Surface Science 602 (2008) 3560–3568

Contents lists available at ScienceDirect

Surface Science

journal homepage: www.elsevier.com/locate/susc

On the existence of a critical perturbation amplitude for the Stranski-Krastanov transition

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ARTICLE INFO

Article history:
Received 17 June 2008
Accepted for publication 18 September 2008
Available online 7 October 2008

Keywords:
Surface roughening
Surface diffusion
Epitaxy
Anisotropy
Surface energy
Monte Carlo simulations
Critical amplitude

ABSTRACT

The stability of an elastically strained thin film is investigated within the context of small and large perturbation theories for the case of an anisotropic surface energy which is a function of the film thickness and the surface orientation. Under typical growth conditions (below the roughening transition temperature) the surface energy function has a sharp (cusped) minima at low energy orientations, i.e. the derivative of the surface energy with respect to orientation is discontinuous. This sharp cusp in the surface energy function is treated explicity here without the normal smoothing assumptions. It is found that the smoothed approximation is unphysical in the sharp cusp limit as it predicts that the film is always stable. It is shown here that full treatment of the cusp disagrees with this finding and predicts that minimum energy surfaces are in fact unstable for perturbations above a critical size. A simple linear model for this critical perturbation size is proposed. Off-lattice kinetic Monte Carlo (kMC) simulations are conducted to test these predictions and good agreement is found. It is demonstrated that roughening in highly strained heteroepitaxial systems is possibly beyond the scope of linear perturbation theory, depending on the exact nature of the surface energy function. A non-linear theory for large perturbations is proposed to this effect. It is also found that the wetting layer in InAs/GaAs(001) and Ge/Si(001) is effectively stable whilst the surface energy of the wetting layer varies with its thickness.

1. Introduction

Heteroepitaxial semiconductor thin films are used in the fabrication of microelectronic and optoelectronic devices. The elastic strain energy inherent in the film due to the lattice mismatch between it and the substrate causes the film to become unstable as the film gets thicker. This can cause the film to break up and grow as a number of discrete entities referred to as islands or quantum dots. This Stranski-Krastanov transition is of great interest as it offers the possibility of manufacturing practical self-organised nanostructures [1]. Technological application of this process requires control of this strain-induced morphological change. The roughening of a strained surface has been widely investigated using the Asaro–Tiller–Grinfeld (ATG) instability theory [2–4]. This shows that the initially flat surface of a strained film is always unstable for an isotropic (constant) surface energy. Reduction in the elastic stored energy of the thin film drives the roughening towards the shortest possible wavelength. The surface energy, meanwhile, wishes to flatten the perturbed surface and drive the system to the longest possible wavelength. This competition between the elastic stored energy and surface energy drives the system towards a particular roughening wavelength. This is critical for the development of self-organised structures with a narrow-size distribution.

Heteroepitaxial growth typically occurs below the roughening transition temperature. This means that the anisotropic nature of the surface is very important in determining the stability of the system [6,7]. In this paper, the effect of a surface energy which is a function of the film thickness and surface orientation is considered. It is widely known that the surface energy of a crystal is highly dependent on the surface orientation [8]. Below the roughening temperature, it is commonly found that surfaces facet at particular orientations which are local minima in the surface energy. At these orientations there is a discontinuity in the derivative of the surface energy. This is referred to as a (functional) cusp. An example of such a cusped function can be seen in Fig. 1, where the cusp is at a surface orientation of $\theta = 0$. ATG theory predicts that the cusp makes such films unconditionally stable against small perturbations [7]. However, Eisenberg and Kandel [7] found in their numerical simulations of strained thin films that such films were unstable to perturbations of a critical size. The origin of this critical perturbation amplitude is considered further in this paper and, for the first time, a simple analytical expression for its magnitude is derived.

For very thin films less than 4–5 monolayers in thickness, it has been shown that the surface energy is also a function of the film thickness [9]. This fact has been used to explain experimental

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doi:10.1016/j.susc.2008.09.036

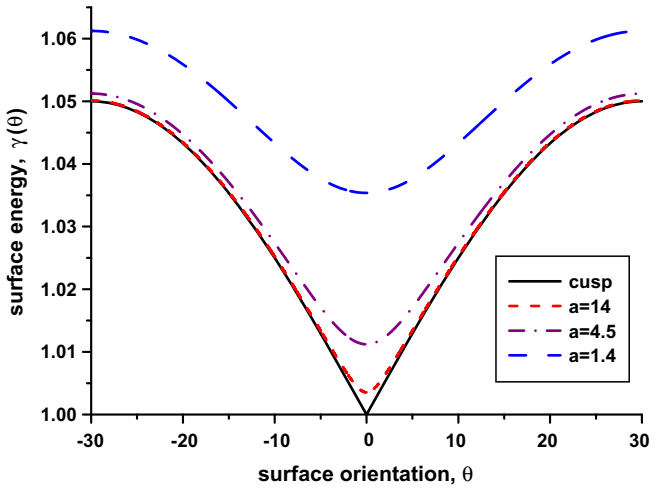

Fig. 1. The surface orientation-dependence of surface energy functions (25) and (26). Visually it appears that the smooth energy function (26) tends towards the cusped function (24) as the rounded cusp radius a get large. However mathematically this is not the case in the limit of small perturbation theory and the two functions give very different predictions for the stability of a surface in the limit $a \to \infty$.

observations of a critical wetting layer thickness, below which the wetting layer is stable [10]. Recent ab-initio calculations have determined this thickness dependence for a number of important systems [11]. The effect of the critical wetting layer thickness is re-evaluated here in the context of this critical perturbation amplitude theory.

A general variational functional is proposed in Section 2 for the analysis of elastically strained anisotropic surfaces in which the surface energy is a cusped function of the surface orientation and a smooth function of the film thickness. A variational method is used to formulate a novel small perturbation continuum theory for the stability of such surfaces in Section 3. Off-lattice kinetic Monte Carlo (kMC) simulations are performed to determine the effect of strain on the roughening wavelength in strained films. These results are then compared with the predictions of the linear continuum theory. This theory is then extended to consider nonlinear large perturbations in Section 4.

2. The variational functional

Consider a two-dimensional thin film on an infinitely thick substrate with similar elastic properties, as shown in Fig. 2. We take the film-substrate interface to coincide with the x-axis and let the thickness of the film be defined by $h(x,t)$. The problem now reduces to predicting how h evolves over time due to diffusion of atoms over the free surface. The mismatch strain between the film and substrate is taken to be ε_0 and plane strain conditions are assumed.

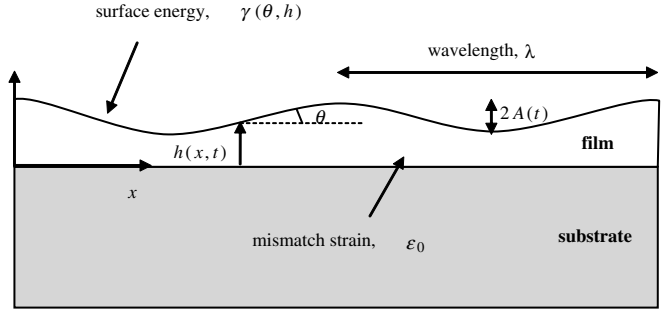

Fig. 2. The lattice mismatch between the thin film and the substrate is ε_0. The height of the surface of the film, $h(x,t)$, is assumed to be sinusoidal with wavelength, λ, and peak-to-peak amplitude, $2A(t)$. The surface energy, $\gamma(\theta, h)$, is assumed to be an anisotropic function of the surface height and orientation, θ.

The details of the problem are now elaborated within the context of a variational framework [12]. This has the advantage that the exact evolutionary morphology of the system does not need to be known. A class of morphologies is proposed. If the system evolves within this class of morphologies then the exact solution to the differential equations is obtained. If it is not, then the best approximation to the exact result within the constraints of this class is obtained. We propose here that the surface morphology evolves as a series of sinusoids. For a surface energy which is a smooth function of orientation, $\gamma(\theta)$, ATG theory shows that this is the exact morphology class for small amplitude perturbations.

In general, we formulate a variational functional

$$\Pi = \Psi + \dot{G} \tag{1}$$

such that the actual kinematic field describing the evolution is the minimal one, i.e. $\delta\Pi = 0$. The kinetics of the surface diffusion process are encapsulated within the dissipation potential, Ψ. The rate of change of Gibbs free energy, $\dot{G}$, provides the driving force for evolution of the system. This analysis is for a two-dimensional system but it is readily extended to three-dimensions [13]. Assuming the surface diffusivity is isotropic [13], the dissipation potential can be written as

$$\Psi = \frac{1}{2}\int_{A_s} \frac{j_s^2}{D_s}\mathrm{d}x \tag{2}$$

where j_s is the volumetric surface flux, A_s is the (one-dimensional) area of the free surface of the film, and D_s is the surface diffusivity. The surface flux is related to the normal velocity of the surface v_n by

$$v_n + \frac{\partial j_s}{\partial s} = 0 \tag{3}$$

where s is a surface ordinate.

The Gibbs free energy has two contributions

$$G = \int_{A_s} \gamma(\theta, h)\mathrm{d}x + \int_V w\mathrm{d}V. \tag{4}$$

The first term is the total surface energy in which the anisotropic surface energy density γ is a function of the surface orientation, θ, and the film thickness, h. The second term is the total elastic stored energy of the system, where $w(x, y)$ is the local elastic strain energy density and V is the (two-dimensional) volume of the film-substrate assembly. Consequently one can write [10]

$$\dot{G} = \int_{A_s} \left[(\gamma(\theta, h)\kappa + w)v_n + \frac{\partial \gamma}{\partial \theta}\dot{\theta} + \frac{\partial \gamma}{\partial h}\dot{h}\right]\mathrm{d}A \tag{5}$$

where κ is the local curvature of the film surface and the dot denotes differentiation with respect to time. The variational functional (1) is now completely defined for a particular class of surface height profiles, $h(x, t)$. In the next section, this is used to determine the stability of this system to small perturbations.

3. Linear stability analysis

To obtain a linear solution to this problem we restrict the spatial gradients in the surface profile to be small so that the amplitude of the perturbation is assumed to be much less than its wavelength. In this case, the mass conservation relation (3) is

$$v_n \approx \frac{\mathrm{d}h}{\mathrm{d}t} = -\frac{\partial j_s}{\partial x} \tag{6}$$