

FINAL PROJECT REPORT

“A HYBRID NANO-IMPRINTING LITHOGRAPHY METHOD FOR NANO-PATTERNING BASED ON INFRARED PULSED LASER HEATING”

Name of Principal Investigator: Professor Yung-Chun Lee

E-mail address: yunglee@mail.ncku.edu.tw

Institution: Department of Mechanical Engineering, National Cheng Kung University, Tainan, Taiwan 701

Mailing address: Department of Mechanical Engineering, National Cheng Kung University, #1 University Rd., Tainan, Taiwan 701. **Phone:** +866-6-2757575 ext. 62177, **FAX:** +866-6-2352973.

Abstract

In this research project we present a novel method of nano-imprinting which adopts important features of conventional nano-imprinting lithography (NIL) and the newly developed laser-assisted direct imprinting (LADI) method. It utilizes an Nd-YAG pulsed laser of wavelength 1064 nm which can easily penetrate and also heat up a silicon mold which is pressed against a resist layer deposited on a substrate. The fast rising temperature in the silicon mold can momentarily melt the resist layer so that the mold is imprinting into the resist layer. After the pattern is transferred, standard nano-imprinting lithography processes can be applied to the substrate for nano-fabrication. This new method has several advantages over existing nano-imprinting methods mostly due to the fast heating-up of silicon mold by high intensity IR laser pulse and therefore has no thermal drifting problem. Both the theoretical modeling and experimental results of this novel IR-laser assisted imprinting method will be investigated.

Report Documentation Page

Form Approved
OMB No. 0704-0188

Public reporting burden for the collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington VA 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to a penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number.

1. REPORT DATE 05 SEP 2006		2. REPORT TYPE Final Report (Technical)		3. DATES COVERED 13-06-2005 to 21-09-2006	
4. TITLE AND SUBTITLE Hybrid Nano-Imprinting Lithography Method for Nano Patterning Based on Infrared Pulsed Laser Heating				5a. CONTRACT NUMBER	
				5b. GRANT NUMBER	
				5c. PROGRAM ELEMENT NUMBER	
6. AUTHOR(S) Yung-Chun Lee				5d. PROJECT NUMBER	
				5e. TASK NUMBER	
				5f. WORK UNIT NUMBER	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) National Cheng Kung University, #1 University Road, Tainan 701, Taiwan, TW, 701				8. PERFORMING ORGANIZATION REPORT NUMBER AOARD-	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) The US Research Laboratory, AOARD/AFOSR, Unit 45002, APO, AP, 96337-5002				10. SPONSOR/MONITOR'S ACRONYM(S) AOARD/AFOSR	
				11. SPONSOR/MONITOR'S REPORT NUMBER(S)	
12. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution unlimited					
13. SUPPLEMENTARY NOTES					
14. ABSTRACT In this research project we present a novel method of nano-imprinting which adopts important features of conventional nano-imprinting lithography (NIL) and the newly developed laser-assisted direct imprinting (LADI) method. It utilizes an Nd-YAG pulsed laser of wavelength 1064 nm which can easily penetrate and also heat up a silicon mold which is pressed against a resist layer deposited on a substrate. The fast rising temperature in the silicon mold can momentarily melt the resist layer so that the mold is imprinting into the resist layer. After the pattern is transformed, standard nano-imprinting lithography processes can be applied to the substrate for nano-fabrication. This new method has several advantages over existing nano-imprinting methods mostly due to the fast heating-up of silicon mold by high intensity IR laser pulse and therefore has no thermal drifting problem. Both the theoretical modeling and experimental results of this novel IR-laser assisted imprinting method will be investigated.					
15. SUBJECT TERMS Nanotechnology, Lasers					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES 11	19a. NAME OF RESPONSIBLE PERSON
a. REPORT unclassified	b. ABSTRACT unclassified	c. THIS PAGE unclassified			

I. BACKGROUND AND INTRODUCTION

Nano-imprinting lithography (NIL) has been developed over a decade [1-5] and is now a promising method for nano-pattern transformation. Figure 1 illustrates the basic concepts of nano-imprinting lithography which includes a mold with some nano-features on its surface, an etching resist layer and a sample substrate. By heating up the resist layer above its glass transition temperature (T_g), the mold can imprint into the resist layer and form a pattern. The nano-pattern is transferred to the resist layer and the substrate by subsequent standard lithography processes. However, conventional NIL methods take quite some times in the thermal heating and cooling processes, and thermal expansion and contraction will affect the dimension accuracy. An alternative approach is to use UV-cured photo-resists and UV light sources. But it requires a UV-transparent mold made from glass or quartz, and it is not easy to fabricate nano-scaled features on these materials. In either approaches, the nano-imprinting lithography still needs subsequent chemical etching processes to finally transfer the patterns to the substrates, usually silicon, and therefore is still complicated and time consuming.

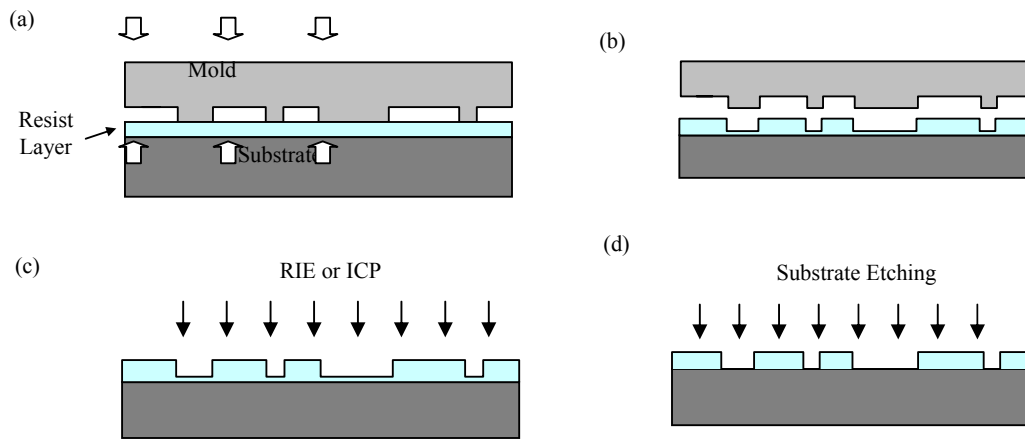


Figure 1. Nano-Imprinting Lithography (NIL) processing

In this work, we propose a new nano-imprinting lithography based on infrared pulse laser heating. The idea comes from the Laser Assisted Direct Imprinting (LADI) method proposed by S.Y. Chou et al. in 2002 [6], but with some substantial modifications. Figure 2 illustrates the basic idea of LADI. A short laser pulse first radiates on the sample substrate through a transparent quartz mold which is pre-loaded against the sample. The quartz mold has some pre-fabricated nano-scaled features. After absorbing the laser energy, near-surface

materials melt and a laser-induced molten layer is formed, which allows the mold to emboss into the sample directly. Subsequent cooling and solidification of the molten layer complete the nano-pattern transformation. Nanostructures are directly formed without any chemical etching. However, the LADI shares the same difficulty as in UV-cured nano-imprinting lithography, that is, the fabrication of a quartz mold with nano-scaled features is quite difficult. Furthermore, the materials subjected to LADI processing will undergo very drastic temperature rising and cooling in short time and phase transitions from solid to liquid and liquid to solid. Such drastic temperature variations and phase transitions may cause some defects and variations in material properties, and may be adverse to their applications.

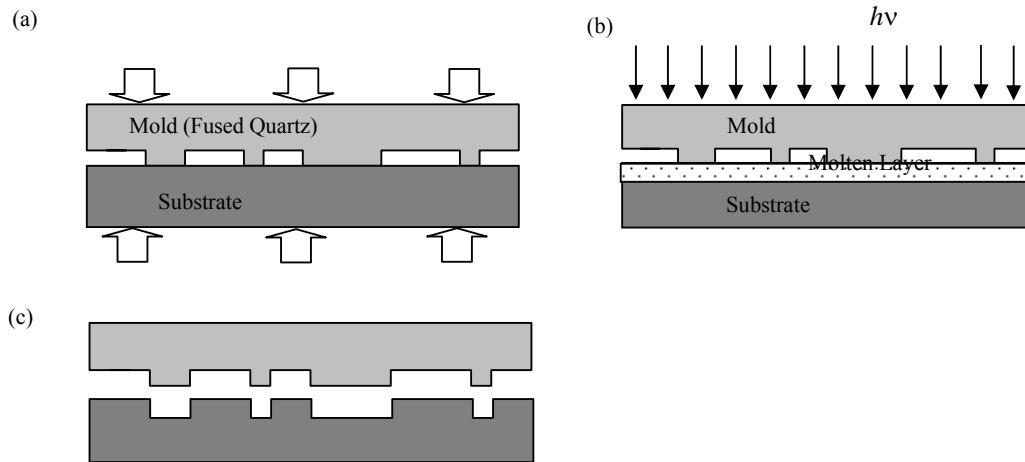


Figure 2. Laser Assisted Direct Imprinting (LADI) processing.

Therefore a new nano-imprinting lithography method is proposed here based on the concept of LADI. As shown in Fig. 3, the fundamental differences are that a polymer resist layer is placed in between the mold and the substrate and a pulsed laser with wavelength in the infrared spectrum is used as a heating source. Since silicon crystals are partially transparent to infrared light, a silicon mold can be used. The infrared laser pulses pass through the silicon mold and some of the laser pulse energy is absorbed by the silicon and the polymer resist layer. The absorbed laser energy turns into heat and results in temperature rising. When the temperature in the polymer layer is above its glass transition temperature, the pre-loaded force applied on the mold against the substrate presses the mold into the resist layer and creates the nano-pattern on the resist layer. Subsequent etching on the resist layer and the substrate will then complete the nano-imprinting process.

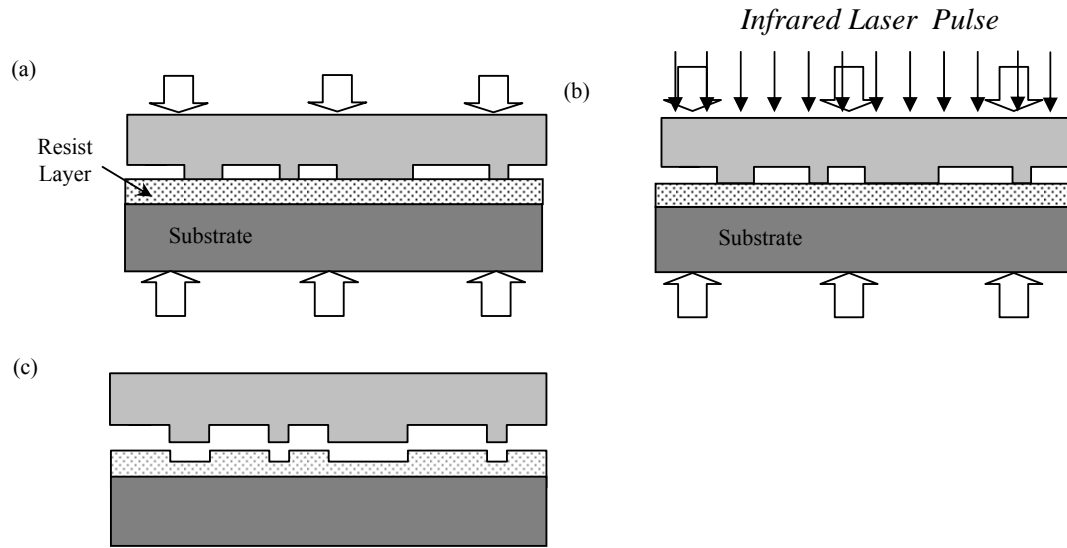


Figure. 3. Nano-Imprinting Lithography with a Infrared Pulsed Laser

II. THEORETICAL ANALYSIS

A. Optical Properties of Silicon in IR Spectrum

The optical properties of a material are usually characterized by optical constants (n , k) or the complicated number $n+jk$, where n is the refraction index and k is related to absorption of light energy in the material. Figure 4(a) shows the dependence of silicon's n and k values on light wavelength. As can be seen from Fig. 4(a), the k value becomes very small for light in infrared spectrum. This is well known and has been used for double-side alignment equipments for photo-lithography. In other words, the infrared light can easily penetrate through silicon with little energy absorption. Figure 4(b) shows the reflection, transmission, and absorption coefficients of a 600 μm thick silicon wafer when subjected to a normally incident light beam. Since a 1064 nm Nd:YAG laser will be applied in this work for laser-assisted hot embossing, a portion of Fig. 4(b) with wavelength centered at 1064 nm is enlarged and shown in Fig. 4(c). At the wavelength of 1064 nm, about 30% of incident light energy is absorbed by the silicon wafer and is transferred into heat to heat up the silicon.

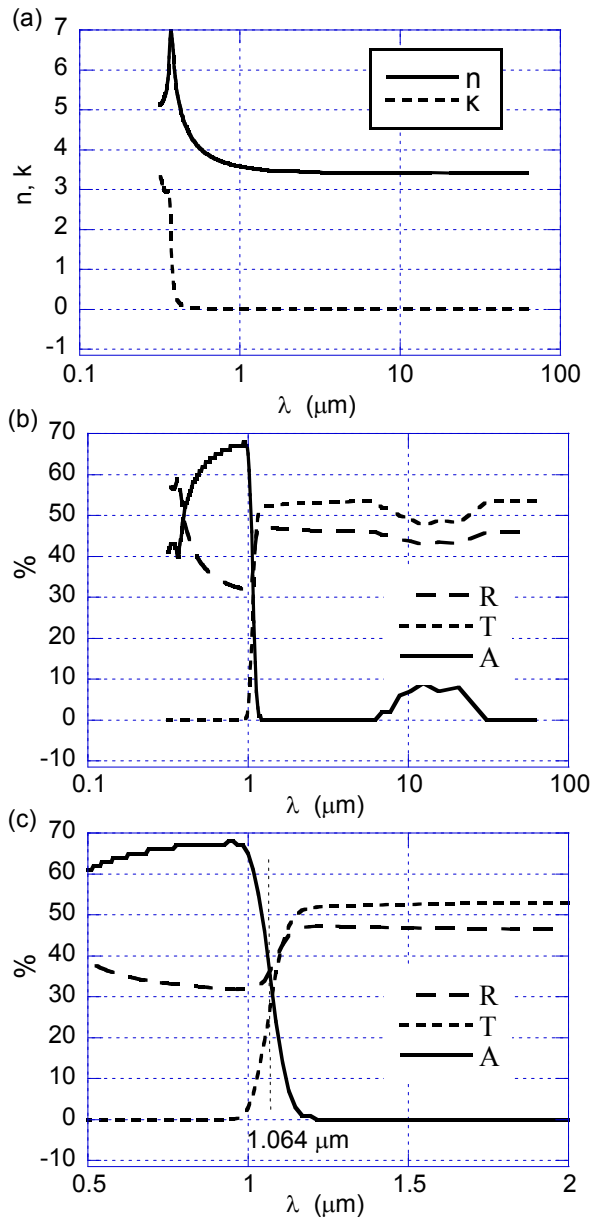


Figure 4. (a). Optical constants (n , k) as functions of wavelength, and (b) the reflection (R), transmission (T), and absorption (A) coefficients for a 600 μm thick silicon wafer.

B. Laser Heat Absorption and Temperature Rising

Figure 5(a) shows the configuration under consider for IR-laser heating and nano-imprinting. A silicon mold of 500 μm thickness with some nano-scaled features is pre-loaded against a resist layer deposited on a 500 μm thick silicon wafer. The resist layer is PMMA of thickness 300 nm. A Nd:YAG laser is normally incident to the silicon/PMMA/Silicon

assembly form the left silicon mold. The Nd:YAG laser is a pulse laser with a pulse duration of 6~7 ns and a laser fluence of 30 J/cm^2 . Due to the n and k values of silicon at the wavelength of 1064 nm, a portion of the laser energy is absorbed by the silicon mold and cause temperature rising in the silicon mold, as well as the PMMA layer and silicon substrate. As shown in Fig. 5(b) is the temperature risings in the silicon mold, the PMMA layer, and the silicon substrate due to the laser heating of the 1064 nm Nd:YAG laser.

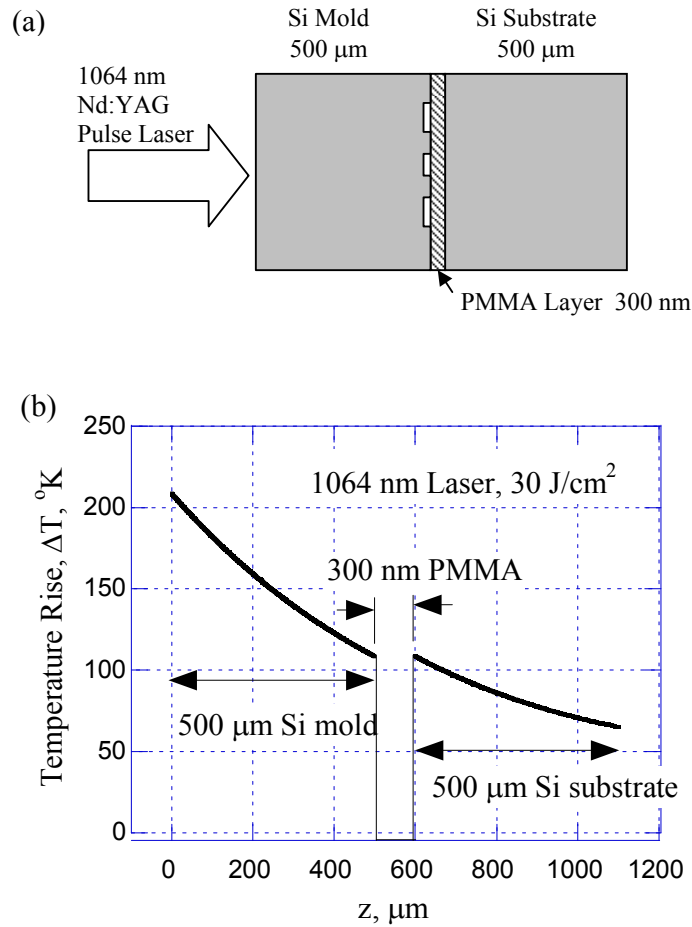


Figure 5. (a) The configuration of IR laser-assisted imprinting, and (b). the temperature rising caused by 1064 nm Nd:YAG pulse laser heating.

The calculation of the laser energy absorption is based on standard optics concerning the interaction between light and a system of 3 dielectric materials, that is, the silicon mold, the PMMA layer, and the silicon substrate. The corresponding optical constants for silicon and PMMA are used [7], as well as their thermal properties for determining the final temperature increases. Since the pulse laser have a very short pulse duration of 6~7 ns, the heat transfer

within such a short period of time may be neglected. In other words, the energy of the laser pulse is distributed into the Si/PMMA/Si system momentarily.

As can be seen from Fig. 5(a), the temperature rising in silicon mold and silicon substrate is basically following the general trend of laser-material interaction, that is, it has an exponentially decaying into the materials. While in the PMMA layer, since the polymer material does not absorb the laser energy, the temperature rising is virtually zero. However, under the specific laser fluence of 30 J/cm^2 , the temperature rising at the silicon/PMMA interface is approximately $100\sim 110 \text{ }^\circ\text{C}$. Given the room temperature as $25 \text{ }^\circ\text{C}$, the temperature can reach $125\sim 135 \text{ }^\circ\text{C}$. The heat conduction between silicon and PMMA can quickly bring heat into the PMMA layer. Once the temperature of PMMA goes beyond its T_g point, which is about $110 \text{ }^\circ\text{C}$, the PMMA layer becomes soft and the silicon mold and impinge into the PMMA layer to form the pattern transformation. Detail calculations on the heat transfer and the final temperature can be quite complicated since it also involves the impinging movement of the mold into the PMMA layer. However, it is reasonable to believe that there is a very good chance to achieve the goal of IR laser-assisted imprinting based on the laser heating method.

III. EXPERIMENTAL RESULTS

From previous analysis and calculation, it is feasible to apply the infrared pulse laser (1064 nm Nd:YAG) for heating up the silicon mold and for completing the embossing on PMMA layer. This section will detail the experimental setup, the mold preparation, and the imprinting results. The two most important parameters in the imprinting process are the laser fluence (J/cm^2) and the contact pressure (Pa) between the mold and the resist-layer/substrate. The two parameters will be closely monitored in the experiments.

A. Experimental Setup

The laser source used in this work for IR laser-assisted imprinting is a Q-switched Nd:YAG laser with 4 different wavelengths, 1064, 532, 355, 266 nm. Only the 1064 nm wavelength is used in this work. The maximum pulse energy at 1064 nm is 800 mJ/pulse and the maximum pulse repetition rate is 10 Hz. The nominal beam size of output laser beam is 8 mm in diameter. For increasing the laser fluence, an optical lens system is applied to reduce the beam size down to 1 mm or so, to achieve very high laser fluence.

Figure 6 shows schematically the experimental setup for the IR laser assisted nano-imprinting. The silicon mold is backed with a glass plate for applying contact pressure. A load

cell is mounted in the test fixture for measuring the loading force and subsequently to obtain the contact pressure by dividing the force by contact area of the mold. The laser fluence can be directly controlled and hence varied from the laser controller, while the applied loading force and hence the contact pressure is controlled by a linear actuator.

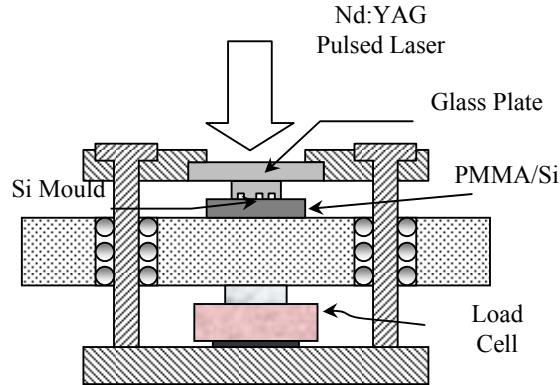


Figure 6. Experimental setup for IR laser assisted nano-imprinting.

B. Mode Preparation

The nano-scaled silicon mold was fabricated with E-beam lithography using an electron beam writer (Raith) attached to a SEM (JEOL SEM-JSM 7000). A 20 nm thick Cr film together with a 75 nm thick Au film was deposited as the etching mask, where the pattern was etched by RIE after definition and completed by lift off method. The size of the mold is $100\ \mu\text{m} \times 100\ \mu\text{m}$ containing a 10×10 square array, where each square is composed of parallel lines of 300 nm in width and $100\ \mu\text{m}$ in length with 1 μm interval spacing as shown in Fig. 7(a) and Fig. 7(b) for an enlarged and tilted view. Figure 7(c) is an AFM image showing that the depth of the features is 300 nm and the side-walls are rough.

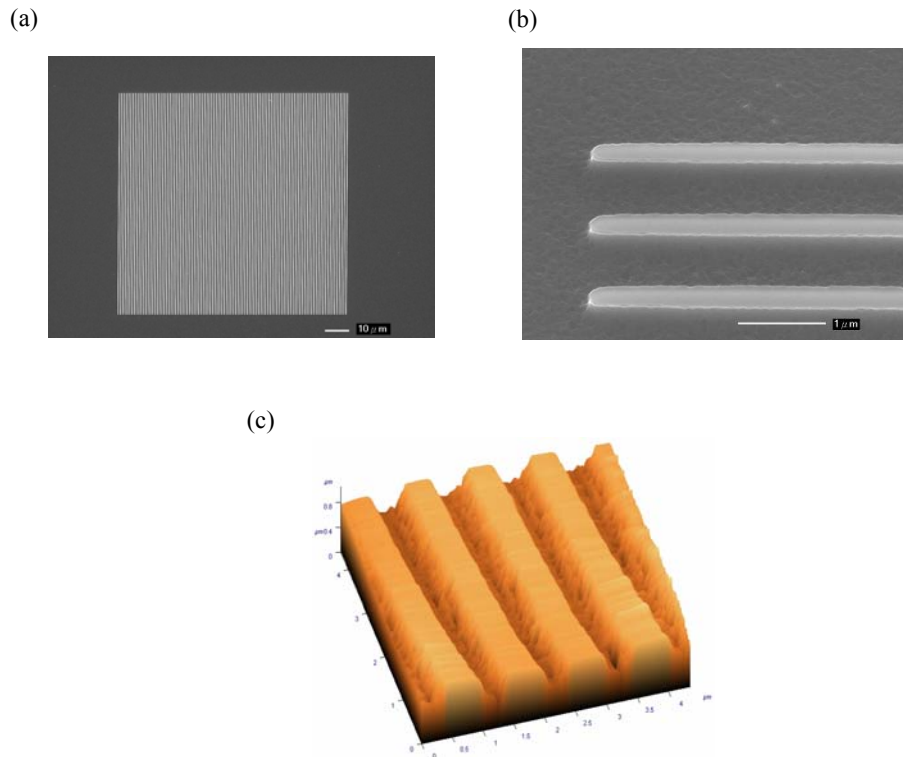


Figure 7. The nano-scaled silicon mold fabricated by E-beam lithography, (a). SEM micrograph, (b). an enlarged view, and (c). 3D topology measured by AFM.

C. Experimental Results

The fabricated nano-scaled silicon mold is mounted into the testing fixture and the pulse laser is fired onto the Si/PMMA/Si assembly under a give pressure. After one laser shot, the PMMA/Si-Substrate is separated from the silicon mold and is retrieved from the fixture. The imprinted sample is observed using a SEM, and the pictures are displayed in Fig. 8. One can see the nano-scaled feature are all transferred to the PMMA layer now and therefore a complete nano-imprinting process is achieved within one laser shot.

The imprinted samples are examined under AFM to measure the imprinting depth. A number of experimental tests are carried out under several different laser fluence and contact pressure. The correlation between the imprinting depth, the applied contact pressure, and the laser fluence is experimentally obtained and is displayed in Fig. 9. It is observed that there is

an energy threshold around 30 to 40 J/cm² and after that threshold the imprinting depth is mainly controlled by the contact pressure.

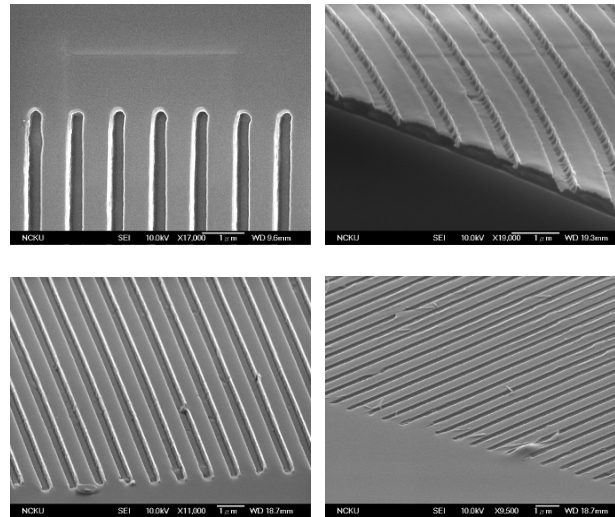


Figure 8. The SEM micrographs of the PMMA/Si after IR laser assisted nano-imprinting.

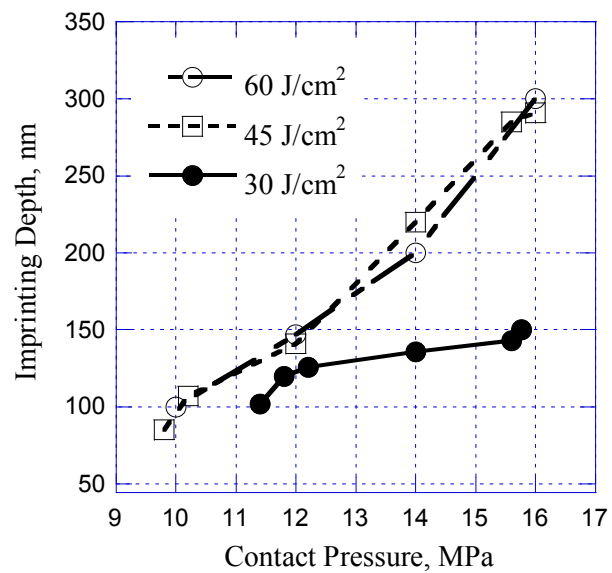


Figure 9. The correlation between the imprinting depth, applied contact pressure, and laser fluence obtained experimentally.

IV. CONCLUSION

We have proposed a new nano-imprinting method based on the infrared pulse laser heating process. The main idea is to take the advantage of optical properties of a silicon in the near infrared spectrum region so that one can heat up the silicon mold quite efficiently using a pulsed Nd:YAG laser at 1064 nm wavelength. A theoretical analysis and calculation indicate the feasibility of this method and point out the necessary laser fluence to accomplish the goal. The sample is a 300 nm PMMA layer deposited on a silicon substrate. Experimental works have been carried out which includes building up the experimental setup and testing fixture, preparing the molds using E-beam lithography, and actually perform the IR laser assisted nano-imprinting processes under various conditions. Experimental data demonstrate that the proposed method indeed work very well for transferring nano-patterns from the mold to the PMMA resist later. Quantitative information is also obtained experimentally which indicates that there exists an energy threshold in the IR laser assisted nano-imprinting. Once beyond the energy threshold, the imprinting depth is mostly dominated by the applied contact pressure. The detail mechanism underlying the proposed imprinting method is quite completed and is beyond the scope of this work.

The most important advantage of this proposed IR laser-assisted nano-imprinting method is the fast speed and throughput. The whole process can be done just in one laser shot. Secondly, the method uses simply silicon mold, which is much easier to prepare as compared to other transparent materials such as glass and quartz, since the technologies are quite mature in semiconductor industries. Finally, the imprinting setup is quite simple and can be implemented in a laboratory easily. However, the disadvantage of the proposed method is that it requires a very high energy level or high laser fluence, and therefore is difficult for large-area nano-imprinting. This is due to the fact that most of the energy is absorbed by the bulk silicon mold and these silicon materials right in contact with the PMMA layer are not efficiently laser-heated. The problem can be overcome by adjusting the optical properties of silicon in the thickness direction so that the laser energy absorption is more concentrated in regions close to the PMMA layer. Another direction will be using polymer materials with lower T_g value for the resist layer. Related studies are under way.

REFERENCES

- [1] S.-Y. Chou, P.R. Krauss, and P.J. Renstrom, "Imprint lithography with 25-nanometer resolution," *Science*, vol. 272, pp. 85-87, 1996.
- [2] L. Guo, P.R. Krauss, and S.-Y. Chou, "Nanoscale silicon field effect transistors fabricated using imprint lithography," *Applied Physics Letters*, vol. 71, pp. 1881-1883, 1997.
- [3] S.-Y. Chou and P.R. Krauss, "Imprint lithography with sub-10 nm feature size and high throughput," *Microelectronic Engineering*, vol. 35, pp. 237-240, 1997.
- [4] W. Zhang and S.-Y. Chou, "Fabrication of 60-nm transistors on 4-in. wafer using nanoimprint at all lithography levels," *Applied Physics Letters*, vol. 83, pp. 1632-1634, 2003.
- [5] W. Wu, , J. Gu, H. Ge, C. Keimel, S.-Y. Chou,"Room-temperature si single-electron memory fabricated by nanoimprint lithography," *Applied Physics Letters*, vol. 83, pp. 2268-2270, 2003.
- [6] S.-Y. Chou, C. Keimei, and J. Gu, "Ultrafast and direct imprint of nanostructures in silicon," *Nature*, vol. 417, pp. 835-837, 2002.
- [7] Donald Rapp, Jet Propulsion Laboratory, California Institute of Technology "Thermo-Optical Properties of Silicon", <http://www.gps.caltech.edu/genesis/Thermal-Coll.html>.

Publications as Results of this Project:

Referred Journal Papers

1. Chun-Hung Chen, Chuan-Pu Liu, Yung-Chun Lee, Fei-Bin Hsiao*, Cheng-Yu Chiu, Ming-Hung Chung, Ming-Hsueh Chiang, Aug. 2006, "IR-Laser Assisted Micro/Nano-Imprinting," *Journal of Micromechanics and Microengineerings*, Vol. **16** (8), pp. 1463-1467. (EI, SCI- 2.499, 4/110 in Mechanics category, 7/27 in Nanoscience/Nanotechnology category, 2/52 in Instruments & Instrumentation, 23/178 in Material Science-Multidisciplinary category).
2. Yung-Chun Lee, Chun-Hung Chen, and Chuan-Pu Liu, "A Hybrid Nano-Imprinting Lithography Based on Infrared Pulsed Laser Heating," *IEEE Review of Advancements in Micro and Nano Technologies* (in press) (SCI)
3. Cheng-Hsin Chuang, Yung-Chun Lee, Huei-Sheng Huang, Fei-Bin Hsiao, You-Ming Hsu¹, Kai-Hsuan Wang, Ling Ding, and Hui-Ju Hsu, "3D Microstructures Array Single-cell-based DEP Chip for Studying Apoptosis of U937 & A431 Cells," *IEEE Review of Advancements in Micro and Nano Technologies* (in press) (SCI)

Invited Conference Talks

1. ***(Invited Speaker,)** Yung-Chun Lee, Chun-Hung Chen, and Chuan-Pu Liu, "A Hybrid Nano-Imprinting Lithography Based on Infrared Pulsed Laser Heating," *2006 IEEE-NEMS Conference*, Zhuhai, China, Jan.18-21, 2006.

Patent

1. Chuan-Pu Liu, Yung-Chun Lee, Fei-Bin Hsiao, Cheng-Yu Chiu, Chun-Hung Chen, "Laser Assisted Hot Imprinting Technique", Patent # I 254367 of Republic of China, 2005.