Stochastic Continuum Modeling Self-Assembled Epitaxial Quantum Dot Formation

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ABSTRACT

Semiconductor epitaxial self-assembled quantum dots (SAQDs) have potential for electronic and optoelectronic applications such as high density logic, quantum computing architectures, laser diodes, and other optoelectronic devices. SAQDs form during heteroepitaxy of lattice-mismatched films where surface diffusion is driven by an interplay of strain energy and surface energy. Common systems are Ge_xSi_{1-x}/Si and $In_xGa_{1-x}As/GaAs$. SAQDs are typically grown on a (001) crystal surface. Self-assembled nanostructures form due to both random and deterministic effects. As a consequence, order and controllability of SAQD formation is a technological challenge. Theoretical and numerical models of SAQD formation can contribute both fundamental understanding and become quantitative design tools for improved SAQD fabrication if they can accurately capture the competition between deterministic and random effects. In this research, a stochastic model of SAQD formation is presented. This model adapts previous surface diffusion models to include thermal fluctuations in surface diffusion, randomness in material deposition and the effects of anisotropic elasticity, anisotropic surface energy and anisotropic diffusion, all of which are needed to model average SAQD morphology and order. This model is applied to Ge/Si SAQDs which are group IV semiconductor dots and InAs/GaAs SAQDs which are III-V semiconductor dots.

1. INTRODUCTION

Heteroepitaxial self-assembled quantum dots (SAQDs) represent an important step in the advancement of semiconductor fabrication at the nanoscale that will allow breakthroughs in optoelectronics and electronics.^{1–11} SAQDs are the result of a transition from 2D growth to 3D growth in strained epitaxial films such as Si_xGe_{1-x}/Si and $In_xGa_{1-x}As/GaAs$. This process is known as Stranski-Krastanow growth.^{3,12–14} If SAQD are to compete with traditional lithography, order and control of SAQDs must be better understood. This understanding can be enhanced and verified through the use of quantitative models.^{12,15–20} At the same time, these models become the design tools for further development. To adequately address control and reproducibility issues, models should include physically based random effects and assess how randomness competes and interacts with deterministic effects to form useful nanostructures. To this end, a continuum stochastic model of SAQD formation is presented and applied to Ge/Si SAQDs and InAs/GaAs SAQDs. Preliminary results are reported.

SAQDs are formed by the deposition of lattice mismatched films such as Ge on Si or InAs on GaAs. When these films reach a critical height, H_c , they become unstable. For example, see Fig. 2. In early work, it was proposed that the initial stages of SAQD formation strongly influence final SAQD formation. In particular, a linear instability theory know as the Asaro-Tiller-Grinfeld (ATG) instability^{21,22} was applied to Ge/Si SAQDs.^{15,23} Although initially these studies included many approximations, they were very fruitful. By finding the growth-rate of each Fourier component (σ_k) or "dispersion relation", the characteristic length and time scales were determined. Later, it was found that elastic anisotropy affects the alignment of Ge/Si SAQDs using both the linear initial formation theory^{24,25} and finite element simulations.¹⁷ Further work found that intermixing and segregation of Ge and Si can be very important.^{26,27} In much of this work, it is held that the initial stages initial undulations in the film surface ultimately grow and mature into an array of SAQDs. Then, depending on the stability of the array, equilibrium is achieved or dynamic ripening takes place. This view appears to be born out in finite element based simulations,^{16,17} and has been formalized using the multiscale-multitime expansion.¹⁸

The ATG-instability picture suggests an interesting way to understand and model SAQD order. Since dots form initially as cross-hatched ripples, it stands to reason that the initial order of ripples should ultimately influence the final SAQD order.²⁸ The linear theory of initial ripple order was worked out for the case of a fast deposit and then anneal considering

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Figure 1. Linear stochastic calculation of fast deposit and anneal of InAs/GaAs after 2.10 time units of annealing. t is in units of characteristic time t_0 ; average film height (\bar{h}) and film height r.m.s. fluctuation (h_{rms}) in nm.

both random initial roughness and the influence of thermal fluctuations in material transport.^{19,20} It was found that the influence of thermal fluctuation was quite strong.

Modeling the influence of random effects such as thermal fluctuations and random deposition on final SAQD morphology and arrangements suggests a more general picture of control and reproducibility of SAQD manufacture. The deterministic dynamics of SAQD formation act as a complicated filter/amplifier of broadband noise. In fact, in the absence of any randomness, SAQDs do not form, and the initially planar films remain planar in an unstable equilibrium. The quality of the filter/amplifier ultimately determines the order of SAQD arrays. The challenge of producing more regular SAQDs is then the challenge of manipulating the interplay of random and deterministic effects. To this end, a stochastic simulation of SAQD formation has been developed that incorporates randomness in material deposition and thermal fluctuations in surface diffusion. Unfortunately, many of the material parameters entering the model are merely guesses, but having a complete simulation with a physical model of random effects will allow more quantitative comparisons with experimental observations and ultimately allow these unknown parameters to be surmised.

Although cumbersome, a fully stochastic model has the additional benefit of allowing all deposition processes to be simulated. Using random initial conditions as a surrogate for ongoing time-dependent fluctuations creates two problems. In the absence of a physical model of the source of randomness, random initial conditions could be anything. Often, in discrete simulations, nodes are moved randomly up or down some small amount, *e.g.* 1Å, but this procedure creates mesh-dependent outcomes; furthermore, the fluctuation amplitudes are arbitrary and non-physical. Second, random initial conditions confound the simulation of the most common experiment, deposition of material at a constant rate. Below the critical film height for SAQD formation, random initial conditions will eventually decay. If deposition is not fast enough, initial roughness can decay to zero before the critical transition film height is reached. Thus, without a fully stochastic model, one is limited to fast deposition simulations, or starting at just above the critical film height

Modeling Ge/Si SAQDs via continuum methods has a long history dating back to 1988.¹⁵ However, this type of modeling has not been applied to InAs/GaAs SAQDs. This imbalance is in part due to the more complicated nature of InAs SAQD formation. In particular, the role of crystal surface anisotropy is more complicated for InAs/GaAs SAQD formation. While elastic anisotropy alone appears to explain spatial patterns of Ge/Si dots. It has recently been worked out via a linear theory that elastic, surface energy and diffusional anisotropy all interact to determine the initial SAQD spatial patterning.²⁹ For example, using the parameters from Secs. 2 for InAs/GaAs, one would predict an initial power spectrum and sample initial configuration similar to Fig. 1. The current work builds on this initial linear theory.

Finally, it should be noted that there are competing theories of SAQD formation. The most well-known is the nucleation and growth model.³⁰ The ATG-based model seems to be a better candidate than nucleation and growth when dense SAQD arrays are formed as occurs for Ge/Si and high temperature InAs/GaAs growth. The second alternative model is the negative surface stiffness model due to surface strain effects on step edges;³¹ however, this model ignores finite temperature and configurational entropy effects³² and might therefore be based on the false supposition that the surface free energy density has a negative stiffness. Further investigation is needed. It is possible that these two models and the ATG-based model all have their place, but that is beyond the scope of the present discussion.

The rest of this proceedings article is organized as follows. Section 2 presents an overview of the physical model of SAQD formation along with numerical implementation. Section 3 presents the simulation results. Section 3 presents a discussion and conclusions.

2. MODEL

Like all computational models, there is both a physical - mathematical statement of the model and then a numerical implementation. The spectral or Fourier method is used to solve the governing equations for a number of reasons: it facilitates the computation of elastic strain energy density, the dynamics of individual Fourier components decouple to linear order, the spectral method is numerically efficient, and the discretized random fluctuations are well understood and simple to simulate using Fourier components. The Fourier components are defined in the usual way for a square periodic simulation system, so that for a function of \mathbf{x} , $f_{\mathbf{k}} = A^{-1} \int d^2 \mathbf{x} f(\mathbf{x}) e^{-i\mathbf{k}\cdot\mathbf{x}}$, and $f(\mathbf{x}) = \sum_{\mathbf{k}} f_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{x}}$, where $\mathbf{k} = (2\pi m/L)\mathbf{i} + (2\pi n/L)\mathbf{j}$ with $m, n = 0, \pm 1, \pm 2, \ldots$, and $A = L^2$ is the area. Since $f(\mathbf{x})$ is in all instances a real function, $f_{-\mathbf{k}} = f_{\mathbf{k}}^*$. Here and below, bold face indicates vectors, and bold face with a ($\hat{\mathbf{j}}$) indicates a rank-2 tensor. Discrete approximations are discussed in Sec. 2.2.1

2.1 Physical Model

The proposed model is built up from existing deterministic models using the celebrated fluctuation dissipation theorem and by assuming a non-equilibrium random deposition process. A nominally uniform thin film is deposited on an (001) surface and then becomes unstable transitioning from 2D planar growth to a 3D rippled configuration and finally to discrete SAQDS with a residual wetting layer. Film height $h(\mathbf{x})$ is treated as a dependent variable with implicit time dependence, and \mathbf{x} is the vector position in the x - y plane parallel to the flat substrate surface (Fig. 2). x increases in the [100] direction, y in the [010] direction. Surface diffusion is driven by gradients in a local diffusion potential $\mu(\mathbf{x})$ that is equal to the variational derivative of the total free energy $\mu(\mathbf{x}) = \delta \mathscr{F} / \delta h(\mathbf{x})$ so that material moves to dissipate energy. In addition, material is deposited at a rate Q that may depend on time. Here, these models are augmented with the appropriate random Brownian motion increments resulting in a stochastic partial differential equation (SPDE) useful for investigating order and control of self-assembly processes. The energy dissipation term is augmented with thermal fluctuations that emulate an atomically discrete deposition process. Thus, there are two contributions to the change in film height, one from thermal fluctuations and energy dissipation (*FD*), and one from random deposition (*Q*). First the dynamics and then the energetics are discussed.

2.1.1 Dynamics

In the language of stochastic differential equations (SDEs), one discusses differential increments rather than time derivatives.³³ In a time increment dt, the height of the film at position **x** changes by an increment,

$$dh\left(\mathbf{x}\right) = dh_{FD}\left(\mathbf{x}\right) + dh_{Q}\left(\mathbf{x}\right),\tag{1}$$

where

$$dh_{FD}(\mathbf{x}) = \left[\nabla \cdot \tilde{\mathbf{D}} \cdot \nabla \mu(x)\right] dt + \left[\sqrt{2k_bT} \nabla \cdot \left(\tilde{\mathbf{D}}^{1/2}\right)^T\right] \cdot \mathbf{dW}_F(\mathbf{x}), \qquad (2)$$

$$dh_Q(\mathbf{x}) = [Q]dt + \left[\sqrt{\Omega_0 Q}\right] dW_Q(\mathbf{x}), \qquad (3)$$

and ∇ is the gradient in the **x**-plane. $\tilde{\mathbf{D}}$ is a constant 2×2 diffusivity matrix; k_b is Boltzmann's constant; T is absolute temperature; $\tilde{\mathbf{D}}^{1/2}$ is the square-root of the diffusivity tensor such that $(\tilde{\mathbf{D}}^{1/2})^T \cdot \tilde{\mathbf{D}}^{1/2} = \tilde{\mathbf{D}}$; $\mathbf{dW}_F(\mathbf{x})$ and $dW_Q(\mathbf{x})$ are a 2D vector and a scalar of local Brownian motion increments; Q is a spatially constant deposition rate measured in height / time; Ω_0 is atomic volume of deposited material. The Brownian motion increments have the following statistical properties: $\langle \mathbf{dW}_F(\mathbf{x}) \rangle = \mathbf{0}$, $\langle dW_Q(\mathbf{x}) \rangle = 0$, $\langle \mathbf{dW}_F(\mathbf{x}_1)^* \mathbf{dW}_F(\mathbf{x}_1) \rangle = \tilde{\mathbf{I}} \delta^2 (\mathbf{x}_1 - \mathbf{x}_2) dt$, $\langle dW_Q(\mathbf{x}_1)^* dW_Q(\mathbf{x})^* \rangle = \delta^2 (\mathbf{x}_1 - \mathbf{x}_2) dt$, $\langle dW_Q(\mathbf{x}_1) \mathbf{dW}_F(\mathbf{x}_1) \rangle = \mathbf{0}$, where $\langle \ldots \rangle$ indicates an ensemble average. This governing dynamics insures that the in the absence of deposition, Q = 0, an ensemble of simulated film height tends towards a thermal equilibrium with a film height probability given by the Gibbs distribution, $P[h] = Z^{-1} \exp(-\mathcal{F}[h]/k_bT)$. Furthermore, in the absence of

diffusion, the film height has the same mean and variance as an atomically discrete deposition process governed by Poisson statistics.

In a more general model, the diffusivity $\tilde{\mathbf{D}}$ may depend on the local configuration, $\tilde{\mathbf{D}} \to \tilde{\mathbf{D}}[h]$. For example, the surface diffusion dynamics of Refs.^{16, 17, 23} results if $\tilde{\mathbf{D}}[h] = D_0 \tilde{\mathbf{I}} / \left[1 + (\nabla h)^2\right]^{1/2}$ (simple proof omitted here). However, a configuration-dependent energy dissipation must be counter-balanced by a configuration-dependent fluctuation term that creates mathematical and numerical challenges, and the theory is not complete for stochastic partial differential equations.³⁴ Thus for simplicity, $\tilde{\mathbf{D}}$ is chosen to be constant resulting in additive thermal fluctuations. A model with more general diffusivity is forthcoming.

Ge/Si has four-fold symmetry of the crystal surface. Thus, within the approximation of a constant diffusivity, $\tilde{\mathbf{D}}$ must be isotropic, $\tilde{\mathbf{D}} = D_0 \tilde{\mathbf{I}}$.¹⁹ For InAs/GaAs, it is commonly believed that diffusion in the [110] directions significantly faster than in the [110] direction;³⁵ thus, for InAs, the diffusivity is set to be $\tilde{\mathbf{D}} = D_0[(1/100)\mathbf{n}_{[110]}\mathbf{n}_{[110]}+\mathbf{n}_{[1\bar{1}0]}\mathbf{n}_{[1\bar{1}0]}]$ where $\mathbf{n}_{[...]}$ is the unit vector in the specified direction, and the repeated unit vectors indicate outer products.²⁹ The numerical value of D_0 is unknown, but it's exact value is somewhat inconsequential for simulations. The important value is the ratio of D_0 to the deposition rate. Here, D_0 is used as a slack variable to ensure that the characteristic exponential growth time for surface undulations, t_0 , is 1.

2.1.2 Energetics

There are three important contributions to the total free energy, $\mathscr{F}[h]$, and the diffusion potential $\mu(\mathbf{x}) = \delta \mathscr{F}/\delta h(\mathbf{x})$. These are the elastic energy $\mathscr{F}_{\text{elast.}}$ that tends to destabilize planar film growth, the surface energy $\mathscr{F}_{\text{surf.}}$ that tends to stabilize planar growth and determine preferred surface orientations, and the wetting energy $\mathscr{F}_{\text{wet.}}$ that initially stabilizes planar film growth and ensures a persistent wetting layer even after SAQD formation. The Fourier components of $\mu(\mathbf{x})$ can be interpreted as the derivative of the free energy density with respect to the given film-height component, $\mu_{\mathbf{k}} = A^{-1} \int d^2 \mathbf{x} e^{-i\mathbf{k}\cdot\mathbf{x}} \left(\delta \mathscr{F}/\delta h(\mathbf{x})\right) = A^{-1} \partial \mathscr{F}/\partial h_{\mathbf{k}}^*$. Each of the three contributions are discussed in turn.

Elastic Free Energy The elastic contribution is calculated by assuming a traction-free surface; thus surface tension effects are neglected. As the surface deviates from planar in the presence of a mismatch eigenstrain, ε_m , additional strain fields must be added to meet the traction-free boundary condition. Additional approximations include linear and homogeneous elasticity. The homogeneous approximation results in about 12-20% offsets in the elastic energy,³⁶ but simplifies calculations a great deal. The elastic strain at the surface is calculated using a method similar to Ref.,¹⁹ but to cubic order. The higher order elasticity calculation method will be described in a forthcoming publication, but it is similar to other higher order perturbation methods in that a Green function is used to solve successively higher order approximations. It is also noted that the rate of change in total elastic energy due to a height increment at **x** is just the elastic energy density at the surface at **x**, $\delta \mathscr{F}_{elast.} / \delta h(\mathbf{x}) = U(\mathbf{x})$.¹⁴ Anisotropic elastic constants used for Ge are $(c_{11}, c_{12}, c_{44}) = (11.99, 4.01, 6.73) \times 10^{11} \text{ dyne/cm}^2$ and for InAs, $(c_{11}, c_{12}, c_{44}) = (8.34, 4.54, 3.95) \times 10^{11} \text{ dyne/cm}^2$. The mismatch strain was set to $\varepsilon_m = -4\%$ for Ge/Si and $\varepsilon_m = -7\%$ for InAs/GaAs.

Surface Free Energy The surface free energy competes with the elastic strain energy to determine the characteristic length scale, $\Lambda_{char.}$ or wavenumber, $k_{char.}$. In addition, the surface energy can be fairly anisotropic and leads to "faceting" of mature dots. "Faceting" appears in quotes because a crystal surface is a true facet if its free energy density is singular or kinked for its orientation. Here, it is assumed that at growth temperatures, there are minima in free energy, but no singularities as previously surmised for Ge/Si,³⁷ and hypothesized here for InAs/GaAs. Surface energy anisotropy can affect SAQD alignment as well.²⁹ The surface free energy is $\mathscr{F}_{surf.} = \int d^2 \mathbf{x} \sqrt{1 + (\nabla h(\mathbf{x}))^2} \gamma(\mathbf{n} [\nabla h(\mathbf{x})])$; thus, the chemical potential, $\mu_{surf.}(\mathbf{x}) = \delta \mathscr{F}_{surf.}/\delta h(\mathbf{x}) = -\nabla \cdot [\delta \mathscr{F}_{surf.}/\delta \nabla h(\mathbf{x})]$.

Ideally, one would like to have the full surface energy profile, but this is not available reliably as configurational entropy effects³² are rarely discussed. Instead, a method similar to Ref.¹⁶ will be used with the energy minima along the observed "faceting" directions.

$$\gamma(\mathbf{n}) = \gamma_0 - \sum_{i}^{N} \gamma_i \exp\left[-\frac{\alpha_i^2}{2} \frac{(1 - \mathbf{n} \cdot \mathbf{n}_i)}{2}\right]$$
(4)

for N different "facets".

Empirical observations, previously used values and zero-temperature surface energy first principle calculations are used to to estimate $\gamma(\mathbf{n})$. For Ge/Si, energy minima are assumed to be in the (001) and {105} orinetations so that $\gamma_0 = 508 \text{ erg/cm}^2$, $\gamma_{1...5} = 7.63 \text{ erg/cm}^2$ and $\mathbf{n}_1 \perp (001)$, and $\mathbf{n}_{2...5} \perp \{105\}$ and $\alpha_{1...5} = 30$. This choice results in a characteristic length scale and wavenumber of $\Lambda_{\text{char}} \approx 39 \text{ nm}$ and $k_{\text{char}} = 0.1611 \text{ nm}^{-1}$.^{19,24,38} For InAs/GaAs, energy minima are assumed to be in the (001) and {136} directions to be consistent with previous observations of SAQD morphology.³⁹ For InAs/GaAs, parameters are estimated to be about $\gamma_0 = 790 \text{ erg/cm}^2$, $\gamma_{1...5} = 158 \text{ erg/cm}^2 \mathbf{n}_1 \perp (001)$, and $\mathbf{n}_{2...5} \perp \{136\}$ and $\alpha_{1...5} = 10$ resulting in $\Lambda_{\text{char}} \approx 25 \text{ nm}$ and $k_{\text{char}} = 0.251 \text{ nm}^{-1}$. For the less symmetric InAs/GaAs system, initially rows alignment closer to the [110] direction(Fig. 1).²⁹ Thus, the anisotropic free energy density effects both large-scale SAQD correlations as well as smaller-scale individual SAQD morphology.

Wetting Free Energy The wetting free energy causes the initial stability of planar film growth until a critical height H_c is reached, and then the presence of a persistent wetting layer even after dot formation, *i.e.* the Stranski-Krastanow growth mode. There has also been some discussion of the electronic origins of the wetting potential.^{40,41} The wetting free energy takes the form of an integral over a local height-dependent density $\mathscr{F}_{wet.} = \int d^2 \mathbf{x} W(h(\mathbf{x}))$ where $W(h) = B/h^a$, a form suggested by Ref.⁴² Thus, $\mu_{wet.}(\mathbf{x}) = W'(h(\mathbf{x})) = -aB/h^{a+1}$. This guessed form is used because the wetting energy is not completely understood or quantitatively determined. Given an assumed critical height, H_c , the coefficient *B* is found by recognizing that the onset of instability occurs when the second derivative of free energy in the Fourier component basis, $\partial^2 \mathscr{F}/\partial h_{\mathbf{k}}^* \partial h_{\mathbf{k}} = 0$ for some non-zero vector \mathbf{k} .^{18,19,41} A soft a = 1 potential and a harder a = 2 potential are tried. Also, two critical film heights are used, $H_c = 1$ or 2 nm.

2.2 Numerical Implementation

The numerical implementation consists of two parts. First, the energetics and governing equations must be spatially discretized via the spectral or Fourier method. Then, the time evolution of the resulting multivariable SDE must be approximated. The random terms in the multivariable SDE are non-differentiable; thus, they render most ordinary differential equation (ODE) integration schemes ineffective.⁴³ An approximately implicit second order weakly convergent integration scheme is used for the time evolution and discussed below.

2.2.1 Spectral Method

In the spectral method, the governing equations are written in the Fourier component basis, and the Fourier series is truncated beyond a smallest wavelength Λ_{\min} or largest wave number k_{\max} . The numerical solution is then greatly facilitated by the use of fast Fourier transforms (FFTs). This method is identical to using Galerkin weighted residuals⁴⁴ with extended sinusoidal interpolation functions.

In the Fourier basis the governing equation becomes

$$dh_{\mathbf{k}} = -\left(\mathbf{k}\cdot\tilde{\mathbf{D}}\cdot\mathbf{k}\right)\mu_{\mathbf{k}}dt + \sqrt{2k_{b}T}i\mathbf{k}\cdot\left(\tilde{\mathbf{D}}^{1/2}\right)^{T}\cdot\left(\mathbf{dW}_{F}\right)_{\mathbf{k}} + Q\delta_{\mathbf{k}-\mathbf{0}}dt + \sqrt{\Omega_{0}Q}\left(dW_{Q}\right)_{\mathbf{k}},\tag{5}$$

Without any loss of simulation fidelity, the thermal fluctuations can be replaced by a term with a scalar noise that has the same covariance,

$$dh_{\mathbf{k}} = -\left(\mathbf{k}\cdot\tilde{\mathbf{D}}\cdot\mathbf{k}\right)\mu_{\mathbf{k}}dt + \sqrt{2k_{b}T\left(\mathbf{k}\cdot\tilde{\mathbf{D}}\cdot\mathbf{k}\right)}\left(dW_{F}\right)_{\mathbf{k}} + Q\delta_{\mathbf{k}-\mathbf{0}}dt + \sqrt{\Omega_{0}Q}\left(dW_{Q}\right)_{\mathbf{k}}.$$
(6)

This replacement is only possible when $\tilde{\mathbf{D}}$ is approximated as constant and the Fourier basis is used as it is the eigenbasis of the dissipation operator $(\mathbf{k} \cdot \tilde{D} \cdot \mathbf{k})$. The new $dh_{\mathbf{k}}$ has the same statistical properties as the old one. $(dW_F)_{\mathbf{k}}$ and $(dW_Q)_{\mathbf{k}}$ are complex modified Brownian motion increments such that $(dW_{F(Q)})_{-\mathbf{k}} = (dW_{F(Q)})_{\mathbf{k}}^*$, $\langle (dW_{F(Q)})_{\mathbf{k}} \rangle = 0$, $\langle (dW_{F(Q)})_{\mathbf{k}_1} (dW_{F(Q)})_{\mathbf{k}_2} \rangle = A^{-1} \delta_{\mathbf{k}_1 + \mathbf{k}_2} dt$, and $\langle (dW_Q)_{\mathbf{k}_1} (dW_F)_{\mathbf{k}_2} \rangle = 0$.

The bulk of calculation is thus finding $\mu_{\mathbf{k}}$ for each successive configuration. The simulations are discretized spatially on a grid $\mathbf{x}_{i,j}$ so that Fast Fourier Transforms may be used. For example, let a discretized function be $\{f\}_{i,j} = f(\mathbf{x}_{i,j})$. Then the approximate gradient is $\{\nabla f\} \approx iFFT [i\{\mathbf{k}\} \times FFT [\{f\}]]$, etc. The numerical calculation of nonlinear functions is improved by padding the Fourier components with zeroes out to at least twice k_{\max} .

Finally, one can obtain reasonable simulations with rather low cutoffs for **k**, for example, $k_{\text{max}} \approx 2k_{\text{char.}}$, where $k_{\text{char.}}$ is the characteristic wave number discussed in Sec. 2.1.2. However, using higher k_{max} values improves the resolution of the simulation and thus one's confidence in the simulation results.

2.2.2 Stochastic Dynamics

Numerical simulation of Eq. 6 is based on the stochastic Euler approximation where $dt \rightarrow \Delta t$ and $(dW_{T(Q)})_{\mathbf{k}} \rightarrow (\Delta W_{T(Q)})_{\mathbf{k}}$,

$$\Delta h_{\mathbf{k}} = -\left(\mathbf{k} \cdot \tilde{D} \cdot \mathbf{k}\right) \mu_{\mathbf{k}} \Delta t + Q \delta_{\mathbf{k}-\mathbf{0}} \Delta t + \sqrt{2k_b T \left(\mathbf{k} \cdot \tilde{D} \cdot \mathbf{k}\right)} \left(\Delta W_T\right)_{\mathbf{k}} + \sqrt{\Omega_0 Q} \left(\Delta W_Q\right)_{\mathbf{k}}$$
(7)

where $(\Delta W_{T(Q)})_{\mathbf{k}}$ is complex and normally distributed with zero mean and variance $= \Delta t/A$, *i.e.* the variance of each complex component is $\Delta t/(2A)$. Each Euler increment is modified to facilitate larger time steps by using a quasi-implicit scheme. The dispersion relation $\sigma_{\mathbf{k}}$ discussed in Sec. 1 is used to give

$$\Delta h_{\mathbf{k}} = -\frac{\left(\mathbf{k}\cdot\tilde{D}\cdot\mathbf{k}\right)\mu_{\mathbf{k}}\Delta t}{1-\left[\sigma_{\mathbf{k}}\right]\Delta t} + Q\delta_{\mathbf{k}-\mathbf{0}}\Delta t + \frac{\sqrt{2k_{b}T\left(\mathbf{k}\cdot\tilde{D}\cdot\mathbf{k}\right)\left(\Delta W_{T}\right)_{\mathbf{k}}} + \sqrt{\Omega_{0}Q}\left(\Delta W_{Q}\right)_{\mathbf{k}}}{\sqrt{1-2\left[\sigma_{\mathbf{k}}\right]\Delta t}},\tag{8}$$

where $\lceil \sigma_k \rceil = \sigma_k$ for $\sigma_k < 0$ and gives zero otherwise. For small Δt , this scheme is identical to the Euler scheme and treats fast fluctuating but bounded Fourier components as appropriately sampled Orhenstein-Uhlenbeck processes.³³ Although somewhat unorthodox, this numerical scheme is in the same spirit as more traditional implicit methods for ODEs and is vastly preferable to the alternative which is fast unrealistically large oscillations of high **k** Fourier components or unacceptably small time increments. The implicit steps are combined into a second order weakly convergent method⁴⁵ that is strongly convergent only to first order.³³ This means that as Δt is reduced, the simulation converges more quickly to an arbitrary but statistically representative solution faster than it converges to a unique solution. The implicit stepping algorithm is implemented in an adaptive step-halving controller where to prevent statistical biasing, Brownian increments are bisected using conditional probabilities but never discarded. Strong step error for Fourier components with $\sigma_k > -1/t_0$ is controled to be less than 12.5%. The weak error should be better. Thus, a reasonably efficient adaptive scheme is implemented. Unfortunately, numerical solution of SDEs and SPDEs is a still developing field. Good generally applicable high-order multivariate strongly convergent schemes are in short supply. The implemented method compromises between expediency and rigor, and the resulting simulations (Sec. 3) are used as a post hoc justification of the method's merits.

3. MODELING RESULTS

The results reported here are preliminary. The purpose of these simulations is to verify the functioning of the non-linear stochastic model, thereby moving beyond the linear stochastic theory^{20, 29} and to study of how the initial stages of SAQD formation influence the final outcome. To this end a series of simulations were performed for Ge/Si and InAs/GaAs. Simulations were started from a uniform film height of $h(\mathbf{x}) = 0.01$ nm, nominally zero. Material was then deposited at various rates. The critical film height H_c and wetting potential hardness *a* were varied somewhat in a couple of simulations. The simulation sizes $l_{\text{sim.}}$ and ratios $k_{\text{max.}}/k_{\text{char.}}$ seem arbitrary, but they have been chosen to allow sufficient oversampling or padding with zeros as well as getting the maximum efficiency from the FFT algorithm which performs best when the number of Fourier components is a power of 2. In each simulation, the unit of time is t_0 , the characteristic time.

Regarding the computational efficiency, each simulation ran in Mathematica⁴⁶ on a single CPU. Each simulation starts off efficiently with fairly large time steps $\Delta t > 1$ when all surface undulations are stable because the average film height is subcritical ($\bar{h} < H_c$). As the critical film height is approached, Δt begins to drop. At supercritical film heights ($\bar{h} > H_c$), Δt drops to about 0.25 as initial surface undulations form. As dots nucleate and grow, Δt drops significantly reaching about 10⁻³ once dots have fully matured. With some patience single CPU simulation can be fairly powerful. A parallel implementation should bring substantial performance improvements.

3.1 Ge/Si Simulations

For Ge/Si, deposition rates vary between $Q = 0.0078125 \text{ nm}/t_0$ and $Q = 0.125 \text{ nm}/t_0$. $H_c = 1$ or 2 nm. System sizes $l_{\text{sys}} = 6\Lambda_{\text{char.}} = 237 \text{ nm}$ or $12\Lambda_{\text{char.}} = 473 \text{ nm}$. Maximum wave number k_{max} varies between $(2.33)k_{\text{char.}} = 0.372 \text{ rad./nm}$ and $(3.25)k_{\text{char.}} = 0.518 \text{ rad/nm}$. Temperature, T = 573.15 K. Except for Simulation 5, the anisotropic surface free energy density is used as described in Sec. 2.1.2. Five simulations were performed with snapshots and links to video shown in Figs. 2–6.

Simulation 1 (Fig. 2) $H_c = 2 \text{ nm}, a = 1, l_{\text{sys.}} = 473 \text{ nm}, k_{\text{max}} = (2.42) k_{\text{char.}} = 0.385 \text{ rad./nm}, Q = 0.125 \text{ nm/}t_0$: Film height and power spectrum snapshots are shown for t = 0, 1, 19, 21, 22, and 25 in units of t_0 . At t = 0, the surface is initially flat. At t = 1, the surface has reached near thermal equilibrium with an rms fluctuation of 10^{-5} nm that resembles white noise (see power spectrum). At t = 19, the average film height $\bar{h} = 2.40$, and the film has taken on undulated structure similar to predictions of linear stochastic theories.^{19,20} The power spectrum appears to corroborate this view showing large support around $k = k_{char}$ in the (100) directions. At t = 21, the initial structure has matured but not in the manner predicted in Sec. 1 and Refs.^{17,18} Each height maximum at t = 19 does not correspond to a developing quantum dot. Only the largest peaks appear to be forming dots. Each of these large dots appears to be nucleating a set of surrounding smaller dots and bumpy rolls in a propagating pattern that does not have an obvious relation to earlier undulations. The power spectrum grows dramatically at higher k values as expected with the development of the new smaller length scale structure. At t = 22, all the intervening surface is broken up into small dots, and some small dots have already coalesced into larger dots. There is perhaps a relation between undulation maxima at t = 19 and the location of coalesced dots at t = 22. The power spectrum is showing new support along the $\langle 110 \rangle$ directions. At t = 25, many of the smaller dots have coalesced into larger dots. The power spectrum has shrunk in radius to smaller k-values. The new octagonal structure of the power spectrum may be indicative of the dot alignments that occur both in the $\langle 100 \rangle$ and $\langle 110 \rangle$ directions. This final configuration has a large distribution in dot size mostly with smaller dots intermingled with larger dots.

Simulation 2 (Fig. 3) $H_c = 1 \text{ nm}$, a = 2, $l_{\text{sys.}} = 237 \text{ nm}$, $k_{\text{max}} = (2.33) k_{\text{char.}} = 0.372 \text{ rad./nm}$, $Q = 0.0625 \text{ nm/}t_0$: This simulation uses a smaller critical height, (H_c) a proportionately smaller deposition rate (Q) and a harder wetting potential than simulation 1, but still seems to share many of the features from Simulation 1. At t = 19.5, the pattern and power spectrum is similar to that in Simulation 1 at t = 19, still consistent with linear theories. At t = 20.7, one sees similar initial dot formation at the sites of largest initial undulations as well as similar radiating smaller-scale patterns as in Simulation 1. However, the mature configuration at t = 23.8 is somewhat different. It still consists of larger dots with intervening smaller dots, but there is much less coalescence and ripening of smaller dots into larger dots. The final pattern is less dense than in Simulation 1, and the dot alignments are predominantly in the $\langle 100 \rangle$ directions, a fact that appears to be reflected by the absence of the significant spectral support in the $\langle 110 \rangle$ directions that was observed in Simulation 1.

Simulation 3 (Fig. 4) $H_c = 1 \text{ nm}$, a = 2, $l_{\text{sys.}} = 237 \text{ nm}$, $k_{\text{max}} = (2.33)k_{\text{char.}} = 0.372 \text{ rad./nm}$, $Q = 0.0078125 \text{ nm}/t_0$: This simulation has a much slower deposition rate than Simulation 2, but follows a similar pattern (compare Figs. 3 and 4). The result of the slower deposition appears to be a lower and less uniform density of dots: fewer dots overall, and smaller dots that radiate like spokes from the larger dots.

Simulation 4 (Fig. 5) $H_c = 1 \text{ nm}$, a = 2, $l_{\text{sys.}} = 158 \text{ nm}$, $k_{\text{max}} = (3.25)k_{\text{char.}} = 0.518 \text{ rad./nm}$, $Q = 0.0078125 \text{ nm}/t_0$: This simulation is the same as Simulation 3, but with a smaller size ($l_{\text{sys.}}$) but higher resolutions, k_{max} . The purpose of this simulation is to test whether the qualitative simulation results are converged for Simulation 3 with respect to k_{max} . There are strong points of qualitative agreement between the two simulations. The mature dot configuration and power spectrum for this simulation at t = 137 has similarities to Simulation 3 at t = 135, but it bears more resemblance to Simulation 2 at t = 23.8. This may indicate that more resolution is needed in the previous simulations or that this simulation has a finite $l_{\text{sys.}}$ size effect where the periodic boundaries are too close. Further study is needed.

Simulation 5 (Fig. 6) $H_c = 1 \text{ nm}$, a = 2, $l_{\text{sys.}} = 237 \text{ nm}$, $k_{\text{max}} = (2.33) k_{\text{char.}} = 0.372 \text{ rad./nm}$, $Q = 0.0625 \text{ nm/}t_0$. This simulation differs from others in that the free energy density γ is set to be isotropic with $\gamma_0 = 1927 \text{ erg/cm}^2$ and $\gamma_1 = 0 \text{ erg/cm}^2$. These values give the same characteristic wavenumbers and times, but alter the later non-linear evolution.: Without the surface energy density anisotropy, the SAQDs form in the manner suggested in Sec. 1. The initial undulation maxima (t = 23) each forms a dot at t = 25 unless the initial maxima were too close or indistinct at the earlier time. These individual dots then mature and ripen (t = 31). In sharp contrast to Simulations 1-4, there are no smaller scale structures. The initial early pattern predicted by linear stochastic theories is strongly reflected in the mature dot configuration.



Figure 2. Video 1: Ge/Si Simulation 1 film heights and power spectra. $H_c = 2 \text{ nm}, a = 1, l_{\text{sys.}} = 473 \text{ nm}, k_{\text{max}} = (2.42)k_{\text{char.}} = 0.385 \text{ rad./nm}, Q = 0.125 \text{ nm}/t_0$. t is in units of t_0 ; average film height (\bar{h}) and film height r.m.s. fluctuation (h_{rms}) in nm. http://dx.doi.org/10.1117/12.795615.1



Figure 3. Video 2. Ge/Si Simulation 2 film heights and power spectra. $H_c = 1 \text{ nm}$, a = 2, $l_{\text{sys.}} = 237 \text{ nm}$, $k_{\text{max}} = (2.33)k_{\text{char.}} = 0.372 \text{ rad./nm}$, $Q = 0.0625 \text{ nm}/t_0$. t is in units of t_0 ; average film height (\bar{h}) and film height r.m.s. fluctuation h_{rms} in nm. http://dx.doi.org/10.1117/12.795615.2



Figure 4. Video 3. Ge/Si Simulation 3 film heights and power spectra. $H_c = 1 \text{ nm}$, a = 2, $l_{\text{sys.}} = 237 \text{ nm}$, $k_{\text{max}} = (2.33)k_{\text{char.}} = 0.372 \text{ rad./nm}$, $Q = 0.0078125 \text{ nm}/t_0$. t is in units of t_0 ; average film height (\bar{h}) and film height r.m.s. fluctuation (h_{rms}) in nm. http://dx.doi.org/10.1117/12.795615.3



Figure 5. Video 4. Ge/Si Simulation 4 film height and power spectrum. $H_c = 1 \text{ nm}, a = 2, l_{\text{sys.}} = 158 \text{ nm}, k_{\text{max}} = (3.25)k_{\text{char.}} = 0.518 \text{ rad./nm}, Q = 0.0078125 \text{ nm}/t_0$. *t* is in units of t_0 ; average film height (\bar{h}) and film height r.m.s. fluctuation (h_{rms}) in nm. http://dx.doi.org/10.1117/12.795615.4



Figure 6. Video 5. Ge/Si Simulation 5 film heights and power spectra. $H_c = 1 \text{ nm}, a = 2, l_{\text{sys.}} = 237 \text{ nm}, k_{\text{max}} = (2.33)k_{\text{char.}} = 0.372 \text{ rad./nm}, Q = 0.0625 \text{ nm}/t_0$. $H_c = 1 \text{ nm}, a = 2, l_{\text{sys.}} = 237 \text{ nm}, k_{\text{max}} = (2.33)k_{\text{char.}} = 0.372 \text{ rad./nm}, Q = 0.0078125 \text{ nm}/t_0$. $\gamma_0 = 1927 \text{ erg/cm}^2$, $\gamma_1 = 0 \text{ erg/cm}^2$. *t* is in units of t_0 ; average film height (\bar{h}) and film height r.m.s. fluctuation (h_{rms}) in nm. http://dx.doi.org/10.1117/795615.5



Figure 7. Video 6. InAs/GaAs Simulation 1 film heights and power spectra. $H_c = 1 \text{ nm}$, a = 1, $l_{\text{sys.}} = 148 \text{ nm}$, $k_{\text{max}} = (2.33)k_{\text{char.}} = 0.593 \text{ rad./nm}$, $Q = 0.00390625 \text{ nm}/t_0$. t is in units of t_0 ; average film height (\bar{h}) and film height r.m.s. fluctuation (h_{rms}) in nm. http://dx.doi.org/10.1117/12.795615.6

3.2 InAs/GaAs Simulations

For InAs/GaAs, deposition rates vary between $Q = 0.00390625 \text{ nm}/t_0$ and $Q = 0.125 \text{ nm}/t_0$. $H_c = 1 \text{ or } 2 \text{ nm}$. System sizes $l_{\text{sys}} = 6\Lambda_{\text{char.}} = 148 \text{ nm}$ or $12\Lambda_{\text{char.}} = 297 \text{ nm}$. Maximum wave number k_{max} varies only slightly between simulations: $(2.33)k_{\text{char.}} = 0.593 \text{ rad./nm}$ and $(2.42)k_{\text{char.}} = 0.614 \text{ rad/nm}$. Temperature, T = 873.15 K. The anisotropic surface free energy density is used as described in Sec. 2.1.2. Three simulations were performed with snapshots shown in Figs. 7–9.

Simulation 1 (Fig. 7) $H_c = 1 \text{ nm}$, a = 1, $l_{\text{sys.}} = 148 \text{ nm}$, $k_{\text{max}} = (2.33)k_{\text{char.}} = 0.593 \text{ rad./nm}$, $Q = 0.00390625 \text{ nm}/t_0$: InAs/GaAs simulations far below the instability transition $\bar{h} = H_c$ begin the same as Ge/Si simulations, with essentially small amplitude white noise. Above the transition threshold, after some structure has developed (t = 272), the surface is undulated in fairly correlated sheared lattice pattern as predicted by linear theories. Compare the film height and power spectrum Fig. 7 (t = 272) with Fig. 1. At t = 283, the larger peaks in the initially undulated surface have grown into mature dots. A propagating wave pattern appears to emanate from each dot and effectively overwhelming previously existing undulations. At t = 303, the wave pattern has coarsened into dots of varying sizes. This sequence is very similar to that observed in Ge/Si simulations.

Simulation 2 (Fig. 8) $H_c = 2 \text{ nm}$, a = 1, $l_{sys.} = 297 \text{ nm}$, $k_{max} = (2.42)k_{char.} = 0.614 \text{ rad./nm}$, $Q = 0.125 \text{ nm/}t_0$: This simulations is larger than Simulation 1 and differs by having a faster deposition rate and a large assumed critical film height (H_c). The initial undulations (t = 20) develop as predicted by linear theory. At t = 24, one can see that the largest undulations have formed blockish dots, and the grooved borders of these blockish dots have propagated outward essentially suppressing preexisting patterns. At t = 28, these grooves have fully propagated filling the intervening spaces between initial dots, and they have themselves broken up into dots. This resulting pattern is similar to the dense dot pattern observed at the end of Ge/Si Simulation 1 (Fig. 2).



Figure 8. Video 7. InAs/GaAs Simulation 2 film heights and power spectra. $H_c = 2 \text{ nm}, a = 1, l_{sys} = 297 \text{ nm}, k_{max} = (2.42)k_{char.} = 0.614 \text{ rad./nm}, Q = 0.125 \text{ nm}/t_0$. t is in units of t_0 ; average film height (\bar{h}) and film height r.m.s. fluctuation (h_{rms}) in nm. http://dx.doi.org/10.1117/12.795615.7

Simulation 3 (Fig. 9) $H_c = 1 \text{ nm}$, a = 2, $l_{\text{sys.}} = 297 \text{ nm}$, $k_{\text{max}} = (2.42)k_{\text{char.}} = 0.614 \text{ rad./nm}$, $Q = 0.0625 \text{ nm}/t_0$: This simulation differs from Simulation 1 by having a faster deposition rate, larger simulation size and a harder wetting potential. Initial undulations form as expected (t = 21). Then, small groups of dots appear to nucleate (t = 22.5). Then, the intervening space between dots groups fills up with rolls aligned obliquely to the $\langle 1\bar{1}0 \rangle$ directions.

4. DISCUSSION/CONCLUSION

The stochastic nonlinear spectral simulation appears to run reasonably well and give reasonable outcomes. The efficiency could probably be improved by using higher order multivariable stochastic SDE integrators, but implementation then becomes difficult. In addition, the most advanced algorithms make assumptions about noise amplitudes that may not apply here.⁴³ Nevertheless, most simulations run in about a day or two clock time on a single CPU, and thus are reasonable candidates for further work. There may be some issues with smaller system sizes as observed in Ge/Si Simulation 4, but for the most part, it appears that one can neglect higher-*k* Fourier components (> $3k_{char}$) without introducing much error. Initial results are promising, but further study is needed.

Three points of progress have been made. First, stochastic effects of thermal fluctuations and random deposition have been incorporated into a non-linear model of SAQD formation. Second, this incorporation allows the simulation of deposition and evolution for a wide range of growth rates and removes concerns about non-stochastic models where random initial conditions are used as surrogates for truly random physical processes. Third, nonlinear models of Ge/Si SAQD growth have been extended to InAs/GaAs SAQD growth by using suitable minima or "facets" in the surface free energy density. These models need to be tuned to experimental observations, but appear to be robust enough that they ought to be suitable and alterable for expanded simulations, such as growth on vicinal⁴⁷ or shallow patterned substrates. Because stochastic contributions have been physically modeled, they should be able to yield quantitative results regarding order and control of SAQD formation. Further improvements of the model might include the introduction of chemical intermixing. This feature would be particularly important for studying alloys such as Ge_xSi_{1-x}/Si SAQD or In_xGa_{1-x}As/GaAs SAQD formation.



Figure 9. Video 8. InAs/GaAs Simulation 3 film heights and power spectra. $H_c = 1$ nm, a = 2, $l_{sys.} = 297$ nm, $k_{max} = (2.42)k_{char.} = 0.614$ rad./nm, Q = 0.0625 nm/ t_0 . t is in units of t_0 ; average film height (\bar{h}) and film height r.m.s. fluctuation (h_{rms}) in nm. http://dx.doi.org/10.1117/12.795615.8

Although the purpose of these preliminary simulations was to test the applicability and robustness of the presented simulation, a few conclusions can be tentatively drawn. The simulations reported in Sec. 3 show fairly diverse growth sequences even though the model is essentially the same for each simulation; only simulation parameters have been altered. In the absence of surface energy anisotropy, the original growth sequence discussed in Sec. 1 appears to hold (Ge/Si Simulation 5) whereby initial surface undulations form distinct dots and then continue to evolve and ripen. However, surface energy anisotropy alters this growth sequence. Initial undulations help determine the locations of a few sparse large dots, but patterns of various sorts emanate from these dots and ultimately form secondary dots that tend to be smaller. The nature of the emanating patterns depends on parameters such as assumed critical film height (H_c) and wetting potential hardness a. They might also depend on deposition rate. This mechanism occurs for both Ge/Si and InAs/GaAs SAQDs. It should be noted that previous deterministic models of Ge/Si SAQDs with surface energy anisotropy have produced growth more like Simulation 5,¹⁷ suggesting that changing the form or parameters can transition the growth behavior from one type of growth sequence to another. Observations of InAs/GaAs SAQDs^{47,48} show alignments close to $[1\overline{10}]$. The presented simulations here suggest multiple mechanisms of how this might occur. First, initial alignments of surface undulations might explain final SAQD chaining,²⁹ but perhaps with a different assumed surface energy density. Second, chains of dots might emanate from original precursor dots. Third, groups of dots might nucleate together as in InAs/GaAs Simulation 3, and then induce surface patterns that suppress nearby SAQD formation leaving spaced chains. Further systematic studies with more quantitative comparison with experimental observations are needed. When making such comparisons care should be taken. For example, in GeSi Simulation 1, SAQDs first appear when $\bar{h} = 3.02$ nm even though the critical film height is actually $H_c = 2$ nm for that simulation. The observed H_c thus lags the actual transition height.

Much of the ambiguity in SAQD formation mechanisms results from the large number of guessed parameters: a, H_c , γ_0 , γ_1 , α_i and the facet locations. The simulations appear to be sensitive to these parameters. On the one hand, this sensitivity means that parameters must be determined to better accuracy before quantitative modeling results can be trusted. On the other hand, it means that bounds can be placed on these parameters by quantitative observation of SAQD morphology and arrangements combined with simulation. If the parameters are important, then they are also measurable.

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