# Deposition pressure effect on the surface roughness scaling of microcrystalline silicon films*

Zhu Zhi-Li(朱志立), Ding Yan-Li(丁艳丽), Wang Zhi-Yong(王志永),
Gu Jin-Hua(谷锦华)†, and Lu Jing-Xiao(卢景霄)

*School of Physical Engineering and Material Physics Laboratory, Zhengzhou University, Zhengzhou 450052, China*

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The scaling behaviour of surface roughness evolution of microcrystalline silicon ($\mu$c-Si:H) films prepared by very-high frequency plasma-enhanced chemical vapour deposition (VHF-PECVD) has been investigated by using a spectroscopic ellipsometry (SE) technique. The growth exponent $\beta$ was analysed for the films deposited under different pressures $P_{\rm g}$. The results suggest that films deposited at $P_{\rm g} = 70$ Pa have a growth exponent $\beta$ about 0.22, which corresponds to the definite diffusion growth. However, abnormal scaling behaviour occurs in the films deposited at $P_{\rm g} = 300$ Pa. The exponent $\beta$ is about 0.81 that is much larger than 0.5 of zero diffusion limit in the scaling theory. The growth mode of $\mu$c-Si:H deposited at $P_{\rm g} = 300$ Pa is clearly different from that of $\mu$c-Si:H at $P_{\rm g} = 70$ Pa. Monte Carlo simulations indicate that the sticking process and the surface diffusion of the radicals are two key factors to affect the growth mode under different pressures. Under $P_{\rm g} = 300$ Pa, $\beta > 0.5$ is correlated with the strong shadowing effect resulting from the larger sticking coefficient.

**Keywords:** microcrystalline Si thin film, spectroscopic ellipsometry, the growth exponent, Monte Carlo simulations

**PACC:** 6855, 8155H, 7840, 7115Q

## 1. Introduction

In the last decade, $\mu$c-Si:H thin films have attracted a great deal of interest, especially for solar cell applications because of its better stability against prolonged illumination.[1,2] The $\mu$c-Si:H thin film can be deposited by radio-frequency plasma-enhanced chemical vapour deposition (RF-PECVD)[3] and very-high frequency plasma-enhanced chemical vapour deposition (VHF-PECVD).[4] Compared to RF-PECVD, VHF-PECVD has the advantages of higher electron density and lower ion energy, and thus the high-quality $\mu$c-Si:H film with high deposition rate can be successfully obtained by using VHF-PECVD.[5] The study of $\mu$c-Si:H films prepared by VHF-PECVD as a novel technique mainly focuses on the process of fabrication,[6−8] the growth mechanism is still not clear.

The deposition pressure is one of the key parameters to determine the property of $\mu$c-Si:H prepared by VHF-PECVD. The property of $\mu$c-Si:H varies with deposition pressure, which may result from the characteristic of the growth process, i.e. the sticking probability, the surface diffusion coefficient of the radical. A systemic study of $\mu$c-Si:H obtained by VHF-PECVD on the growth mechanism under different deposition pressures is helpful to have a better insight into the growth process.

The growth of Si thin films using VHF-PECVD at low temperatures (∼200 °C) is a non-thermal equilibrium process. The thin film growth fronts under nonequilibrium conditions always show the scaling behaviour.[9−11] In the scaling regime, the root-mean-square surface roughness $w$ increases as a power law of the film thickness $d$, $w \sim d^{\beta}$, where $\beta$ is the growth exponent related to the film growth mechanism. The values of $\beta$ are 1/2 for zero diffusion, 1/3 for finite diffusion and 0 for the step flow growth.[12] Therefore, scaling analysis of thin-film surface is an effective approach to study the film growth mechanism.

In this paper, two sets of $\mu$c-Si:H films grown for various lengths of deposition time are prepared by VHF-PECVD at different deposition pressure $P_{\rm g}$ of 70 and 300 Pa. We investigate the evolution of the surface roughness in the process of the $\mu$c-Si:H growth by spectroscopic ellipsometry (SE), derive the $\beta$ values from the scaling analysis, and discuss the possible

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†Corresponding author. E-mail: gujinh@zzu.edu.cn

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growth mechanisms from the $\beta$ values. We also simulate the growth process by the Monte Carlo method and indicate what factors influence the growth process under different pressures.

## 2. Experimental details

Two sets of $\mu$c-Si:H films for various lengths of deposition time were prepared by VHF-PECVD with the deposition pressures $P_g$ of 70 and 300 Pa. Substrates were the glass. Other deposition parameters were as follows: excitation frequency 75 MHz, distance between two electrodes 1.5 cm, silane concentration (SC = $[SiH_4]$ / ($[H_2]$ + $[SiH_4]$)) 4%, substrate temperature 220 °C, electrical source power 140 W, and total gas flow rate 250 sccm. For $P_g$ = 70 Pa, films were deposited for the growth time being 1, 2, 3, 5, 10, 20, 30, 60, 74 min. For $P_g$ = 300 Pa, films were deposited for the growth time of 1, 2, 5, 7, 10 min. The ellipsometry spectra ($\psi$, $\Delta$) of Si films were obtained using VASE variable angle spectroscopic ellipsometer (J. A. Wollam) in the spectral range of 204∼1000 nm. The angle of incidence is fixed at 70°.

## 3. Results and discussion

Figure 1 shows the SE spectra for the growth time $t$ = 20, 30, 60 and 74 min at $P_g$ of 70 Pa. The dashed lines are the fit spectra using the two-layer model. In the two-layer model, the dielectric function of the surface roughness layer ($\varepsilon_{surf}$) was modeled as a 50/50-vol % mixture of the dielectric functions of bulk layer ($\varepsilon_{bulk}$) and voids using the effective medium theory (EMT). The surface roughness layer thickness $d_s$ and bulk layer thickness $d_b$ for $P_g$ of 70 Pa was determined using the Wvase32 software. The film thickness $d$ is calculated by $d_b + d_s$. The study indicates that $d_s$ is linear with $w$,[13,14] so $d_s$ and $d$ have the relation of $d_s \sim d^\beta$. The log–log plot of surface roughness layer thickness $d_s$ versus the film thickness $d$ is shown in Fig. 2. The linear fit of Fig. 2 yields $\beta \sim 0.22$, the data of $d < 34$ nm are not included in the fitting because the film growth in this regime does not obey the scaling theory. The parameter $\beta \sim 0.22$ corresponds to the definite diffusion growth, namely, after the radicals arrive at the growth surface, they may diffuse around and stick at a position with lower energy.

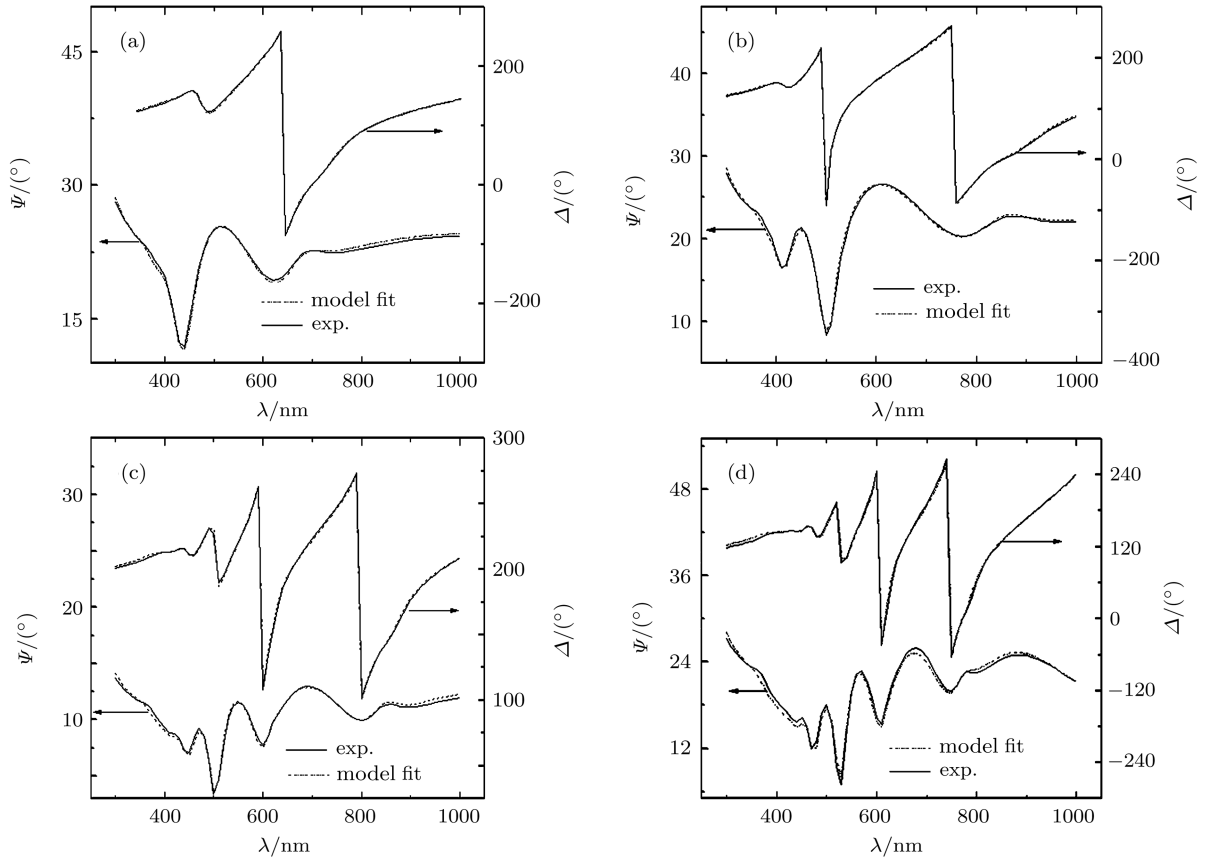



**Fig. 1.** SE spectra of $\mu$c-Si:H films deposited at $P_g$ of 70 Pa for (a) 20 min, (b) 30 min, (c) 60 min and (d) 74 min. The dashed lines are the fit spectra using the two-layer model.

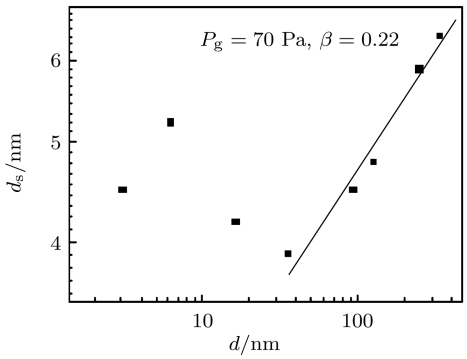



**Fig. 2.** The variation of surface roughness layer thickness $d_s$ with the film thickness $d$ for films deposited at $P_g = 70$ Pa.

Figure 3 shows the SE spectra for the growth time of $t = 2$, 5, 7 and 10 min at $P_g = 300$ Pa. The log–log plot of surface roughness layer thickness $d_s$ versus the film thickness $d$ is shown in Fig. 4. The linear fit of Fig. 4 yields $\beta \sim 0.81$, which is much larger than 0.5 of zero diffusion limit in the scaling theory. Raman spectra of $\mu$c-Si:H films with film thickness of about 200 nm deposited at $P_g$ of 70 and 300 Pa are shown in Fig. 5. It shows that the films deposited at different pressures are both crystallized, and the crystalline fraction decreases with the deposition pressure.

The $\mu$c-Si:H deposited at different pressures shows two distinct surface scaling behaviours. To figure out what factors affect the surface scaling behaviours of $\mu$c-Si:H film deposited at different pressures, we simulate the growth process of $\mu$c-Si:H using Monte Carlo method. The simulation program was provided by Zhao, the details were given in Refs. [10] and [11]. The basic growth process is as follows: A particle falls on a random position on the surface in the $x$ and $y$ plane with a 1024×1024-lattice size, and it is either stuck on the surface with a certain probability or re-emitted. The average probability of sticking is defined as the sticking coefficient $s$. Once the particle is stuck on the surface, it can diffuse on the surface. The surface diffusivity of the radical depends on the energy of the radical, the hydrogen coverage of the surface, and the substrate temperature. $D/F$ is introduced to describe the surface diffusion, where $D$ denotes the number of the surface diffusion atoms within a unit time, in which $F$ atoms are deposited on the surface. We also take into account the shadowing effect, i.e. some surface points are shadowed from the incident fluxes of particles due to nearby high surface features. The shadowing effect results in the distribution of the incident flux on the growing surface, being not uniform, i.e. the local low surface receives the fewer incoming particles than the high surface.

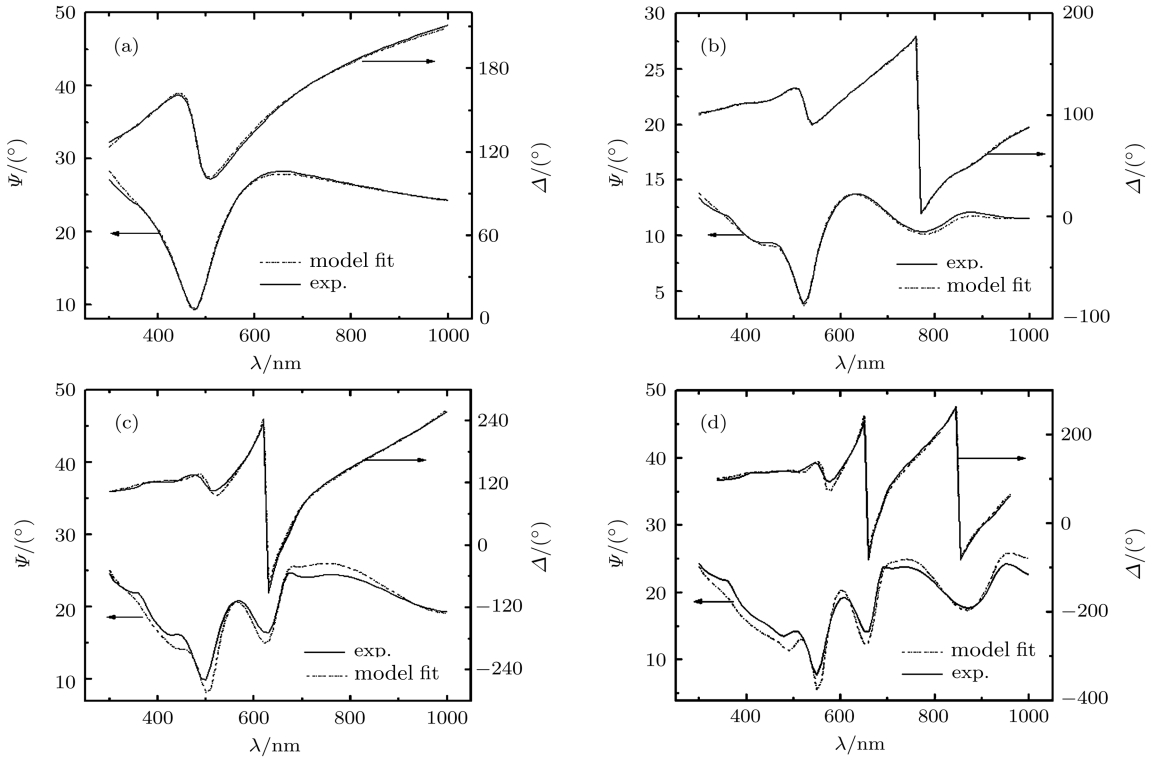



**Fig. 3.** SE spectra for $P_g$ of 300 Pa measured at (a) 2 min, (b) 5 min, (c) 7 min, and (d) 10 min. The dashed lines are the fit spectra using the two-layer model.

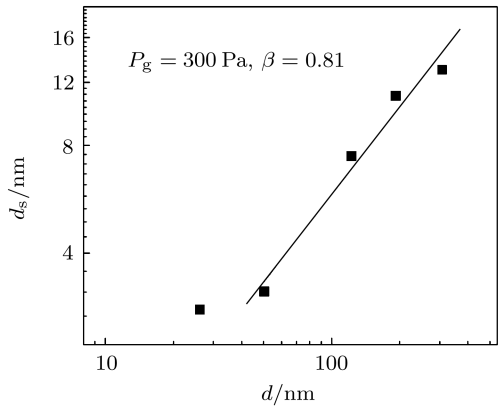



**Fig. 4.** The variation of surface roughness layer thickness $d_s$ with the film thickness $d$ for films deposited at $P_g = 300$ Pa.

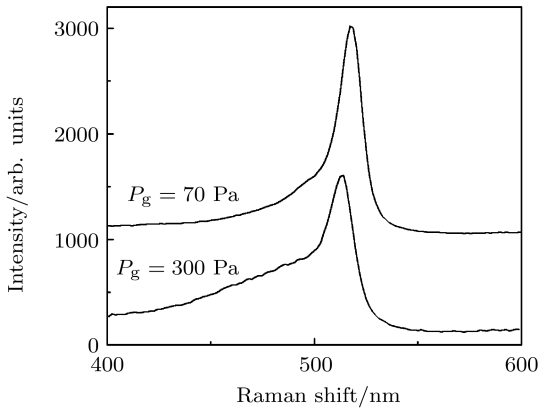



**Fig. 5.** Raman spectra for $\mu$c-Si:H films with film thickness of 200 nm deposited at $P_g$ of 70 and 300 Pa.

As for the value of $s$, we refer to the experimental data of Hamers.[15] Hamers reported the surface reaction probability $P_{sr} = 0.2$ for $\mu$c-Si:H with the deposition rate lower than 1 Å/s (1 Å = 0.1 nm) by PECVD. For $P_{sr} = s + \gamma$, where $\gamma$ is the probability of desorption, the desorption process will not occur when the substrate temperature is below 400 °C. In our case, the substrate temperature is 220 °C, so $P_{sr} = s$. For the $\mu$c-Si:H deposited at 70 Pa, the deposition rate is 0.5 Å/s, close to the value in the experiment of Ref. [15], so we set $s = 0.2$, $D/F = 10$. For the $\mu$c-Si:H deposited at 300 Pa, the deposition rate is 5 Å/s far higher than the value in the experiment of Ref. [15], so we have to consider the effect of the deposition rate. At 300 Pa, the particles fall so fast that some of the re-emitted particles have not enough time to re-emit before being covered by the particles of the next layer. This is equivalent to an increase in sticking probability of particles, so we set $s = 0.6$ for the $\mu$c-Si:H deposited at 300 Pa. The lower electron temperature and higher deposition rate at high pressure of 300 Pa, compared with 70 Pa, decreases the surface diffusion of the adatom, thus we set $D/F$ =5 at 300 Pa deposition pressure.

Figures 6 and 7 show the simulated surface images for $\mu$c-Si:H deposited at 70 and 300 Pa, respectively. From Figs. 6 and 7, we see that the $\mu$c-Si:H deposited at different pressures show distinct surface morphology. The clusters are larger and the surface is rougher for $\mu$c-Si:H films deposited at 300 Pa than those deposited at 70 Pa. Figure 8 illustrates the variation of the root-mean-square surface roughness $w$ with film thickness $d$. From the linear fit, we obtain $\beta = 0.23$ and $\beta = 0.81$ for $\mu$c-Si:H deposited at 70 and 300 Pa, respectively, very close to the values of $\beta$ obtained from the experiments. In the Monte Carlo growth simulations, only the parameters $s$ and $D/F$ are varied. This implies that sticking process and surface diffusion of the radicals are two key factors to affect the growth mode at different pressures.

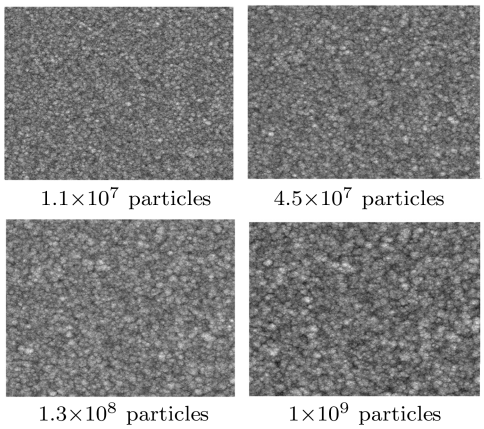



**Fig. 6.** Simulated surface images of Si films for $1.1\times10^7$, $4.5\times10^7$, $1.3\times10^8$ and $1\times10^9$ particles on the growth surfaces at $P_g = 70$ Pa.

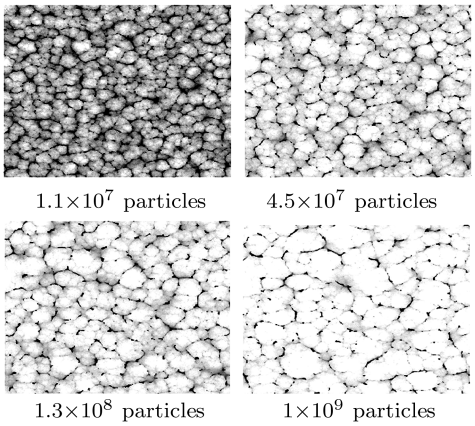



**Fig. 7.** Simulated surface images of Si films for $1.1\times10^7$, $4.5\times10^7$, $1.3\times10^8$ and $1\times10^9$ particles on the growth surfaces at $P_g = 300$ Pa.

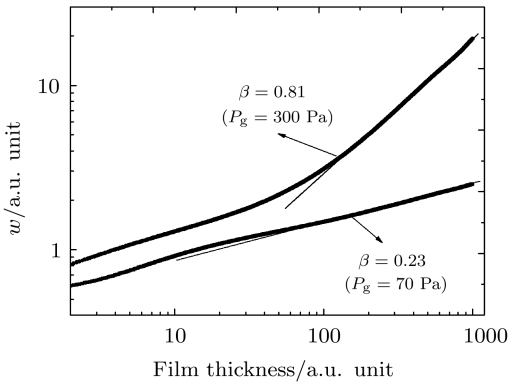



**Fig. 8.** The simulated surface roughness $w$ versus film thickness $d$ for $\mu$c-Si:H deposited at $P_g$ of 70 and 300 Pa.

In the KPZ theory,[12] the maximum $\beta$ is 0.5 on considering that the incident flux falls uniformly on the growing surface. In our case, considering the shadowing effect, the high point receives more incoming particles than the lower point, which increases the surface roughness, so $\beta$ may be larger than 0.5. Zhao[10] reported that the strength of shadowing effect increases with the sticking coefficient, and the value of $\beta$ increases abruptly larger than 0.5 when $s > 0.5$ owing to the strong shadowing effect. At 300 Pa, the sticking coefficient $s$ is 0.6 and the shadowing effect is strong, so we obtain the much greater $\beta = 0.81$.

# 4. Conclusion

The scaling behaviour of surface roughness evolution of $\mu$c-Si:H deposited under different pressure $P_g$ by VHF-PECVD has been investigated. For films deposited at $P_g = 70$ Pa, the growth exponent $\beta$ is about 0.22, which corresponds to the definite diffusion growth. However, films deposited at $P_g = 300$ Pa show abnormal scaling behaviour with the exponent $\beta$ of about 0.81 that is much larger than 0.5 of zero diffusion limit in the scaling theory. In the Monte Carlo growth simulations, the parameters $s$ and $D/F$ were varied, the values of $\beta$ obtained from the simulations are very close to the experimental results. This indicates that sticking process and surface diffusion of the radicals are two key factors to affect the growth mode under different pressures. For films deposited at $P_g = 300$ Pa, $\beta$ larger than 0.5 is related to the strong shadowing effect owing to a larger sticking probability.

# References

[1] Yan B J, Yue G Z, Banerjee A, Yang J and Guha S 2004 *Mater. Res. Soc. Symp. Proc.* **808** A **9** 41

[2] Meier J, Dubail S, Fliickiger R, Fischer D, Keppner H and Shah A 1994 1994 *IEEE First WCPEC* Hawaii

[3] Hou G F, Xue J M, Sun J, Guo Q C, Zhang D K, Ren H Z, Zhao Y, Geng X H and Li Y G 2007 *Acta Phys. Sin.* **56** 1177 (in Chinese)

[4] Guo X J, Lu J X, Cheng Y S, Zhang Q F, Wen S T, Zheng W, Shen D H and Cheng Q D 2008 *Acta Phys. Sin.* **57** 6002 (in Chinese)

[5] Sobajima Y, Nakanoa S, Nishinoa M, Tanakaa Y, Toyamaa T and Okamoto H 2008 *J. Non-Cryst. Solids* **354** 2407

[6] Niikura C, Kondo M and Matsuda A 2004 *Proceedings of the 19th European Photovoltaic Solar Energy Conference* Paris, France, p. 1637

[7] Mai Y, Klein S, Carius R, Stiebig H, Geng X and Finger F 2005 *Appl. Phys. Lett.* **87** 073503

[8] Mai Y, Klein S, Geng X and Finger F 2004 *Appl. Phys. Lett.* **85** 2839

[9] Rezek B, Stuchlík J, Fejfar A and Kočka J 2002 *J. Appl. Phys.* **92** 587

[10] Zhao Y P, Drotar J T, Wang G C and Lu T M 2001 *Phys. Rev. Lett.* **87** 136102

[11] Karabacak T, Zhao Y P, Wang G C and Lu T M 2001 *Phys. Rev.* B **64** 085323

[12] Karder M, Parisi G and Zhang Y 1986 *Phys. Rev. Lett.* **56** 889

[13] Fujiwara H, Kondo M and Matsuda A 2001 *Phys. Rev.* B **63** 115306

[14] Teplin C W, Levi D H, Lwaniczko E, John K M, Perkins J D and Branz H M 2005 *J. Appl. Phys.* **97** 103536

[15] Hamers E A G, Morral A F I and Niikura C 2000 *J. Appl. Phys.* **88** 3674