Computational Studies of Thin Film Growth on Patterned Substrates

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The purpose of high quality thin film growth on patterned substrates is to maintain the pattern during the deposition process. A simulation growth model is used here to determine appropriate conditions under which the pattern can remain for a long time. Two noise reduction techniques, *multiple hit* and *long surface diffusion length*, are introduced into the original model. The longer surface diffusion length (*d*) in computational simulations is equivalent to higher substrate temperature in experiments. We find that the pattern persists for longer periods of time when *d* is increased. However, there is a limit to how large *d* can be. We conclude that the pattern has long-lived persistence when the film is grown at sufficiently high, but not too high, substrate temperature.

Key words: patterned substrate growth, long surface diffusion length, multiple hit noise reduction technique, persistence probability.

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การศึกษาด้านการคำนวณของการปลูกฟิล์มบางบนแผ่นรองรับที่มีรูปแบบ

สุวกัลป์ เพียรกรณีย์ และ ปัจฉา ฉัตราภรณ์ (2548)
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จุดมุ่งหมายในการปลูกฟิล์มบางบนแผ่นรองรับที่มีรูปแบบให้มีคุณภาพสูงคือ ต้องรักษารูปแบบให้อยู่ได้นานตลอดกระบวนการปลูกฟิล์ม แบบจำลองทางคอมพิวเตอร์ได้ถูกนำมาใช้ในการหาเงื่อนไขที่เหมาะสมที่ทำให้รูปแบบคงอยู่ได้เป็นเวลานาน ในที่นี้เป็นการคำนวณปริมาณการคงอยู่ของรูปแบบที่เวลาหนึ่งๆได้จากความน่าจะเป็นของการคงอยู่ เพื่อให้รูปแบบคงอยู่ได้นานได้เพิ่มเทคนิคการลดสิ่งรบกวนในแบบจำลองดั้งเดิม อันได้แก่ มัลติเพิลฮิตและการเพิ่มระยะการแพร่บนพื้นผิว การเพิ่มระยะการแพร่บนพื้นผิวในการจำลองทางคอมพิวเตอร์นั้นจะเทียบได้กับการเพิ่มอุณหภูมิของแผ่นรองรับในการทดลอง พบว่ารูปแบบจะคงอยู่ได้นานเมื่อระยะการแพร่บนพื้นผิวเพิ่มมากขึ้น อย่างไรก็ตามวิธีนี้ก็มีข้อจำกัด ซึ่งสรุปได้ว่ารูปแบบจะคงอยู่ได้นานเมื่อปลูกฟิล์มด้วยอุณหภูมิของแผ่นรองรับที่สูงเพียงพอ แต่ต้องไม่สูงมากเกินไป

คำสำคัญ การปลูกฟิล์มบนแผ่นรองรับที่มีรูปแบบ การเพิ่มระยะการแพร่บนพื้นผิว เทคนิคการลดสิ่งรบกวนแบบมัลติเพิลฮิต ความน่าจะเป็นของการคงอยู่

INTRODUCTION

Thin film growth from molecular beam epitaxy (MBE) is an important technological process used to produce high quality thin films. Here, the purpose of high quality patterned growth is to maintain the pattern during deposition. In order to determine appropriate conditions in which the pattern can remain for a long time, simulation growth models are used.

In the past decades, many non-equilibrium, limited mobility discrete models have been introduced to study the MBE growth process.[1-4] Generally, limited mobility models produce very rough thin films because of the presence of unavoidable stochastic noise. To obtain high quality thin films, the noise must be reduced. In this paper, two noise reduction techniques are used: the multiple hit noise reduction technique[5-8] and the long surface diffusion length noise reduction technique.[2, 9]

MODEL AND METHODS

Das Sarma-Tamborenea Model

All our results presented in this paper are based on the use of the Das Sarma-Tamborenea (DT) model.[1,2] This is a non-equilibrium, limited mobility, conserved model, *i.e.* desorption, bulk vacancies and overhangs are neglected. These conditions are known as the solid-on-solid (SOS) constraint. In previous works, The DT model was used extensively for studying thin film growth process on flat substrates.[1, 4, 7, 10] It was shown that this model can explain MBE growth reasonably well. Moreover, this growth model is the low to intermediate substrate temperature version[1,2] of the full temperature dependent activated diffusion MBE growth model.[2,4] Because the morphologies and scaling exponents of the DT model agree quantitatively with the MBE model in this temperature range, we chose to use it the DT model to determine the appropriate conditions for thin film growth on patterned substrates.

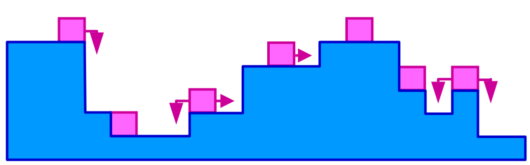

Figure 1. Diffusion rule for DT model with *d* = 1 on one-dimensional substrate.

In the DT model, an atom is assumed to be a simple cubic. Step deposited atoms are dropped vertically, randomly and sequentially (one atom at a time at a randomly chosen spatial position) on a substrate. After the deposition, each atom is allowed to move immediately only once within a finite lateral surface diffusion length, *d*, to its final incorporation site according to the diffusion rule. After the atom chooses its final site, it is permanently incorporated at that site and cannot move for the rest of the growth time. For the original DT model (d = 1), deposited atoms are dropped on the substrate and move following the DT diffusion rule (see Figure 1). A deposited atom is allowed to move only if it has no lateral nearest neighbor in the same layer. The deposited atom must have at least one nearest neighboring bond with the atom underneath it in order to satisfy the SOS constraint. If a deposited atom has more than one nearest neighbor bond at its deposition site, it will be stuck at that place. For a deposited atom which has one nearest neighbor bond, it diffuses instantaneously by one site within the surface diffusion length to its final position. At this position, the atom must have more local coordinate numbers than at the deposition site. This is equivalent to increasing the number of nearest neighbor bonds it forms with other atoms. If there are many possible final sites that satisfy the requirement to increase the number of bonds compared with the deposition site, the deposited atom chooses one of those sites with equal probability. If there is no other site within the surface diffusion length that satisfies the diffusion rule, then a deposited atom is incorporated at its deposition site. Once atoms are incorporated, they cannot diffuse.

It is important to emphasize that deposited atoms in the DT model search for final sites with higher coordination numbers compared to the original deposition site. The final sites are not necessarily the local sites with the maximum coordination numbers. In other words, atoms try to *increase*, but *not necessarily maximize*, the local coordination number. The DT diffusion rule also allows only downward diffusion of the atoms (can move on the same layer or move down), but the diffusion process is not necessarily to search for a site with the minimum height (following gravitational rule) – because in this case the electronic force has much more effect than the gravitational force.

Note that in this work, we measure time in units of monolayer (ML). Since our deposition rate (growth rate) is fixed as one ML per second, it means that in one second, the film is filled up with an average of one layer with $N = L$ atoms, where L is substrate size. Moreover, all of our simulations are done with periodic boundary conditions along the substrate to avoid the effects of the substrate edges. Hence, the topology of the substrate in a one-dimensional substrate is a circle.

Noise Reduction Techniques

In DT growth simulation, there is an unavoidable stochastic noise during the growth processes. The noise causes kinetic roughness on the surface. To obtain a high quality film, the noise must be reduced so the effects of the diffusion can be enhanced. In our work, two noise reduction techniques were used: *long surface diffusion length* and *multiple hit* noise reduction techniques. The former is to increase the lateral surface diffusion length ($d > 1$), which means the atoms have more chance to search for the most appropriate site. This is an obvious technique which can improve the smoothness of the growth surface. Typically, a larger d corresponds to a higher substrate temperature T in experiments. However, we found that in higher substrate dimensions, it is difficult and extremely time consuming to use $d > 1$ in simulations. To this end, the multiple hit noise reduction technique was introduced.(7, 8)

The multiple hit noise reduction technique involves the acceptance of only a fraction of allowed atomistic deposition events in simulations.(7, 8) It is characterized by multiple hit factor m, which is defined as an integer number. Each lattice site has an assigned counter. When a deposition event occurs and the deposited atom moves to its final preferred site according to the DT diffusion rule, the counter is increased by one. In original the DT model with $m = 1$, the height of the preferred site is increased by one. However, in the DT model with multiple hit noise reduction technique, its diffusion rule is slightly modified in such a way that the height of the preferred site remains the same and the counter of that preferred site is the quantity that is increased instead. The deposition event at a particular site is accepted and the height is increased only when a counter of that site reaches a multiple hit factor $m > 1$. Therefore, this technique is called multiple hit noise reduction technique since deposition events become a true deposition process only if that site is hit m times. After the true deposition, the counter at that particular site is reset to zero, and the whole multiple hit process is repeated. We note here that, in contrast to the long surface diffusion length technique, this multiple hit technique does not correspond to any physical mechanism in experiments. It is purely a trick in computational modeling that has been used successfully in the study of nonequilibrium growth phenomena.(7, 8)

RESULTS AND DISCUSSION

In the following we will show and discuss the simulation results of two different initial patterned substrates: (1) a completely flat substrate and (2) a periodic pattern substrate with a fixed feature size. Before presenting our results, we discuss the quantity that we use in our study: persistence probability(11) $P(t)$. The persistence probability $P(t)$ is a quantity used to determine fractions of the pattern that survive through time t MLs. It is defined as

$$P(t) \equiv \left\langle \prod_{s=1}^{t} \delta_{h(x,s),h(x,0)+s} \right\rangle_L ,$$

where $h(x,s)$ is the height of the growing surface at time s at lattice site x, δ is the Kronecker delta function where $\delta_{i,j} = 1$ if $i = j$ and 0 otherwise, and the angular brackets represent the average over substrate site L. By definition, $P(t)$ means *survival* of the pattern through time t is counted only when the initial patterned configuration, $h(x,0)$, is reproduced everytime after each monolayer deposition until the film is grown to t MLs.

All of our results shown here are simulated on a one-dimensional substrate of size L = 10,000. In Figure 2, we show the calculated $P(t)$ as a function of growth time t on flat substrates for various values of d (Figure 2(a)) and m (Figure 2(b)). We found that $P(t)$ decays exponentially in time. This is because in very early time the original patterns can be reproduced nearly perfectly with only minimal amount of surface roughness. At later times, however, the growing surfaces become rougher and the original pattern cannot survive. It can also be seen in Figure 2 that when noise reduction techniques are used, the pattern can persist for a longer time. According to previous works, these techniques allow thin films to grow in layer-by-layer mode in which each layer reproduces its original pattern.[7,9] Hence, the pattern has long-lived persistence when *d* or *m* is large. Moreover, we found that results from the long surface diffusion length technique are equivalent to results from the multiple hit technique. Therefore, we can replace the former technique with the latter in order to save computational time in higher substrate dimensions.

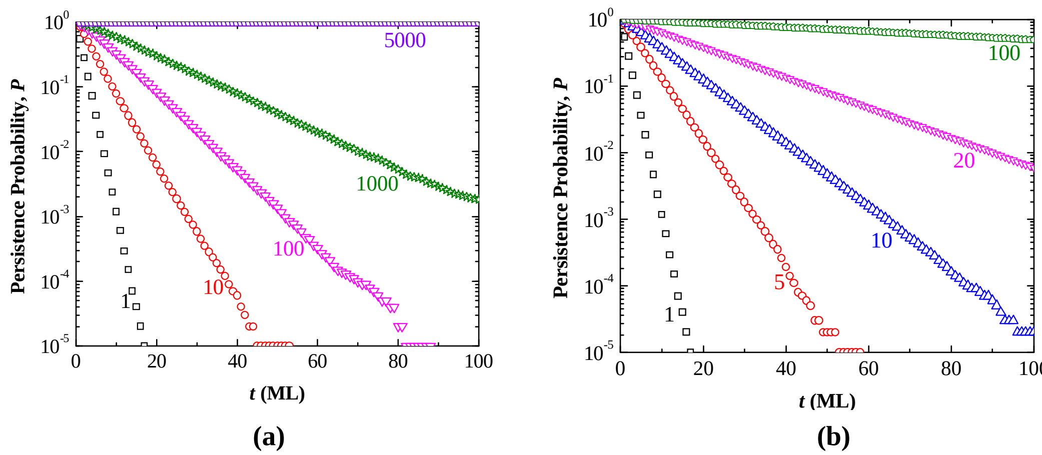

Figure 2. Persistence probability *P(t)* of flat substrates as a function of growth time *t* : (a) the plot for different diffusion lengths *d* and (b) for different multiple hit factors *m*.

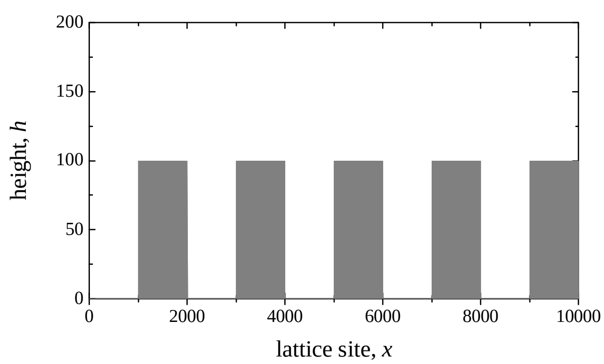

Figure 3. Schematic plots showing morphotogy of the periodic substrate.

Next we show our results from a simulation of growth on periodic patterned substrates with an initial condition that creates a series of blocks of width 1,000 lattices. The initial height of each of these blocks is 100 layers and the blocks are placed at an equal interval throughout the substrate (see Figure 3). In Figure 4(a) we found that *P(t)* can persist longer when *d* is larger up until *d* = 500. However, when *d* > 500, *P(t)* decays dramatically. This can be explained that since the feature sizes of our periodic pattern are fixed at a width of 1,000 lattice sites per block, when *d* > 500 the deposited atoms can move along the entire block, and so they do not nucleate on top of the blocks anymore. They have to be incorporated at the edges of the blocks where they can form more lateral bonds and therefore the information of the pattern is lost. For the multiple hit noise reduction technique, the results as shown in Figure 4(b) are not the same as Figure 4(a). Results presented in Figure 4(b) show that the survival of the pattern continues improving as the multiple hit factor *m* is increased. There is no apparent limit to the value of *m*. This is because multiple hit noise reduction technique is a process that considers only local configuration. The size of the pattern does not have any effect on the diffusion process in this case, which is not realistic. Hence, we cannot use this technique in thin film growth on periodic patterned substrates.

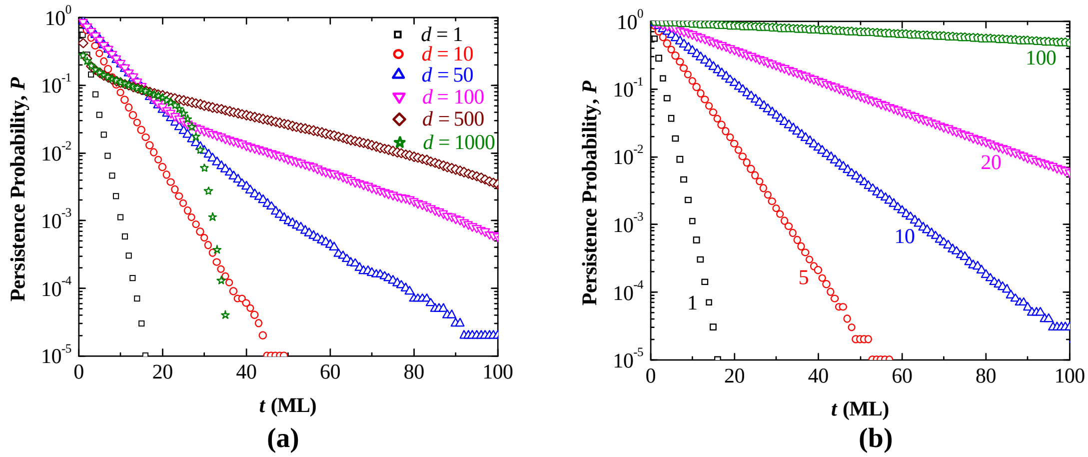

Figure 4. Persistence probability *P(t)* of periodic substrates: (a) the plot for different diffusion lengths *d* and (b) for different multiple hit factors *m*.

SUMMARY

In summary, we have shown that the flat patterned thin film persists for a long time when the value of either d or m is increased and there is no limit for these values in simulations. These results show that we can use the multiple hit noise reduction technique ($m > 1$) to replace the long surface diffusion length noise reduction technique ($d > 1$). In a physical meaning, larger d corresponds to higher substrate temperature T. Hence, we can say that the flat substrate has long-lived persistence when the film is grown at sufficiently high T. For periodic patterned substrates with fixed feature size, the multiple hit noise reduction technique is not a good choice. Moreover, we found that when d is very large (compared with the width of the feature size), persistence of the pattern decays dramatically since atoms cannot nucleate on the top of the blocks but rather hop down to fill empty space between each block. Increasing d, therefore, is limited by the width of the feature size of the initial pattern. From our results, it seems the optimal value for d is approximately half of the size of the pattern. So we conclude that the pattern can have long-lived persistence through the growth process when the film is grown at sufficiently high, but not too high, substrate temperature.

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