Effect of Incident Intensity on Films Growth in Pulsed Laser Deposition *

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Incident intensity, defined by the amount of particles deposited per pulse, is an important parameter in the film growth process of pulsed laser deposition (PLD). Different from previous models, we investigate the irreversible and reversible growth processes by using a kinetic Monte Carlo method and find that island density and film morphology strongly depend on pulse intensity. At higher pulse intensities, lots of adatoms instantaneously diffuse on the substrate surface, and then nucleation easily occurs between the moving adatoms resulting in more smaller-size islands. In contrast, at the lower pulse intensities, nucleation event occurs preferentially between the single adatom and existing islands rather than forming new islands, and therefore the average island size becomes larger in this case. Additionally, our results show that substrate temperature plays an important role in film growth. In particular, it can determine the films shape and weaken the effect of pulse intensity on film growth at the lower temperatures by controlling the mobility rate of atoms. Our results can match the related theoretical and experimental results.

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In the last decades, the thin film growth process has received a great deal of attention because it can help people to understand the natural mechanism of film growth and prepare more good-quality film materials. The simple diffusion-limited-aggregation (DLA) model was used to simulate fractal growth at lower temperatures.[1,2] The effect of substrate temperature on island aggregation was investigated in detail by Bruschi *et al.*,[3] Wei *et al.*,[4] and Gu *et al.*,[5] discussed the influence of microstructure of substrate surface on early stage of film growth. Some authors established Monte Carlo models to simulate the concrete film growth processes or give a review to this research field.[6−11] These models were established for describing the effect of a continuous incident flux on thin films growth in deposition techniques such as molecular beam epitaxy (MBE) or thermal evaporation.[12−14]

As is known, PLD is a versatile technique to prepare many kinds of functional materials. A pulsed incident flux rather than a continuous flux is just used in the PLD process. Figure 1 gives a schematic illustration of a pulsed flux. According to the characteristic of the PLD process, we have investigated the number density of point defects on substrate surface and its influence on film growth varying with laser energy and substrate temperature.[15] Recently, Hinnemann *et al.*[16] proposed a simple irreversible model for the PLD growth process without considering pulse duration. Zhang and Chu[17] established an irreversible growth model considering pulse duration to simulate the pulsed beam deposition process. Tan *et al.*[18] developed a reversible model to disclose the influence of substrate temperature and pulse duration in the PLD process. Their results showed that the behaviour of PLD at the relatively low temperature is similar to that of MBE.[18] Experimentally, PLD is rarely used for simple metal systems, although it is an interesting alternative technique to MBE.[19,20] In Ref. [20], Jubert *et al.* have successfully prepared Fe films on a Mo substrate and established kinetic equations to calculate island density in the PLD film growth process.

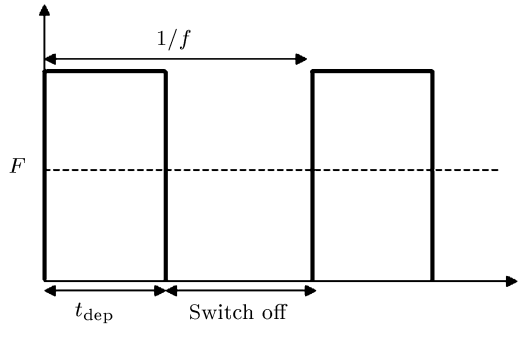


Fig. 1. Schematic diagram of the pulsed flux in the PLD process. Here f is pulse frequency; t_{dep} is pulse duration and F is the average flux. Pulse intensity I varies with pulse frequency f at the same average flux F.

In this Letter, a kinetic Monte Carlo model is developed to simulate the PLD film growth process. Different from the previous models, the influences of pulse intensity and pulse duration on film morphologies are considered in the present model. The film growth is defined as a layer-by-layer growth process on a defect-

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free square lattice. Also, in order to quantify the growth characteristics of PLD, we propose to analyse island aggregation at the submonolayer stage. A periodical boundary condition is used in the simulations. The processes are introduced in our model as follows: (1) Deposition. As shown in Fig. 1, during the ON fraction $t_{\rm dep}$ of a cycle, particles are deposited on the substrate in regular pulses with intensity I. At the rest of the cycle, no particle reaches the substrate surface, and the average flux F given in monolayers/s (ML/s). The instantaneous flux v_{de} can be calculated by $F/(ft_{\rm dep})$. (2) Diffusion. The adatoms diffuse on the surface along random directions and the hopping rate v_{di} is agreement with the Arrhenius equation $v_{di} = (2k_BT/h)\exp[-(nE_n + E_d)/k_BT]$, where T is substrate temperature, $2k_BT/h$ is the vibration frequency of substrate surface atoms, E_d is the adatom diffusion activation energy for a single adatom on an empty surface, E_n is the second-neighbour interaction energy, namely pair bond energy, n is the number of second-neighbours around the adatom on the same terrace, k_B is Boltzmann's constant, and h is Planck's constant. If two particles come to occupy neighbouring sites during the diffusion process, they stick and form an island. Here an island means a cluster made up of some adatoms in the same terrace.

During the switch-on process, we calculate the ratio of $p = v_{di}/(v_{de} + v_{di})$ at the beginning of a new cycle. We produce a random number R in the interval $(0, 1)$. If $R < p$, the diffusion process is selected; otherwise, the deposition event is selected. If the deposition event is selected, a new particle is randomly placed on the empty sites of surface. If the diffusion event is selected, the adatom will move along the direction determined by a hopping probability proportional to $\exp[-(nE_n + E_d)/k_BT]$.

The kinetic Monte Carlo simulation is carried out on 400×400 sites. In order to compare with the experimental results, the same parameters are used in the present calculations: a mean deposition rate is 5×10^{-3} ML/s, pulse duration $t_{\rm dep} = 10 \times 10^{-6}$ s, $E_d = 0.11$ eV and $E_n = 0.3$ eV.[20]

Figure 2 shows the evolution of film morphology for different pulse intensities at $D/F = 10^6$. According to the pulse frequency used in Ref. [20], we select the incident intensities $I = 0.005$, 0.0005 and 0.0001 to investigate the film growth process. From Figs. 2(a)–2(c), we can obviously see that at $I = 0.005$, the island density is larger and the average size of islands is smaller. When I is gradually reduced, island density N begins to decline while the island becomes larger. Because at higher incident intensities, more atoms arrive at the surface simultaneously, most of them can meet each other in a larger proportion and form more new islands. However, when the pulse intensity is decreased, new particles easily adhere to the existing islands rather than forming new islands, and therefore the average island size becomes larger in these cases. Figure 3 clearly shows the island density N and the average island size S versus incident intensity. A comparison between our simulated island density and Jubert *et al.*'s theoretical calculation is also given in Fig. 3. It can be seen that our results are in good agreement with the theoretical results.

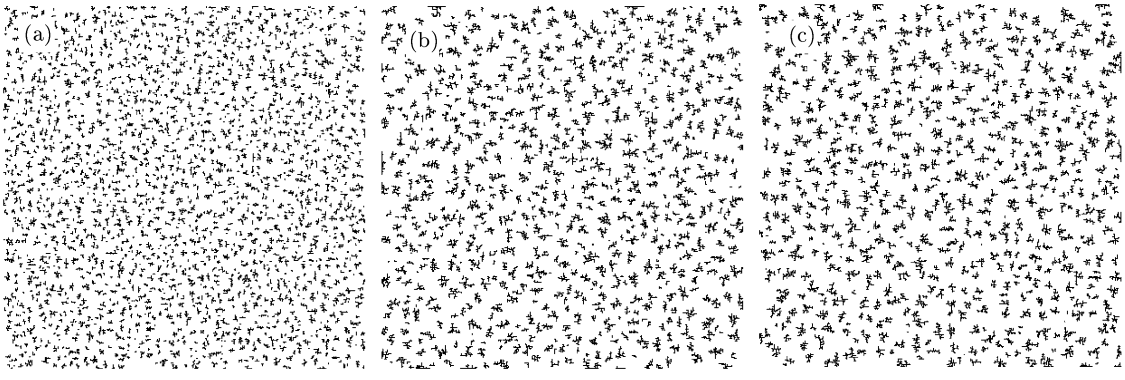


Fig. 2. Snapshots of film morphologies for different pulse intensities at the coverage 0.12 ML at pulse intensities $I = 0.005$ (a), 0.0005 (b) and 0.0001 (c) for $D/F = 10^6$.

To justify our model and to obtain a clear comparison between our simulation results and the experimental data, we simulate the film growth at the different substrate temperatures $T = 150$ K, 300 K, 400 K and 500 K, respectively. Figure 4 shows an Arrhenius plot of N as a function of substrate temperature. It is obvious that substrate temperature plays an important role in the film growth because temperature can determine the diffusion rate of surface adatoms D, which can be approximately calculated by $(2k_BT/h)\exp[-E_d/k_BT]$. When substrate temperature is elevated, the island density is reduced and contrarily the average island size is increased. Our simulated result is comparable to the experimental result and the difference between the slopes possibly can ascribe the perfect conditions used in our simulations.

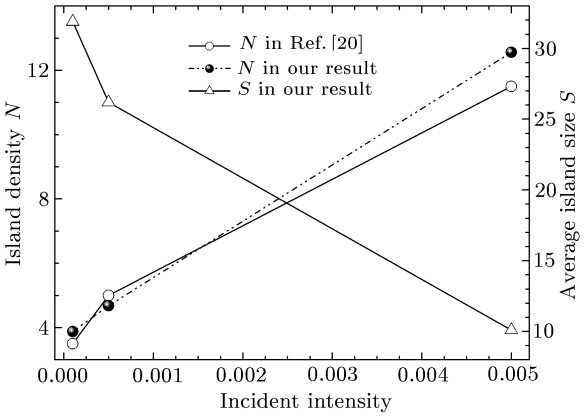


Fig. 3. Comparison between our result and other theoretical calculation of island density N and average island size S versus incident intensity.

Previously, the irreversible and reversible models are often discussed separately. In fact, when the pair-bond energy E_n is relatively smaller than E_d, and/or substrate temperature T is high enough, the irreversible model then effectively becomes a reversible growth model. The reversible model allows the atoms with lateral nearest neighbours to move, or the dimmers dissociate to single adatoms. Therefore, we select other group of parameters to carry out Monte Carlo simulations in order to display an obvious reversible growth regime. The typical average deposition flux is 0.1 ML/s, and we set $E_d = 1.3$ eV and $E_n = 0.3$ eV, the values used by Ratsch *et al.*[21]

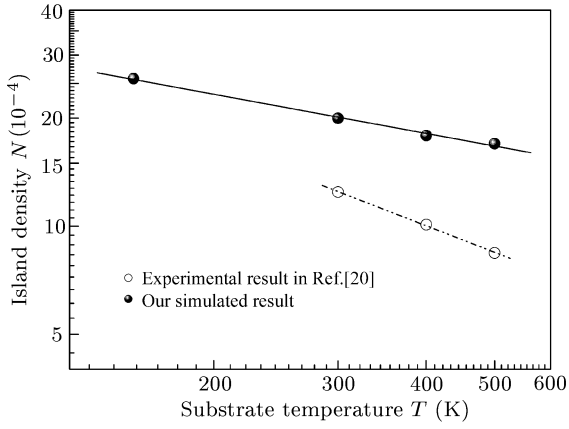


Fig. 4. Comparison between the experimental result and our simulated result of island density.

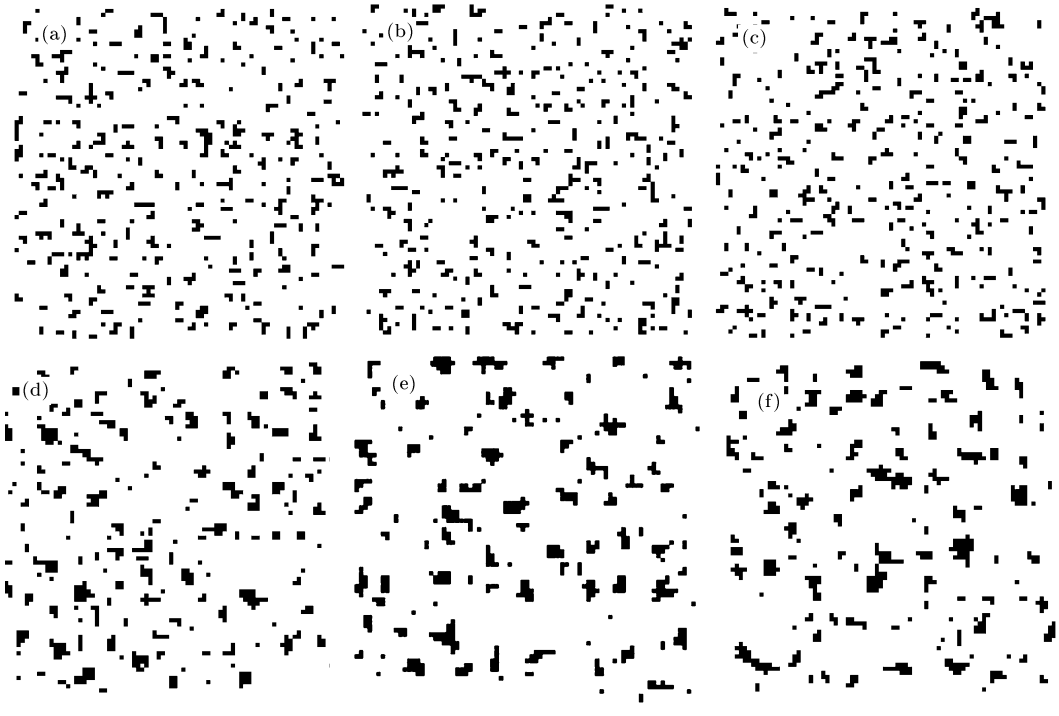


Fig. 5. The evolution of film morphology for different pulse intensities at the coverage 0.1 ML for pulse intensities $I = 0.1$ (a), 0.01 (b) and 0.002 (c) at substrate temperature 550 K, and at the same coverage for pulse intensities $I = 0.1$ (d), 0.01 (e) and 0.002 (f) at 650 K.

Figure 5 depicts the evolution of film morphology for different pulse intensities at substrate temperatures $T = 550$ K and 650 K, respectively. In Figs. 5(a)–5(c), the substrate temperature T equals 550 K and it is found that the islands grow in a dispersed regime, and especially the film morphologies at the different intensities $I = 0.1$, 0.01 and 0.002 advance in the similar way. The result suggests that the influence of pulse intensity is not obvious at the lower substrate temperatures. That is to say, the behaviour of the PLD at relatively low temperature is similar to that of the MBE[18] and the substrate temperature

can weaken the effect of pulse intensity on the growth process by controlling the diffusion rate of adatoms. In comparison, Figs. 5(d)–5(f) display the different island nucleations at the three intensities for temperature $T = 650$ K. The two series of panels in Fig. 5 show that the island nucleation and shape are evidently different due to varying substrate temperatures. The results are similar to the theoretical results obtained by Ratsch *et al.*[21] and demonstrated by the STM images for Ni/Ni(001).[22] Nucleation density N as a function of pulse intensity I and coverage θ for substrate temperature $T = 650$ K is clearly shown in Fig. 6.

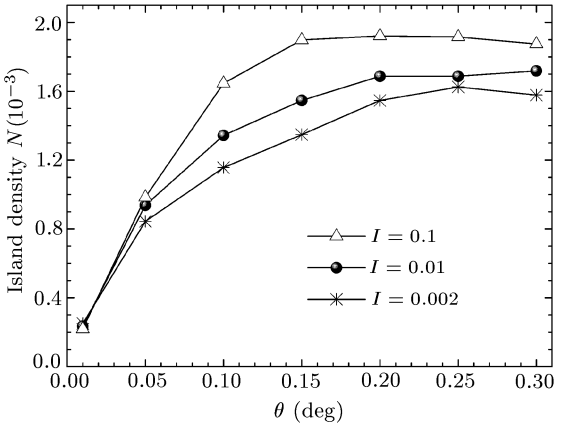


Fig. 6. Influence of incident intensities $I = 0.1$, 0.01 and 0.002 on island density at $T = 650$ K.

In conclusion, using the kinetic Monte Carlo method, we have simulated the initial film growth in a PLD process, and the role of pulse intensity on island aggregation is stressed. The obtained results show that the incident intensity per pulse can influence island density and film morphology. In particular, for the higher substrate temperatures, the effect of pulse intensity on film growth is more obvious. In spite of irreversible and reversible growth regimes, the incident intensity is an important technological parameter in the PLD process. At the same time, substrate temperature influences the growth shape by determining the adatom diffusion ability on surface. Consequently, in the PLD process, the two technological parameters should be appropriately selected at the same time to obtain the required film material.

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