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Nanoimprint lithography - the past, the present and the future

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Abstract: Nanoimprinting lithography technique uses a very simple concept of transferring pattern of nanoscale features from a mold to a target substrate. In the past two decades, this technique has successfully broken through the barrier of laboratory scale production and become an industrial scale production technique. This article reviews major types of nanoimprint lithography techniques which are mainly base on thermal treatment, ultraviolet light exposure, laser light irradiation or electro chemical reaction. The machining mechanism and components of major nanoimprinting methods are described. A technical comparison of these methods is presented through the analysis of their state-of-the-art research advances. Moreover, the advanced applications of nanoimprint lithography in the fields of optoelectronic devices, solar cells, memory devices, nanoscale sensors and biology are presented. Finally, the article investigates the challenges faced by nanoimprint lithography and its future trend, and discusses the possible solutions to meet these challenges.

Keywords: nanofabrication, nanoimprint lithography, throughput, resolution, roll-to-roll

1. Introduction

Currently, the research on high throughput, high precision and low cost fabrication approach for nanostructures has attracted a lot of attentions in the field of nanoscience and nanotechnology. Such fabrication technique is of crucial importance for the widespread and

commercialisation of nanotechnology as nanostructures are the key building blocks and driving forces for microchips, microelectronic devices, biosensors and high density magnetic storage media [1–5] to reach their ultimate capacities. As such the integration density of the semiconductor devices and circuitry is continuously growing, the dynamic random-access memories (DRAMs) and microprocessors are experiencing a steady growth by 50% and 35% per year [6].

In early 1990's, scanning probe microscope (SPM) lithography [7, 8] were proposed to fabricate nanostructures. Even with further technical improvements [9], SPM lithography still suffers from low throughput and is therefore only applicable in a laboratory scale. On the other hand traditional high throughput optical lithography approaches such as, X-ray lithography and extreme ultraviolet (EUV) lithography are limited by the requirement of expensive equipment and mask, as well as the attainable resolution [10] due to Abbe diffraction limit. The breakthrough in mass production of nanostructures was made by Prof. Chou who proposed a nanofabrication technique, named as nanoimprint lithography (NIL) in 1995-1996 at NanoStructure Laboratory [11], University of Minnesota, USA. Since then NIL has evolved quickly to become one of the most promising nanopatterning processes and is considered as the future of nanofabrication.

Similar to other step and stamp approaches, the principle of NIL is to transfer the patterns of a given sample (such as mold) to a target substrate. This can be achieved by using mechanical contact, or heating, or curing [12], or by electrochemical reaction. NIL is a fast and low cost production process. With the help of modern NIL techniques, pattern of sub-5 nm resolution can be easily achieved [13]. NIL has some key advantages over other existing lithographic processes [14–16] which are summarized as follows:

- NIL is a cost-effective fabrication technique which is essential for commercial production.

- NIL can provide high resolution structures. The resolution of a fabricated pattern is limited by the mold only.
- NIL is a fast process and NIL can be used to fabricate nanopatterns at a large scale in a short time.

Due to these advantages manufactures of hard disk drive and semiconductors find NIL as the most convenient production process. NIL has also been widely used in the fabrication of nanostructures for light emitting devices, memory devices, solar cell, flat panel display, and plasmonic devices, etc [17–20]. Because of its high-resolution dense pattern capability, NIL is also considered as the enabling fabrication technique for future flash based memory devices [16]. The conventional nanoimprint techniques are facing many challenges due to some imperfections. Firstly, the throughput of current approaches should be further improved to meet the dynamic market demand for commercial production. Moreover, even if the desired patterns have very small variation, the molds used in physical nanoimprint must be remanufactured by using reactive ion etching (RIE) [9] or electron beam lithography (EBL) [21]. Thirdly, the adhesion between mold and resist is a significant problem, because the mold and patterns may be broken during the procedure of mold release. This review article briefly walks through the history of imprinting process, and then discusses the major techniques developed for NIL and their applications. Finally, it gives a detailed discussion on the challenges and the future aspects of the nanoimprint lithography technique.

The article is segregated into the following sections and subsections:

- History of imprinting process
- Major NIL techniques developed
    - Thermal NIL
    - Ultraviolet (UV) NIL
    - Laser-assisted direct imprint(LADI)

- Nanoelectrode lithography
- Application of NIL
- Challenges and the future of NIL

2. History of imprinting process

Even though the term “nanoimprint lithography” is new, the concept of imprinting to replicate structure is old. Casting is a manufacturing process where a liquid is poured into a mold and subsequently allowed to solidify it. It has been one of the most popular manufacturing processes estimated to be used by the human civilizations since 6000 BC. The process of inserting a mold into a plastic material to replicate structure has been used by the old civilizations. This process is generally termed as embossing process and has been used to replicate and produce coins. It was also used for letterpress. Later the process of hot embossing was added to create structures on polymer materials [22, 23], ultra-thin teflon-like films [24], DNA separation and detection [25] and micro lens array [26].

The realization of creating nanostructures at a large scale using imprinting process came from Prof. Chou’s work in 1995-1996 [10, 27]. He showed that nanostructures of 25 nm resolution could be achieved using imprinting processes. As shown in the schematic picture (Fig.1), a mold with feature size of few tens nm is used to press on a thermoplastic material substrate which is heated up during the molding process. If the temperature of the material increases above its glass transition temperature, the polymer will behave as a viscous material which helps the molding process. With subsequent RIE, the thickness contrast of the imprinted patterns can be transferred through the entire resist region. In the article published by Chou's group, a silicon dioxide mold was used. The patterns, which had a feature size of 25 nm, were created using EBL technique. Polymethyl methacrylate (PMMA) layer of thickness 50-200 nm was prepared on a Si substrate in order to replicate the pattern [10]. The prepared PMMA layer must have the property of desired mold release (should not adhere to the mold after the

release of the mold). After the imprinting, RIE was used to etch away the thin resist layers on the compressed portions to achieve a high quality pattern as shown in Fig. 2.Using this approach, patterns of different shapes of nanometre feature size were created (Fig. 3).

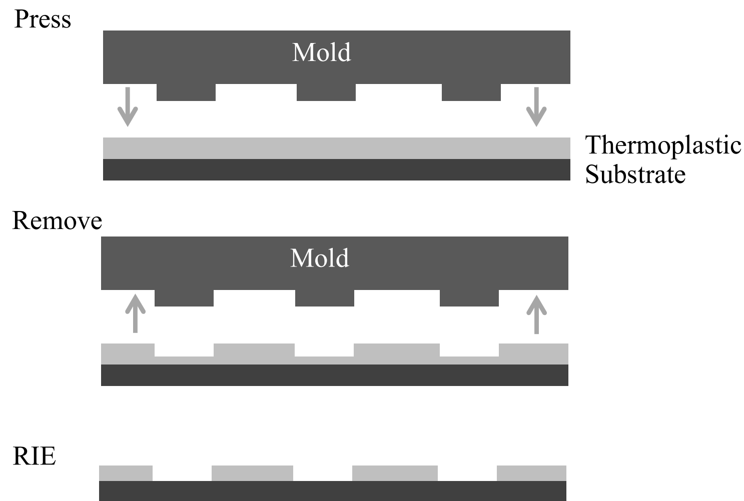

Fig.1 Schematic of the imprinting approach implemented by Prof. Chou [27]

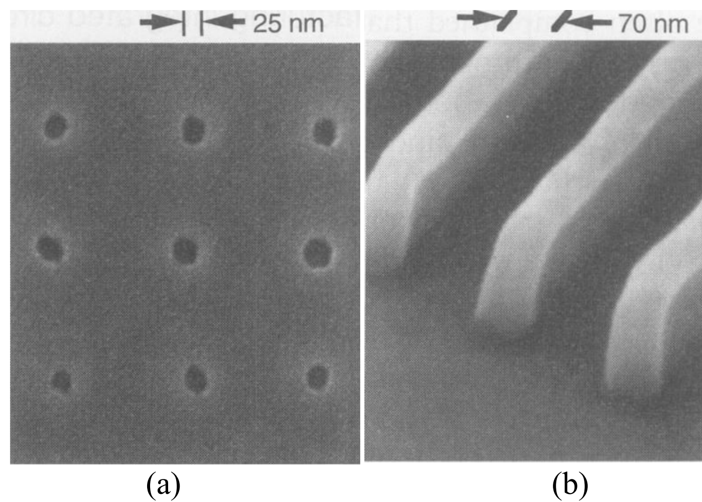

Fig.2 Scanning electron microscopy image of a (a) PMMA film having 25 nm hole structure (top view), (b) strip pattern formed on a PMMA film using the imprinting process (perspective view) [10] (Reprinted from ref. [10] with permission)

Using this flat imprint lithography technique, fabrication of nanostructures at a large-scale has been successfully demonstrated. Prof. Chou's group subsequently has introduced the concept of roller imprint lithography in 1998, a continuous nanoimprinting process [28].Two types of approaches have been discussed for roller imprint lithography in his paper, which are

cylindrical mold method and flat mold method. As shown in Fig.3, in the cylindrical mold method, the mold is prepared on the surface of a cylindrical roller. In the second method, a flat mold of Si is placed on the substrate and a smooth roller is rotated over the flat mold.

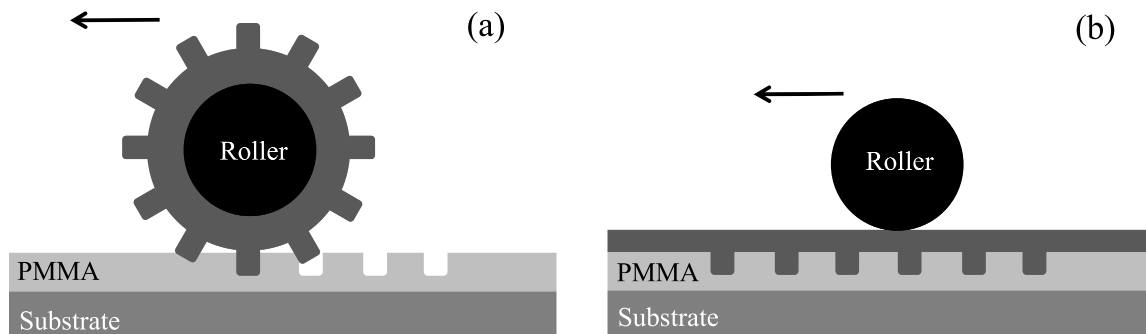

Fig.3 Schematic picture showing the roller NIL methods as perceived: (a) cylindrical mold method and (b) flat mold method [28]

In roller imprinting process, the temperature of the roller is kept above the glass transition temperature of the resist and the temperature of the resist is kept below its glass transition temperature. This ensures that only the contact region between the mold and the resist is suitable for the imprinting process. Using the first generation of roller NIL, resolution of sub-100 nm was achieved which was shown in Fig.4 [28].

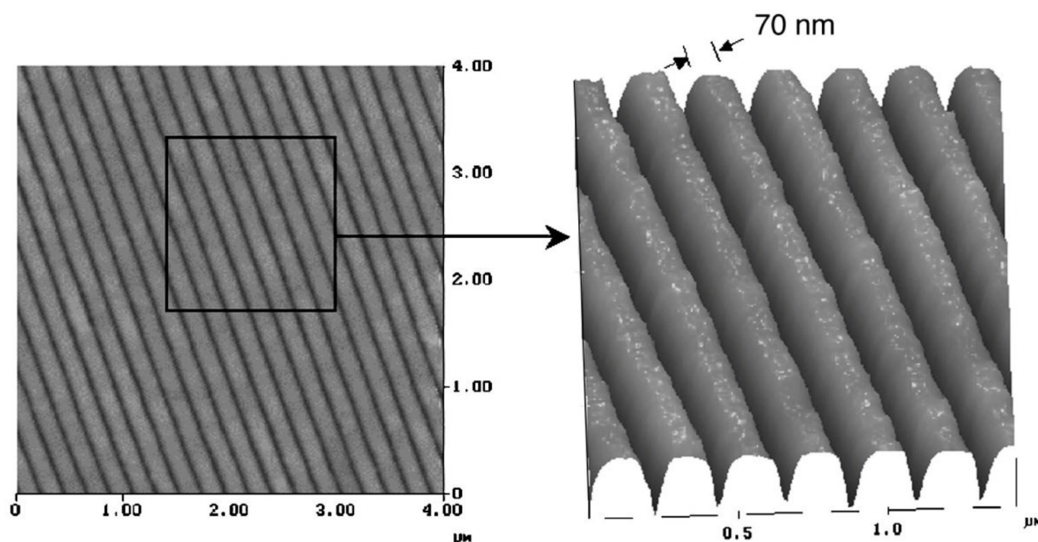

Fig.4 PMMA nanostructure of sub-100 nm resolution created using roller imprint method [28] (Reprinted from ref. [28] with permission)

3. Major NIL techniques developed

Since the birth of NIL process two decades back, several research groups have contributed in developing several nanoimprinting methods and NIL has grown rapidly within a short time. Depending on the process used for nanoimprinting, major NIL techniques can be classified into four categories, which are discussed below.

3.1 Thermal NIL

3.1.1 Machining mechanism of thermal NIL

In order to print a pattern on the target sample using NIL process, the target material must be softened to take the pattern. One of the ways to achieve this condition is to heat the sample surface layer above the glass transition temperature (T_g) of the material. Achieving the required temperature through heating can be approached through different ways. The resist can be directly heated up as discussed by Chou [10]. The resist is heated up above the T_g and then the mold is pressed against the resist layer in order to imprint. Subsequently, the sample is cooled down and the mold is separated from the target sample. In order to achieve proper imprinting, the temperature and the pressure on the mold, and the time duration for holding the mold on the surface have to be controlled properly.

Instead of heating the sample, the mold can also be heated up. During the imprinting process, the mold touches the sample surface and only the contact region of the sample is getting sufficient temperature. In general, the molding temperature (T_m) is kept above the glass transition temperature (T_g) and the de-molding temperature (T_{dm}) is kept below T_g. The local pressure on the sample has to be varied accordingly. In case of thermal roller NIL, the heating and the cooling procedure may become tricky.

3.1.2 Components of Thermal NIL

A typical thermal NIL system includes the following components:

- A plate or roller mold with nanopatterns;

- A feeding and de-molding system;
- A heater is integrated with platform or roller installed on the moving stage to heat up the substrate or roller;
- The roller is also adapted to enhance the press force between the stamp and the substrate;
- In order to achieve proper heating and imprinting, the precise control system of pressure, temperature and cooling is needed;
- Substrate such as polymer.

3.1.3 Advances in thermal NIL

Hirai et al. [29] have studied the fracture defect of the polymer in thermal nanoimprint lithography. They concluded that the stress concentration at the corner of the pattern is the reason to induce the fracture defect. The strain is caused by the applied pressure below the glass transition temperature and the difference of the thermal expansion coefficients between the mold and the substrate. Bilenberg et al. [30] have established the Topas-based lab-on-a-chip (LOC) microsystems by using the thermal nanoimprint lithography and analysed the effect of the environmental stress cracking. In order to achieve highly efficient optoelectronic devices, the thermal nanoimprint lithography has been used to develop a direct indium tin oxide (ITO) nanopatterning technique to improve the transmittance and conductivity of the TCO layers [31]. Youn et al. [32] described a thermal roller NIL approach in their article. The schematic picture (Fig.5) shows the major components of the system developed by Youn et al. The stamp is connected with two moveable springs through the pullers.

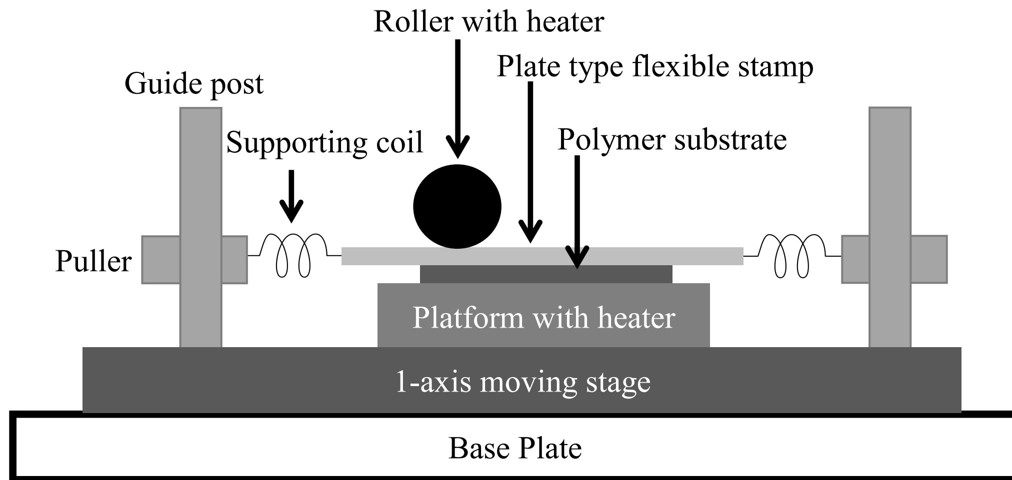

Fig.5 Schematic picture showing the thermal roller NIL approach [32]

The flow chart of the thermal roller NIL technique is described schematically in Fig.6. The stamp, which is supported by the coils, is pushed down to touch the polymer. The roller temperature is set above the T_g and the temperature of the platform is set below the T_g. The stamp receives the heat from the roller. The roller is rolled over the stamp and as soon as the stamp touches the polymer with pressure, the imprinting process starts. Following this, the stage holding the polymer is moved parallel as shown in the schematic picture. The technique of thermal roller imprinting process developed in this way shows good results in replicating ultra precision micron scale structures at the scan speed of 0.1–10 mm/s [32].

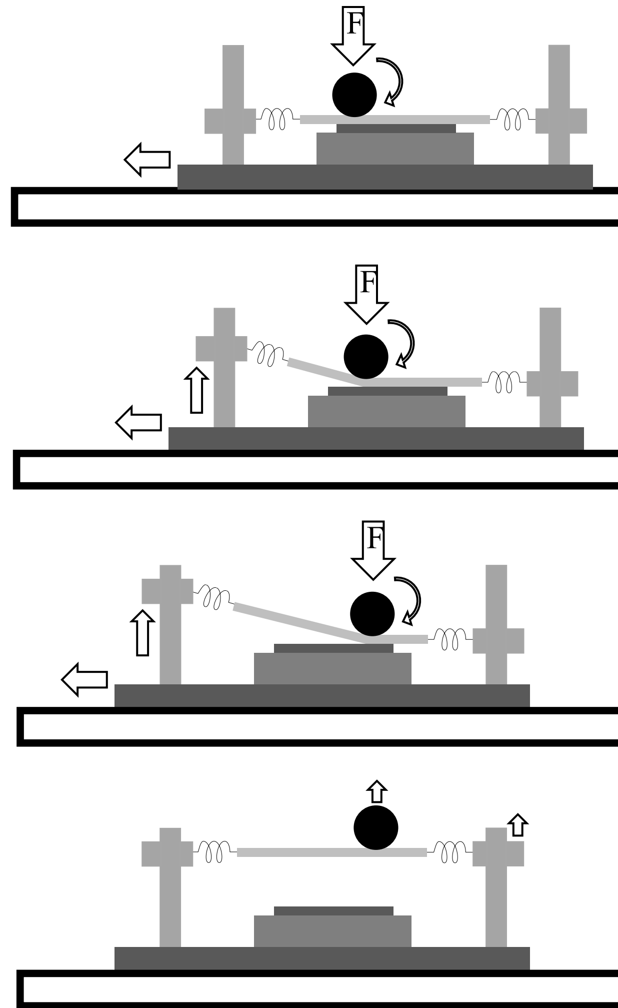

Fig.6 The schematic drawings of the steps in the thermal roller NIL process [32].

In thermal NIL, the mold surface should be hard enough and have the ability to avoid any distortion, even if it withstands the mechanical contact at high temperature and high pressure during the molding and demolding process. Hence, some hard materials such as Si, fused silica, SiC and diamond can be chosen as the mold material used in thermal NIL. Meanwhile, the thermal expansion coefficient of the mold, polymer and substrate is a significant factor which can cause some defects such as distortion and asymmetric structure, especially when the difference of the thermal expansion coefficient is large. Also the demolding process has to be well-controlled in order to avoid the generation of distortion or damages on the imprinted structures. In addition, the effect of adhesion, friction, shrinkage and trapping

which can happen between the surface of the mold and the imprinted structure has to be eliminated or controlled in a small scale [12].

3.2 Ultraviolet (UV) NIL

3.2.1 Machining mechanism of UV NIL

UV light has been used for a long time to create micro/nanostructures on photoresist material. The resist material is exposed to the UV light and chemical reaction is initiated. Subsequently, the sample is developed to extract the patterns. This technique can be used to develop patterns in a batch process.

Ahn et al. has developed a system to integrate UV lithographic capability with a roll nanoimprinting system [33]. The schematic picture of the continuous UV roll NIL system is shown in Fig.7. As shown in the figure, there is a dispensing unit to supply UV-curable polymer material on a transparent substrate, a UV illumination unit to process the pattern formation and a stamp roller unit. As the polymer-coated substrate passes through the roller stamp, the UV light is illuminated on the curing region and in this way patterns are generated. Fig.7(a) shows the design to work on a flat substrate, whilst further modified to work on flexible substrate is shown in Fig.7(b). Using the UV roll NIL technique, nanopatterns of 500 nm line structure and 50 nm pillar structure have been successfully fabricated [33].

3.2.2 Components of UV NIL

A typical UV NIL system includes the following components:

- A dispensing unit is needed to coat the UV-curable resin onto the substrate;
- A roller stamp with micro- or nanopatterns;
- In order to control the thickness of the final replica, a contact roller is adapted to control the pressure between the roller stamp and substrate;
- Rotating control system of roller and feeding system of substrate;
- The rigid or flexible substrate such as Si or polyester;

- UV-curable resin;
- A UV light illumination unit is needed to cure the UV-curable resin.

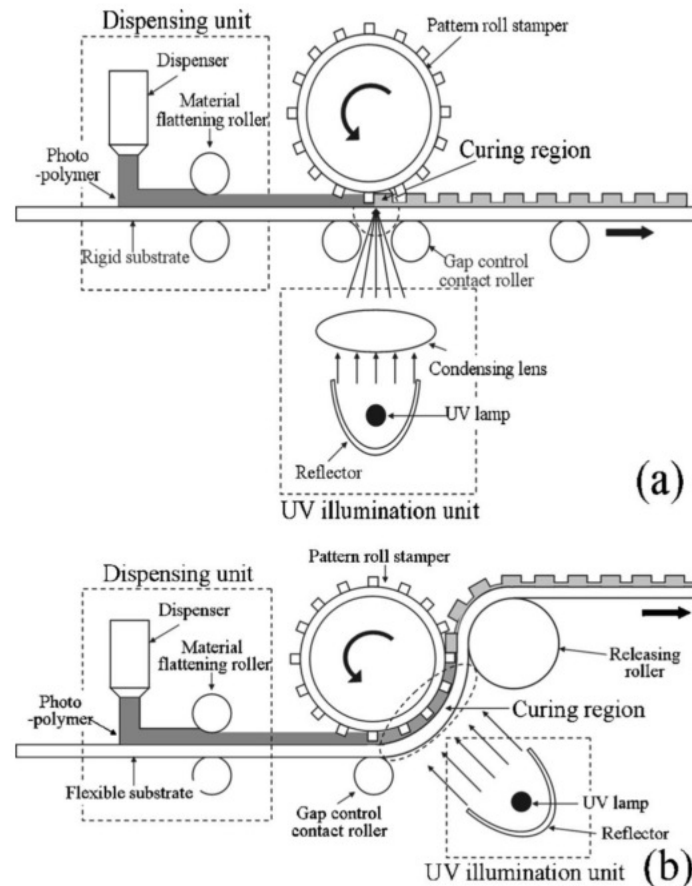

Fig.7 Schematic diagrams of the UV roll nanoimprinting system: (a) for a rigid substrate and (b) for a flexible substrate developed by Ahn et al. [33] (Reprinted from ref. [33] with permission)

3.2.3 Advances in UV NIL

The roll-type UVNIL process has been proposed by Lee et al.[34]. In this method, a flexible transparent thin stamp is used for the imprinting process on Si as well as on plastic substrates. The flexible PDMS (polydimethylsiloxane), polycarobontate and polyester (PET) film have been fabricated from Ni master stamp using thermal imprinting process. These fabricated stamps are set up on quartz cylinder and replicated on Si using the roll typed UV NIL process. 100-300 nm DVD patterns and 75 nm line-width patterns have been achieved using this

process[34]. Later, Ahn and Guo [35] have demonstrated a high-speed (7.94 mm/s) and large-area (4 inch wide) roll-to-roll (RTR) and roll-to-plate (RTP) NIL apparatus to continuously imprint grating patterns with 300 nm linewidth. In addition to greatly improving the throughput, the apparatus has the ability to maintain the pressure uniformity and successfully de-mold in large-area imprinting, which are big challenges faced by conventional NIL. Moro et al. [36] have transferred the antireflection (AR) structure onto a polymer film by RTR UV nanoimprint lithography. As a result, the high-performance AR film with the 0.1% reflectivity and about 95% transmittance at visible wavelengths has been produced at a feed rate of 1.8 m/min. In general, it is difficult to transfer structure having high aspect ratio using RTR method. The Japanese research group, Hiroshi et al., have developed a technique based on RTR UV process to overcome this problem. A tough replica mold has been used in the RTR UV NIL process to fabricate high aspect ratio (AR = 5) line-and-space (L&S) pattern [37]. The L&S pattern (height = 500 nm, width = 100 nm) was imprinted on a polyester film.

One of the challenges in nanoimprinting technique is the use of a master mold. Even though NIL is a cost effective method, the cost of manufacturing a master mold is relatively higher, because it is usually fabricated by other expensive fabrication techniques such as, EBL, RIE, deep-UV lithography, chemical etching, etc. A quick solution for this problem is to manufacture the mold using a NIL method. In fact, many groups have demonstrated to use NIL for the fabrication of molds. Jarrett et al. [38] have demonstrated a technique to use RTR UV process to fabricate resin mold. The mold has been subsequently used as a master mold in a thermal nanoimprinting system.

UV NIL process can be realized at room temperature, and the required mechanical force for imprinting is smaller than thermal NIL because the mold is normally controlled at a constant distance from the substrate, on which liquid film or droplets are coated. By using RTR method, UV NIL process can increase the imprinting speed and ease the demolding process.

Similar to thermal NIL, local shrinkage is an unsolved problem for UV NIL due to the reorientation of polymer chains during curing.

3.3 Laser-assisted direct imprint (LADI)

3.3.1 Machining mechanism of LADI

A rapid imprint technique which is named as laser-assisted direct imprint (LADI) was introduced by Chou et al. in 2002 [39]. Fig.8 shows the schematic of the LADI processing. Firstly, a single excimer laser pulse that passes through a quartz mould is adopted to melt the surface layer of silicon substrate. The solid-state silicon will be in liquid phase after absorbing the laser energy. Then, the molten silicon layer is embossed by the quartz mould. The nanoscale patterns with sub-10 nm resolution on the silicon wafer could be imprinted within the 250 ns embossing time. LADI is a promising approach of NIL as it can be applied to more materials (polysilicon, Ge, and dielectrics) and larger samples (more than four inches diameter or one inch square) [39]. However, precisely controlling the pressure between the quartz mould and silicon wafer must be estimated for different features of different area.

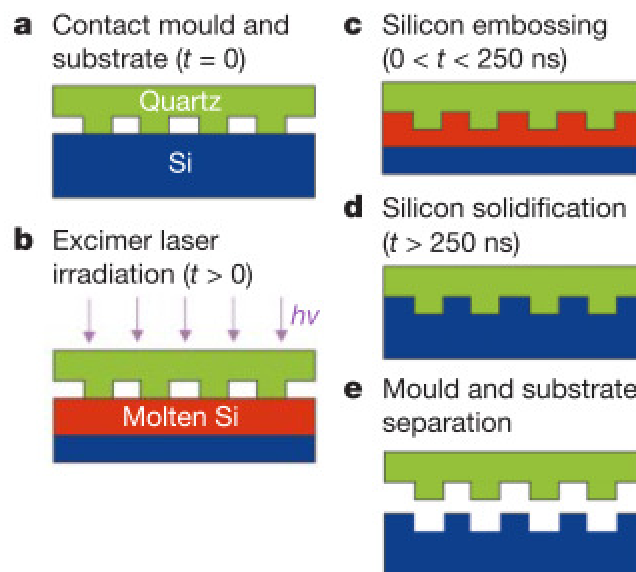

Fig.8 Schematic of LADI of nanostructures in silicon [39]. (a) A contact is made on the Si substrate with a quartz mould, (b) an excimer laser pulse is used to melt the upper layer of Si,

(c) embossing on Si is done, (d) the Si solidifies instantly retaining the patterns, (e) the mould is detached finally. (Reprinted from ref. [39] with permission)

3.3.2 Components of LADI

A typical laser-assisted direct imprint system mainly includes the following four parts:

- Laser source (for example, an excimer laser pulse of 308nm wavelength and 20 ns pulse duration [39]);
- A mould with patterns such as quartz which does not absorb the laser energy, or some metal which has to possess a higher stiffness and strength than the workpiece;
- Substrate which has a lower melting point than mould;
- A precise pressure control system between mould and substrate.

3.3.3 Advances in LADI

Xia et al. [40] have used the laser-assisted nanoimprint (LAN) lithography to pattern nanostructures on various polymers within a short heating and imprinting time. The result showed that the 200 nm pitch gratings with line width of 100 nm and height of 90 nm could be achieved with high fidelity. An approach to generate the high aspect ratio metal nanotips by means of nanosecond pulse laser melting was developed by Cui et al.[41]. As shown in Fig.9, an excimer laser pulse shines through the quartz wafer on which a thin chromium (Cr) film is evaporated. The Cr film becomes melted and can self-form discrete metal pillars toward the top of the lower wafer due to the attractive electrostatic force. After Cr solidification and separation, the nanotips would be achieved both on the quartz wafer and Si/SiO_2 substrate. The nanotips with diameter of 10 nm and height of 180 nm could be created by using nanosecond pulse laser melting. However, it may be difficult to control the shape evolution of molten metal apex and the uniformity of nanotips array also needs to be improved. Cui et al. [42] pointed out that metallic gratings with 100 nm linewidth, 100 nm depth and 200 nm pitch could be imprinted even the temperature was higher than the melting

point of the quartz mold by using LADI. Therefore, LADI is a good tool to pattern the nanostructures directly without etching, especially for metallic materials. This novel method can swiftly overcome the patterning problem on hard solid-state metal materials.

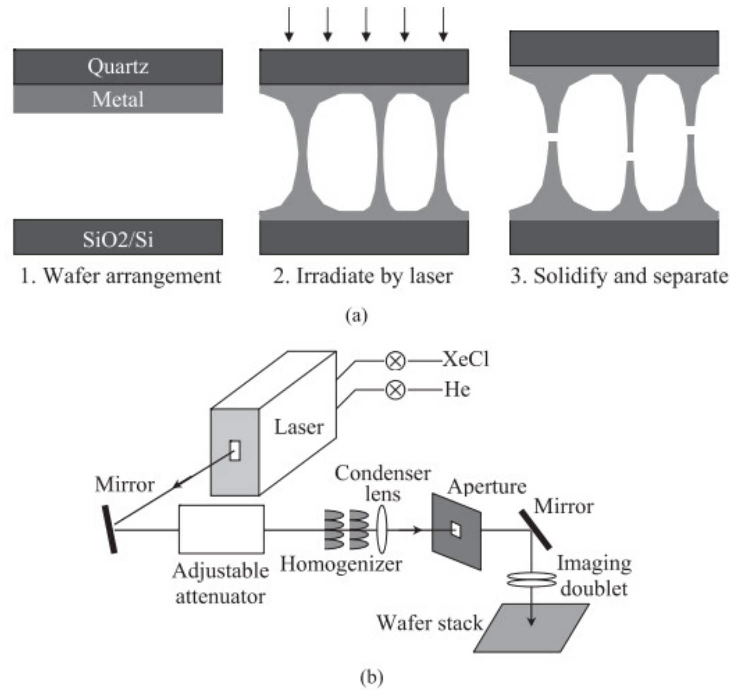

Fig.9 (a) Schematic laser-induced metal nanotip formation process; (b) schematic set-up of the excimer laser and optical components [41] ((Reprinted from ref. [41] with permission)

Lee et al. [43] have developed a laser-assisted high-speed (200 mm/s) one-step direct nanoimprinting technique for semiconductor and metal nanoparticles (NPs). As direct imprinting using NP solutions can eliminate the processes of etching, lift-off and long cooling steps, this approach can reduce the imprinting time and improve the throughput. Ehrhardt et al. [44] have used laser embossing to pattern sub-micrometer-scaled gratings into solid copper samples, and the analysis results obtained from transmission electron microscopy (TEM) and electron backscatter diffraction (EBSD) studies show that there are no significant structural modifications of monocrystalline copper. However, the effect of the residual stress on the patterned surface induced by the laser embossing process cannot be negligible. Nagato et al. [45] have proposed a laser-scanning replication on PMMA film by spot irradiation. The

diamond-like carbon (DLC) is chosen as the light-absorption layer, which is directly irradiated and heated by a laser through the quartz. The high power density and high scanning speed of the laser lead to a high throughput. In additon, the thermal conduction is considered to be the most important factor which limits the scanning rate. Ye and Cheng [46] have demonstrated a scaleable patterning approach on NiTi shape memory alloy by laser shock assisted direct imprinting (LSADI). A high energy pulsed laser beamshots onto the copper grid through the BK7 glass, which can induce the ionizationon on the top surface of the copper grid and thus plasma is generated. The plasma creates a shock pressure [47], which pushes the copper grid into the NiTi substrate. The obvious advantage of LSADI is that any heating or etching process is not required to pattern metals.

LADI introduced by Chou et al is an exciting nanoimprinting approach. Compared with photolithography, the imprinting equipment of LADI is much cheaper than photolithography device because there is no need for expensive focusing optics. LADI is realized by using laser illumination and mechanical imprinting. At the moment, the resolution of photolithography is limited around 100 nm and it is very difficult to achieve smaller structures [48]. Compared with imprinting in a thin resist film, the LADI approach is able to pattern silicon or metals directly with sub-10 nm resolution and there is no need for etching process. Thus, it is likely that in the future LADI may take over costly optical lithography for silicon chips.

3.4 Nanoelectrode lithography (NEL)

Energetic focused electron beam can be used to fabricate nanostructures on polymer materials. This fabrication technique has been popularly known as (EBL) [49, 50]. In EBL, a chemical reaction is initiated on the sample using the E-beam. In EBL systems, an electron gun is normally used to generate electron beam. In 1992, Majumdar et al. have used the underlying concept of EBL in an AFM system where he used the AFM tip as an electron source for

creating nanostructure. Line pattern of 68 nm periodicity and 35 nm linewidth were successfully fabricated on PMMA samples [8].

3.4.1 Machining mechanism of NEL

Yokoo [51, 52] has conceived the idea to use nanoelectrodes for nanostructure fabrication which is called as NEL. The principle of NEL is to use a patterned nanoelectrode to induce an electrochemical reaction on the target surface. Hence, a conductive mold with patterns is needed in NEL. As shown in Fig. 10, the conductive mold is firstly controlled to contact with the target, and only the projecting part of mold can touch the top surface of the target. As it defines, a voltage is applied and current flow occurs between the mold and the target. The projecting part of the mold is the conductive area in which an electrochemical reaction is induced. As a result, the reacted part on the target will possess the patterns transferred from the conductive mold. Finally, the target is developed or etched to achieve the nanostructures like the conventional NIL. The target material can be varied. If the Si substrate is chosen, SiO_2 patterns will be produced after the anodic oxidation which can be etched with HF solution. The SiO_2 patterns with pitch of 500nm and thickness of over 5 nm was successfully generated in a uniform area of over 400 × 300 μm^2 by using NEL [51].

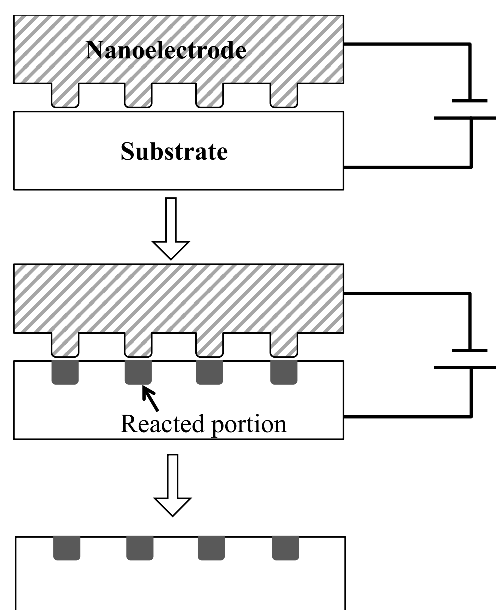

Fig.10 Principle of NEL [51]

3.4.2 Components of NEL

A typical NEL system mainly includes the following parts:

- A patterned nanoelectrode which consists of a conductive area and an insulating area such as a conductive substrate with projections on its surface;
- Target substrate (Si, organic material, GaAs, anodic oxidation of semiconductor or metal);
- Current source (voltage of 15–25 V and current of 2 A [51]);
- A mold pressure adjustment system to control the pressure on each part of the mold;
- A constant temperature-humidity controller to manage the reaction environment and a highly accurate positioning stage;
- A development or etching process is required.

3.4.3 Advances in NEL

As a flexible patterning technique, the NEL can be employed repeatedly to modify the patterns. It enables multiple patterning to overwrite a patterned target [52]. Fig.11 shows the procedure of multiple patterning by NEL. In the first procedure, the electrochemical reaction on the target surface is induced by a line-and-space-pattern nanoelectrode. Second, the nanoelectrode is rotated by 90° and the process is repeated to obtain a checked pattern. Multiple patterning can also be used on GaAs substrate [53]. A fabricated pattern of 400 nm pitch has been successfully generated on a 6×8 mm^2 GaAs surface within 30s of reaction time, which can be directly used as a wet- or dry- etching mask. It is found that the pattern on the flat mold could also be transferred to a target Si substrate [54]. The flat mold includes two parts: conductive area and insulating area, which has a similar function with the master mold in NEL. Hence, the relief pattern on the mold is not necessary, so this approach can simplify the mold fabrication and modification processes.

The development of newly emerging NEL has increased the diversity of nanofabrication techniques. NEL is not only a reasonable nanopatterning technique, but also a good choice of mold manufacturing[55]. Multiple patternings can be performed on a patterned target surface because the electrochemical reaction does not need mechanical force and will not cause any residual stress on a patterned surface. A significant issue to be improved is the reaction time which has limited the productivity of NEL. If the reaction time is decreased to meet the requirement of RTR processing, the combination of RTR process and NEL will offer superior performance in throughput in nanofabrication.

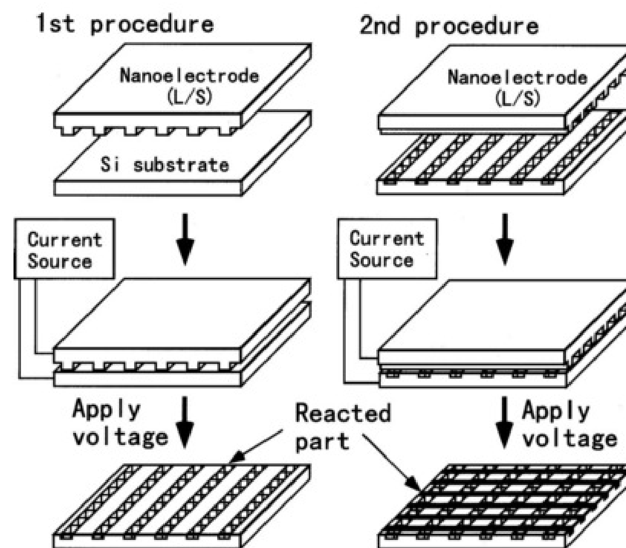

Fig.11 Experimental procedure for multiple patterning by NEL [52] (Reprinted from ref. [52] with permission)

4. Applications of NIL

Nanopatterns (for example, nanoholes, nanotips, nanolines and nanogrids) have been recognised as the essential components in various fields of solar cells, optoelectronic devices, nanoscale sensors and actuators, high density magnetic disks, biological devices, micro fluidics etc. in the future manufacturing. Currently, NIL technology is one of the most successful technologies that are being used commercially for the fabrication of structures at the nanoscale. Due to its economic aspect and mass fabrication capability, NIL technique has

been explored continuously to fabricate devices at both academic and commercial platform. Some of the successful applications of NIL in commercial products are discussed below.

4.1 Optoelectronic devices

Light-emitting diode (LED) in the current era has come out as one of the most efficient energy saving electric appliance. Its efficiency depends upon the accuracy of the nanostructures in it. NIL has been used to fabricate highly efficient nanostructures of varieties shapes to meet the designed efficiency specification of LED [56, 57]. An ideal miniaturized laser light sources at 385 nm, with an emission linewidth less than 0.3 nm is achieved when the semiconductor nanowires is under optical excitation [58]. ZnO is one of the high-efficiency phosphor materials, even at a low-voltage excitation. ZnO nanostructures open the widely applications in the fields of light-emitting display [59, 60], and optoelectronic devices [61]. Plasmonic Bragg mirrors with the nanohole arrays are utilized as sub-micron resolution surface plasmon resonance (SPR) imaging pixels to enhance the resolution, scalability and isolation [3]. Since NIL is a cost effective and mass fabrication technique, NIL can meet all the prerequisites to be used for the fabrication of LED devices.

4.2 Solar cells

Solar energy is regarded as one of the most important renewable energy sources in the future. Although it has been extensively studied for many years, cost effective manufacturing with high efficiency remains a bottle neck in utilizing solar energy. Since the birth of NIL, various groups have been using NIL for patterning nanostructures to be used for solar cells [62, 63]. Ultrathin crystalline silicon solar cells can offer high energy conversion efficiencies, but they face a big challenge of reduced optical absorption in the red and near-infrared spectrum regions [64]. Nanoscale interface in an organic solar cell or periodic photonic nanostructures on thin-film c-Si solar cells are reported to enhance light trapping capabilities and thus improve the power conversion efficiency [64–66].

4.3 Memory devices

One of the finest uses of NIL is in magnetic devices. Such devices in general should have single-domain magnetic structure, high density patterned magnetic media. NIL has been used by various researchers to pattern magnetic nanostructure and investigate the efficiency. For example, Lee et al. have fabricated 60 nm $Ge_2Sb_2Te_5$ nanostructures in order to design a PRAM (Phase-change Random Access Memory) device [67]. UV NIL has been adapted by Yang et al. to pattern the PRAM in nanoscale [68]. Meier et al. have successfully used NIL technique to understand the capacity of next generation memory and logic devices [69]. Currently, commercial companies are on their way to employ NIL technique for the development of next generation semiconductor lithography system which can be used to fabricate memory devices.

4.4 Nanoscale sensors

NIL can produce stable and large-area nanostructures with high throughput. This technology is widely applied in the fabrication of sensor chips, biochemical sensors and waveguide sensors [70–72]. The nanoimprinting technique is able to improve the throughput with low-cost, and enhance the uniformity and sensitivity of the sensors. Moreover, during the nanoimprinting process, the fine adjustment of sensor structures can be achieved by controlling the imprinting conditions [72].

4.5 Hydrophobic surfaces

The functional surfaces with hydrophobicity which are characterized by the 'lotus effect' have attracted many researchers' attention, because the hydrophobic surfaces can exhibit unique properties such as self-cleaning, anti-fogging and drag reduction. UV NIL is an effective tool for mass-producible replication of highly hydrophobic surfaces from plant leaves [73]. The nano-scaled structures of superhydrophobic surfaces have been well reproduced and the contact angle has been increased noticeably using UV NIL[74]. In

addition, the fully-covering hierarchical surface with micro/nanostructures which turn the surface into a super-hydrophobicity is achieved by NIL and modified laser swelling [75].

4.6 Biology application

Biological microarrays such as genomics, proteomics and tissues are essential tools to broaden the understanding of surface-mediated biological behaviours [5]. These biological microarrays have opened the wide application in the field of biosensors, biochips, tissue engineering, and bioassays [76–79]. The ability to precisely immobilize the tiny objects in well-defined patterns has been a significant challenge [80]. NIL is a parallel patterning method to fabricate biological microarrays which will increase the sensitivity and accuracy of the bio-tech devices. It is advantageous in terms of its high throughput, flexibility and simplification and low cost compared with photolithography technique. Capillary electrophoresis chips for DNA separation and nanoelectrochemical transducers for bio-chemical sensor is achieved using NIL [81, 82]. Moreover, NIL and molecular self-assembly have been combined to generate the protein nanopatterns [83].

5. Challenges and the future of NIL

5.1 Comparison of current NIL techniques

Table 1 shows the comparison of current NIL techniques. Thermal NIL and UV NIL can achieve a two-dimensional (2D) or two and a half-dimensional (2.5D) nanoimprint processing. Large-area RTP or RTR thermal NIL and UV NIL is a kind of continuous nanoimprinting method, which offers a superb high-throughput, short imprint time and low cost. Hence, these two techniques are widely used to fabricate nanostructures for commercial production. LADI is a physical nanoimprint process and it offers high resolution, short heating and imprinting time, no need of etching process. 2D nanopatterns with sub-10 nm resolution can be fabricated by using LADI. The power uniformity of the large-diameter laser beam is a big limitation for LADI to process the larger samples. Compared with the

conventional NIL, NEL is categorized as a chemical nanoimprint process, and this approach offers many merits. Firstly, there are no defective patterns caused by mold release because no resist is needed in patterning process. In addition, NEL can be used to produce the flat mold, which eliminates the etching process in mold manufacturing and reduces the costs. Finally, NEL does not deform the target surface during the patterning process and there is no residual stress on the target surface. Hence, multiple patterning for complex nanostructures can be realised by using NEL or combine it with other patterning methods such as EB lithography, SPM lithography. However, the influence of the current and power-on time on the features of anodic oxidation remains unclear. The pattern resolution and uniformity by NEL should be further improved.

Table 1 Comparison of current NIL techniques

Items	Thermal NIL	UV NIL	LADI	NEL
Principle	Physical process	Physical and chemical process	Physical process	Chemical process
Machining capability	2D, 2.5D	2D, 2.5D	2D, 2.5D	2D
Resolution	Width of 70 nm and depth of 40 nm [28]; Width of 0.8–5μm and depth of 1μm [32].	Pitch of 150 nm and height of 35 nm [33]; Pitch of 101 ± 5 nm and height of 274±3 nm [36].	Width of 10 nm and depth of 15 nm [39]; Width of 100 nm and depth of 100 nm [42]; 500 nm [45].	Pitch of 500 nm and depth of 240 nm [51]; Pitch of 500 nm and depth of 70 nm [53]; Width 300 nm and depth of 20 nm[54].
Size of sample	Width of 0.25-2 cm [28]; 100 mm × 100 mm [32].	80 mm width [33]; 4 cm^2 [36].	1.5 mm × 1.5 mm[39]; 1 mm^2 [42]; 10 mm × 10 mm[45].	400 × 300 μm^2 [51]; 6 × 8 mm^2 [53].
Imprinting speed / Imprinting time	0.5 to 1.5 cm/min [28]; 0.1-10mm/s [32]	13 mm/s [33]; 1.8 m/min [36].	Hundreds of nanoseconds [39, 42]	15 to 60 minutes [51]; 30 s [53]; 20 minutes [54].
Machinable materials	Polymer	Photopolymer	Silicon, metal, polymer, semiconductors, dielectrics	Si, organic material, GaAs, anodic oxidation of semiconductor or metal
Etching	No need	No need	No need	Need
Defective patterns caused by mold release	Yes	Yes	Yes	No

5.2 Challenges and the future of NIL

Soon after the invention of NIL as a simple and economical nanofabrication method, NIL was able to draw attention from researchers and engineers and quickly became a popular commercial nanofabrication tool. The reason behind the success of NIL is that NIL is a cost-effective and fast nanofabrication method. However, despite of its commercial success, NIL has several issues, which need to be addressed and resolved in order to fulfil the demand of the dynamic market.

Thermal NIL is a physical nanoimprinting process and UV NIL is a physical and chemical nanoimprinting process. They are both very efficient in nanofabrication on polymer materials and can provide high-resolution structures. The fracture defect generated during demolding process and surface residual stress of the polymer substrates are unavoidable problems in thermal NIL. UV NIL is expected to have the ability to pattern more kinds of materials in addition to the UV-curable resin. LADI is also a physical nanoimprinting process and can be very useful to fabricate nanostructures on silicon, polymer, metals and semimetals. However, the power uniformity of the laser beam (having a large diameter) has been a big limitation in processing bigger samples. NEL is categorized as a chemical nanoimprinting process, and this approach offers many advantages to NEL. Nevertheless, NEL is a time-consuming process. It is because of the fact that NEL takes finite amount of time in order to complete the chemical process. Thus, if the reaction time can be reduced, NEL can eventually replace conventional EBL technique. Provided the reaction time is reduced down to few seconds or even lower than this, RTR NEL can be designed to fabricate nanostructures with higher resolution and throughput. Hence, the influences of the current and power-on time on the features have to be studied deeply. The formability mechanism in NIL process is still not fully understood, and the effect of the imprinting parameters such as temperature, loading force, aspect ratio and imprinting velocity on formability requires a

thorough and systematic study. To make up for the inconsistency and turbulence of experimental research, molecular dynamics (MD) simulation is an effective tool for studying material behaviour and thermodynamic properties at the atomic level[84]. In addition, the attainable resolution and the uniformity of patterns should be improved.

One of the disadvantages of NIL over other nanofabrication techniques is the flexibility of patterning. For example, if the designed pattern is changed slightly, the mold has to be remanufactured. NIL has been very successful in nanopatterning regular shapes. However, NIL might not be a suitable tool in fabrication of complex structures. Integrating NIL with other existing nanofabrication techniques can be helpful in order to overcome such issues. Each existing nanofabrication methodology has its advantages and imperfections. The key is to find a stage and provide full play to the advantages of different nanotechnologies, which consist of EBL, AFM lithography, SPM lithography, Focused Ion Beam (FIB), optical lithography, NIL, molecular self-assembly, etc [85–87]. The integrated use of the physical and chemical nanopatterning approaches is highly anticipated to improve the pattern resolution, uniformity and throughput, as well as to fabricate desired 3D nanostructures.

In the past decades, the semiconductor and circuitry industries have witnessed the rapid development of nanoimprint lithography, whilst research on NIL in biological application is limited and worthy of an in-depth study in the future. The development of biochips, artificial organs, diagnostic system, and fundamental research in cell biology will be benefited from fabricating protein arrays at a molecular level. These potential applications will attract researchers to push nanoimprint lithography forward at a resolution of 10 nm or less in the future.

6. Concluding Remarks

Despite of the late invention of nanoimprinting technique, NIL has quickly broken through the barrier of laboratory fabrication scale to industrial production and become one of the most

employed commercial platform for nanofabrication. The reasons are the simplicity it adopts, the resolution of the structures it can offer, high throughput and the cost-effectiveness involved in the fabrication approaches.

Through this article, an overall view of the major commercially successful NIL techniques (Thermal and UV) along with the still-in-laboratory but promising (LADI and NEL) NIL techniques has been provided. The machining mechanism, components and advances of each method have been discussed in order to provide a clear understanding of the techniques. Moreover the methods have been compared in terms of machining capability, speed, sample materials and size, resolution of the fabricated structures, etc. Through this direct comparison, readers can quickly access to the advantages and limitations of each technique.

Despite of the huge commercial success, there are still many challenges which lie in NIL fabrication processes and some of the key points have been discussed in this article. Possible solutions to those challenges have been discussed in order to provide a multi-directional approach for the researchers to think and pursue on the issues.

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