

# Nanoimprint Lithography

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## 1. Introduction

The Nanoimprint lithography (NIL) is a novel method of fabricating micro/nanometer scale patterns with low cost, high throughput and high resolution (Chou et al., 1996). Unlike traditionally optical lithographic approaches, which create pattern through the use of photons or electrons to modify the chemical and physical properties of the resist, NIL relies on direct mechanical deformation of the resist and can therefore achieve resolutions beyond the limitations set by light diffraction or beam scattering that are encountered in conventional lithographic techniques (Guo, 2007). The resolution of NIL mainly depends on the minimum template feature size that can be fabricated. Compare with optical lithography and next generation lithography (NGL), the difference in principles makes NIL capable of producing sub-10 nm features over a large area with a high throughput and low cost (Chou et al., 1997). Therefore, the charm of NIL largely comes from its capability for patterning with high resolution, high fidelity, high throughput, and low cost. In addition, nanometer sized patterns can easily be formed on various substrates, e.g., silicon wafers, glass plates, flexible polymer films, and even nonplanar substrates. The process has been added to the International Technology Roadmap for Semiconductors (ITRS) for the 32 and 22 nm nodes. Toshiba, moreover, has validated it for 22 nm and beyond. What is more significant is that NIL is the first sub-30 nm lithography to be validated by an industrial user (Yoneda et al., 1997).

Nanoimprint lithography was first invented by Chou and his students in 1995 as a low-cost and high throughput alternative to photolithography and e-beam lithography (EBL) for researchers who need high resolution patterning, motivated by the high expense and limited resolution of optical lithography. Due to historical reasons, the term NIL initially refers to a hot embossing lithography (HEL) process, and was also used as a synonym for thermal NIL (Chou et al., 1995). However, NIL has now an extended meaning which includes not only two fundamental types (Hot Embossing Lithography and UV-based Nanoimprint Lithography, UV-NIL) but also many different variations developed such as roll imprint process, laser-assisted direct imprint, reverse imprint lithography, substrate conformal imprint lithography, ultrasonic NIL, etc. Compared to other lithography processes and next generation lithography with nanoscale resolution, such as e-beam lithography and extreme ultraviolet lithography (EUVL), the most prominent advantage of NIL is its ability to pattern 3D and large-area structures from micron to nanometer scale and

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its potential to do so at a high throughput and low cost. This paves the way for many applications in the area of data storage, nano-optoelectronic, optical elements, NEMS and NOEMS, etc (Balla et al., 2008). Furthermore, NIL has currently demonstrated great potential and commercial prospects in many application fields, such as Hard Disk Drive (HDD), LED, OLED, flexible display, optical and biological devices, etc. After more than ten years of NIL process development, a range of standard tools and materials is available from various industrial providers.

NIL technology involves two fundamental aspects: the basic research and the application research, as shown in Fig.1. The basic research consists of the process, tool, template (mold), material (resist, functional material, etc) which aim to meet the different application requirements, namely the micro/nano structures or devices fabrication. NIL applications mainly cover nanoelectronics, nano-optoelectronics, nanophotonic, nano-biology, optical components, etc.

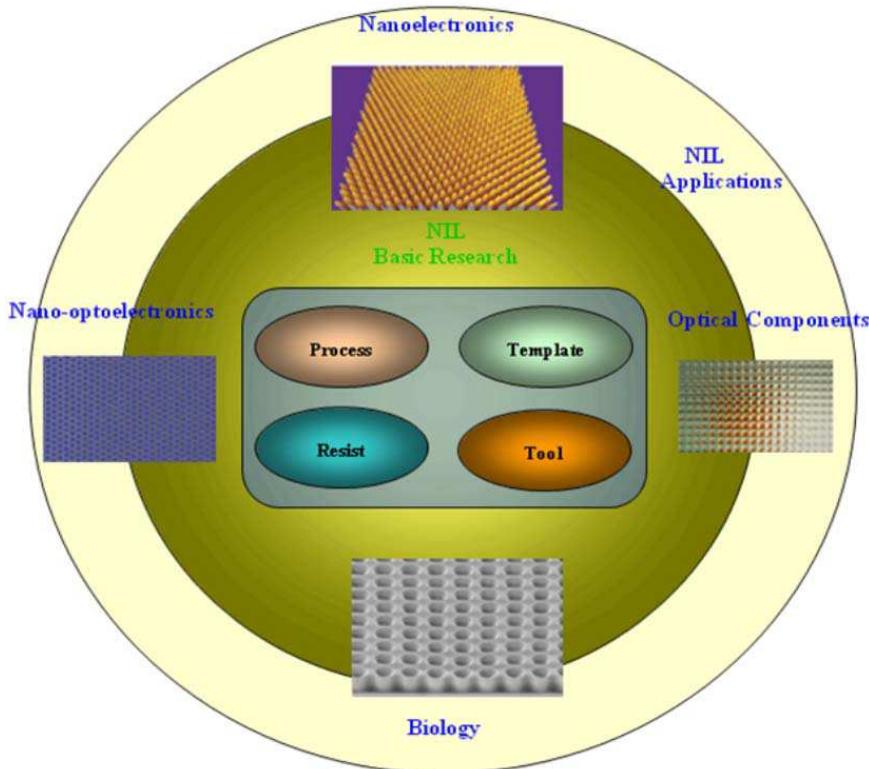


Fig. 1. Overview of the NIL

The chapter systematically presents the NIL technique and various applications. Moreover, some key issues and recent progresses for NIL are discussed in detail. Finally, the prospect and challenges in NIL are also addressed. The aim of this chapter is to make readers to systematically and deeply understand NIL, and better apply the new technique.

## 2. NIL process

### 2.1 Principle of NIL

NIL is based on the principle of mechanically modifying a thin polymer film (mechanical deformation of the resist) using a template (mold, stamp) containing the micro/nanopattern, in a thermo-mechanical or UV curing process. In other words, NIL uses the direct contact between the mold (template) and the thermoplastic or UV-curable resist to imprint (or replicate) the pattern, unlike optical lithography, does not require expensive and complex optics and light sources for creating images. The switch from using light to using contact to pattern brings some advantages. For instance, it can therefore achieve resolutions beyond the limitations set by light diffraction or beam scattering that are encountered in conventional techniques, simplifies process and largely reduces cost. However, that will also indeed bring new challenges and issues, the most important of which are alignment and the 1x mask/template fabrication. Since NIL can be considered as such a process based on squeeze flow of a sandwiched viscoelastic material between a mold and a substrate, the property of interface between the two materials has to be considered throughout the entire process, both from topographical, chemical, and mechanical points of view. Furthermore, the characteristics of the interface and surface have a great impact on the demolding capability and filling behavior which can strongly influenced pattern quality and throughput (Schiift, 2008, Bhushan, 2007, Guo, 2004).

The patterned polymer can even act as a functional device, e.g. lens for imaging sensors, micro fluidic chip, biomedical array etc. It can also be used as a high resolution mask for subsequent steps of the process (metal deposition, electroplating, etching and lift-off process). Moreover, various substrates, including silicon wafers, glass plates, flexible polymer films, polyethylene terephthalate (PET) polymer film, and even nonplanar substrates can be utilized for NIL (Costner et al., 2009). The ultimate resolution of the patterns fabricated by NIL is primarily determined by the resolution of the features on the surface of the mold. Because of the 1X nature of NIL compared with 4 X for photolithography, the 1X template must be more accurate than conventional masks.

As a result, distinct features for NIL involve two points: (1) the contact nature of the process; (2) direct mechanical deformation of the resist. Two crucial steps, namely the resist filling rheology behavior and demold capabilities, have decisive influence on transferred pattern quality and throughput for NIL. The particular advantage of NIL compared to other lithography techniques and NGL is the ability to fabricate large-area and complex three-dimensional (3D) micro/nanostructures with low cost and high throughput.

### 2.2 Two fundamental processes for NIL

Currently, there are a great variety of NIL process types, but two of them are most important and fundamental: Hot Embossing Lithography (HEL) or thermal nanoimprint lithography (T-NIL), UV-based Nanoimprint Lithography (UV-NIL), as shown in Fig. 2 (Steward & Willson, 2005). Both thermal and UV-NIL have demonstrated a sub-10 nm resolution.

T-NIL is the earliest NIL developed by Stephen Chou's group. In a standard T-NIL process, a thin layer of imprint resist (thermoplastic polymer) is spin-coated onto the substrate. Then the mold, which has predefined topological patterns, is brought into contact with the substrate and they are pressed together under certain pressure. When heated up above the glass transition temperature ( $T_g$ ) of the polymer, the feature pattern on the mold is pressed into the melt polymer film. After being cooled down, the mold is separated from the

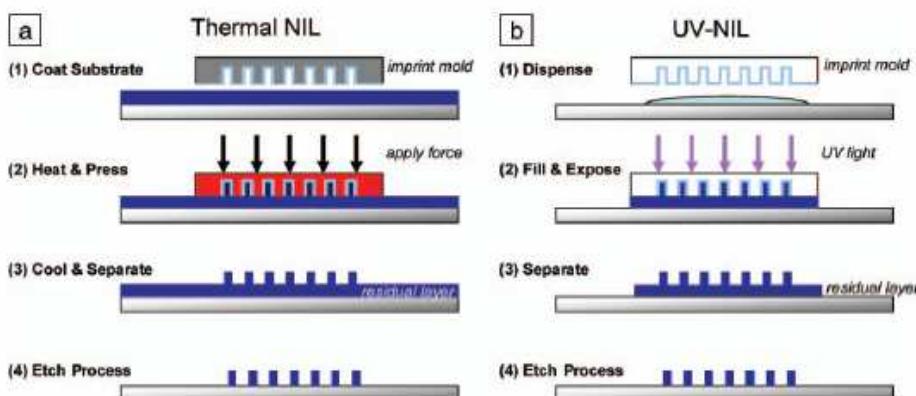


Fig. 2. Two fundamental process types for NIL (Steward & Willson, 2005)

substrate and the pattern resist is left on the substrate. A subsequent pattern transfer process (e.g. reactive ion etching) can be used to transfer the pattern in the resist to the underneath substrate. In UV-NIL, a UV-curable liquid photopolymer instead of thermoplastic as resist is applied to the substrate and the mold is normally made of transparent material like fused silica, quartz mold. After the mold and the substrate are pressed together and the cavities (trenches) are fully filled by resist, then the resist is cured in UV light and becomes solid. After demolding, a similar pattern transfer process can be used to transfer the pattern in resist onto the underneath material. The polymer residual layer is removed (Nanoimprint lithography, 2009). The basic difference between UV-NIL and T-NIL is that a resin, which is liquid at room temperature, is shaped by a moderate pressure, which is then crosslinked and hardened by curing. Each process has of its own prominent advantages, e.g. while UV-NIL can be performed at room temperature and low pressure, hot embossing is low-cost since nontransparent molds can be used (Less restrictions on mold). Schift and Kristensen provide a comparison of T-NIL and UV-NIL, with typical parameters of current processes (Bhushan, 2007). However, UV-NIL has established itself as a promising alternative to NIL in which imprint lithography is conducted at room temperature under low pressure conditions. UV-NIL is one of the most important NIL technologies for structuring of large wafer areas up to 300 mm in diameter. The process offers several decisive technical advantages concerning overlay alignment accuracy, simultaneous imprinting of micro- and nanostructures and tool design due to the absence of high imprint pressures and thermal heating cycles (Fuchs et al., 2008, Bender et al., 2006).

UV-NIL offers two approaches for patterning using either rigid quartz glass molds (Hard UV-NIL) or soft molds (Soft UV-NIL) for structuring of UV sensitive resists resulting in an etching mask for the substrate to be patterned (Glinsner et al., 2007, Plachetka et al., 2004). For UV-NIL using a rigid mold, the hard mold brings about two weaknesses, one is the sticking characteristic which can lead to the following shortcomings that a release agent or surfactant is necessary, and the demolding force is especially large. Another is the limitation in imprint area due to having the surface waviness onto the mold and the substrate surfaces. Furthermore, it is rather difficult to ensure uniform and parallel surface contact between a template and a wafer during imprinting process. Compared with the hard mold, using a soft or flexible mold can avoid the conglutinating of resist, acquire high precision feature,

enlarge the pattern transferring area, reduce the parallelism error between the mold and the substrate and lengthen the life-time of the master. In particular, high flexibility of the mold enabled conformal contact as well as imprinting at significantly reduced imprint pressure. However, the elastomeric behavior of the soft mold has both, positive and negative attributes. On one side it offers numerous advantages, but on the other hand some defects such as resolution limitations and non-uniformity of the transferred patterns, etc., will have to be considered and resolved. In addition, Swelling is a commonplace issue with PDMS based molds since most organic liquids will swell PDMS. In the case of flexible mold local deformations limit the resolution of soft UV-NIL principally. Therefore, compared to HEL and UV-NIL used rigid molds, it is particularly important for the soft UV-NIL to understand and reduce as much as possible the mold deformation for the practical application of the technique. The current capability for the process from AMO and Süss can ensure nanoscale resolution down to sub 50 nm and perfect pattern stability (SCIL, 2009). However, the deformations of the soft mold during imprinting process which can cause serious consequences have to be considered for the practical application of the process and further improving the pattern resolution (Lan et al., 2009).

The Jet and Flash™ Imprint Lithography (J-FIL®) process, formerly called Step and Flash® Imprint Lithography, (S-FIL®) developed at the University of Texas at Austin, is a typical and fundamental UV-NIL process. The fused silica surface, coated with a release layer, is gently pressed into a thin layer of low-viscosity resist. The resist is deposited in a customized pattern matching the template using the IntelliJet™ Drop Pattern Generator. When illuminated by a UV lamp, the surface is polymerized into a solid layer. Upon separation of the fused silica template, the pattern is left on the substrate surface. A residual layer of polymer between features is removed by an etch process, and a perfect replica of the pattern is ready to be used in subsequent processing for etch or deposition. Fig.3. demonstrated the Jet and Flash™ imprint lithography process. Sub-10nm features have been made that exceed the present requirements outlined in the ITRS, as well as most patterned media roadmaps (J-FIL, 2009).

In addition, the NIL process can also be divided into two broad categories: single-step imprinting and multi-step imprinting based on the size of a template and imprinting time. The former is to imprint resist on a wafer using a wafer-sized template at a time, whereas the latter is to use a chip-sized template by the step and repeat process. For the single-step NIL, the wafer-sized template consists of multiple groups of chip-sized patterns that are uniformly distributed with the equal in between space. According to the layer number of an imprinting pattern, it can be further divided into the monolayer patterning process (single level) and multilayer patterning process (multilevel). High-resolution overlay is considered to be an important challenge for multilayer imprinting processes. Therefore, there are four types of corresponding imprinting processes: (i) single-step and monolayer, (ii) single-step and multi-level, (iii) multi-step and monolayer, (iv) multi-step and multi-level (a real commercialization process). The current NIL machines mainly involve first these forms of (i), (ii) and (iii).

### 2.3 Variants of NIL

In the recent years, a variety of new processes have been proposed and investigated, such as reverse NIL, soft UV-NIL, Laser assisted direct imprint (LADI), Sub-10 nm NIL, chemical nanoimprint, electrical field-assisted NIL, etc., which aim to implement the micro/nano structures fabrication with large area, 3-Dimension, high throughput, high resolution, free

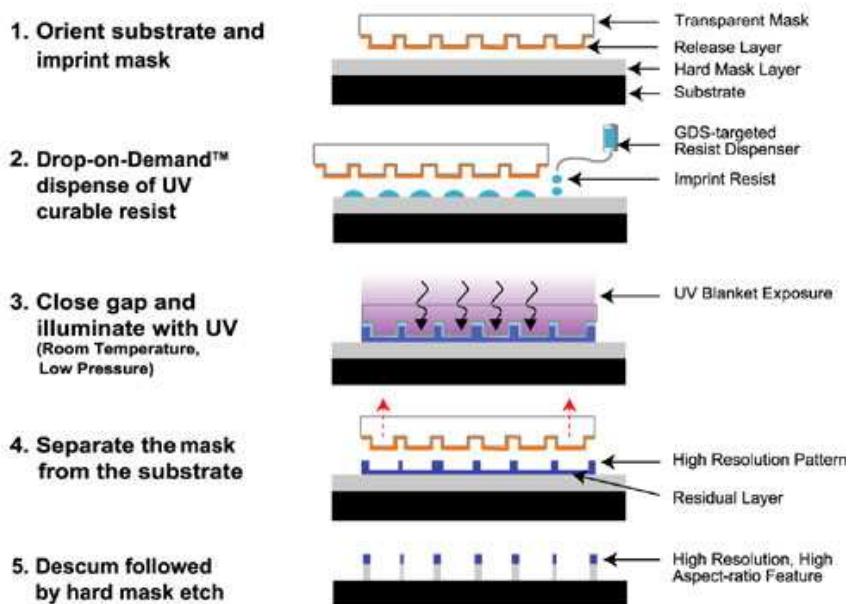


Fig. 3. Jet and Flash™ imprint lithography process (J-FIL, 2009)

defect, and to directly make various functional structures, as well as to improve the throughput and pattern quality.

### 2.3.1 Combined thermal and UV-NIL

STU® (Simultaneous Thermal and UV) technology from Obducat enables simultaneously combined thermal and UV-NIL, as shown in Fig.4, allowing the complete imprint sequence into UV-curable thermoplastic pre-polymers to be performed at a constant temperature. By using the unique STU® technology, problems related to thermal expansion mismatch between stamp and substrate are avoided. The method allows the use of spin-coated UV-curable polymers with a homogeneous thickness distribution on wafer scale, crucial for CD control and enabling pattern transfer to an underlying substrate. Obducat has proved the ability to imprint 17nm features with its proprietary IPS (intermediate polymer stamp)-STU process. Furthermore, 17 nm dots have been printed uniformly with a residual layer below 7 nm (Key Technologies, 2009).

### 2.3.2 Reverse imprint process

For the reverse NIL process, a polymer film is firstly spin-coated onto the mold (rather than substrate), the polymer will fill up the trench regions of the surface relief patterns. This means that a replica of the mold pattern is formed in the polymer film simply by spin coating. Subsequent, this film can be transferred from the mold to a substrate, patterned structures are obtained. The key to the successful film transfer lies in the fact that the mold has a lower surface energy than does the substrate, and so the polymer film has better adhesion to the substrate and therefore can be detached from the mold. The process has the ability to construct the three-dimensional and multilayer micro/nanostructures.

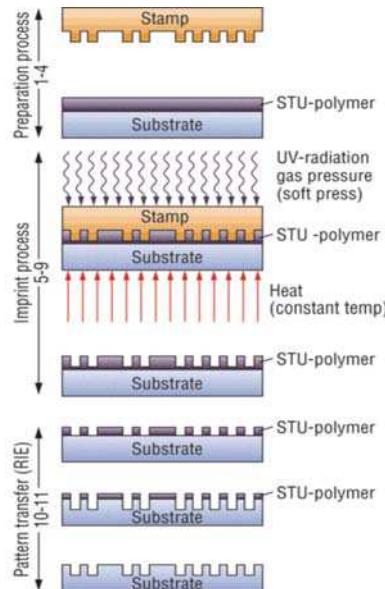


Fig. 4. A simultaneous thermal and UV (SUV®) imprint process (Key Technologies, 2009)

Furthermore, the crucial advantage of this technique is the possibility to construct three-dimensional device-like structures without having to etch polymer residual layer at any intermediate step. Some devices such as three-dimensional photonic crystals, multi-layered nano-channels, polymer optical devices, gold gratings (metallic nanostructures) have been fabricated using the reverse NIL process or the combination of the reverse imprint process and other micro-fabrication technologies (Guo, 2004, Kehagias et al., 2006, Han et al., 2007). Fig.5 shows the schematics of a reverse UV contact NIL for 3D Nanofabrication (Kehagias et

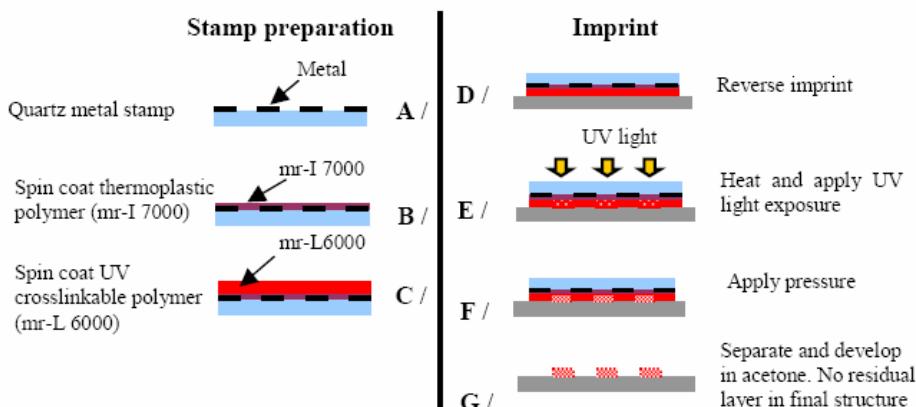


Fig. 5. Reverse UV contact NIL process (Kehagias et al., 2006)

al., 2006). Combined the UV-curable reverse NIL process with a water soluble PVA (polyvinyl alcohol) based removable template and home made UV-curable glue, Lee's team implement the successful fabrication of multi-stacked 2D nano patterned slabs on various substrates including flexible polymer film. The highlight for the process is to develop a PVA mold which be dissolved by water (Han et al., 2007).

### 2.3.3 Laser-assisted direct imprint

Laser assisted direct imprint (LADI) is a rapid technique for patterning nanostructures that does not require etching. LADI is based on the following principle: a single excimer laser pulse melts a thin surface layer of the functional materials, and a mold is embossed into the resulting liquid layer, as shown in Fig. 6. It has been used for making nanostructures in silicon and metals with a resolution better than 10 nm. LADI offers direct patterning without etching for compound semiconductors which are hard to be etched. Using this method, applicants have directly imprinted into silicon large area patterns with sub-10 nanometer resolution in sub-250 nanosecond processing time. The method can also be used with a flat molding surface to planarize the substrate. The high resolution and speed of LADI could open up a variety of applications and be extended to other materials and processing techniques (Chou al., 2002).

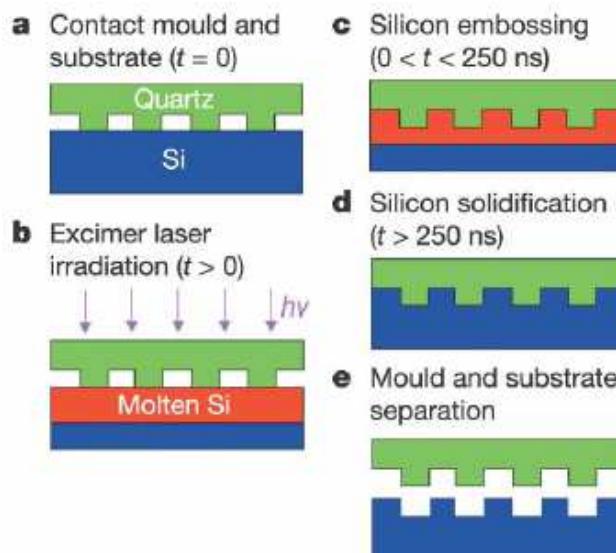


Fig. 6. Laser assisted direct imprint (LADI) (Chou al., 2002)

### 2.3.4 Roll imprint process

For conventional NIL processes, one of the most important problem is that it cannot significantly improve the throughput in the patterning of large area product with low cost because it is not a continuous process. To overcome this limitation, roller-type nanoimprint lithography (RNIL) has been developed and is becoming the most potential manufacturing method for industrialization of nanoimprinting process, due to its prominent advantage of continuous process, simple system construction, high-throughput, low-cost and low energy

consuming. Compared to other NIL processes, the unique advantage for the RNIL is only a continuous process with a high throughput to fabricate the large-area patterns. The RNIL involves three essential steps: deposition, patterning and packaging (Fig. 7 (II)). Two molds (roller mold and flat mold, Fig. 7 (III)) and two substrates (flexible substrate and rigid substrate, Fig. 7 (I)) can be used for the RNIL (Lan et al., 2008, Ahn et al., 2006, Ahn & Guo, 2008, Youn et al., 2008, Kao et al., 2005, Lee et al., 2008, Chang et al., 2006). Lan et al., have presented a general literature review on the RNIL (Lan et al., 2008). Guo et al., demonstrated a Roll-to-Roll NIL process in which polymer patterns down to 70nm feature size were continuously imprinted on a flexible web (Ahn & Guo, 2008). A thermal roller imprint lithography (RIL) system was developed and applied to RIL tests to evaluate its feasibility for the large area replication of an optical micro device. The system has the capacity to replicate ultra-precision structures on an area of 100mm×100mm at the scanning speed range of 0.1–10 mm/s. A light guide plate (LGP) for a back light panel was fabricated. The system is suitable for the fabrication of various optical micro devices such as flat panel displays, electronic papers, functional films, and others (Youn et al., 2008). A combination method of the roller-type imprinting lithography and photolithography (CRIP), followed by wet chemical etching was used to fabricate the patterned organic light emitting devices (OLEDs) with pixels of 500 μm × 300 μm on the flexible PET substrates. Compared with the conventional imprint lithography or photolithography, CRIP using the hybrid mold has the advantages of better uniformity, less force, less time-consuming, lower cost and higher aspect ratio. This technique is potentially cost-effective, offers high throughput, less time-consuming and is suitable for fabrication on flexible substrate (Chang et al., 2006).

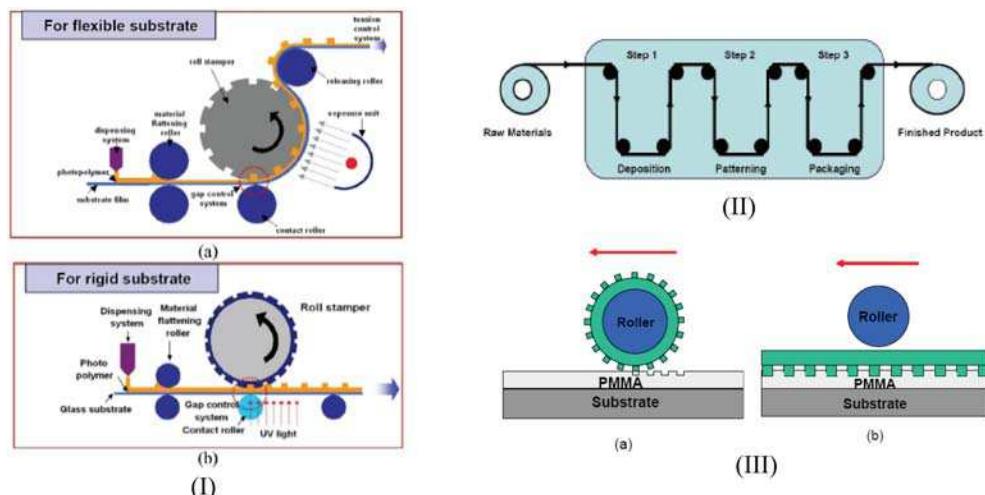


Fig. 7. Roller-type Nanoimprint Lithography (Lan et al., 2008)

### 2.3.5 Large area imprint

Large area imprint over the full wafer surface is needed often in single layer imprinting when high throughput is needed. In a full wafer nanoimprint scheme, all the patterns are contained in a single nanoimprint field and will be transferred in a single imprint step. This allows a high throughput and uniformity. To ensure the pressure and pattern uniformities

of full wafer nanoimprint processes and prolong the mold lifetime, a pressing method utilizing isotropic fluid pressure, named Air Cushion Press (ACP) by its inventors, was developed and being used by commercial nanoimprint systems. ACP applied air pressure for conformal contact and imprint, therefore achieved ultra-uniformity over the whole imprint field. It has advantages over traditional parallel plate press: capable of handling the substrate with uneven back and patterning on curved surface. Presently, Nanonex provides three series of NIL tools (NX-1000, NX-2000, and NX-3000) for T-NIL and P-NIL, with and without alignment. All of them use the ACP to achieve excellent pattern uniformity (Tan et al., 2004, Gao et al., 2006). Pelzer et al., studied full wafer replication of nanometer features, and presented results on full wafer imprints up to 200mm with high-resolution patterns for microelectronic applications. There are no physical limitations encountered with imprinting techniques for fully replicated structures, in the sub-10nm range. The real challenge for the technique is its utilization for dense structured full wafer imprints up to 200mm (Pelzer et al., 2005).

The step and repeat process is another approach to pattern on large areas (e.g. 300 mm wafers scale). The imprint field (die) is typically much smaller than the full wafer nanoimprint field. The die is repeatedly imprinted to the substrate with certain step size. This scheme is good for nanoimprint mold creation. It is currently limited by the throughput, alignment and street width issues (Jeong et al., 2005).

### **2.3.6 Substrate conformal imprint lithography**

SCIL (Substrate conformal imprint lithography), that was developed based on a cooperation between Philips Research and SUSS MicroTec in 2008, is an enabling technology offering large-area soft stamps with repeatable sub-50nm printing capability, while avoiding stamp deformation as no contact force is applied, non-UV based curing at room temperature and allowing high aspect ratios even up to 1:5 and more. The new SCIL technology has been designed for sub-50nm patterning and is bridging the gap between small rigid stamp application for best resolution and large-area soft stamp usage with the usual limited printing resolution below 200nm. Süss believes that the SCIL represents an enabling new technology that paves the way for further commercialization of NIL (SCIL, 2006).

### **2.3.7 Nanoelectrode lithography**

Nanoelectrode lithography, which is a pattern duplication method that combines nanoimprint with an electrochemical reaction. The conductive mold pattern undergoes an electrochemical reaction that enables an oxide pattern to be fabricated directly on the surface of a semiconductor or metal layer. Since this technique transfers the mold pattern to a target surface chemically, it is categorized as chemical nanoimprint, while conventional nanoimprint physically transfers a mold pattern having peaks and valleys to the target (Fig.8). This patterning phenomenon gives nanoelectrode lithography some advantages such as resistless patterning and multiple patterning, which will improve the accuracy and flexibility of nanoimprint (Yokoo & Namatsu, 2009).

### **2.3.8 Hybird NIL process**

Mix-and-match approaches are used to combine the advantages of two or more lithographic processes or simply to avoid their mutual disadvantages. This is also a way to improve throughput and reliability, e.g. since the fabrication of large-area nanostructures is often costly, the definition of microstructures can be done with optical lithography, while the

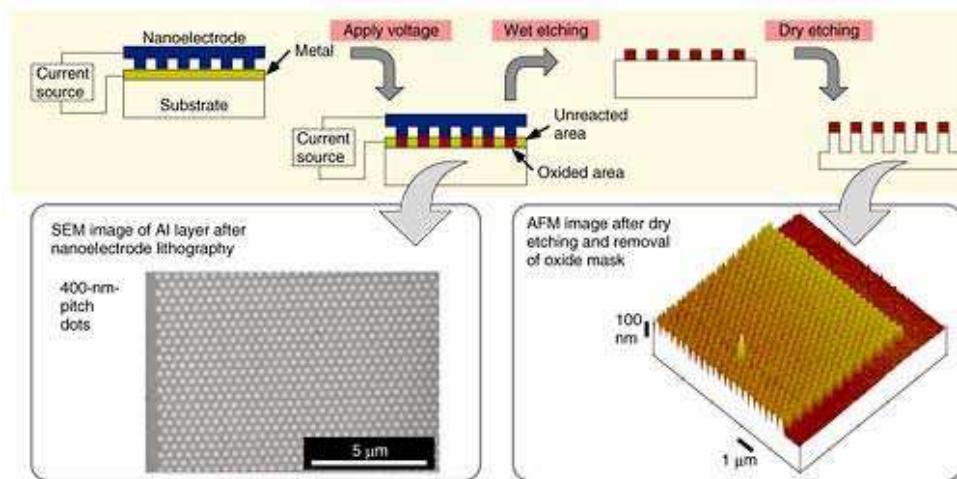


Fig. 8. Chemical nanoimprint (nanoelectrode lithography) (Yokoo & Namatsu, 2009)

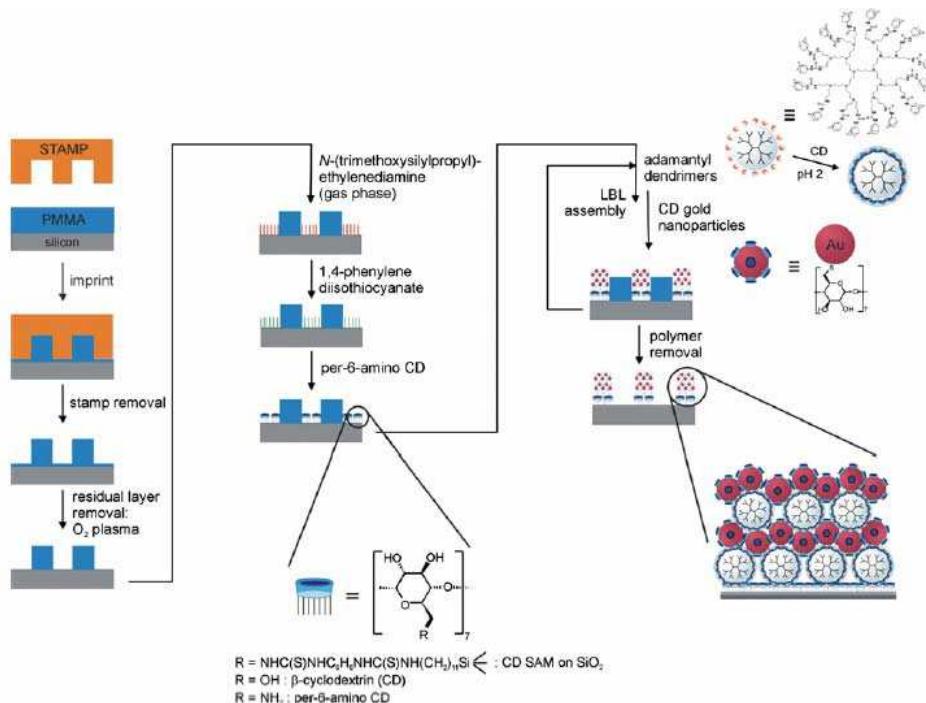


Fig. 9. Integrated nanofabrication scheme on nanoimprinted pattern (Plachetka et al., 2008)

nanopatterning of critical structures in small areas can be done by NIL. An integrated process combining top-down nanoimprint lithography and bottom-up layer-by-layer (LBL) self-assembly was applied to the fabrication of 3D hybrid nanostructures, as shown in Fig.9.

The method can implement the fabrication of 3D nanoobjects of arbitrary shapes on substrates, where the x,y dimensions are determined by NIL and the z dimension by the LBL assembly. The process can be used to fabricate the photonic components. Passive photonic devices in silicon waveguide technology have been fabricated with quite acceptable results in comparison with other lithography methods (Plachetka et al., 2008). Cheng and Guo proposed a combined-nanoimprint-and-photolithography (CNP) technique which introduced a hybrid mask mold made from UV transparent material and with a light-blocking metal layer placed on top of the mold protrusions. The CNP method using such a hybrid mold can achieve resist patterns without residual layer, and the resist patterns can have higher aspect ratio than the feature on the mold. In addition, the photoresist used in the CNP technique can provide higher etching durability compared with thermal plastic polymers that are commonly used in NIL. Compared with contact photolithography techniques, the CNP can achieve much higher resolution by reducing the effective resist thickness down to tens of nanometers (Cheng & Guo, 2004).

### 2.3.9 High resolution NIL

HP and MIT developed a sub-10 nm NIL by wafer bowing, introduce the concept of wafer bowing to affect nanoimprinting, as shown in Fig. 10. In the scheme, the imprint force is applied uniformly and systematically from center to edge, preventing air from being trapped. More importantly, it shortens the mechanical path between the mold and wafer; this makes the imprinter less susceptible to ambient vibration and helps to preserve the alignment during mold-wafer approach. After an UV exposure step, air can be let into the module to effect mold-wafer detachment. These will enable achieving excellent patterning and overlay at much lower cost (Wu et al., 2008).

In order to meet the manufacturing requirements of a variety of micro/nano devices and structures, lots of new NIL processes are being proposed and developed in recent year. Here only presents some principal and typical NIL processes. As the rapid development of the NIL technique and micro/nano fabrication technologies, much more innovative processes or methods concerning NIL will emerge in the future.

## 2.4 Crucial process issues for NIL

### 2.4.1 Thickness and uniformity of residual layer

A key characteristic of NIL is the residual layer following the imprint process. It is preferable to have thick enough residual layers to support alignment and throughput and low defects. However, this renders the NIL step less critical for critical dimension (CD) control than the etch step used to remove the residual layer (Nanoimprint lithography, 2009). In addition, as a practical technology for the mass production of nanosized patterns, NIL must have an ability to produce a uniform layer with minimal residual layer thickness. Reducing the residual layer is important as it limits the effect RIE has on the resist mask improving tolerance control. Therefore, having a thin and uniform residual layer plays an important role for various NIL processes. In order to satisfy the requirements, various methods have been proposed. Among these include (1) the addition of some aspects of photolithography such as selective UV-curing through a hybrid mask- mold followed by a development step, (2) contrast-modified exposure followed by development, (3) reducing the initial volume of resist to induce incomplete filling of the mold, (4) using high pressure to squeeze excess resin out from between the mold and the substrate, and (5) optimization of droplet positioning in the case of a liquid resin system (Jun et al., 2005, Dumond & Low,

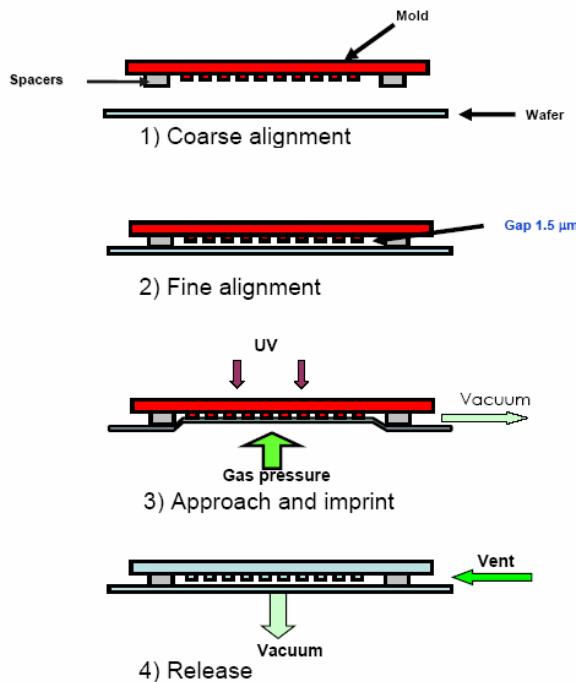


Fig. 10. Sub-10 nm NIL by wafer bowing (Wu et al., 2008)

2008, Lee & Jung, 2004, Bogdanski et al., 2007, Hiroshima, 2008). Jun et al., from HP Lab, devised a novel technique for dispensing resist that takes advantage of the opposing surface energies of the mold and the substrate to produce a uniform and air-free resist film (Jun et al., 2005). Dumond and Low developed a method of imprinting resist structures wherein the residual layer is self-removed via failure while desirable resist features are transferred to an external substrate. The uniqueness of this technique is further enhanced by the relative ease with which it can be used to fabricate overhang structures useful for shadowing evaporated materials (Dumond & Low, 2008). Balla et al., discussed the relationship between the residual layer and initial resist thickness. Lee and Jung investigated the factors affecting residual layer thickness in UV-NIL. To obtain imprinted patterns with no residual layer, a thin and uniform layer of imprint resin must initially be applied. Effective rearrangement of the resin during imprinting is also essential to obtain no residual layer. The use of pressurized imprinting and a more fluidic imprint resin are very helpful. The high fidelity transfer of various patterns as small as 150 nm with no residual layer was successfully demonstrated by controlling the initial resin thickness at an imprint pressure of 15 atm (Lee & Jung, 2004). Investigators at University of California at Berkeley have developed a zero residual layer NIL process. This technology is applicable to large-scale nanoimprinting, flexible electronics, printing of nanoscale metal electrodes, patterning photoresist for subsequent processing, among other applications. The method provides a new way to change the fluid and solid interaction so as to exclude more fluid and eliminate the residual layer. Preliminary results show that the gold nanoparticles are confined to patterned

features, and excluded from all other areas. The ability to pattern micro/nanoscale features without residual layer formation makes NIL significantly simpler and reduces processing time as compared to prior methods. This technology is application to all NIL n needs (Zero residual layer nanoimprint lithography, 2009).

#### 2.4.2 Pattern fidelity

In most cases, NIL involves two transferring steps for whole imprint course. Firstly, the patterns in the template is replicated to the resist, subsequently, the resist pattern is to be further process for transferring the substrate applying the etching process or functional materials using combination of deposition process and the lift-off technique. Therefore, it is very important to ensure pattern fidelity in NIL process for achieving high-quality pattern. There are two problems to overcome to preserve the pattern fidelity in transferring the polymer pattern to the underlying substrate. One of them involves removing the residual resist from the recessed parts (windows) of the pattern by RIE, depositing a metal layer and applying a lift-off technique such that the metal layer left only in the windows can be used as the etch mask for further processing. Another approach is the use of a multilayer, typically a bilayer of one polymer on another polymer layer or a trilayer in which a layer easily deformable at relatively low temperature is added to a metal-polymer bilayer. This bilayer reversal imprint lithography offers a distinct advantage over other imprint techniques in allowing for a high aspect ratio of the pattern transferred onto a substrate, which has been difficult to obtain for small feature sizes. The method requires only one etching step as opposed to the two etching steps typically needed in the imprint lithography, which can degrade the pattern fidelity (Suh et al., 2004). Li et al., studied the pattern transfer fidelity of NIL by patterning sub-micron MESFET gates on six-inch wafers. The CDs of gate patterns in resist are 5.2% (or 37 nm) on average larger than those on the mould with a standard deviation of 1.2% (or 8 nm), and the CDs after oxygen RIE and metal lift-off are 42% (or 296 nm) on average larger than those on the mould with a standard deviation of 8% (or 30 nm). Compared with conventional photolithography, NIL has higher resolution and better pattern transfer fidelity with CD controls about four times smaller (Li et al., 2003). The primary measure of process quality in NIL is the fidelity of pattern transfer, comparing the dimensions of the imprinted pattern to those of the mold. A rapid, nondestructive technique termed critical dimension small angle X-ray scattering (CD-SAXS) is used to measure the cross sectional shape of both a pattern master, or mold, and the resulting imprinted films. CD-SAXS data are used to extract periodicity as well as pattern height, width, and sidewall angles. Films of varying materials are molded by thermal embossed NIL at temperatures both near and far from the bulk glass transition (Jones et al., 2006).

#### 2.4.3 Defect control

In NIL, defects roughly could be divided into two groups: random distributed and repeated defects. Random distributed defects include particle-associated defect, gap (or void) associated defect, and separation related defects, and the residual after imprint, which are not repeatable in terms of location and amount. Repeated defects include those existing defects on mold and substrate, which are repeated in the process. The gap-associated defect is a unique phenomenon in NIL, which is induced by the incomplete contact between mold and substrate. Defects have been one of the biggest obstacles for NIL to be the real

nanofabrication process in the industry. To the nanodevice manufacturer, defects are one of the key issues to product quality and yield (Chen et al., 2005). Unlike in other lithography, a particle induced defect in NIL is larger than the particle itself. To remove the particles, a dry clean process for the nanostructure-patterned surface can be used (Chen et al., 2005). When vacuum is not used during the imprint process, air can get trapped, resulting in bubble defects. This is because the imprint resist layer and the template features are not perfectly flat. There is an elevated risk when the intermediate or master stamp contains depressions (which are especially easy air traps), or when the imprint resist is dispensed as droplets just before imprinting, rather than pre-spun onto the substrate. Sufficient time must be allowed for the air to escape. Bubble defects are unavoidable when UV-NIL is carried out in air. Hiroshima et al.; proposed two methods to eliminate bubbles, i.e., resin squeezing by which bubbles are transferred out of the mold, and gas condensation by which bubbles are considerably decreased in volume. The resin squeezing method, which requires no additional cost, is effective for a thick initial film, but not for a thin initial film because resin flow transporting bubbles out of the mold is insufficient. The gas condensation method does not involve such restrictions. Due some limitations of the gas condensation method, they studied the elimination of bubble defects by gas condensation method using pentafluoropropane. When UV nanoimprint is carried out in an environment of a pentafluoropropane flow higher than 150 sccm with a hold time longer than 20 s, no bubble defects are generated in the entire imprint area. Bubbles are eliminated within a few seconds under an imprint pressure of 0.5 MPa. They consider that bubble elimination using pentafluoropropane is very useful and realizes UV nanoimprint of the same quality as that using vacuum but at a much lower cost (Hiroshima & Komuro, 2007). Liang et al., reported an experimental and theoretical study of two most critical yet still to-be-answered issues in dispensing-based nanoimprint lithography (D-NIL): air bubble formation and absorption, and discuss their impact on NIL yield and throughput. Their study shows that the key factors that affect the air dissolution time (and hence the air bubble shrinking time) are air bubble initial size, imprinting pressure, air solubility, and resist residue layer thickness. One of the key conclusions from the study, which has significant practical importance, is that although the air in a bubble can be completely dissolved in a resist liquid as long as the bubble is smaller than a certain size, the air absorption time might be too long for the dispensing-NIL operating in atmosphere or poor vacuum to have a necessary throughput in mass manufacturing. In addition, when the residual layer thickness is close to zero or a critical thickness, the bubble dissolution process can be significantly slowed down (Liang et al., 2007).

#### 2.4.4 Filling process

The NIL involves two crucial process steps including the resist filling rheology behavior and demold characteristics which have decisive influence on pattern quality and throughput. In order to better understand the NIL mechanism and obtain the optimal imprint conditions (e.g., pressure, temperature, pattern layout of the mold, and time), it is necessary and important to investigate the flow and filling rheology behaviors of the resist. Two fill mechanisms have been observed: simple flow of the PMMA from the borders and formation of polymer mounds. A simple theory was used to estimate the embossing time required to fill a given stamp geometry. In both cases there is evidence of compression causing buckling of the polymer and also capillary action drawing the polymer up to the top of the stamp cavity (Scheer & Schulz, 2001). Heyderman et al., thought that the time to fill a nanostructure array

with a large unstructured surrounding area is the same for a microcavity with the same surrounding area and cavity volume. The fastest embossing times for complete fill of the cavities were obtained at temperatures greater than 100 °C above  $T_g$  with PMMA viscosities in the range 300 to 3000 Pas (Heyderman et al., 2000). Jeong et al., study simulated hot embossing micro/nano-manufacturing with thin films, including the effects of capillary force and width of stamp groove on the flow behavior during embossing. Simulations showed double peak flow for regions of large width, slow printing speeds and with surface tension as a significant factor (Jeong et al., 2002). Macintyre and Thoms developed a method to directly observe the flow of resist arising from NIL. A fiducial grid was embedded in the polymer layer being imprinted, and they observed its distortions after imprinting (Macintyre & Thoms, 2005). Scheer and Schultz recorded wave-like fronts of resist moving between the stamp and the substrate during imprinting processes operating at more than 100 bar (Scheer & Schulz, 2001). The mechanism in the low-pressure/high-temperature imprinting depends on the type of mold used. When a rigid mold is used, capillarity is the mechanism. In the case of a flexible film mold, however, both capillarity and viscous flow are responsible for the imprinting. As time increases, only capillarity is operative. An understanding of the mechanisms involved in various imprinting methods is valuable on its own. More important is the insight it can provide into possible modifications of the imprinting process that can be tailored for specific applications (Khang & Lee, 2008).

### 3. NIL mold (Template)

The mold (template, stamp) is one of the most critical elements for the NIL process. The ultimate resolution of the patterns fabricated by NIL is primarily determined by the resolution of the features on the surface of the mold. Because of the 1X nature of NIL compared with 4 X for photolithography, the 1X template fabrication has now been considered the greatest challenge for NIL process. This section will mainly discuss three issues regarding NIL molds: material, fabrication method and crucial process issues.

#### 3.1 Mold material

A variety of materials such as Silicon,  $\text{SiO}_2$ , Fused Silica (bulk), Quartz (fused), Glass, Silicon Nitride ( $\text{Si}_3\text{N}_4$ ), Diamond, Nickel, PDMS, etc., have been utilized to make molds for NIL. The material chosen affects the mold lifespan and reliability. Harder materials provide better wear characteristics, while soft moulds may have a limited lifespan, but can simplify stamp creation (Pfeiffer et al., 2002). Not only the mechanical characteristics, but also optical and chemical properties are important when choosing a mold material for NIL. Critical mechanical parameters and their implications for NIL are hardness and thermal stability (lifetime and wear), thermal expansion coefficients and Poisson's ratio (dimension mismatch leading to distortions during demolding), roughness (higher demolding force and damage), Young's modulus (bending), and notch resistance (lifetime and handling). Issues related to fabrication are processability (etching processes, selectivity, clean room environment), and surface quality (resolution) (Bhushan, 2007). The handbook gives a brief overview of the mechanical and thermal properties of materials used for molds (Bhushan, 2007). The use in a NIL process is also determined by additional properties such as transparency, conductivity, anti-sticking properties (with/without anti-adhesive coating, e.g. by covalent coating), availability and cost (standard materials and sizes, tolerances, processing equipment and

time), and how easy it is to employ in NIL (e.g. fixing by clamping, thermobonding, gluing). Currently, Silicon, Quartz, Nickel and Silicon Nitride ( $\text{Si}_3\text{N}_4$ ) are typical materials frequently used for hard molds. Various polymeric materials, including polydimethylsiloxane (PDMS), polyurethane acrylate (PUA), polyvinyl alcohol (PVA) and polyvinyl chloride (PVC), have UV transparency, mechanical hardness and formability and thus can be used as the material for soft UV-NIL templates. Among these polymeric materials, PDMS is highly UV-transparent and has a very low Young's modulus which gives it the flexibility required for conformal contact. It has a very low reactivity and interfacial energy toward the polymeric materials and is sufficiently elastic that it can be separated from the polymeric structure without destruction or distortion. In addition, PDMS mold has a low surface energy at the polymer interface, eliminates the problem of the polymer sticking to the surface of the mold during detachment, which has proved a critical defect of NIL. Currently, PDMS has been considered as standard soft mold material due to its favourable properties concerning flexibility, UV-transparency and low surface energy. However, the main drawback of PDMS materials is the high viscosity and swelling. It is necessary to develop much more new mold materials with better performances to meet new NIL requirements (e.g. conductive mold for electrical field assisted NIL, release agent-free mold) (Guo, 2007, Costner et al., 2009, Yokoo & Namatsu, 2009, Pfeiffer et al., 2002, Bender et al., 2004, Choi & Park, 2004).

### 3.2 Mold fabrication

Mold fabrication is one of the biggest hurdles to imprint acceptance due to the 1X pattern resolution required. The three major challenges for mold manufacturing are resolution, quartz etch uniformity, and defect inspection. For many of the early adopters of this technology, the first two items are of greatest interest, as their designs are not defect sensitive. The minimization of defects and the ability to verify template quality are ultimately necessary for CMOS type applications. Patterning of most templates in a commercial mask making facility is done using e-beam lithography. There are two types of e-beam writers in use today, shaped beam tools and spot beam tools. Shaped beam tools are used predominately by mask makers due to their higher throughput and superior image placement accuracy. Spot or Gaussian beam tools offer finer resolution, but write times tend to be significantly longer. This requires a mask maker to determine the requirements of each template job and choose the appropriate e-beam writing strategy (Maltabes & Mackay, 2006, Maltabes et al., 2005).

For NIL to be accepted across the industry, a new infrastructure for 1X template fabrication, inspection, and repair needs to be established. This is a challenge. Obducat uses its own e-beam lithography technology to support and develop its template technology. Molecular Imprint has been working with many industrial partners, including Dupont Photonics, Toppan Photomasks (template fabrication), Motorola, KLA-Tencor (template inspection), and Carl Zeiss (template repair), to establish a template infrastructure. Recently, BenchMark Technologies, Inc. started offering e-beam written standard nano-imprint test templates. Motorola Labs has been focusing on developing the template and wafer-level processes while optimizing the imprinting process and collaborating with external partners to optimize both the inspection and repair of imprint templates (Hussain et al., 2007). Dauksher et al., reviewed recent results of template fabrication (including template repair and inspection), the imprinting process, the wafer-level pattern transfer processes, and applications (Dauksher et al., 2006).

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