A study of morphology of silicon with femtosecond laser based on phase field model

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Keywords: femtosecond laser, surface bumps, phase field model, atomic diffusion

Abstract

In this paper, phase field model is employed to simulate the process of femtosecond laser which irradiates on silicon surface. The numerical model is mainly used to study the micro/nano evolution process and the atomic diffusion motion of the system. Meanwhile, the Semi-Implicit Fourier spectral method and Semi-Implicit Backwards Differentiation Formula are applied for high efficiency and numerical stability. Further, the rules of surface bumps are controlled by different thermal conductivities which are systematically revealed. Our work provides a new prospect for fabricating the bumps of the probe, to obtain high sensitivity sensors in chemical and biological fields.

1. Introduction

Recently, femtosecond laser plays an important role in micro/nano field because of ultra-high laser power and ultra-short laser duration [1]. Due to the advantages of femtosecond laser, it could be used to fabricate many micro/nano-structures for the sensors [2–4]. When a laser pulse hits the surface of the substrate of silicon, it produces periodic micro/nano-structures which are similar to the classical ripples and bumps [5, 6]. In the medical field, cantilever resonance has been used as a probe to detect the quality of biological cells, viruses and antibodies [7].

In the experiments of femtosecond laser controlling surface evolution, many researchers mainly show the evolution of micro/nano-structures by changing laser parameters, such as laser wavelength and laser polarization [8]. Majority experiments of laser inducing periodic micro/nano-structures are carried out by one-dimensional or two-dimensional temperature field [9–11]. The changes of local temperature field drive the movement of silicon atoms by diffusion, and then micro/nano-structures are formed on the surface of silicon [12]. Within the sensor region, the fabrication of the bump could be used as the probe in micro/nano-cantilevers to determine the sensitivity of the detection process [13]. Micro/nano-cantilevers probes are widely used in micro/nano-sensors and atomic force microscopes due to their unique mechanical, electronic and optical properties [14, 15]. The micro/nano-cantilever probes are mostly prepared by low-pressure chemical vapor deposition, sputtering and chemical corrosion. However, the disadvantages of the above methods are unstable molding, complicated manufacturing processes and low molding efficiency. Femtosecond laser pulses have attracted extensive attention from the scientific community in the field of micro/nano processing [16]. The advantages of the femtosecond laser are ultra-short pulse width and ultra-strong instantaneous power. The energy density of the femtosecond laser is adjusted to be approximately equal to the ablation threshold of silicon, a periodic temperature field will be generated on the surface of the substrate. Due to the effect of the Gaussian temperature field, silicon atoms will migrate and form a periodic morphology on the surface of silicon [17].

In order to obtain scientific and accurate observation results, numerical models are provided as the kernel theory to simulate the evolution process of micro/nano fabrication. In recent decades, phase field model has been widely used in the modeling to predict the micro/nano-structures evolution of many materials. It was
firstly proposed by Vander Walls and Cahn Hilliard [18] on the basis of the traditional diffusion interface model. In the phase field model, the evolution of each variable is driven by the decrease of total free energy [19]. The phenomenon of micro/nano fabrications was explained by the perspective of energy. Phase field model incorporates multiple kinetics and energetics in the multi-component system [20, 21]. Zhang [22] indicates that the phase field model is employed to simulate the dendritic solidification of Ti-6Al-4V alloy during the additive manufacturing process. The primary results that are obtained by phase-field simulation are in good agreement with experimental data and analytical predictions. In traditional classical sharp interface model, it involves explicitly tracking the location of the diffused interface region. The interface tracking method reduced computing complexity in one-dimensional system, but it limits us understanding complex mechanisms of the evolution in three-dimensional microstructures [20]. We aim to reveal the dynamic evolution process of femtosecond laser induces micro-convex structures on the surface, which involves multiple energetics and dynamics, such as bulk free energy, surface energy, thermodynamic heat transfer and surface diffusion. Compare with traditional model, phase field model can predict the evolution of arbitrary morphology and complex microstructure without explicitly tracking the interface position. Phase field model is increasingly relying on existing or future thermodynamic, kinetic, and crystallographic databases obtained from empirical modeling. The interface is not explicitly treated, but implicitly given by the concentration field, where the interface is represented by a thin continuous transition region. Therefore, any additional computational difficulties are avoidable in phase field model, and it improves computational efficiency and numerical stability [23–25].

In our computational work, we provide a visual impression of the development of micro/nano-structures on silicon, the bumps on the substrate of silicon are appealing. Therefore, we propose to study the effect of the thermal conductivity of the materials on the fabrication, and observe the changes in the height of the bumps on the surface.

2. Model and numerical implementation

Phase field model is employed here to simulate the changes on the surface of silicon during the substrate that is irradiated by femtosecond laser. After laser irradiation, the absorbed heat developed a temperature gradient throughout the substrate. The temperature gradient induced the thermophoresis force, to affect the mass transport of atoms on the surface. The evolution of micro/nano-structure is driven by the minimization of free energy of the system under the constraint of mass conservation for the surface modification of silicon. According to the wavelength, the temperature field of the femtosecond laser is divided into a single Gaussian temperature field. The upper part of figure 1 shows that the femtosecond laser induces the temperature field.

In the phase field model, a conservative variable \( c(x, y, z, t) \) is assumed a time-dependent and spatially continuous function. We define \( c = 1 \) as silicon phase, \( c = 0 \) as the air phase. All the expressions are normalized with a characteristic length \( L_0 \), and a characteristic time \( t_0 = L_0^2 / M_0 f_0 \). Here \( f_0 \) is a positive constant, \( M_0 \) is a material constant. The real time is equal to the characteristic time multiply by time-step of the simulation. The Cahn-Hilliard model [23] could clearly express the free energy of the whole system as follows:

\[
G = \int_V \left[ f(c) + \frac{1}{2} h(\nabla c)^2 \right] dV
\]  

(1)

Here, \( f(c) \) is the local free energy density [24, 25], \( f(c) = f_0 c^2 (1 - c)^2 \), which induces different phase separation and could be calculated by the double well function. The second term \( \frac{1}{2} h(\nabla c)^2 \) means the surface energy of silicon, the parameter \( h \) is the gradient energy coefficient. In this model, a driving force that relates the
silicon flux for mass transport to the chemical potential is given by $E_l = -\nabla \mu$. The chemical potential is defined by $\mu = \delta G / \delta c$, where $\delta G / \delta c$ is related to the total free energy of the system.

During laser irradiation, the substrate is placed in a temperature field gradient. The mass flux can be rewritten as $J = -M\nabla \mu - \theta Mc \nabla T$. Within the adopted diffuse interface framework, we define the mobility of silicon $M$ is:

$$M = M_0 \left( \int c^2 (1 - c)^2 dc / \int c^2 (1 - c)^2 dc \right) (1 - c) \right\}^{26}.$$  

Mobility is restricted within the interfacial region, it vanishes outside the interfacial region of the substrate of silicon. According to the Cahn-Hilliard nonlinear diffusion equation and mass conservation law, $\frac{\partial c}{\partial t} = -\nabla \cdot J$, the expression of $c$ could be written as:

$$\frac{\partial c}{\partial t} = \nabla \cdot (-M\nabla \mu_1).$$  

$$\mu_1 = [4c^3 - 6c^2 + 2c - \text{CH}^2 \nabla^2 c + \alpha c]$$

Here, CH is the Cahn number, which represents the relative significance of the surface energy of silicon, $\alpha = \theta / f_0$. The temperature evolution is as follows:

$$\rho \varepsilon \frac{\partial T}{\partial t} = k \left( \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} \right) + Q$$

$\rho$ and $\varepsilon$ are the density and specific heat of silicon, respectively. $k$ is the thermal conductivity of silicon. The temperature field of the femtosecond laser is simplified to a single Gaussian temperature field. The heat source of the femtosecond excitation is set to be $Q$. The expression of heat source is as follows:

$$Q = \lambda_0 \exp \left[-\beta \left( \frac{x^2 + y^2 + z^2}{\lambda} \right) \right]$$

$\lambda$ is the wavelength of the femtosecond laser. The parameter $\beta$ is the absorption efficiency of silicon for the temperature. $I_0$ is assumed to be the intensity of the femtosecond laser, which is related to the phenomenon of femtosecond laser irradiates on the substrate of silicon, and could be written as:

$$I_0(x, y, z, t) = \frac{2p}{\pi \sigma} \exp \left[ -\frac{2(x^2 + y^2 + z^2)}{\sigma^2} \right]$$

Here, $p$ is the power of the femtosecond laser, and $\sigma$ is the radius of the beam. In the three dimensional dynamic numerical model, the mechanism of the fabrication is temperature gradient field inducing thermal diffuse in the substrate. The temperature gradient induces the thermophoresis force, which could affect the mass transport of atoms on the surface. The atoms of silicon are moving from low temperature to high temperature \cite{27}. As shown in figure 2, the femtosecond laser irradiates the top surface of silicon, and high temperature induces thermal diffusion flow.

In the temperature field, an air convection boundary condition is employed to indicate the free end of the substrate, and equation (4) can be rewritten as:
Here, \( W_1 = k t / \mathcal{L}^2 \rho c \), \( W_2 = \mathcal{L}^2 / k \). A Semi-implicit Fourier spectral method and Semi-implicit Backwards Differentiation Formula (SBDF) scheme are applied to obtain effective and accurate solutions. The right-hand side of equation (2) needs to handle the variable mobility, which is given by:

\[
\nabla \cdot (M \mathcal{V} \mu_i) = A \nabla^2 \mu_i^{(t)} + s_{\mu_i}
\]

The term \( A \nabla^2 \mu_i^{(t)} \) is explicitly treated, \( \mu_i^{(t)} \) is a linear segment of \( \mu_i \), \( \mu_i = - \mathcal{V} c \frac{c}{c^2} \mathcal{V} H \), \( A \) is assumed to be 1. The evolution of the variables \( c \) and \( T \) are as follows:

\[
\frac{3}{2} c^{n+1} - 2c^n + \frac{1}{2} c^{n-1} = A \Delta t (2 \nabla^2 c^{n+1} - \mathcal{H}^2 \nabla^2 c^n) + 2 P^n - P^{n-1}
\]

\[
P^n = A \Delta t [\nabla \cdot (M \nabla \mu_i^{(t)}) - 2 \nabla c^n + \mathcal{H}^2 \nabla c^n]
\]

\[
\frac{3}{2} T^{n+1} - 2T^n + \frac{1}{2} T^{n-1} = \Delta t \nabla^2 T^{n+1} + Q^{n+1}
\]

As a result, equations (9)–(11) are applied with the Fourier transform and modified to be:

\[
\hat{c}^{n+1} = \frac{1}{\text{Dev}} (4 \hat{c}^{n} - \hat{c}^{n-1} + 4P^{n} - 2P^{n-1})
\]

\[
\hat{T}^{n+1} = \frac{4T^n - T^{n-1} + Q^{n+1}}{3 + 2\Delta t \xi^2}
\]

Here, \( \text{Dev} = 3 + 4 \Delta t \xi^2 + 2 \Delta t \mathcal{H}^2 \xi^4 \). The caret ‘\( \hat{\cdot} \)’ and \( \xi \) stand for the Fourier transform. The numerical model of the laser irradiates on silicon substrate has been established as above. The solution of variables \( c \) (x, y, z, t) could be listed as follows: Firstly, calculate the chemical potential \( \mu_i \) of the numerical model. Then, by Fourier transform to get the evolutional process of \( c \). Finally, repeat the above steps until the prescribed time steps are reached. By calculating of the above equations, the morphological changes of silicon at different spatial coordinates and different times are obtained until the system reaches equilibrium.

### 3. Results and discussion

A numerical model of micro/nano fabrications of silicon is established. The substrate of silicon is irradiated by femtosecond laser, we define the basic dimensions of silicon substrates to be 10.0 × 10.0 × 1.5 \( \mu m \), and the characteristic length was taken to be 50 \( \mu m \). The femtosecond laser (power approximately 75 mW, wavelength 532 nm, the repetition frequency 1 kHz, and the pulse width 120 fs) irradiates directly on the surface of silicon, the spot diameter of the collimated laser was 8 \( \mu m \), which was exposed to the surface of substrate for 10 s. A Gaussian heat source generated by a femtosecond laser, which is applied to the center of the substrate. In the numerical model, the parameters of the laser are used to irradiate on the surface of silicon are listed in table 1.

![Figure 3](image_url)

**Figure 3** shows the simulated evolutions of the temperature field which is produced by the laser at different times. The center temperature of the substrate is up to 1500 K. Based on the effect of thermal diffusion, the temperature gradient is generated, an extension of the interior space towards the exterior. The temperature gradient induces the thermophoresis force, which affects the mass transport of atoms on the surface of silicon. The atoms of silicon make mass migration from low temperature to high temperature. Figure 3(b) shows the simulated observations on the effect of temperature field and the morphology of the surface of silicon. The modification of the bump on the surface is growing as time increases. The thermophoresis force induces the aggregation of atoms of silicon in the high temperature field, the height of the bump is changed with the different temperature. High temperature triggers more atoms gathering, so the phenomenon of the growth on the surface of silicon is more visible than at the low temperature field. Our numerical model has been validated by the experiment [28, 29], these experimental parameters were established same as our simulated work, the
morphological changes on the surface of silicon are expressed. Qualitative and quantitative observations indicate a good agreement between the experimental and simulated results.

We also propose to study the growth of the bump on the surface by using different materials. Thus, the different thermal conductivity of the substrates is assumed in our computational model, to observe the different changes on the bumps. Since the thermal conductivity of the materials is modulated, which corresponds to the femtosecond laser irradiates on the other different silicon compounds, such as silicon dioxide. The thermal conductivity of non-conducting material (asbestos) is $0 \times 10^{-9}$ W nm$^{-1}$K$^{-1}$, silicon dioxide (SiO$_2$) is $8 \times 10^{-9}$ W nm$^{-1}$K$^{-1}$. In the figure 3(a), we choose the thermal conductivity: $K = 4 \times 10^{-9}$ W nm$^{-1}$K$^{-1}$ and $K = 25 \times 10^{-9}$ W nm$^{-1}$K$^{-1}$ to represent different silicon compounds (silicon dioxide) with different thermal conductivities. In both cases, the simulation time equals to 10 seconds. The lower thermal conductivity; $K = 4 \times 10^{-9}$ W nm$^{-1}$K$^{-1}$, which induces that the height of the bump on the surface is 200 nm. But the height of the
bump grows to 900 nm when the thermal conductivity becomes $25(10^{-6} \text{W nm-K})^{-1}$. Compared with these two cases, the morphological change on the surface is more obviously which is induced by higher thermal conductivity. The quantitative values of the observation with different thermal conductivity are presented in figure 4(b). With the increase of thermal conductivity, the height of surface bumps gradually increases. Through the above work, we verify that the thermal conductivity of the materials could control the height of the bumps on the surface. The different thermal conductivity determines the different gradient of the temperature to influence the shape of the bump. Base on considering the effect of the thermal conductivity of the materials on the aspect ratio of the bump, which provides a necessary way to design the resultant morphology by using different materials. These observations give an idea on preparing high aspect ratio of a probe on the cantilever, and provide an inspiration to fabricate high quality micro/nano-cantilever sensors in actual.

4. Conclusions

This paper mainly introduces micro/nano-structures of silicon which is induced by femtosecond laser. The phase field model is employed to simulate the evolution of the fabrication of micro/nano-structures. The femtosecond laser induces the temperature gradient on the surface of silicon, and the temperature field drives the atoms to make diffusion motion and induce conical bump. The thermal conductivity is proposed to control the height of the bump. Our work provides an inspiration for fabricate high aspect ratio of a probe of the sensor manufacturing in biological and chemical fields.

Acknowledgments

This work was supported by ZheJiang Provincial Natural Science Foundation of China under Grant No. LY21F040005, GF22H205926 and LY21A020009, and National Natural Science Foundation of China under Grant No. 52175460.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Conflicts of interest

The authors declare no conflict of interest.

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