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Growth Mechanism of Microcrystalline Silicon Films by Scaling Theory and Monte Carlo Simulation*

Zi Wei, Zhou Yuqin†, Liu Fengzhen, and Zhu Meifang

(*College of Physical Science, Graduate University of the Chinese Academy of Sciences, Beijing* 100049, *China*)

Abstract: Hydrogenated microcrystalline silicon (μc-Si:H) films with a high deposition rate of 1.2nm/s were prepared by hot-wire chemical vapor deposition (HWCVD). The growth-front roughening processes of the μc-Si:H films were investigated by atomic force microscopy. According to the scaling theory, the growth exponent $\beta \approx 0.67$, the roughness exponent $\alpha \approx 0.80$, and the dynamic exponent $1/z = 0.40$ are obtained. These scaling exponents cannot be explained well by the known growth models. An attempt at Monte Carlo simulation has been made to describe the growth process of μc-Si:H film using a particle reemission model where the incident flux distribution, the type and concentration of growth radical, and sticking, reemission, shadowing mechanisms all contributed to the growing morphology.

Key words: μc-Si:H; growth mechanism; scaling theory; Monte Carlo simulations; reemission process
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1 Introduction

μc-Si:H thin films have been widely applied to solar cells, thin film transistors, and flat panel displays owing to their good opto-electronic properties. It is desirable to understand the growth mechanism of μc-Si:H film in order to improve its opto-electronic properties further. Low-temperature growth of μc-Si:H thin films generally leads to surface roughening. The roughening process can be modeled by the scaling theory. In the scaling theory, the root-mean-square surface roughness (w) has an exponential relation with film thickness (d) as $w \sim d^{\beta}$, where β is the growth exponent that describes how the vertical width of the surface scales with time[1]. The main growth process determines the surface morphology of the film, which can be characterized by specific values of scaling exponents. For example, β has the universal value of 1/2 for zero diffusion, 1/3 for finite diffusion, and 0 for step flow growth mode. Therefore, scaling analysis of the growth front provides an effective approach to study the growth mechanism of thin film.

The surface roughness studies of amorphous Si thin films prepared by plasma enhanced chemical vapor deposition[2~5], low-pressure chemical vapor deposition[6], sputtering[7], and thermal evaporation[8] have been investigated to get a basic understanding of film growth process. However, little is known about the growth mechanism of device-quality μc-Si:H films with a high deposition rate at low temperature in a HWCVD process.

In this paper, we report the surface roughness evolution of μc-Si:H thin film prepared at a high deposited rate by HWCVD. The scaling behavior from AFM images and the surface morphology simulated by the Monte Carlo method for μc-Si:H film growth are presented.

2 Experiment

The μc-Si:H thin films were deposited on glass (corning 1737) at the pressure of 5Pa by HWCVD. The distance between the filament and the substrate was 4cm, the hydrogen dilution ratio $R = H_2/(H_2 + SiH_4)$ was 88%, the silane flow rate was 18sccm, the substrate temperature was 250℃, and the filament temperature was 1900℃. The thicknesses of films were measured by Dektek 3st, and the deposition rate of 1.2nm/s was obtained from the thicknesses of films and the deposition time. The crystalline volume fraction (X_c) was estimated by Raman spectroscopy. X_c of all the films is about 50%. The surface morphologies of the films were observed by AFM (Nanonscope IIIa). The surface height and the w value of the film can be obtained by AFM images.

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† Corresponding author. Email: yqzhou@gucas.ac.cn

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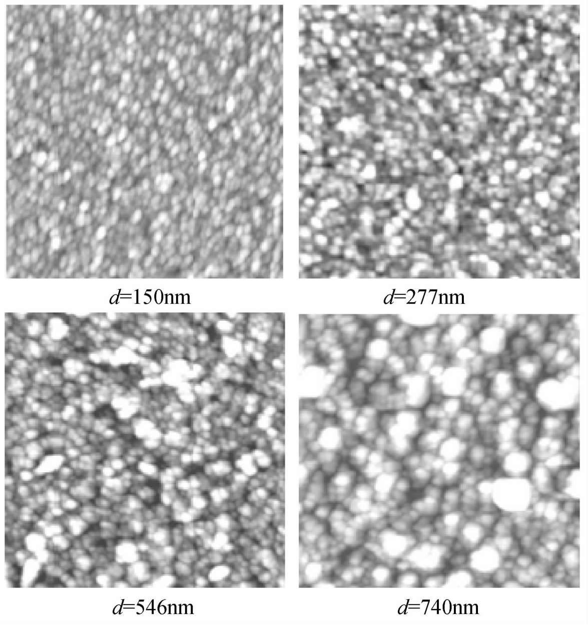
d=150nm d=277nm

d=546nm d=740nm

Fig.1 AFM images ($1\mu m \times 1\mu m$) for the films with d = 150, 277, 546, and 740nm, respectively

3 Results and discussion

Figure 1 shows the AFM images of the surface morphological growth of thin films with different film thicknesses (d). The size of the grain becomes larger as d increases. The d dependence of w in log-log scale is shown in Fig. 2, and the linear fit gives $\beta = 0.67$. The data for $d < 30$nm are not included in the fitting because the initial film growth does not obey the dynamic scaling theory. Further information on the surface morphology, i.e., the local and the lateral characteristics, can be extracted from the height-height correlation function $H(r) = \langle [h(r) - h(0)]^2 \rangle$, where $h(r)$ is the surface height at position r obtained by AFM, and the notation $\langle \cdots \rangle$ indicates a statistical average. The scaling hypothesis requires that $H(r) \sim r^{2\alpha}$ for $r \ll \xi$, and $H(r) = 2w^2$ for $r \gg \xi$[8], where α is the roughness exponent describing the surface texture in the short scaling range, and ξ is the lateral correlation length representing the lateral size of the mountains and valleys in the surface. ξ grows as $d^{1/z}$, where $1/z$ is the dynamic exponent. Dynamic scaling requires $z = \alpha/\beta$ for stationary growth.

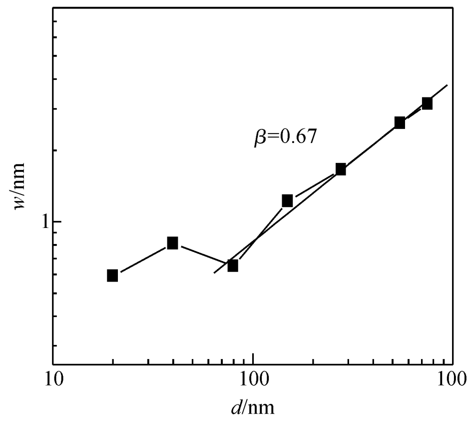

Fig.2 Dependence of w on d in log-log scale

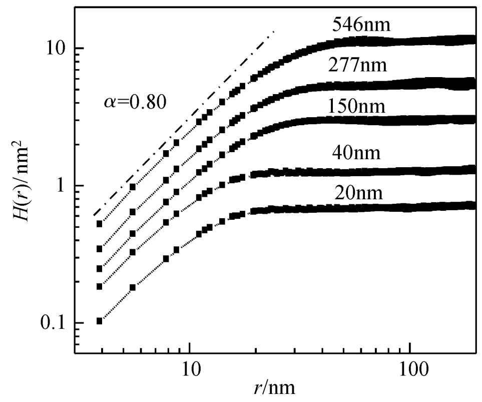

Fig.3 r-$H(r)$ curves in log-log scale for different d

The r-$H(r)$ curves for different d are plotted in Fig. 3. The overall behaviors of $H(r)$ curves are similar without regard to d. A kink appears between the linearly increasing and the saturation regime of the $H(r)$ curves, and then the values of ξ are determined. In the region $r \ll \xi$, the values of α can be deduced from $H(r)$-$r^{2\alpha}$. Although d is different, all the values of α are the same at ~0.80. The dependence of ξ on d is plotted in Fig. 4, which gives $1/z = 0.40$ by a linear fit. The value of $1/z$ ($= \beta/\alpha$) calculated from the parameters α and β obtained above is 0.84, which is not consistent with the fitting value of $1/z = 0.40$. This indicates that the μc-Si film growth cannot be scaled by the conventional scaling law, which requires a stationary growth mechanism.

The growth process caused by the non-stationary growth is more complicated and strongly dependent on the deposition conditions. A wide range of scaling exponents for Si thin films has been reported, and the reported exponents seem to depend on the deposition

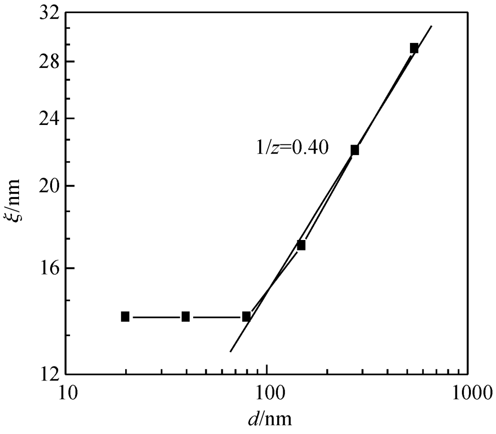

Fig.4 Dependence of ξ on d

methods as well as the experimental conditions. For example, Karabacak *et al*.[7] observed $\beta = 0.41 \pm 0.01$ by sputtering, Yang *et al*.[8] obtained $\beta = 0.26 \pm 0.02$ by thermal evaporation, Kondo *et al*.[5] found $\beta = 0.36$ and Dalakos *et al*.[10] got $\beta \approx 1$ by PECVD, and Richardson *et al*.[11] reported $\beta = 0.82$ by HWCVD. Roughness evolution of the film surface depends on type, concentration, energy, charge of the growth radicals, and sticking, reemission, shadowing, surface diffusion, and desorption processes. Therefore, Monte Carlo simulations may be helpful to understand the growth processes further.

4 Simulations

The simulation details were reported by Zhao *et al*.[12]. Briefly, the surface is described by a height function $h(x, y)$ defined on a $N \times N$ lattice ($N = 1024$). The periodic boundary condition is adopted in the simulation. One particle is deposited once and more than 10^8 particles are assumed to be deposited. A random site in the x-y plane is chosen for deposition. As a particle arrives at this position, it either sticks on a site with a certain probability or is reemitted. The average probability of sticking, referred to as the sticking coefficient (s), is a result of the complicated interactions between the incident radical and the surface. The s values of the species can range between 0 and 1, and are believed to be dominant in our simulations.

If reemitted, this particle may re-fall down to another site of the surface and then repeat the same process again. Nucleation forms island structures first, and the reemitted behavior means the incoming particle flux has an angular re-distribution that affects the surface morphology of the growth front. We call incident particles zeroth-order particles, while an nth-order particle that has been reemitted is called an $(n+1)$th-order particle. An nth-order particle has a probability s_n of sticking, where s_n is called the nth-order sticking coefficient. In fact, a reemission process is characterized by, among other things, its sticking coefficients. This reemission process has been applied to interpret growth front roughening in LPCVD successfully[6].

If all the factors and growth processes are considered in the simulations, the simulations will be difficult to carry out. At low substrate temperature, the surface diffusion and desorption of radicals can be neglected. Here, we assume that the different radical proportions and the sticking coefficient (s) are the main input parameters in the simulations.

Table1　Model, input parameters, and output results of Monte Carlo simulations

Model	Parameter		Result	
	Proportion of radicals (SiH/SiH_3)	s values	β	α
1st reemission	0.05/0.95	SiH: $s = 1$ SiH_3: $s_0 = 0.54, s_1 = 1.0$	0.49	0.70
4th reemission	0.05/0.95	SiH: $s_0 = 1$ SiH_3: $s_i = 0.54\ (i = 0 \sim 4)$ $s_5 = 1.0$	0.67	0.75

First, we determine the proportions of radicals. It is difficult to determine the absolute concentration of radicals under a certain deposited condition. According to the results reported by Nozaki *et al*.[13], the main growth radicals are SiH and SiH_3 under the deposition pressure of 6.7Pa and gas flow ratio of $F_{SiH_4}/F_{H_2} = 10/150$. The concentration of SiH_3 is about one order of magnitude higher than that of SiH. Our deposition conditions are similar to that of Nozaki and the ratio of particle numbers of SiH/SiH_3 is adopted from Ref.[13] to be 0.05/0.95.

Second, the values of s for SiH and SiH_3 need to be determined. According to the experimental results reported by Ho, Ramalingam, and Hamers *et al*.[14~16], we selected the s values of SiH and SiH_3 to be 1.0 and 0.54 as the input parameters in the simulations. All the input parameters are listed in Table 1.

The scaling exponents obtained through simulations are summarized in Table 1. By comparing the simulation results with the scaling exponents obtained from AFM images, we found that it is necessary to consider the high-order reemission process. Simulation results indicate that the scaling exponents converge as $n \geqslant 4$. For the 4th reemission, $\beta = 0.67$ and $\alpha = 0.75$ are obtained, which are in agreement with $\beta = 0.67$ and $\alpha = 0.80$ deduced from the AFM images. Figure 5 shows four two-dimensional simulated surface images obtained by considering the 4th reemission model. The grains become larger as the number of deposited particles increases. These images seem to be similar to the AFM images presented in Fig. 1.

Because of the shadowing effect, for the 1st reemission process with small s_0, more particles are remitted to the valleys than to the peaks, so the surface will be tend to be smooth. With the introduction of the nth reemission, the particle flux at the valley will have a greater probability of reemitting. Therefore, the smoothing effect will be weaker. Thus, the growth exponent β will be larger.

5 Conclusion

μc-Si:H thin films with a deposition rate of

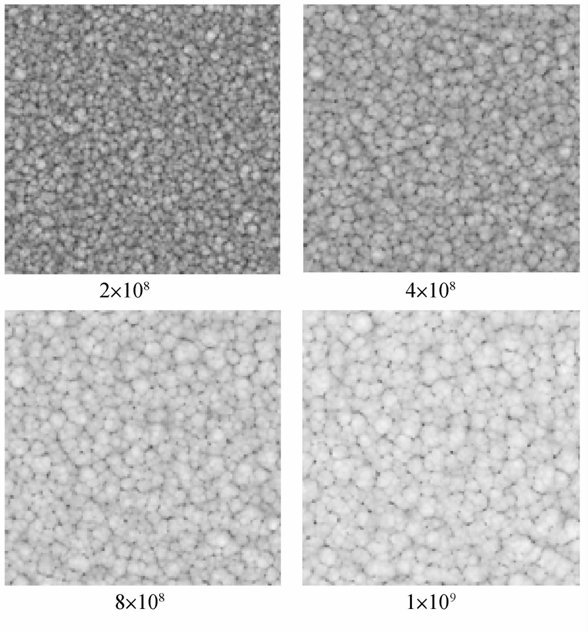

Fig.5 Two-dimensional images for 2×10^8, 4×10^8, 8×10^8, and 1×10^9 particles deposited on the growth surfaces by simulations

1.2nm/s were prepared by HWCVD. From the scaling analysis, the scaling exponents $\beta=0.67$, $\alpha=0.80$, and $1/z=0.40$ are obtained. The results of Monte Carlo simulation with the 4th reemission model are consistent with the experimental results. Thus, the incident flux distribution, the type and concentration of growth radical, and sticking, reemission, shadowing processes all affect the growth morphology of μc-Si:H thin films by HWCVD.

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用标度理论和蒙特卡洛方法研究微晶硅薄膜的生长机制*

訾 威 周玉琴[†] 刘丰珍 朱美芳

（中国科学院研究生院 物理科学学院，北京 100049）

摘要： 使用热丝化学气相沉积技术制备微晶硅薄膜(沉积速度为1.2nm/s)，通过原子力显微镜研究了薄膜前期生长的粗糙化过程.按照标度理论获得微晶硅薄膜的生长因子为$\beta\approx0.67$，粗糙度因子为$\alpha\approx0.80$，动力学因子为$1/z=0.40$.这些标度指数不能用一般的生长模型来解释.通过蒙特卡罗模拟给出与实验一致的结果.模拟表明，入射流方向、生长基元的类型和浓度、生长基元的粘滞、再发射和影蔽过程都对微晶硅薄膜的表面形貌有比较重要的影响.

关键词： 微晶硅薄膜；生长机制；标度理论；蒙特卡罗方法；再发射模型

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[†] 通信作者.Email:yqzhou@gucas.ac.cn

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