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Study of Stranski–Krastanov growth using kinetic Monte Carlo simulations with an atomistic model of elasticity

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ABSTRACT

We have analyzed the Stranski–Krastanov growth mode in heteroepitaxial thin films using lattice-based kinetic Monte Carlo simulations with an atomistic model of elasticity. In this growth mode, elastic effects due to the lattice mismatch between the film and the substrate cause a transition from two-dimensional layer-by-layer growth to three-dimensional island growth. In our simulations on a simple cubic lattice model in a 3-dimensional system with nearest neighbor interactions only, we see very little tendency towards islanding. On modifying the anisotropy using next-to-nearest and next-to-next-to-nearest neighbor interactions, the system shows a greater tendency towards islanding. When the calculations are carried out in a 2-dimensional system, islanding is fairly pronounced. To gain insights into the possible reasons for these observations, we evaluate the elastic energy and bond energy of different configurations in a 2-dimensional system. Our calculations show that island growth in 2-dimensions also involves a significant nucleation barrier. This suggests that the barrier for island formation is more easily overcome in a 2-dimensional system as compared to a 3-dimensional system.

1. Introduction

Stranski–Krastanov (SK) is the mode of growth for certain strained heteroepitaxial thin films like Ge/Si [1–4] and InAs/GaAs [5–12]. In such films, the first few layers grow flat over the substrate; and the subsequent layers rearrange to form 3-dimensional mounds. This transition is referred to as the 2D/3D growth transition or the Stranski-Krastanov transition. These defect-free mounds are referred to as quantum dots (QDs) and have potential applications in optoelectronic devices. In particular, it is found that the photovoltaic cell efficiency is highly increased in devices made with QDs of InGaAs/GaAs [13,14]. The flat layers below the mound are referred to as the wetting layer (WL). The thickness of the WL is found to be about 3 ML for Ge on Si(100) [1,2,15,16–18] and about 1 ML for InAs on GaAs [9–11,19].

The driving force for SK growth is the strain in the system due to the lattice mismatch between the film and the substrate. The total strain energy increases as more layers of the film are deposited. When this strain energy becomes very large, the film becomes unstable towards island formation. Since the mismatch for the InAs/GaAs system (7%) is greater than Ge/Si (4%), the strain energy is greater and thus the 3D mounds to appear at lower coverage. However, it is the larger islands that are able to relax strain more efficiently, so they are more stable than the smaller islands. Thus, SK growth involves nucleation of a critical island and is believed to go through a nucleation barrier [20]. The SK growth transition is different from the longer wavelength Asaro-Tiller-Grinfeld (ATG) instability of a planar stressed film [21,22], though both have their origins in strain. For lower mismatched systems ($\in < 2\%$), a nucleationless route to SK growth is proposed via surface undulations [23] caused by the ATG instability.

From a thermodynamic perspective, SK growth is explained by the variation of the surface chemical potential μ_S with thickness n of the film, measured in number of layers [24]. If $d\mu_S/dn > 0$, the film wets the surface; otherwise it dewets it and forms mounds. In SK growth, the derivative $d\mu_S/dn$ changes sign from positive to negative as the number of layers of the film are increased above some critical value. μ_S has contributions from the misfit strain energy, the surface energy and the wetting layer interactions. In continuum models, each of these terms is modeled independently. The surface energy is modeled using either surface curvature [25–27] and surface energy anisotropy [28,29] or step formation and interaction energies [30,31]. The wetting layer interactions are attributed to the chemical interaction between the film and substrate atoms and are modeled using empirical wetting potentials. The elastic energy is calculated by solving the equilibrium equations of elasticity in the film and the substrate with appropriate boundary conditions. An alternate method involves the use of either partially or fully atomistic models to calculate the surface chemical potential. It was shown that an appropriately tuned Keating model for Ge on Si(100) showed wetting layer of thickness up to 3 ML is stable [32]. A

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similar conclusion was arrived at using quantum calculations performed with density functional theory [33] and molecular dynamics simulations [34].

From a kinetic perspective, SK growth is studied by modeling the time evolution of a growing film. In continuum theories, this is done using the local surface chemical potential under a linear kinetic framework [31,35–37], with an additional term for the deposition flux. This evolution equation involves the derivative of the surface height function and is typically a nonlinear partial differential equation. In the usual spirit of nonlinear differential equations, the evolution equation has been analyzed for both linear [26] and nonlinear stability [27]. Additionally, numerical simulations of the height evolution have been carried out by using techniques involving Fourier or Hilbert transforms [27,29,38].

The wetting layer and islands are not completely homogeneous. There is some amount of Si present in the Ge islands and some amount of Ge enters into the substrate material. This intermixing has consequences on the growth and stability of the islands [39,40]. Continuum models with intermixing have also been implemented to study the effect of compositional inhomogeneity in these islands. In [41], the effect of composition on the evolution equations is a modification of local values of some kinetic parameters. An alternative prescription uses a composition field coupled to the strain field together in the variational approach to calculate the composition maps of island of different shapes [42].

Recently, the formation and stability of the wetting layer were investigated using kMC simulations with an atomistic model of elasticity [43]. These calculations were performed using a model 2D system, where growth was in the Z-direction and the growing surface was a line in the X-direction. This work proposed three mechanisms involved in SK growth - the kinetic notion of an apparent critical thickness, a stable wetting layer and an entropic effect. In all these mechanisms, intermixing plays an important role. Using these mechanisms, the authors are able to explain a number of experiments at different temperatures. There have been a few calculations where SK growth has been studied in 3D systems where the growing surface is a 2-dimensional area in the XY plane [44–46], however, they lack the detailed analysis of Ref. [43].

Surface energy anisotropy is believed to play an important role in the SK growth transition [47] and the shapes of the islands [59]. In particular, for the Si–Ge system, the quantum dots are formed with facets in the 105 direction. This anisotropy has been simulated in kMC simulations either using next-to-nearest and next-to-next-to-nearest interactions [46], or using an explicit facet stabilization term in the bond energy [48]. These simulations involve a combination of surface anisotropy and elasticity and our interest is to study these effects independently.

In this work, we first carry out calculations on 3D(2 + 1D) systems with varying contributions from elasticity and surface energy anisotropy. Our calculations indicate that a 3D system with only nearest neighbor interactions does not show SK transition at 3 ML as observed for the 2D system with the appropriately scaled parameters. However, similar systems with interactions involving next-to-nearest neighbors and next-to-next-to-nearest neighbors shows SK growth more readily, as seen in earlier work [44,46]. To understand the origin of this difference, we carry out a detailed energetic analysis of the 2D system to see whether the islands can become more stable than flat layers. Our calculations indicate that island formation in the 2D system is observed even though it involves a significant nucleation barrier.

The rest of this article is organized as follows. In Section 2, we describe the SOS model and the elastic model used in our calculations. Section 3 contains the results of elastic energy calculations for some configurations in a 3D system. In Sections 4 and 5, we show the results for kMC simulations of multilayer growth for a 3D system and the 2D system respectively. In Section 6, we carry out an energetic analysis of the 2D system and conclude in Section 7.

2. Model description

We simulate heteroepitaxial systems using a multicomponent simple cubic solid-on-solid model wherein each site can be occupied by one of the two species corresponding to the film and the substrate [49]. In a 3D(2D) model system, the sites belong to a 3D(2D) simple cubic lattice. The usual no-overhang condition is implemented in the direction of crystal growth, i.e. the positive Z-axis. Periodic boundary conditions are implemented in the X and Y directions (for 3D systems) or the X direction (for 2D systems). Elastic effects are modeled using a ball and spring model. Such models have been widely used in literature [50–55] and offer an advantage over continuum elastic models in kMC for shorter length scales, but are computationally much more expensive. In this model, all the atoms on the lattice sites of the SOS model are allowed small local displacements from their positions on a reference lattice. The reference lattice is chosen to be identical to the substrate lattice in the X and Y directions, consistent with epitaxial growth. In the Z direction, the reference lattice is chosen to be identical to the substrate for layers below the film-substrate interface, and equal to the lattice formed by a completed flat film for layers above the interface. This choice of the reference lattice ensures that the displacement of atoms is small for most of the calculations performed, but, does not affect the results in any way. All atoms are connected by harmonic springs between their nearest neighbors and their next to nearest neighbors. The spring constants depend on the nature of atoms forming the bonds, and are chosen to reproduce the bulk elastic moduli. The equilibrium lengths of the spring between two film (substrate) atoms are chosen to be equal to the lattice constant of the film (substrate). For bonds involving one film and one substrate atom, the choice of spring constants and equilibrium lengths is described in Section 3.

To calculate the elastic energy of a given lattice configuration, we minimize the total elastic energy with respect to atomic displacements. The atoms of the film and the top few layers of the substrate are treated numerically, whereas the remaining semi-infinite substrate is treated analytically using Fourier transforms and linearized elasticity theory [49,55]. The atomic displacements obtained as a result of this minimization are used to calculate the elastic energy contribution to the barrier energy.

This model is simulated using a typical kinetic Monte Carlo (kMC) scheme consisting of a succession of deposition and diffusion moves consistent with experimental conditions. Surface diffusion is modeled by hopping to nearest neighbor sites on the lattice at a hopping rate given by

$$r = \nu_0 exp\left(-\frac{E_b}{k_B T}\right)$$

where E_b is the barrier energy for this process, ν_0 is the vibration frequency which is taken as a constant, k_B is the Boltzmann constant and T is the temperature. The barrier energy is the total energy required to detach the atom, and is written as a sum of the energy of the bonds and the elastic energy. Thus the calculation requires frequent calculation of the elastic energy, which is an expensive calculation. To speed up these calculations, we approximate the elastic energy contribution to the barrier energy for adatoms in a given layer to be a constant, independent of the configuration of other atoms in the layer. Further, the elastic energy of surface atoms (which are not adatoms) is approximated using the local displacements around the atom. This approximation actually underestimates the elastic contribution and numerical correction factors of about 1.33–1.5 in 3D [44] and about 2.4–3.5 in 2D [57] have been suggested to improve this accuracy. However, these values are based on the spring constants and the other parameters used. In this work, we ignore these factors and focus instead on the effects of varying elastic energy and surface energy anisotropy. For simplicity,

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