S., 2021. Roll-to-roll nanoimprint lithography using a seamless cylindrical mold nanopatterned with a high-speed mastering process. *Nanotechnology* 32(15), p. 155301.
- Kinoshita, S., Yoshioka, S., & Miyazaki, J., 2008. Physics of structural colors. *Reports on Progress in Physics* 71(7), p. 076401.
- Lang, V., Rank, A., & Lasagni, A. F., 2017. Large Area One-Step Fabrication of Three-Level Multiple-Scaled Micro and Nanostructured Nickel Sleeves for Roll-to-Roll Hot Embossing. *Advanced Engineering Materials* 19(8), p. 1700126.
- Rank, A., Lang, V., & Lasagni, A. F., 2017. High-Speed Roll-to-Roll Hot Embossing of Micrometer and Sub Micrometer Structures Using Seamless Direct Laser Interference Patterning Treated Sleeves. *Advanced Engineering Materials* 19(11), p. 1700201.
- Rank, A., Lang, V., Voisiat, B., & Lasagni, A. F., 2019. Roll-to-roll hot embossing process: A way to scale up the fabrication speed of micro-nano structures formed by direct laser interference patterning. *International Society for Optics and Photonics. Laser-Based Micro- and Nanoprocessing XIII 10906*, p. 109060V.
- Rosenkranz, A., Hans, M., Gachot, C., Thome, A., Bonk, S., & Mücklich, F., 2016. Direct laser interference patterning: Tailoring of contact area for frictional and antibacterial properties. *Lubricants* 4(1), p. 2.
- Soldera, M., Wang, Q., Soldera, F., Lang, V., Abate, A., & Lasagni, A. F., 2020. Toward High-Throughput Texturing of Polymer Foils for Enhanced Light Trapping in Flexible Perovskite Solar Cells Using Roll-to-Roll Hot Embossing. *Advanced Engineering Materials* 22(4), p. 1901217.
- Storm, S., Alamri, S., Soldera, M., Kunze, T., & Lasagni, A. F., 2019. How to Tailor Structural Colors for Extended Visibility and White Light Generation Employing Direct Laser Interference Patterning. *Macromolecular Chemistry and Physics* 220(13), p. 1900205.
- Taniguchi, J., & Aratani, M., 2009. Fabrication of a seamless roll mold by direct writing with an electron beam on a rotating cylindrical substrate. *Journal of Vacuum Science & Technology B: Microelectronics and Nanometer Structures Processing, Measurement, and Phenomena* 27(6), p. 2841–2845.

Voisiat, B., Gedvilas, M., Indrišiūnas, S., & Račiukaitis, G., 2011. Picosecond-laser 4-beam-interference ablation as a flexible tool for thin film microstructuring. *Physics Procedia* 12, p.116–124.