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TECHNICAL PAPER

Nanoimprint lithography and micro-embossing in LiGA technology: similarities and differences

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Abstract LiGA and NIL are both techniques with origination by lithography, using molding techniques for upscaling or even stamp copying. The main difference is the size range for which they were seen to have most of their applications. The technology toolboxes contain similar processes and concepts, but due to historical differences, NIL and LiGA were not seen as twins, but rather children from different families. LiGA, as the more mature microtechnology, has found its application in microfluidics, -optics and -mechanics. NIL has found its place in photonics and sub-wavelength gratings, and is considered as a candidate for patterned media and next generation lithography for IC manufacturing. In this review I will discuss similarities and differences of the two technologies, tackle questions from pattern transfer, size effects including the need for hard and soft elements for molding and discuss points where LiGA and NIL might find a common basis for further cross-fertilization and joint applications.

1 Introduction

Nanoimprint lithography (NIL) was developed in the 1990s as an alternative to existing high throughput nanolithographies for microchip manufacturing. This was because its sub-10 nm resolution capabilities were (and still are) far beyond the reach of established photon based lithographies and due to its parallel, large area approach it also enabled upscaling of substrate areas while keeping cost issues low (Chou et al. 1995; Chou and Krauss 1997; Haisma et al. 1996). Although its concept of imprinting a stamp surface topography into a moldable material by "conformal" contact is seen critical by high-end lithography engineers due to its proneness to defects and contamination, it was exactly this unconventional mechanical approach which made NIL attractive in contrast to all methods relying on diffraction limited exposure. At the same time replication by molding was well established in Compact Disc manufacturing, roll embossing of security features, polymer sheets with Fresnel gratings and integrated optics in polymers. The main difference of NIL to other established embossing techniques is that it was intended for pattern transfer, i.e. by using the thin imprinted structure as a masking layer a pattern transfer into the underlying substrate was possible, like with a resist in photolithography. For LIGA technology, German acronym for lithography (Li = lithographie), electroforming (G = galvanik) and molding (A = abformung), that is only a variant of already established processes, e.g. pattern transfer was demonstrated by molding of microstructures on processed wafers (Both et al. 1995), followed by electroplating on locally opened substrate windows (Becker et al. 1986). This window opening is similar to NIL, because the first step after imprint, to enable pattern transfer, is a thinning of the resist without a material-related etching contrast by global, but anisotropic RIE etching in O_2 plasma. This means that only after the thinning down of the resist topography to remove the residual layer below the stamp protrusions, the substrate is cleared and ready for etching, lift-off or electroplating. This process is therefore often called breakthrough etching. NIL established itself as candidate for next-generation lithography (NGL) in manufacturing of integrated circuits (IC), but was always seen as a nanopatterning solution for many other applications (see Table 1; Stewart et al. 2005; Ruiz et al. 2008; Ahn et al. 2005; Ji et al. 2010).

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Table 1 Applications in NIL

	NIL-part	Challenge	Comment
Next generation lithography in IC manufacturing, e.g. for transistor interconnect layer (Stewart et al. 2005)	Single resist level step and repeat imprint with multilevel stamp + pattern transfer by etching	"Mask" overlay, resolution defectivity, only 1:1 mask with contact, S&R stitching, air bubbles	3D stamps (2-levels in one step) may reduce process steps, drop-on-demand resist dispensing enables compensation of structure density variation
Patterned media (Ruiz et al. 2008)	Imprint + etching of pillars into surface of aluminum plate for creation of isolated magnetic islands	Expensive high resolution (20 nm) and large area (2.5″ disk) master with high uniformity by electron beam patterning	Density multiplication by block copolymer lithography (directed self assembly)
Polarizers for flat panel display (Ahn et al. 2005)	Imprint of large area sub-wavelength period grating in thin Al-film on glass	Large 100 nm period master, uniformity in pattern transfer, visible stitching errors	Master by interference lithography, possible upscaling by roll embossing
High brightness light emitting diodes (LED) (Ji et al. 2010)	Patterning of sapphire substrate before growth or enhancement of light extraction by photonic crystal etched into semiconductor surface	Imprint over 80,000 LEDs by conformal printing, substrates with point defects and high vertical unevenness	Conformal printing using surface conformation imprint lithography (SCIL) technique and UV-NIL with flexible stamp

Recently, the term NIL has also been applied for roll embossing, termed roll-to-roll (R2R) NIL, which is another sign that not the specifics of the molding process, but the structure size and application defines how the process is named (Ahn and Guo 2009). For NIL this is sub-100 nm large area patterning circumventing the shortcomings of photolithography and for LIGA the high-aspect-ratio structuring of microstructures aimed towards replacement of traditional precision tooling. In this respect, from the beginning, NIL was more related to the more popular microcontact printing (µCP) than to embossing of microtopographies. Its nanopatterning capabilities opened a new range of applications (Wilbur et al. 1996), where either the limits of photolithography are overcome, or nanoscale effects become visible. Therefore hot embossing and thermal imprint may exist as synonyms, but to coin the name "Hot embossing lithography" instead of NIL failed long ago.

Due to the small size of structures, not only surface effects have to be considered (e.g. surface modifications by dense monomolecular coatings), but also size ranges up to a few nm where polymer molecules are subjected to confinement between boundaries, or where films become so thin that surface and interface properties become dominant over bulk properties. Therefore this is the area where the meaning of "nano" becomes clearer: it is not only defined by the size of the pattern, but also by the presence of effects which might not be relevant or at least not observable with larger structures. At the same time, these structures have to be replicated over large areas from a few mm^2 up to wafer sizes. Tolerances of a few 10 s of nm (in vertical and lateral direction) can only be achieved if bending of substrates is taken into account, a concept which is contradictory to hot embossing of microstructures, which continues to rely on highly rigid structures in a stiff setup with reproducible precision in the µm range.

I am referring here to major publications from the author of this contribution with review-like character (Schift et al. 2003; Schift and Kristensen 2010; Schift 2008), from which part of this article was taken, furthermore newer publications on NIL (NaPa Library of processes 2012; Zhou 2013). Concerning LiGA, I use the up-to-date information presented in (Menz et al. 2005; Heckele and Schomburg 2004; Saile 2009; Worgull 2009). It has to be emphasized that LiGA is not only meant to be DXRL, but a range of other processes aiming to fabricate High Aspect Ratio (HAR) microstructures. Here, however, we will focus on the ability to use the technology toolboxes from LiGA and NIL in the area where both techniques show a strong overlap, which is molding. For both, LiGA and NIL, we use embossing and imprint as synonyms. In case of NIL, we are restricting ourselves to the case of thermal NIL and will only point out specific issues related to UV-assisted NIL, a low pressure room temperature alternative to thermal NIL, if this is necessary for understanding actual problems between LiGA and NIL.

2 Technological differences

2.1 Design related issues: "proximity effects" during molding

The strength of photolithography is its high process robustness and stability within a large range of structure sizes, film thicknesses and polymer chemistries. Both LiGA and NIL have demonstrated similar potentials from the beginning, however, since they were in competition with other established or emerging techniques from the beginning, and had therefore to compare in terms of speed, precision and robustness, process challenges were soon visible. This often led to intensive iterations and process optimization. Their origins were often identified to be design-related. LiGA has demonstrated its ability to create vertical sidewalls and aspect ratios of 100, NIL its ability to achieve 10 nm structures with an aspect ratio of 4, both already in their initial phase after their invention. However, real applications either require highest aspect ratio or smallest resolution with high reproducibility in arbitrary designs, in which these aspect ratios or resolutions have to be created along with other structures—often with contrasting process requirements. Particularly difficult is the situation when both small and large structures have to be patterned, or when the density of structures varies from one side of the substrate to the other. In LiGA this often leads to deformation or cracks due to excessive localized stress in large resist areas after wet development or after demolding (i.e. the separation of the mold from the molded structure after hardening). In NIL this is, because the local diversity of structures and ratio of cavities to stamp protrusion may cause inhomogeneous flow and consequently bending of stamps. Although it is possible to deal with this using appropriate post-processing strategies, satisfactory results can often be achieved if designs allow being adapted. Often this can be done according to some simple design rules.

These design rules for NIL seem to be mostly related to so-called imprint related "proximity effects", which are caused because the molding of a pattern is influenced by the flow of polymer in neighboring patterns. Typical proximity effects in probe-based lithography are due to scattering in electron beam lithography (EBL) and blurred exposure in oblique areas in proximity shadow lithography with a constant gap. In NIL the proximity effect is mostly caused by stamp topography and the limited ability of the thin resist layer to flow between stamp and substrate until the cavities are filled. It is due to the high ratio of structural depth to resist thickness. This means that single structures can lead to starvation of polymer flow or that density variations can cause differences in cavity filling and thus stamp sinking, since equilibration and transport of material cannot take place over larger distances or when residual layers become too thin to allow significant squeeze flow. However, although the range of this proximity effect is larger than in probe-based lithographies, due to the ability of a stamp to bend, these effects are often limited to a few 100 s of µm or mm (see Fig. 1). Therefore optimization is often to be done on a few square mm, but can then be applied to an entire, wafer-like substrate. By pre-compensation of designs, within bending distances, e.g. an

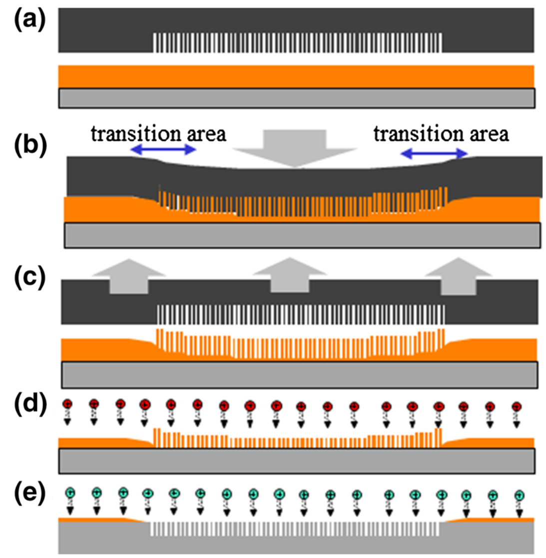

Fig. 1 Thermal NIL of non-uniform patterns (local density variations) relies on stamp bending (**a–c**) during molding. Special care has to be taken for residual layer variation in transition areas (e.g. at the borders of large gratings to non-structured areas), to enable full pattern transfer by reactive ion etching after resist thinning (**d–e**)

equilibration of density variations or simply by adding auxiliary cavities to avoid long distance flow of material, bending can often be reduced to a level where variation in residual layer does not impair pattern transfer.

2.2 The role of size ranges and aspect ratios

The main difference between LiGA and NIL is in the size range for the intended applications, both laterally (structure size) and vertically (vertical structure sizes, including the initial resist height), in which the process is stable. LiGA addresses sizes from 10 to 1,000 µm, NIL from 10 to 1,000 nm, which is roughly a factor of 1,000 between the two technologies. However, since LiGA also advances towards smaller structural sizes and NIL designs often contain structures in the range of several 10 s of µm, there are regions of overlap. This is particularly the case in applications which combine functionalities in the micro- and nanorange, e.g. in microfluidical setups with nanochannels for DNA sequencing and analysis (Fernandez-Cuesta et al. 2011). In principle, apart from these basic size ranges, the ratio between these sizes and their correlation with substrate sizes and thicknesses play an even larger role. For LiGA this is the aspect ratio of structures, i.e. the ratio of height to width of single structures with the ability to form high vertical sidewalls. For NIL this is the ratio of structure size with respect to the residual layer thickness and the ratio of the lateral extension of thin films or wafer like substrates with respect to their thickness and hence the ability of the stamp and substrate to bend. In Fig. 2 we can

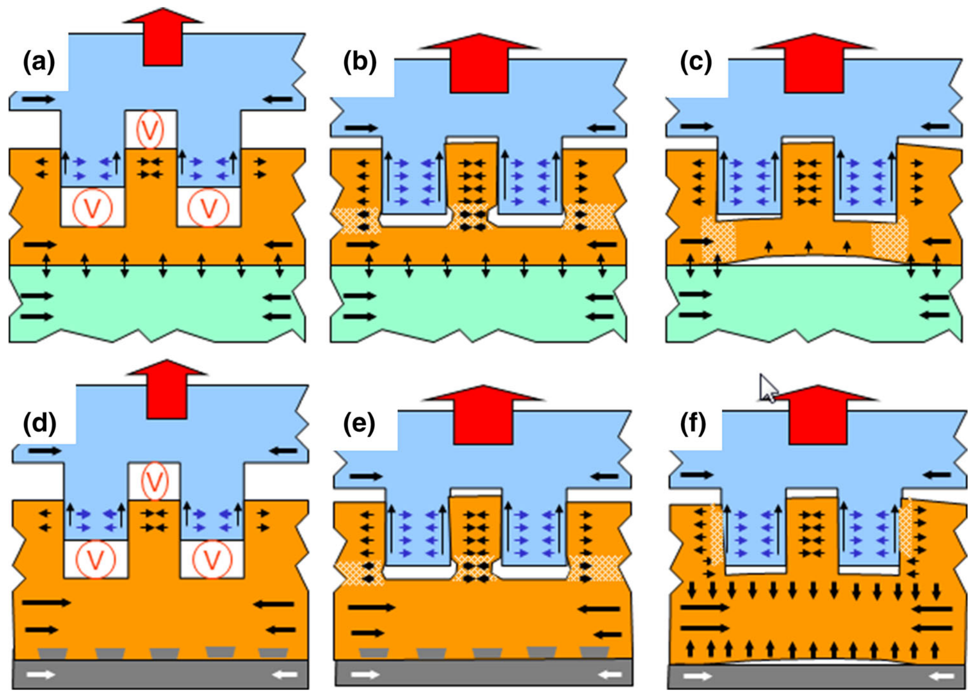

Fig. 2 Demolding issues in LiGA and NIL: the ratio of structure size to film thickness and residual layer determines the effect of thermal expansion on structural deformation. Thin **a**–**c** polymer layers accommodate to both stamp and substrate, thick **d**–**f** plates can cause high stresses due to bulk contraction. Particularly for thermal processes, differences in thermal expansion during demolding and the inability to average out demolding stresses lead to different defects

compare two cases of thin (a–c) and thick (d–f) polymer layers, which demonstrate the influence of thin polymer layers on substrates and thick polymer films or plates where a surface topography is imprinted.

2.3 Demolding

Demolding of high aspect ratio structures with microdimensions is often a two-step process: When the mold begins to be retracted vertically, i.e. along the orientation of the structure walls, it first detaches from the flat protrusions and cavity flats. Afterwards it begins to slide along the sidewalls. This means that the polymer structure has to overcome adhesion forces of the entire surface of the mold towards the molded structure, but only a part of it detaches. During sliding, the sidewall area between mold and molded structures continuously decreases until the mold is totally released from it. Already a small inclination of the sidewalls would eliminate this sliding process and the entire structure would detach at once. In this case the ratio of total surface area to the vertical sidewall area is important, as well as the lateral shrinkage of structures during cooling and hence the force of the aggregate sidewall areas. Demolding in low aspect ratio nanostructures is often disruptive. We do not have any indication that there is an extensive sliding, even if there are smooth vertical sidewalls (Trabadelo et al. 2008). Therefore it is also difficult to distinguish between surface adhesion forces and forces stemming from friction. Furthermore, due to the bendability of the stamp, demolding by delamination is possible, i.e. by creation of a gap on one side of the stamp and a local transition of the gap from one substrate side to the other. Then, similar to crack propagation, part of the structures is demolded while others are still in intimate contact with the stamp topography. Due to this, large area demolding becomes possible with the consequence that roll embossing is not different from the continuous demolding of a bent stamp from an extended large area substrate. The main difference, however, is the possibility to control the relaxation of material after cooling, while in roll-to-roll, molding and demolding are part of a dynamic process.

3 Tool and machine concepts in NIL and LiGA

3.1 Hard and soft tool concepts

Imprint is a highly dynamic process where the vertical sinking movement of a stamp is transformed into a 3-D flow with large lateral flow components. In thermal NIL the speed of the pressure buildup at the back side of substrate and stamp and equilibration of local inhomogeneities over a large surface can influence the mode of cavity filling. This determines how the stamp will bend during sinking. To cope with these inhomogeneities, different machine and tool concepts were developed which involve hard and soft elements. Two typical machine types are presented in Table 2.

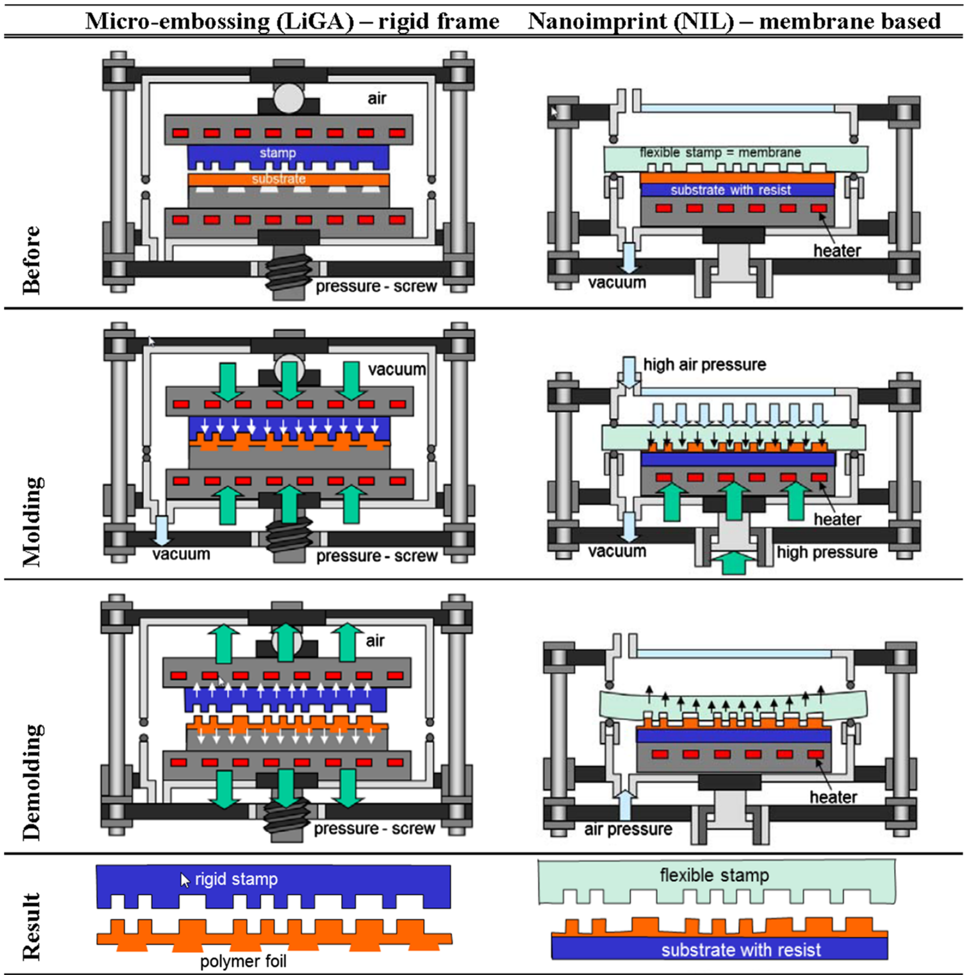

Table 2 (a) Embossing machine with rigid frame for time/distance controlled molding and demolding of microstructures, (b) imprint machine with flexible stamp and compliance membrane for pressure controlled molding of nanostructures

3.2 Nickel and silicon: material choice for stamps

While LiGA has found electroplating of Nickel (Ni) and its alloys to be the right choice for fabricating metal molds with high mechanical and thermal stability, NIL is still relying on crystalline or glass-like materials like silicon (Si) and fused silica (quartz, SiO_2). Brittle stamps are prone to breaking, particularly because of their susceptibility to small defects and notching. Although silicon exhibits the same Young's modulus as steel (Si used as micromechanical elements such as springs in watches), it therefore is not the first material of choice for a mold in high throughput manufacturing. However, it has some striking assets:

- It can be patterned by standard cleanroom based micromachining techniques, such as etching and lift-off down to the nm range. Etching of metals in RIE processes is either much more isotropic or performed with Argon ions (i.e. purely physical etching). In contrast to this, despite of the nanocrystalline property of Ni molds, electroplating results in reproducible copies of the surface topography with sub-5 nm resolution, similar to molding.
- Silicon can be easily modified by silane chemistry, i.e. a dense assembly of nm-long functional molecules on top of the oxidized Si surface, which barely modify its topography. By using fluorinated silanes, this can be used for over 1,000 imprint cycles before the surface has to be recoated. For Ni, related chemistries such as fluorinated alkyl phosphates or phosphonates can be applied. However, in contrast to silanes on SiO_2, they do not enable a covalent binding. Furthermore NiO is known to be much less defined than SiO_2, and therefore these coatings are proven to be less stable. Hybrid solutions, such as a SiO_2 coating by PECVD (ca. 15 nm thick) on top of a freshly prepared Ni surface, enable silane chemistry but represent a significant alteration of the surface profile of nanostructures.
- Due to the need to pattern thin polymer films on Si substrates, the thermal expansion of stamp and

substrate needs to be similar, while the film on top of the substrate adapts to the lateral expansion or contraction of the substrate upon heating and cooling. Therefore, in similar way than by imprint of polymer plates with metal stamps, imprint of resists using Ni shims is not so much a problem because of the thermal expansion mismatch of metal and polymer, but of the stamp and the substrate material.

3.3 Machine concepts for molding and demolding

Hot embossing of high aspect ratio patterning in LiGA requires a high degree of stiffness of both microstructures and tools, enabling a defined unidirectional movement in vertical direction without lateral shear both for pressing (molding) and retraction (demolding). The only element with intended flexibility and possibility to damp the sinking of the mold is the polymer material in its visco-elastic or viscous state to be molded between stamp and bottom plate. Therefore embossing machines were originally built by integrating a heating and cooling device into the rigid load frame of a material testing machine (realized in the Jenoptik HEX concept). It enabled precise positioning within µm resolution based on screw movement, and embossing cycles with controlled pressure and position. This is in contrast to machines for wafer anodic bonding, where two hard (wafer) substrates are pressed together without any movement wafers (a concept favored by EV-Group and SÜSS), while the bonding is induced between the two surfaces applying heat and high electrical voltage. Due to the non-ideal flatness up to a few 10 s of µm over the two wafer surfaces, thin compliance layers behind the wafers enable an intimate conformal contact by bending, which has to be maintained over some minutes. Additionally to this, in NIL the pressing mechanism has to compensate for the vertical movement of the stamp due to the squeeze flow and mold cavity filling, both globally (over the entire wafer surface) and locally (over a few mm). Both the global movement of up to a few 100 nm and the compensation of local height variations of a few tens of nm are easy to implement with a compliance-type mechanism. Both stamp and compliance layer are therefore considered as membranes with ability to generate a conformal contact by bending. A purely pressure driven setup is therefore nearer to thermal NIL and therefore bonders with appropriate compliance layers have been favored by many researchers in semiconductor industry, which was well adapted to other cleanroom equipment, particularly mask aligners with their alignment capabilities. In presses with a stiff mechanism based on hydraulic, air, and screw driven hard stampers, the buildup of the whole stack includes the use of an elastic compliance layer, e.g., flexible graphite, rubber, or Teflon, of about 1–2 mm thickness, which is needed for the surface equilibration due to the lack of flatness of common substrates. Other concepts use an air-pressurized membrane as a soft cushion, which equilibrates local pressure variations during the sinking of the stamp in a more controlled way, or even flexible stamps used as a membrane. For example, while a pressurized membrane (made from 50 to 200 µm thick Al or polymer foil) will equilibrate local pressure variations due to stamp sinking within a fraction of a second, an elastic element, e.g., a 1 mm thick rubber layer, will build up pressure with a short delay when compressed with a screw-driven press. In contrast to this, the constant speed for compression and demolding, in the Jenoptik HEX 03, 0.2 mm/min (i.e. 3 µm/s), can be used for continuous molding of microstructures with several 100 µm of height when the rigid stamp is attached to one of the press stampers. Because of this difference in ability to perform micro- and nm movements in a controlled way, not all presses are equally suitable for the molding of micro- and nanostructures, or micro-embossing and NIL, but can be adapted to do both cases reasonably well. In NIL, the stamp as a whole, however, has to be flexible that it can accommodate surface undulations and the pressure inhomogeneities by structure density variations. In (Schift 2008), further machine concepts are presented. This includes presses suitable for stamps of a few mm^2 or cm^2 only. Because of this small size they are considered as rigid (bending is limited), a spring-based setup equilibration, e.g. for wedge compensation, can be used. Here, large surface patterning is achieved using segmented imprints, e.g. by a step and repeat approach.

4 Combinations of LiGA and NIL

LiGA has found major applications in optics, fluidics, mechanics and combinations of them with sizes often larger than 10 µm, and even larger structures for interface. Vertical sidewalls with perfect straightness and surface quality down to sub-10 nm are essential for prisms and cylinder lenses, and gears in a purely 2½ dimensional shadow exposure setup. NIL is seen mostly in areas of sub-wavelength, molecular-scale applications, where traditional high end lithographies are not available or too expensive. The latter is often the case in large area surface patterning with dense nanostructures for structures below 100 nm. Due to its proneness to stamp bending, patterning of regular large surface structures with low requirements on alignment between layers is most promising. Particularly interesting are cases, where the surface decoration can be “decoupled” from the micromold fabrication process. Two main combinations can be seen: (1) the most common case is the direct combination of micro- and nanostructures in the same mold, e.g. by interconnects between microfluidic and nanofluidic channels. Here, hybrid approaches using mix- and match of different lithographic methods are needed. Particularly difficult is if structures have to be connected, e.g. by smooth (tapers) or abrupt (steps) transitions. Once a mold is created, it can be treated like a normal LiGA mold, particularly if no pattern transfer is required. A typical example is the combination of micro- and nanofluidic channels, e.g. as needed in DNA sequencing analysis (Fernandez-Cuesta et al. 2011). Another example is the fabrication of combined processes such as NIL and grayscale lithography (Schleunitz et al. 2011). (2) The combination of different functionalities, e.g. a micro-element or device with specific outlines which is decorated with nanostructures on one or several of its surfaces. Also here the aim is to create a mold which exhibits both micro- and nanostructures. But in contrast to (1), the surface of the microstructure needs to be modified by the nanostructures, e.g. by etching the surface, by thermoforming a nanostructure into the microcavities, or simply by generating a hybrid mold where the two molds are assembled in a single mold insert. A specific advantage of mold making is given if the nanostructured surface is, e.g. on the top or the bottom of a 2½-D structure. Then it can be added to molded structure by a mix- and match process or simply by attaching it to different sides of the molding tool. In all cases, special care has to be taken that the nanostructures are well replicated and demolded, particularly when they are decorating sidewalls of microstructure. Examples for applications where the fabrication is hybrid molds is needed are gears with integrated holograms as anti-counterfeit labels or simply the decoration of large microstructures with nanostructures, e.g. by manufacturing structures in the µm-regime and add functionality possible by patterning. A good example of a combined mold is the fiber interconnect elements with diffractive optical elements on the surface and mechanical registration e.g. by the outlines of a micro-element (Schift and Söchtig 1998). This is a prerequisite for the coupling of light from optical fibers into integrated optics or manipulation of light transmission between entire fiber connectors. An example for a hybrid mold, in which the micro-mold and surface decoration mold are attached to two opposite sides of an injection molding tool is the fabrication of micro-cantilevers with surface decoration. They may serve for cell force measurement, if its surface can be patterned with linear gratings suitable for cell attachment (Urwyler et al. 2011). Finally, a microfluidic channel with small high aspect ratio micro- or nanopillars becomes hydrophobic (instead of applying hydrophobic chemical coatings) and can be used for fuel cells where liquid inputs and gaseous outputs have to be separated by localized anti-wetting properties (Senn et al. 2011). This can be done by thermoforming of nanostructured foils into micromolds. In this contribution, two examples of combining molds are shortly presented in Fig. 3. In the LiGA fabricated micro-element the lenses are integrated into the mold by clamping two electroplated LiGA and a grayscale lithography fabricated molds together (Schift and Söchtig 1998). The lenses of the eight fibers are aligned with respect to a MT fiber connector by clamps sitting on 700 µm steel rods (here only for demonstration) (Schift et al. 2013). In the polymer micro-cantilevers in polypropylene the linear grating was created by attaching a replicated polymer stamp at the mirror side of a molding tool opposite to the mold with the cantilever cavity (Urwyler et al. 2011).

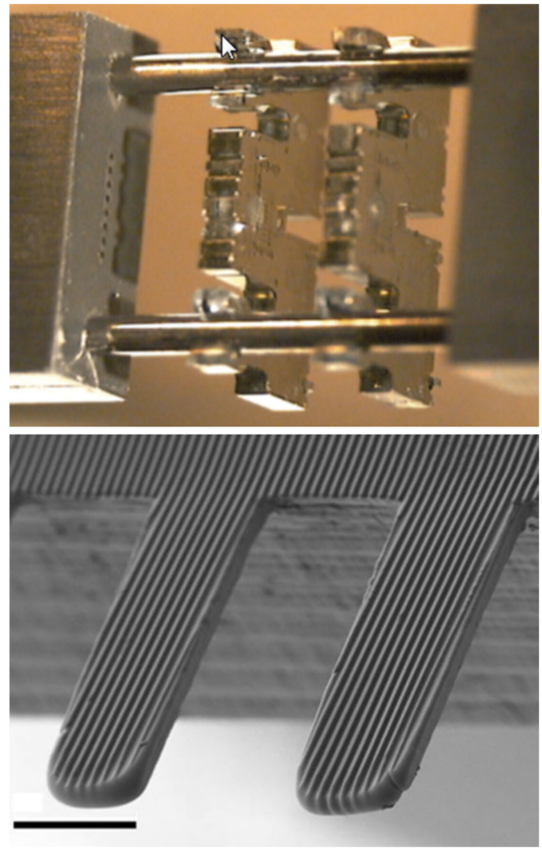

Fig. 3 Examples for combined surface patterns with micromechanical elements by using hybrid mold approaches in polymer injection molding: (*top*) LiGA fabricated micro-element with spring structures sitting on 700 µm steel rods with surface diffractive optical elements (pitch of fibers 250 µm). *Bottom* polymer micro-cantilevers in polypropylene (dimensions $l \times w \times t = 500 \times 80 \times 35$ µm) with linear grating with a period of 10 µm. Reprinted with permission from Figs. 3 and 4 in ref. Schift et al. (2013). Copyright 2013, AVS Science and Technology of Materials, Interfaces, and Processing

5 Conclusion

Is NIL different or similar to LiGA? From a physical and technical point of view, the differences are marginal. The physics is the same down to a few nm, but the concepts of thinking are different. It is not the same if engineers simply want to downscale existing processes, starting with precision mechanics and laser micromachining and always competing against the easiness to implement these techniques without mask design and exposure techniques. In the molding step of LiGA, reproducibility, e.g. demolding problems due to small deviations from vertical sidewalls, is often bound on design and sidewall quality. Vertical sidewalls with perfect straightness and surface quality down to sub-10 nm are essential for prisms and cylinder lenses, and gears in a purely 2½-D shadow exposure. NIL is seen mostly in areas of sub-wavelength, molecular-scale applications, where traditional high end lithographies are not available or too expensive. The latter is often the case in large area surface patterning with dense nanostructures for structures below 100 nm. In the future, more and more applications will need both qualities and dimensions. There is a strong overlap with LiGA and already existing process routes (in R2R and Compact Disc injection molding) where metal molds are used for production. The highest promise is given by application where a hybrid mold is fabricated, which can be done by mix- and match processing or electroplating. Apart from the mold fabrication, however, also molding processes have to be adapted to fit both the requirements of high aspect ratio molding and nanoscale surface replication. This is particularly difficult if high aspect ratio micro- or nanostructures have to be molded.

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