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On the dependence of surface undulation on film thickness

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ABSTRACT

This paper investigates the dependence of surface undulation on a film thickness considerably greater than the critical value of a thin film system. It considers that surface tension and residual stress are the main cause of surface undulation. The study found that there is a critical undulation wavelength that minimizes the free energy of a thin film system, that this critical wavelength depends on the film thickness, and the effect of undulation amplitude is insignificant. The research also found that the surface undulation has a negligible influence on the residual stresses in the thin film system.

1. Introduction

The surface of a vapour deposited thin film is not flat, but undulate. Fig. 1 shows some room-temperature scanning probe microscopy (SPM) images of three silicon film surfaces which were epitaxially grown on a sapphire substrate under a same temperature. It can be seen that the wavelength and amplitude of the surface undulation depend on the thickness of the film: increasing the film thickness aggravates both the wavelength and amplitude of the surface undulation. When the film thickness t is 500 nm, the undulation wavelength and amplitude are around 0.5 μm and 4 nm, respectively. As the thickness increases to 5 μm, the wavelength and amplitude increase to around 1.5 μm and 15 nm.

Intuitively, the undulate surface is the consequence of the epitaxial crystal growth. However, different growth theories, such as the Volmer–Weber growth [1–4], the Frank-van der Merwe growth [5,6] and the Stranski–Krastanov growth all conclude that the undulation wavelength does not change with the film thickness when the thickness is beyond a critical value. Mathematical models such as the KPZ model [7] and Eden model [8] do not show the relationship between the undulation wavelength and film thickness. Tiedje and Ballestad [9] argued that the critical wavelength may increase with film thickness at the initial stage of film growth, and that undulation in the surface tends to smooth out during the growth. There is a lack of understanding of the dependence of undulation wavelength and amplitude on film thickness. The existing film growth theories only indicate that if surface undulation occurs during a growth process, its wavelength should be the same or only slightly different from that of the substrate surface. Experimental results, however, have demonstrated significant amplifications of wavelength and amplitude of the surface undulation when film thickness increases. It is therefore clear that the existing growth theories are unable to justify the phenomenon exhibited in Fig. 1.

A way to understand the surface undulation is based on the rationale that surface undulation is to minimize the free energy of the thin film system and in turn to stabilize the surface morphology. Freund and Suresh [10] calculated the critical wavelength for the small amplitude sinusoidal surface. By assuming that there was no surface traction, they obtained the free energy density χ as:

$$\chi = U_m + 4\pi\frac{A}{\lambda}\left[\frac{\pi}{\lambda}\gamma-(1+\nu)U_m\right]\cos\frac{2\pi x}{\lambda} \tag{1}$$

where $U_m=\sigma_m^2/M$ is the uniform strain energy density of the material due to a remote biaxial stress field σ_m; M is the biaxial elastic modulus; γ is the surface energy density; A and λ are the amplitude and wavelength, respectively, and ν is Poison's ratio. If A/λ is small, the change rate of the total free energy associated with the variation of the amplitude can be written as

$$\dot{F}(t)=\int_0^\lambda \chi \nu_n dx = 2\pi A\dot{A}\left[\frac{\pi}{\lambda}\gamma-(1+\nu)U_m\right] \tag{2}$$

where $\nu_n=\dot{A}\cos(2\pi x/\lambda)$ is the undulation growth rate. According to Eq. (2), if $(\pi/\lambda)\gamma>(1+\nu)U_m$, the system can lower its free energy by reducing the amplitude towards naught. If $(\pi/\lambda)\gamma<(1+\nu)U_m$,
it can lower its energy by increasing the waviness amplitude. Therefore, Freund and Suresh [10] concluded that the critical wavelength is

$$\lambda_c=\frac{\pi\gamma}{(1+\nu)U_m} \tag{3}$$

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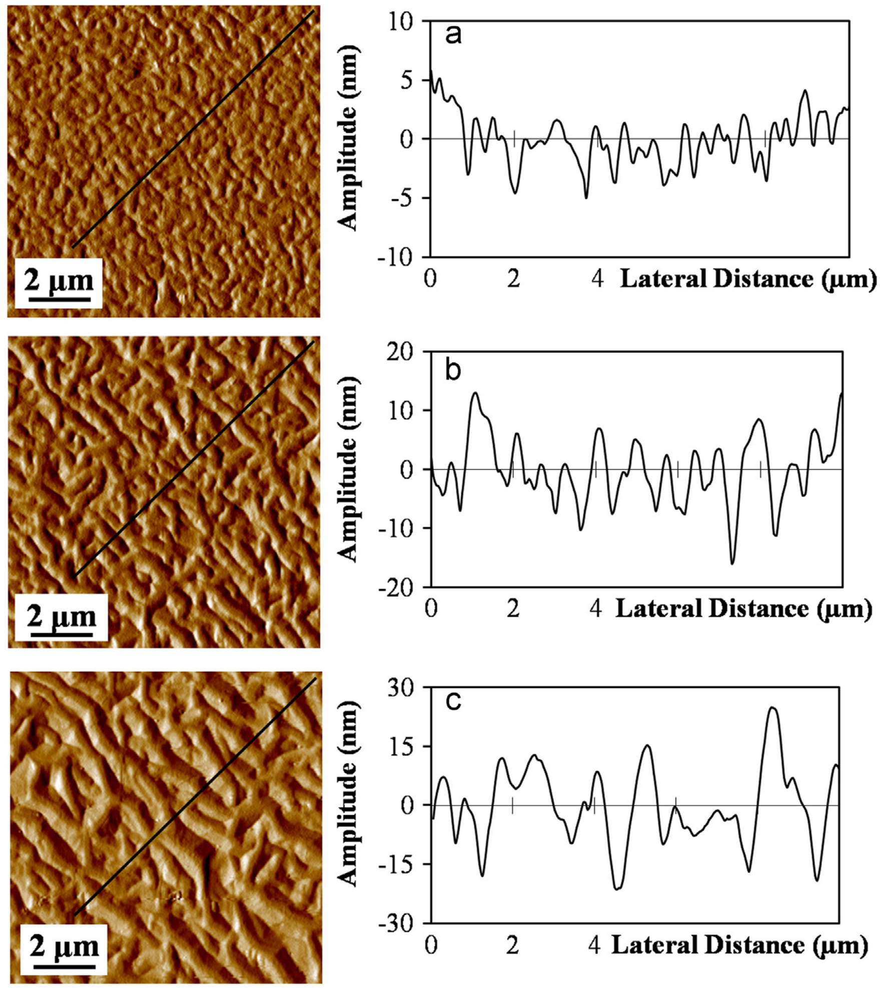

Fig. 1. Effect of film thickness t on surface undulation. (a) $t=500$ nm, (b) $t=2$ μm and (c) $t=5$ μm.

which stabilizes the surface. This critical wavelength is not thickness-dependent; but the undulation is caused by the interplay of surface energy and residual stress in the material.

Involving the effect of surface tension, Jonsdottir and Freund [11,12] calculated the surface undulation resulted from the gradient of chemical potential of the material along the surface. They developed a numerical model to estimate the magnitude and profile of surface undulation and concluded that the undulation wavelength corresponds to the distance between misfit dislocations and the amplitude depends on the mismatch strain. Spencer at al. [13] performed an analytical analysis of morphological instability of a growing epitaxially strained dislocation-free solid films. They derived an equation for surface morphology based on a stress-dependent chemical potential, by assuming that the film-substrate bonding is perfect and that a stress forms merely due to misfit strain. They then concluded that the instability wavelength is long compared to the film thickness, and that the critical thickness of a growing film is determined by the kinetic stabilization. Although they theoretically achieved a model for calculating critical wavelength, they did not discuss the thickness-dependent critical wavelength. Gao [14] studied the formation of misfit dislocations due to surface evolution in the film-substrate systems. He concluded that morphological instability which causes undulation on the film surface may eventually result in defects such as misfit dislocations and the stress mitigation in the film. Kim and Vlassak [15] calculated the critical wavelength for multi-layer structures which again arises as a result of the competition between surface and strain energy. Yang and Srolovitz [16] performed a numerical study of stress-assisted surface morphology evolution. They investigated the formation of crack-like morphology in the film by calculating chemical potential along the surface. Although there have been many other investigations [17–19] try to understand the details on the strain energy density and chemical potential in a hetero-epitaxial structure, the cause of thickness-dependence surface undulation has not been addressed.

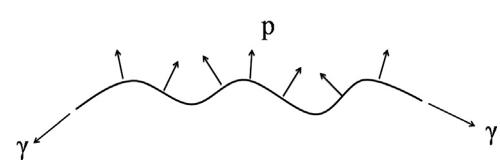

Fig. 2. A schematic of the equivalent surface traction due to surface tension.

The aim of this study is to resolve this phenomenon by considering the effects of both surface energy and residual stresses.

2. Modelling

Suppose that γ is associated with the unit area of a free surface, which leads to the tendency of reducing the free energy of the system via reducing the surface area. This amounts to the surface normal traction p associated with the surface curvature, κ, i.e.,

$$p=2\gamma\kappa \tag{4}$$