Thin film models for quantum dot structures in solid films

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Background and Introduction

Epitaxial growth of self-organizing nanostructures ("quantum dots") from a thin solid film demonstrates fascinating patterning, and the geometrical structure of the growing solid film has intrinsic interest [1]. However these nanocrystals can also be very valuable for industrial applications, because their semiconducting properties make them useful for electronic and optoelectronic devices, such as blue laser diodes [2]. The photovoltaic industry hopes to incorporate the structures—called "artificial atoms" because of their discrete energy states—inside new thin layers in order to increase thermodynamic conversion efficiency [3]. For widespread applications, comparatively cheap, but controllable self-assembly growth processes are desired.

In a typical process, atoms of germanium are deposited on top of a silicon substrate in a hot chamber, and after a pseudomorphic growth phase the Asaro-Tiller-Grinfeld (ATG) instability leads to formation of small, initially round, structures ("pre-pyramids") [4]. These later transform to pyramids, the quantum dots. Further deposition can lead to multi-faceted domes and eventually introduce dislocations that cause the quality of the quantum dots to deteriorate.

A main mechanism for the growth is the misfit strain arising from lattice mismatch. The film and substrate are crystalline materials with differing lattice constants. From a bare substrate, a film initially grows uniformly ("layer by layer growth"), its lattice straining to that of the substrate, inducing elastic energy that competes with surface energy. As the film thickness increases, strain energy is released by the formation of quantum dots, while wetting effects ensure film material covers the entire substrate. This is the Stranski-Krastanov growth mode. During growth both materials elastically deform and the observed dots initially form rounded pre-pyramids. Anisotropy is responsible for pyramidal shapes or multi-faceted domes that are visible at later times. We consider only the anisotropy of the surface energy, though in principle anisotropic elastic properties may also be significant.

Stochastic methods that act on atomic scales are limited to small domains with few dots. To treat larger

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domains for longer times, we use a continuum model based on the concept of surface diffusion [5]. Spencer and co-workers derived similar partial differential equations without wetting or anisotropy e.g. [6]; later work has included these to differing extents [7, 8, 9, 10]. However, for simulations of reduced models the important effects of stress and wetting were neglected. Finite element methods allow for the solution of the Navier-Cauchy equations in the elastic substrate in three dimensions but are computationally intensive. To avoid the high computational costs, a small slope approximation similar to lubrication theory for liquid films is applied.

Model overview

We consider the dislocation-free evolution of a thin solid film on a substrate as sketched in Fig. 1. The film-substrate interface is at $z=0$, and the film surface is at $z=h(x,y,t)$ at time t and location (x, y) in the periodic spatial domain. In the absence of deposition the film evolution satisfies

$$h_t = D \sqrt{(1 + |\text{grad } h|^2)}\ \text{laplacian}(\mu)$$

with the diffusion constant $D = \Omega^2 D_S \sigma/(k T)$ (Mullins [M57]) . Here $\mu(x, y, t)$ is the chemical potential at the free surface of the film, Ω is the atomic volume, σ the surface density of atoms, D_S the diffusion coefficient, k the Boltzmann constant and T the absolute temperature. Following Tekalign and Spencer [10] we consider a chemical potential μ that consists of two terms

$$\mu = E_{\text{sed}} + E_{\text{surf}}$$

representing the competing contributions from elastic and surface energies, respectively. The first term is the strain energy density evaluated at the surface $E_{\text{sed}} = \Sigma_{ij=1,2,3}\ [(1/2)\ \sigma_{ij}\ \varepsilon_{ij}]_{z=h}$ with the stresses σ_{ij} and strains ε_{ij} related by Hooke's law. A standard solution for the elastic response of a semi-infinite solid subjected to a point force allows computation of E_{sed}. The surface free energy $\gamma(h, \text{grad } h)$ is assumed to depend on orientation in addition to film thickness. The total surface energy E_{surf} is found by taking the functional derivative

$$E_{\text{surf}} = (\delta/\delta h) \int \gamma(h, h_x, h_y)\ dS = -\gamma\kappa + E_{\text{wet}} + E_{\text{anis}}\ .$$

where κ is the mean free surface curvature. It can be decomposed into three components including surface energy, wetting potential, and an additional term due to the anisotropy of γ. After model reduction assuming small slopes, the resulting evolution equation for the surface profile h is quasilinear and of fourth order, with a non-local contribution due to E_{sed}.

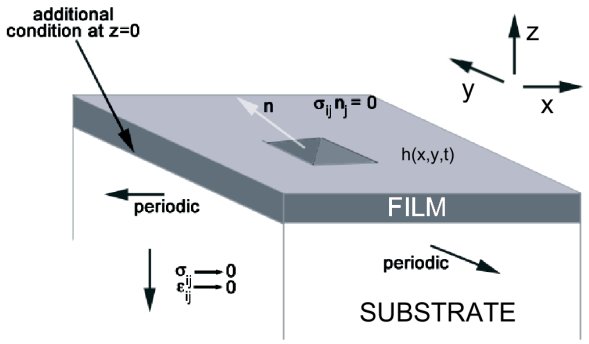

Fig. 1: A quantum dot forming on a substrate and the coordinate system used.

Results

Linear stability analysis shows that a flat film of thickness H is unstable over a range of finite wavelengths, when H exceeds a critical thickness. Furthermore, anisotropy acts to destabilize the surface. It lowers the critical height at which flat films become unstable due to wetting interactions, and there exists an anisotropy strength G above which all thicknesses are unstable (Fig. 2). A spectral numerical method is used to perform simulations in two and three space dimensions. In a typical simulation a film which is nearly flat evolves to form small "islands" or pyramids separated by a thin wetting layer; at longer times the smaller of these loses material to the larger via the wetting layer, resulting in coalescence as a single pyramid (Fig. 3). When anisotropy is included our simulations clearly show faceting of the growing islands (Fig. 4) and a power law coarsening behavior [11].

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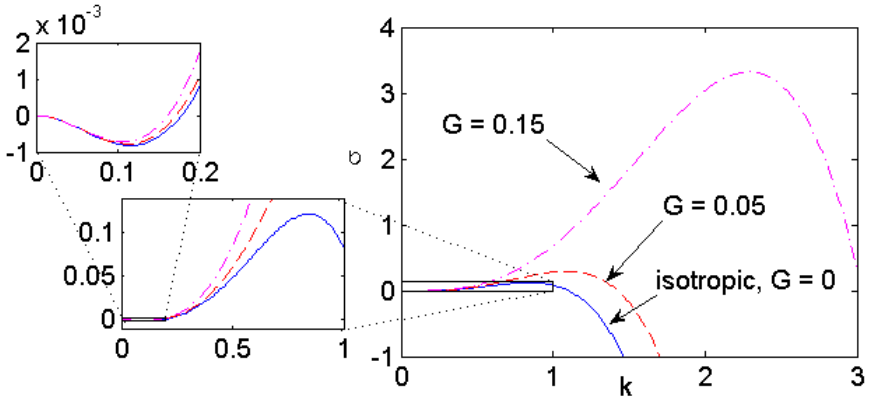

Fig. 2: Dispersion relation for the isotropic (G=0) and the anisotropic cases G=0.05 and G=0.15 with H = 0.8, where $k = 2\pi/\lambda$ is the wavenumber. Close-ups show the wavenumber regime near the minimum growth rate [11].

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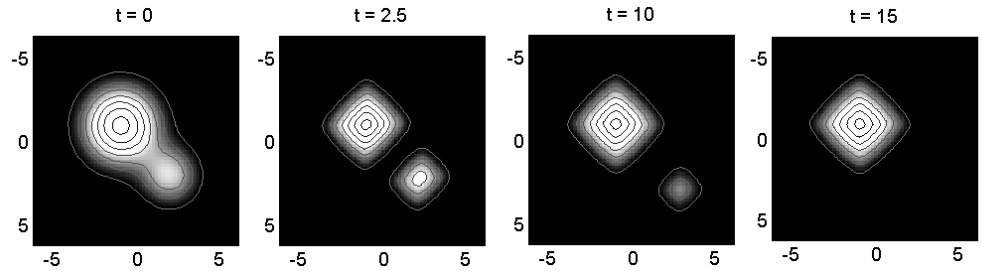

Fig. 3: Collapse of a Ge/Si quantum dot. Lighter shades correspond to larger film thicknesses. The initial condition for these two islands was two rounded Gaussian humps. These first evolve into faceted structures before the bigger dot "eats" the smaller one and survives. Here G=0.2; time and space are in dimensionless units [11].

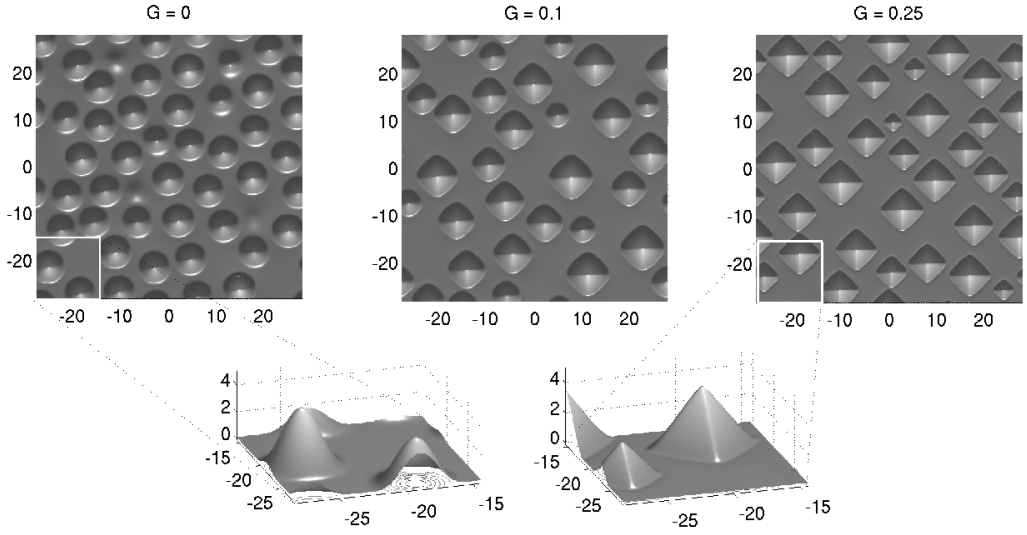

Fig. 4: Anisotropy effect on the Ge/Si system. Each picture shows the film at the same time after evolution from the same initial condition (random perturbation of a flat state). The upper left shows a simulation with isotropic surface tension (G=0) and the dots have round tips. In the other two upper pictures (G=0.1 and G=0.25) we observe faceting which is stronger for the bigger anisotropy coefficient G. Lower pictures are perspective views of subregions of the domain, showing the transition from bell shapes (G=0) to pyramids (G=0.25) [11].