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Molecular Dynamics Study of the Quenching Effect on Direct Nanoimprint of Gold

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Abstract

Nanoimprinting of gold has the potential to make significant impact in the fabrication of miniaturized devices including optical devices, photonic crystals and biochemical sensors. The deformation behavior and recrystallization of gold structures has profound impact on the accurate replication of the imprinted pattern. An important step in the nanoimprint lithography (NIL) process is the cooling of the mold and resist assembly to room temperatures. The rate of cooling, commonly termed as quenching determines the final shape of the NIL features. Molecular dynamics simulations were implemented to study the nanoimprinting of gold substrates using a silicon mold. Simulations were conducted at room temperature (298 K), gold recrystallization (473K) and melting point (1000K) temperatures. The effect of different cooling rates (quenching) on the deformation behavior and spring back of gold was investigated. These include 0.01ns, 0.05ns, and 0.2ns time steps of quenching at 1fs per time step. The modified embedded atom method (MEAM) potential was used for the system molecules. Fast cooling at 298 K and 473 K whereas, intermediate cooling at 1000 K resulted in good pattern transfer. High fidelity nanoscale features were achieved with a gradient cooling proportional to the initial heating temperatures. The findings of this research provide insight on the effect of quenching in the material deformation and spring back during direct metal nanoimprinting.

Keywords

Molecular Dynamics; Nanoimprint Lithography; Spring back Effect; Modified Embedded Atom Method; Quenching.

1. Introduction

Nanoimprint lithography has emerged as a cost effective and high throughput manufacturing process to produce nanoscale patterns. Nanoscale patterns have applications in miniaturized electronic components, optical devices, and biomedical research [1]. In nanoimprinting, a mold with a nano pattern is pressed against the silicon wafer that is spin-coated with polymer resist to achieve a pattern replication [2]. Traditionally, nanoimprint lithography was developed for nanopatterning of polymers but, nanoimprinting of metals is vital due to their potential applications. Generally, metal patterning process involves lithography followed by secondary processes such as electroplating to deposit a metal layer within the nanopatterns. Metal nanopatterning also requires post-processing operations such as etching or lift off which is expensive and time consuming [3]. Nanoimprinting is a very attractive technique due to its single step procedure. Metal direct nanoimprinting has applications in gratings for optical components, electromagnetic relays, and biomedical implants [4]. Nanoimprinting has been applied to fabrication of Micro Electro Mechanical Systems (MEMS). Among metals, gold is an attractive candidate for nanoimprinting because it has extraordinary electrical, thermal properties, outstanding chemical stability and is amenable to forming processes. Moreover, it has widespread use in diverse fields such as electronic, photonics, and medical devices. Recently, gold has been used in nanofabrication of sensors for medical applications to detect biomolecules for disease diagnostics [5].

Nanoimprinting has been used to fabricate gold and silver nanopatterns by forging the gold and silver plates against the previously patterned silicon wafer for photonics [6]. Researchers have studied various NIL process steps such as lift-off process and their effect on the resolution of the final imprinted nanopatterns [4]. In the case of polymers, the effect of temperature on the imprint force was studied, wherein a decrease in the imprint force was observed with increase in the temperature [7, 8]. The Finite Element Method was used to explore the mechanism of nanoimprinting and study the effect of velocity and temperature on the mold deformation [9]. However, FEM is limited to the macroscale phenomenon and the continuum approach does not give insights at the nanoscale. Molecular dynamics simulations deal with atomistic models and are suitable to explore a phenomenon at nanoscale regime [10]. Several researchers have studied the nanoimprinting phenomenon with respect to mold geometry, temperature and velocity effects on the polymer filling mechanism during nanoimprinting [11, 12]. In addition, studies have been conducted on dislocation behavior of gold during nanoindentation with respect to various process conditions [13]. Recently, research has been done to investigate nanoimprinting mechanism of copper substrate [14]. However, the effect of cooling cycle during nanoimprint of gold is under explored and needs further study.

This paper focusses on the quenching effect during the cool down phase of the nanoimprinting with a silicon mold and gold substrate. Quenching effect is the cooling phase after pattern transfer and is vital to the accuracy of the nanopattern transfer. This affects the spring back phenomenon of the gold imprint and directly impacts the dimensional accuracy of the nanopattern transfer from the silicon mold to the gold substrate.

2. Method

In this research, molecular dynamics simulations were performed using Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) open source code [15]. LAMMPS is well-known for its robustness for parallel computing and compatibility with most force fields. Open Visualization Tool (OVITO) platform was used to visualize the simulations and analyze the results [16]. The simulations were performed on the Extreme Science and Engineering Discovery Environment (XSEDE) resource [17]. Figure 1 shows the molecular dynamics model and nanoimprinting stages for gold substrate and silicon mold. The gold substrate had 11250 atoms with dimensions of 10 nm x 6 nm x 3 nm. The Si mold had 3090 atoms with a tooth depth of 3.5 nm and tooth width of 3.5 nm. Periodic boundary conditions were applied in both x and y directions. Two layers of atoms i.e. 1125 at the bottom of the gold substrate were fixed. All the simulations were conducted at a velocity of 55 m/s and temperatures of 298 K, 473 K, and 1000 K, respectively. The cooling cycle was varied at 10000 (0.01ns), 50000 (0.05ns) and 200000 (0.2ns) time steps for each temperature. The Modified Embedded Atom Method (MEAM) potential was used to simulate atomic force interactions.

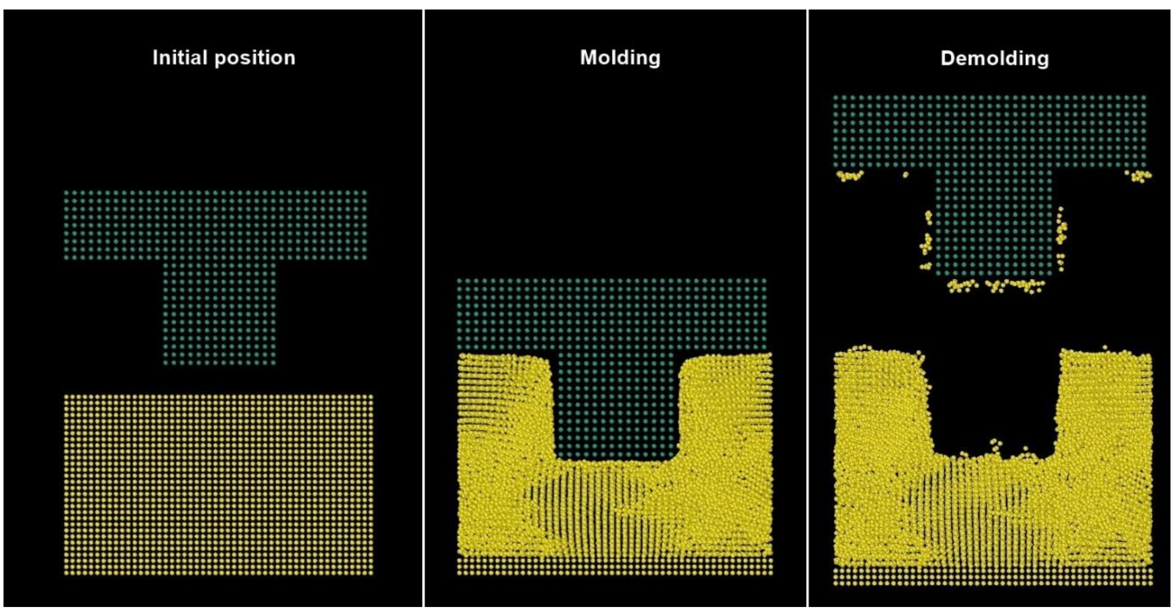

Figure 1: Molecular dynamics model for nanoimprinting of gold substrate with silicon mold

Figure 2 represents the phases of nanoimprint process followed for gold substrate and silicon mold. The nanoimprint process was divided into 5 steps which include heating the system, molding, cooling the system (quenching), relaxation of the system, and demolding. Among all these steps in nanoimprinting, the cooling phase has a significant influence on the final shape and accuracy of the transferred pattern. In this research, studying the dynamic behavior of substrate during the cooling steps is important for successful pattern transfer.

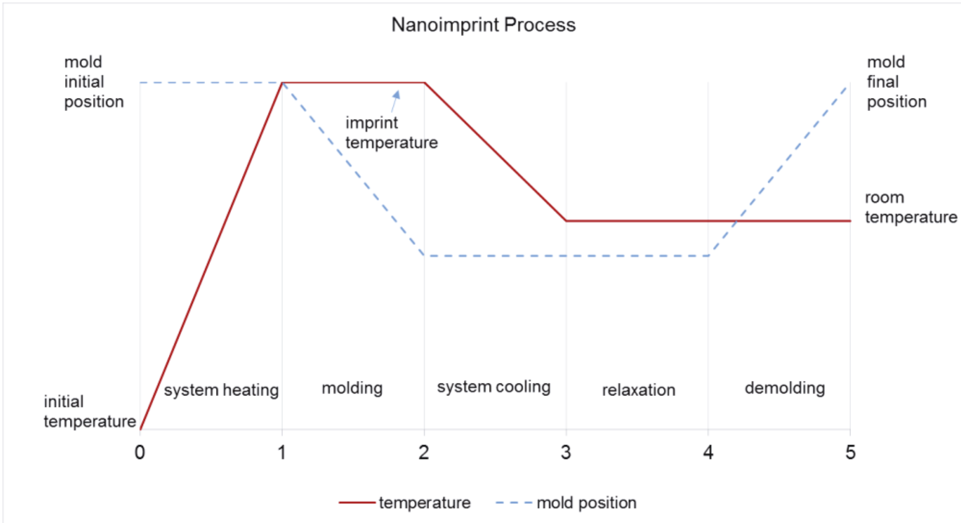

Figure 2: Phases in the nanoimprint (NIL) process

3. Results and Discussion

The simulations were performed for gold substrate and silicon mold at three temperatures of 298K (room temperature), 473K (recrystallization temperature), and 1000K (temperature close to melting point of gold). For each temperature, the cooling time was varied at three ranges such as 10000, 50000 and 200000 time steps, respectively. These variations in the cooling times were conducted to understand the rapid cooling or quenching effect on the spring back phenomenon and imprint quality. The spring back in the vertical (depth) direction was calculated by equation 1.

$$R = \frac{L - L'}{L} * 100 \tag{1}$$

Where, R is the recovery or spring back value in the depth direction, L is initial dimension during molding, and L' is final dimension after demolding. Figure 3 depicts the effect of cooling time on the spring back observed in the gold substrate for the depth direction. The negative values shows the presence of spring back whereas positive value indicates the lack of spring back effect. Figure 3 shows that spring back exists at 298K and 473K whereas, there is no spring back observed at 1000K. At 1000K the temperature is high enough to permit motion of the gold atoms during the imprinting phase and thereby minimal residual stresses exist to offer spring back phenomenon.

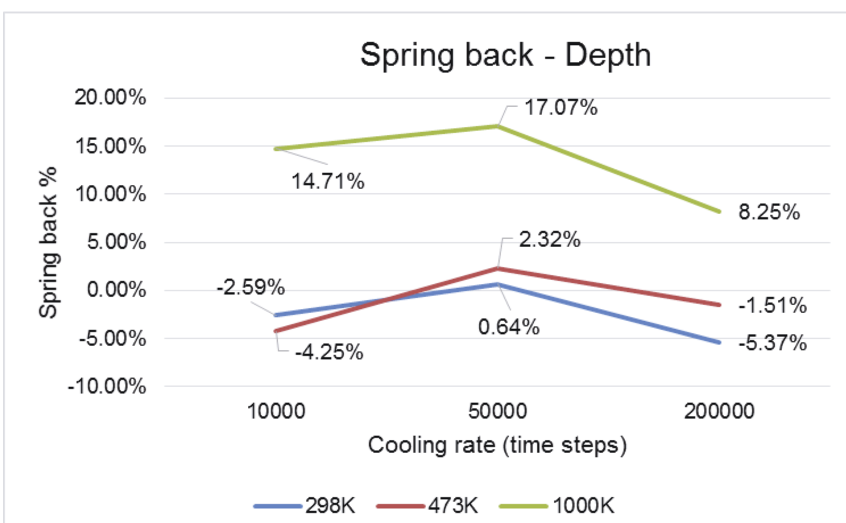

Figure 3: Spring back observed in the vertical (depth) direction.

Figure 5, 6 and 7 demonstrate the deformation of the gold substrate at 298K, 473K and 1000K for different cooling cycles. Figure 5 shows that as the cooling cycle time increases from 10000 to 200000 time steps the spring back is reduced. This can be attributed to the increase in the contact time of mold and substrate during cooling cycle allowing relaxation of the gold atoms. The relaxation of the residual stress during the imprinting process allows the imprinted cavities to become wider and deeper after demolding.

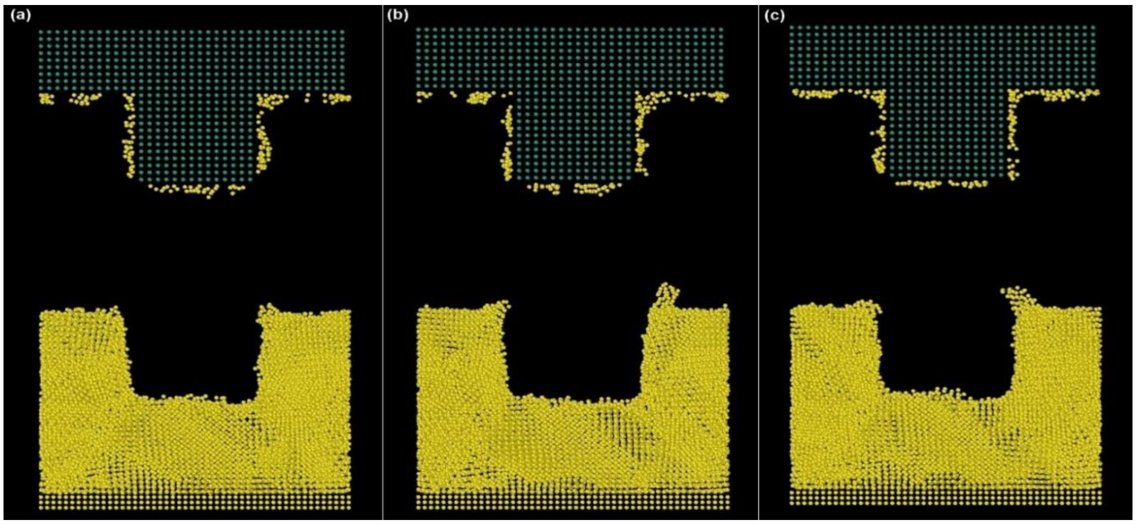

Figure 5: NIL simulation at 298 K with cooling cycles at (a) 10000 time step (b) 50000 time (c) 200000 time step

Figure 6 shows the differences in the imprinting pattern at the recrystallization temperature (473K) for different cooling time steps. An increase in the cooling cycle time from 10000 to 50000 time steps resulted in lower spring back. This can be attributed to the recrystallization of the gold atoms which allow them to reorient and lower the residual stresses within the substrate. However, prolonged cycle times result in the adhesion of the gold atoms to the mold which results in a lower quality imprint pattern. Figure 6 shows that as the cooling cycles increase to 50000 and 200000 time steps the more gold atoms adhere to the mold caused wider and distorted imprints. Also, the gold atoms on the top surface of the substrate are torn apart when the mold releases from the substrate.

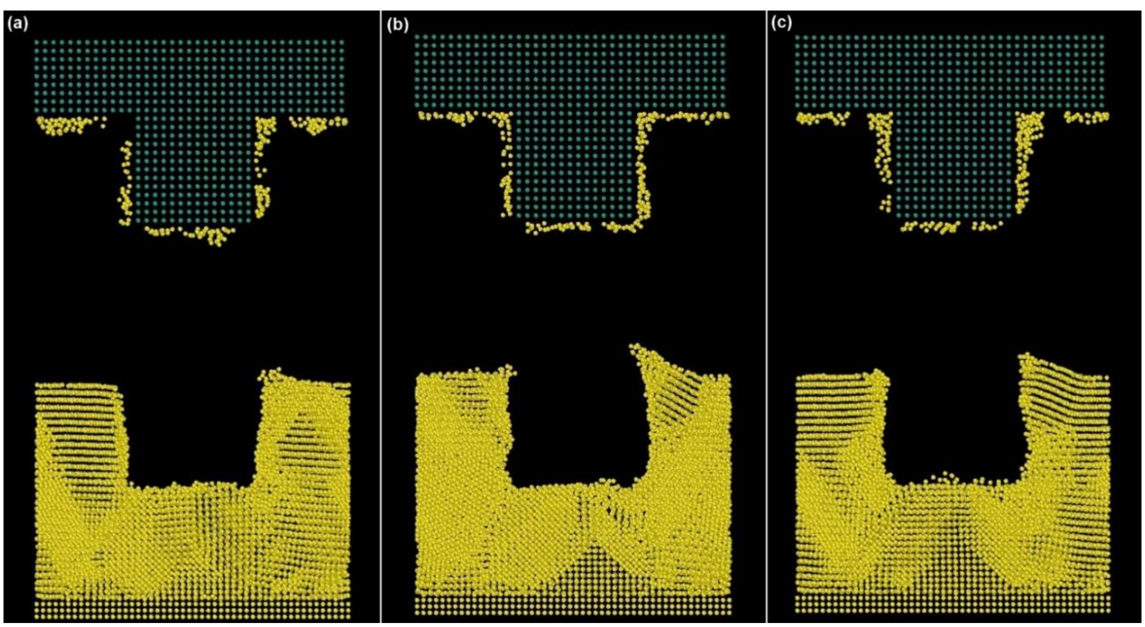

Figure 6: NIL simulation at 473K with cooling cycles (a) 10000 time step (b) 50000 time step (c) 200000 time step

Figure 7 represents the NIL process at 1000 K which is close to the melting temperature of gold. Significant adhesion of gold atoms is observed on the silicon mold at 10000 steps. This can be attributed to the "chilling effect" based on the rapid cooling of the mold and substrate. The steep cooling rate from 1000K to 298K results in the instantaneous solidification of the gold atoms on the silicon mold. Thus, as the mold is retrieved from the substrate it tears apart gold atoms along with it (Figure 7(a)). However, the cooling cycle at 50000 time steps allows adequate time for the gold atoms to recrystallize among themselves. Thus, the imprinted profile of the substrate replicates the mold profile accurately. At 200000 time steps, the mold is in prolonged contact with the gold atoms resulting in more adhesion and removal of gold atoms from the imprinted substrate. Thus, the 50000 time steps provides optimal imprinting results for 1000K temperature.

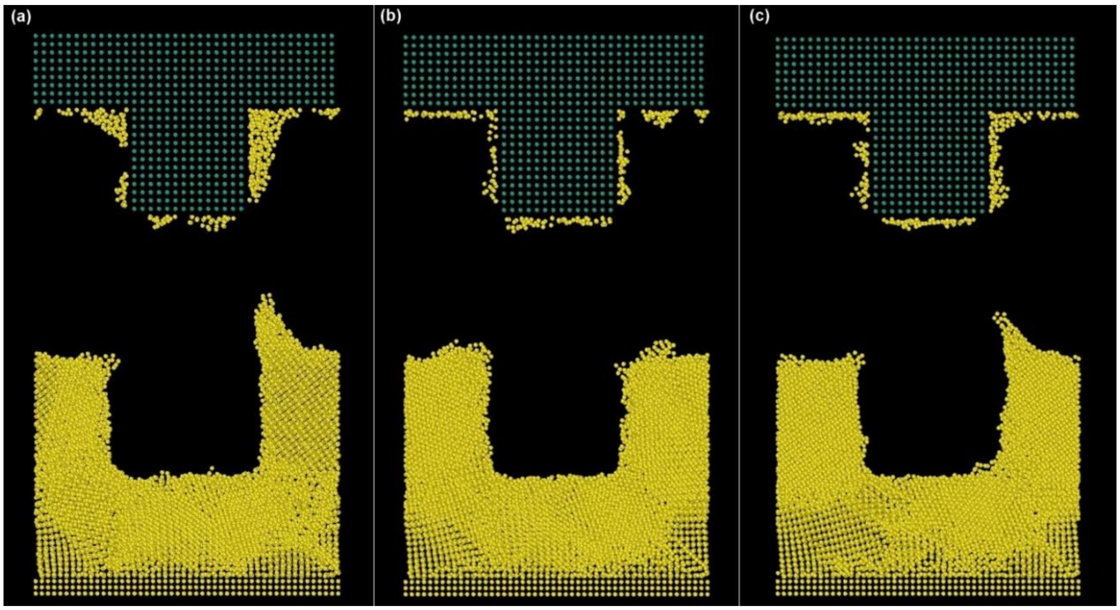

Figure 7: NIL simulation at 1000 K with cooling cycles at (a) 10000 time step (b) 50000 time (c) 200000 time step

4. Conclusion

This paper investigates the effect of cooling cycles in the nanoimprinting process with gold substrate and silicon mold. The cooling phase in the nanoimprint process plays a significant role in the final output quality of imprint with regards to dimensional accuracy. Nanoimprinting process simulations were conducted at temperatures of 298 K, 473 K and 1000 K, respectively. The cooling cycle (quenching) was conducted for 10000, 50000, and 200000 time steps. Spring back phenomena was investigated for different cooling cycles as it is very crucial in high resolution pattern transfer. Higher spring back was observed at lower temperatures 298K and 473K due to limited mobility of gold atoms resulting in higher residual stresses during imprinting. The 10000 time step cooling cycle displayed higher dimensional accuracy for both 298K and 473K, respectively. However, at 1000K an intermediate cooling rate of 50000 time steps resulted in good pattern transfer accuracy. This can be attributed to the minimal adherence of gold atoms to the silicon mold during the stamp demolding phase. Heating the NIL setup close to the melting point of gold (1000K) allowed the flow of gold atoms and the relaxation of the internal stresses, thereby, no spring back was observed. Rapid cooling (10000 time steps) at 1000K resulted in the "chill effect" on the silicon mold causing adherence of gold atoms resulting in a poor quality imprint. This research explains the relationship between cooling cycle, spring back phenomenon and its effect on high fidelity nanoimprint pattern transfer for several practical applications.

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