

Dynamic scaling phenomena and universality classes in growth of iron nitride thin films deposited by direct current magnetron sputtering

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Abstract

In this paper, dynamic scaling approach has been used to investigate the growth of iron nitride films, which were deposited by direct current magnetron sputtering using an Ar/N₂ gas mixture (N₂/(N₂+Ar)=30%) at room temperature and 250 °C substrate temperature. The structure of the deposited films was determined by using X-ray diffraction. The perpendicular fluctuations in the height $h(x, t)$ of the surface were analyzed by atomic force microscopy and grazing incidence X-ray scattering in the light of dynamical scaling approach, and the two dependent nontrivial exponents, roughness exponent α and growth exponent β , were determined. For the iron nitride films grown at room temperature and 250 °C, $\alpha=0.39\pm 0.01$ and 0.30 ± 0.02 and $\beta=0.29\pm 0.03$ and 0.28 ± 0.07 , respectively, which were in agreement with a type of universality that was suggested by Kardar, Parisi and Zhang. It might be concluded that it was desorption rather than the surface diffusion that dominated the relaxation process in both the case of room and higher substrate temperatures for the deposition of iron nitride thin films. © 2005 Elsevier B.V. All rights reserved.

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1. Introduction

The growth of variety of films under inhomogeneous deposition will lead to the formation of rough surface. The quantitative description of rough surfaces far from equilibrium has been an important aspect for years [1] because of its practical importance for the growth of solid films and the great scientific and practical interest [2,3]. Solid films growth under far from equilibrium conditions exhibit scale-invariance phenomenon and naturally evolve into self-affine fractal [4,5], which can be described by the dynamic scaling form:

$$\sigma(L, t) = L^\alpha f\left(t/L^{\alpha/\beta}\right) \quad (1)$$

The exponent α in Eq. (1) is known as the “static scaling” exponent or “roughness exponent” which characterizes the L dependence of the saturated width σ , for $t \gg L^{\alpha/\beta}$. L is the sample size perpendicular to the growth direction. The parameter β is the growth exponent, for $t \ll L^{\alpha/\beta}$. Much work [6–12] has been done to determine the values of the growth exponents analytically and numerically. It has been shown that models with locally very different growth rules can almost give similar values for the exponents of α and β , which suggests the existence of universality [5]. Some models [13] are representative of the Edwards–Wilkinson universality classes of linear growth; while much more of the others belong to the universality class of Kardar–Parisi–Zhang (KPZ) equation [14,15], which is given as follows:

$$\partial h / \partial t = v \nabla^2 h + \lambda (\nabla h)^2 + \eta(r, t) \quad (2)$$

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where surface height $h(x, t)$ is a function of position x (in the substrate) and time t . λ determines the average growth velocity and ν makes the surface smoother, while η is a Gaussian random noise, which makes the surface rougher. KPZ equation provides a convenient basis for the discussion of universality among different lattice growth models. It is well known that growth processes belonged to the KPZ universality all allow the formation of overhangs or voids, and desorption of the adsorbed particles should not be negligible. The exponents have the relationship of $\alpha + \alpha/\beta \approx 2$. Among these models, solid-on-solid model has been extensively studied and used in both equilibrium and nonequilibrium crystal growth [16]. Kim and Kosterlitz [17] have studied the growth of the interface in an irreversible restricted solid-on-solid model and values of $\alpha = 2/(d+2)$, $\beta = 1/(d+1)$ ($d-1$ is the substrate dimension) have been obtained, which belongs to the KPZ universality class. In their model, $(d-1)$ -dimensional lattice is randomly selected and particles are added to or removed from a lattice subject to solid-on-solid with a restriction on neighboring heights. However, according to Sarma and Ghaisas [16], local atomistic relaxation rules and its dependence on the atomic coordination number determine the universality class of the growth model, i.e. the morphology of the rough films surface and their growth mechanism. They indicated that at higher substrate temperature, various crossover effects would be observed as surface diffusion dominates the relaxation process and growth under this situation may not belong to KPZ universality class.

Fe_xN alloys and compounds have been widely studied since they are excellent materials due to their variety of structural and magnetic properties, high corrosion resistance, and high wear resistance. It has been reported, Fe_xN phases are ferromagnetic at room temperature when $x \geq 2.26$ [23–24]. In particular, among the different phases, γ' - Fe_4N and α'' - Fe_{16}N_2 phases are the most important compounds. It may be a possible candidate for high-density magnetic recording media owing to its very high magnetic moment. However, up to now, there may still be problems needed to be resolved [18–21]. As to our knowledge, there is little report on the investigation of the dynamical scaling behavior of iron nitride films although dynamic scaling may be a useful method for understanding the formation of structure of the thin films. In this work, we obtain iron nitride films by dc magnetron systems at room and 250 °C substrate temperature. Using dynamic scaling approach, the growth of iron nitride thin films deposited at different substrate temperatures has been discussed, and the universality class of such Fe–N thin films has been determined.

2. Experimental details

Iron nitride films were deposited on Corning glass substrates in mixed Ar/ N_2 discharges at different substrate

temperatures (room temperature and at 250 °C substrate temperature, respectively) using dc magnetron sputtering high purity (99.99%) α -iron (60 mm in diameter). The distance between the substrate holder and the target was 6.5 cm. The base pressure was 5×10^{-5} Pa. Prior to deposition, the substrates were cleaned ultrasonically in acetone and alcohol consecutively, and then baked in a vacuum chamber at 250 °C for 2 h and cooled down to room temperature. During sputtering, the dc power and the magnet exciting power were kept constant at 110 W and 45 W, respectively, and the total pressure was fixed at 2.0 Pa. Pure argon (99.999%) and nitrogen (99.999%) gases were fed into the chamber, controlled by two independent mass-flow controllers. The argon gas flow was fixed at 47.4 sccm, while the nitrogen fraction in whole gas flow was 30%. The thickness of all the iron nitride thin films was about 240 nm, as measured using Surface Profilometry (DekTak 3).

The structure of the films was analyzed using X-ray diffraction (XRD) with Cu $K\alpha$ radiation using a current of 150 mA and voltage of 40 kV (Rigaku, D/MAX-rA). The diffraction patterns were recorded using a monochromatic wavelength $\lambda = 0.15406$ nm, filtered by a graphite monochromator. The measurements were done in the range $20^\circ \leq 2\theta \leq 100^\circ$ in the $\theta/2\theta$ mode with a 2θ angle scanning rate of $0.02^\circ/\text{sec}$. The experimental errors of the $\Delta 2\theta$ was about 2%. The surface morphology of the films was characterized using atomic force microscopy (AFM) (Digital Instruments Nanoscope III scanning probe microscopy), using Si_3N_4 as tip, which is 4 μm long and 10° with central line. The probe is non-contacted to a cantilever spring, which has a length of 180 μm , a width of 38 μm , a thickness of 1.0 μm , a force constant of 0.4 N m^{-1} . The scan area was 1–5 μm , the scan speed was 1 Hz and the sampling rate was 256 points per line. Grazing-incidence X-ray analyses, including small-angle scattering and asymmetry diffraction, were performed using a Philips X'Pert diffractometer with Cu $K\alpha$ radiation. The parameters used were: X-ray wavelength $\lambda = 0.15406$ nm, voltage 40 kV and current 40 mA. Both specular and diffuse X-ray scattering tests were carried out with a fixed incidence angle of 0.5° .

3. Results and discussion

3.1. Film structure

Structures of the iron nitride thin films have been analyzed using XRD. Fig. 1 shows the XRD patterns of the Fe–N films deposited on glass substrates at different substrate temperatures. The diffraction pattern of the samples was indexed as belonging to ϵ - Fe_{2-3}N and ζ - Fe_2N . At the 250 °C substrate temperature, the intensity of the XRD diffraction pattern for ϵ (300), ζ (040) (023) is much higher than that of the samples prepared at room

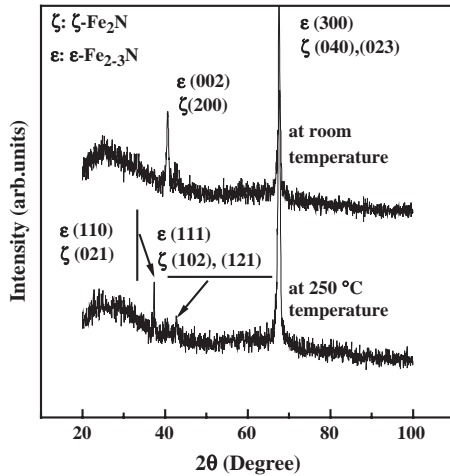


Fig. 1. The XRD patterns for iron nitride films grown at different substrate temperatures.

temperature, which indicates that the preferred orientation is formed more easily at higher growth temperature.

3.2. Film growth dynamic scaling

To determine the scaling exponents and the universality class of iron nitride thin films deposited by DC magnetron sputtering, AFM, X-ray specular scattering, and X-ray diffuse scattering were used. Surface roughness, $\sigma(t)$, roughness of the samples increase with deposition time for the two deposition temperatures and as a function of deposition time for samples grown at room temperature and at 250 °C substrate temperature are given in Fig. 2. The $\log(\sigma(t))$ increases linearly with $\log(t)$. The slopes of Fig. 2 yield β values of 0.29 ± 0.03 and 0.28 ± 0.07 , respectively, for the samples grown at room temperature and 250 °C substrate temperature.

From the specular reflectivity data of the films deposited at different substrate temperatures we can obtain the values

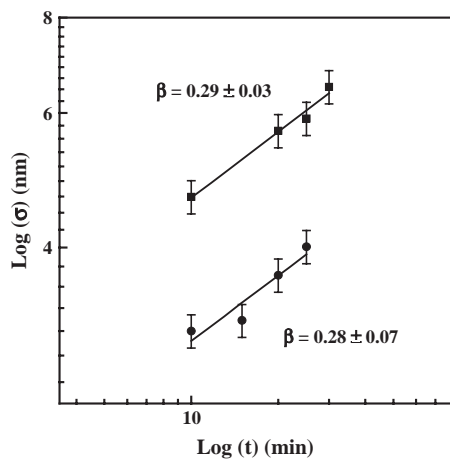


Fig. 2. AFM surface roughness, as rms, of samples prepared at room temperature and at 250 °C as a function of deposition time. The slopes of the $\log(\sigma(t))-\log(t)$ are 0.29 ± 0.03 and 0.28 ± 0.07 , respectively.

of surface roughness width, which are in agreement with the results from AFM. For diffuse scattering the angle of incidence is not equal to the angle of reflection and the component of scattering is related to the height–height correlations of the rough surface. Sinha et al. [12] have calculated the diffuse scattering produced by surface roughness. The diffuse cross-section of self-affine surfaces with no cut-off has the asymptotic form

$$I(q_z) \sim (L_x L_y) e^{-2/\alpha} q_z^{-(2+1/\alpha)} \quad (3)$$

where $q_z = 4\pi/\lambda \sin\theta$ is the wave vector transfer perpendicular to the surface and $L_x L_y$ the area illuminated by the beam. The data obtained using diffuse reflectivity are shown in Fig. 3, in the form of an Ln–Ln plot of intensity versus q_z for the films grown at different N_2 fraction. According to Eq. (3) the $\text{Ln}(I) \sim \text{Ln}(q_z)$ plots will be straight lines with slopes $-(2+1/\alpha)$ if the surface is self-affine. Thus, α can be determined. The scaling exponent $\alpha = 0.39 \pm 0.01$ and 0.30 ± 0.02 for samples grown at room temperature and 250 °C substrate temperature, respectively, which were obtained from the best fit of the linear portions of the graphs.

A survey of the exponents observed for a variety of systems [4] reveal scattered data. The exponents in general fall in the range of $0.2 \leq \alpha \leq 1.0$, $0.2 \leq \beta \leq 0.56$. Our results also lie in this range. As to the sample deposited at room temperature, the values of $\alpha = 0.39 \pm 0.01$ (roughness exponent), $\beta = 0.29 \pm 0.03$ (growth exponent), and $\alpha + \alpha/\beta \approx 2$ are quite consistent with those of the computer simulations [1,22], which means that the growth of sample deposited at room temperature belongs to the restricted solid-on-solid model (RSOS) and also, the growth of iron nitride thin films deposited at room temperature belong to the KPZ universality class. In this model, Incident atoms stick to the tops of the already existing substrate atoms and a restriction on the maximum height difference between

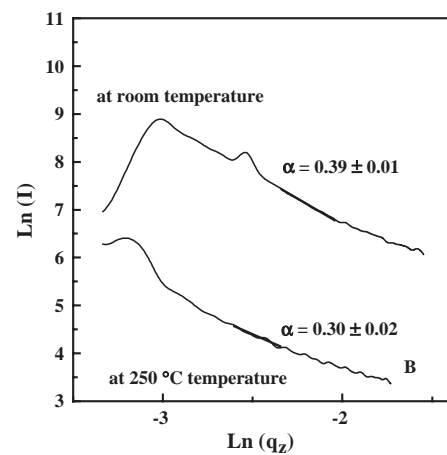


Fig. 3. The data obtained at the diffuse reflectivity conditions for samples prepared at room temperature and at 250 °C. The scaling exponent α is obtained from the best fit of the linear portions.

neighboring columns is imposed. Desorption is the main atomistic relaxation rule in the growth process. For the films grown at 250 °C substrate temperature, overhangs or voids also formed, and although the exponents of $\alpha=0.30\pm 0.02$ and $\beta=0.28\pm 0.07$ do not belong to any model available, the value of $\alpha+\alpha/\beta$ is near to 2, which implied that the growth of sample deposited at 250 °C substrate temperature also belong to the KPZ universality class. The above results show that for the deposition of iron nitride thin film, higher substrate temperatures have little influence on the atomistic relaxation rule and desorption still dominates the relaxation process.

4. Conclusions

The iron nitride films with different phases can be deposited on Corning glass by D.C. magnetron sputtering using an Ar/N₂ working gas mixture at various Ar/N₂ gas mixture ratio and/or substrate temperatures. Using the dynamic scaling approach, both the roughness exponent α and the growth exponent β can be obtained. It is found that the magnetic properties and the values of α and β of Fe–N thin films strongly depend on the nitrogen pressure [25,26]. When the N₂/(N₂+Ar)=30%, the Fe–N thin films are nonmagnetic, and the values of roughness exponent and growth exponent are $\alpha=0.39\pm 0.01$, $\beta=0.29\pm 0.03$ and $\alpha=0.30\pm 0.02$, $\beta=0.28\pm 0.07$, according to different substrate temperatures (at room temperature and 250 °C, respectively). For the sample deposited at room temperature, the exponents are quite consistent with those of the computer simulations reported, which means that the growth of sample deposited at room temperature belongs to the RSOS model. For samples prepared at higher temperature, although the exponents of α and β changed, $\alpha+\alpha/\beta$ is still near to 2, which means that under different growth temperature, the particles deposition might belong to different model. However, as desorption is the dominant relaxation process, the growth of samples deposited at room temperature and 250 °C substrate temperature belongs to the KPZ universality class.

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