Direct Molecular Dynamics Simulation of Ge Deposition on Amorphous SiO₂ at Experimentally Relevant Conditions

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Abstract

Molecular dynamics simulations were used to study Ge island nucleation and growth on amorphous SiO₂ substrates. This process is relevant in selective epitaxial growth of Ge on Si, for which SiO₂ is often used as a template mask. The islanding process was studied over a wide range of temperatures and fluxes, using a recently proposed empirical potential model for the Si-SiO₂-Ge system. The simulations provide an excellent quantitative picture of the Ge islanding and compare well with detailed experimental measurements. These quantitative comparisons were enabled by an analytical rate model as a bridge between simulations and experiments despite the fact that deposition fluxes accessible in simulations and experiments are necessarily different by many orders of magnitude. In particular, the simulations led to accurate predictions of the critical island size and the scaling of island density as a function of temperature. The overall approach used here should be useful not just for future studies in this particular system, but also for molecular simulations of deposition in other materials.

Keywords: molecular dynamics, atomic deposition, amorphous SiO₂, germanium, silicon.

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1. INTRODUCTION

The need for high-efficiency, yet cost-effective, advanced semiconductor devices has fueled interest in the production of high quality Ge films on Si wafers. The optical and electronic properties of Ge allow for many applications including photodetectors [1, 2] and transistors [3-6]. The lattice compatibility of Ge with the III-V materials also makes it an ideal substrate choice for high-efficiency III-V multijunction solar cells [7-10], but the feasibility of Ge substrates is limited by the high cost of bulk Ge. High-quality Ge on Si virtual substrates, on the other hand, preserve the advantages of Ge substrates while reducing costs significantly.

The primary challenge for heteroepitaxial growth of Ge on Si is the lattice mismatch strain between Ge and Si, which drives formation of misfit dislocations in the highly-strained Ge layer [11]. Many approaches have been proposed to reduce the impact of lattice mismatch strain in epitaxial Ge films, including compositional grading [12], cyclic thermal annealing [13], and selective epitaxial growth [14-17]. Selective epitaxial growth (SEG), in particular, has received much attention because of its potential to produce strain-free, high-quality Ge film on Si without the need for costly thermal treatments or thick, compositionally-graded Ge layers. In SEG, an amorphous masking material (usually SiO_2 or Si_3N_4) is applied on top of the Si wafer surface. Nanoscale cavities are then introduced in the mask layer either by high-resolution lithography [15, 16] or by self-limiting chemical reactions [17]. Subsequent Ge deposition leads to the formation of epitaxial Ge islands at the base of these cavities (“seeding pads”) which then overgrow onto the amorphous masking layer. The epitaxially adhered islands eventually grow and coalesce into a contiguous crystalline film on top of the amorphous mask. A key aspect of SEG is that the epitaxial contact area between Ge and Si layers is restricted to the nanoscale seeding pads, which significantly lowers the mismatch strain in the Ge layer and suppresses the formation of misfit strain defects. Indeed, various studies have reported threading dislocation densities in SEG films as low as $\text{O}(10^6) \text{ cm}^{-2}$, making such films suitable substrates for the fabrication of III-V structures [18]. While the low defect densities in SEG films reported to date are promising, further improvements will require a

more fundamental understanding of the defect formation processes that lead to stacking faults and threading dislocations.

Here, we consider the direct nucleation of Ge on the amorphous SiO_2 (a- SiO_2) mask. While secondary nucleation of Ge is not a primary technical challenge in SEG, we focus on this process because it has been characterized experimentally in great detail [19]. In particular, Leonhardt *et al.* [19] have studied quantitatively the kinetics of Ge island nucleation using scanning electron microscopy. Overall, it was found that Ge islands grow on a- SiO_2 by direct impingement from the vapor, rather than by adatom surface diffusion, principally because Ge adatoms tend to desorb before they diffuse an appreciable distance on the surface – this growth mode corresponds to the so-called *extremely incomplete condensation* regime. As a result, the condensation coefficient, defined as the amount of adsorbate condensed on the surface versus the total amount deposited, was found to be small across all temperatures considered in ref. [19]. The small characteristic Ge surface diffusion distance on SiO_2 was corroborated by the absence of Ge adatom exclusion zones around the seeding pads that typically arise as a consequence of diffusion-limited island growth [19].

We use the experimental results in ref. [19] to (1) establish the overall quantitative reliability of the empirical potential-based atomistic simulation framework introduced previously in ref. [20] and (2) determine the feasibility of meaningfully studying defect formation in SEG with fully-resolved atomistic simulations such as molecular dynamics (MD). The latter challenge is a well-known one in the broader context of deposition processes. The difficulties are two-fold. First, experimental atomic deposition rates, typically $\text{O}(10^{13}\text{-}10^{14})$ atom/ cm^2s , are many orders-of-magnitude too slow to be simulated directly with molecular dynamics, which is limited to timescales on the order of hundreds of nanoseconds. Second, sufficiently large substrate areas need to be considered to allow for the proper capture of island size and density distributions. Empirical potential MD simulations currently are limited to tens to hundreds of nanometers per spatial dimension, and increasing the number of atoms usually comes at a cost of a further reduction in the accessible timescale. Together, these restrictions make the direct MD simulation of

deposition rather challenging, and constrain simulation conditions to unrealistic operating conditions, which may or may not meaningfully represent relevant experimental ones.

We note that alternatives to direct MD simulations exist for atomic deposition processes. A powerful example is the kinetic Monte Carlo (KMC) method [21-23], which has been applied in various forms to the study of deposition. The most computationally efficient variant of KMC, typically defined on a lattice, require *a priori* knowledge of all possible events and their rates, so assumptions have to be made on choosing processes to be included in the model [23-34]. However, the amorphous nature of the system of current interest makes it very difficult (or even impossible) to specify a comprehensive event catalog. The development of ‘on-the-fly’ atomistic KMC methods [35-37] addresses such limitations by building dynamically a catalog of events during the simulation via searches over all energy saddles accessible from each current configuration. However, this type of KMC is much more computationally expensive, and may not be necessarily much faster than MD simulation. Lastly, we briefly mention the variety of techniques that have been proposed to accelerate MD simulations. Examples include temperature-accelerated dynamics (TAD) and hyperdynamics [38, 39]. These methods aim to speed up MD simulation by increasing the rate at which the system escapes from one energy basin to another. While accelerated MD simulations have been successfully applied to study island growth in some cases [40, 41], the identification of transitions between energy basins in rough-landscape systems such as the present one can pose challenges and reduce efficiency.

The remainder of the paper is organized as follows. In Section 2, a brief summary is provided of the analytical rate equation approach developed by Venables and others [42-46]. The rate equation theory is used to provide a quantitative ‘bridge’ between simulations and experiment, which, as we will show, would otherwise be difficult to achieve. The molecular dynamics simulation method used to simulate the deposition process and associated calculations are discussed in Section 3. In Section 4, results are provided for various deposition simulations, along with analysis based on rate equation theory and comparisons to experimental data. Finally, conclusions are presented in Section 5. Reinforcing our previous conclusions [20], we demonstrate that the empirical potentials used to describe the interatomic

interactions between Si, Ge, and O atoms appear to provide an excellent quantitative picture for Ge island deposition on a-SiO₂. We also demonstrate that it is possible to directly access experimentally relevant deposition regimes with molecular dynamics simulation, even if the actual deposition rates are unrealistically high. The latter finding may be true for the simulation of atomic deposition in a variety of systems.

2. RATE EQUATION THEORY FOR ISLAND NUCLEATION AND GROWTH

Analytical descriptions based on rate equations provide a powerful framework for understanding various features of island evolution during deposition. Here, we briefly summarize the framework proposed by Venables and others [45-47] that describes island growth in terms of low-order moments, namely the island density and surface coverage. More detailed descriptions also have been proposed to include additional measures of island growth dynamics, such as island size and/or capture zone distributions [48-55], but these are not addressed here.

Several key assumptions underpin the Venables framework. First, subcritical clusters are assumed to exist in a quasi-steady state relative to stable (supercritical) species, i.e.,

$$\frac{n_j}{N_0} = C_j \left(\frac{n_1}{N_0} \right)^j \exp(\beta E_j) \quad (1)$$

for all j less than or equal to the critical nucleus size, i [43]. In Eq. (1), N_0 is the number of adsorption sites, n_1 and n_j are the concentrations of monomers and clusters of size j , respectively, C_j is a statistical weight factor representing the orientation degeneracy for j -sized clusters, and E_j is the binding energy of j -sized clusters relative to j individual monomers adsorbed on the surface. In addition, spatial variations of monomers are assumed to be negligible, and only monomers are assumed to be mobile. With these assumptions in place, the monomer and cluster balances may be expressed as

$$\frac{dn_1}{dt} = F - \frac{n_1}{\tau_a} - 2U_1 - \sum_{j>1}^i U_j - U_x, \quad (2)$$

$$\frac{dn_j}{dt} = U_{j-1} - U_j, (1 < j \leq i) = 0, \quad (3)$$

$$\frac{dn_x}{dt} = U_i - U_c, \quad (4)$$

where F is the deposition flux, n_x is the number of supercritical clusters, and $\tau_a^{-1} \sim \exp(-\beta E_a)$ is the monomer desorption rate with E_a representing the adsorption energy barrier. The various U terms represent different processes: $2U_1$ represents the rate of monomer dimerization, U_j is the net rate of formation of subcritical clusters (of size j) via monomer addition, U_i is the rate of monomer addition to critically sized clusters, U_x is the rate at which monomers join all supercritical clusters ($x = \{i+1, i+2, \dots, \max\}$) either by surface diffusion or by direct impingement, and U_c is the rate of coalescence between supercritical clusters.

For clusters that are of critical size and smaller, it is assumed that direct impingement by monomer from the vapor is negligible so that the monomer addition rate is given by

$$U_k = \sigma_k D n_1 n_k, \quad (5)$$

where σ_k is the capture number for clusters of size k , and $D \sim \exp(-\beta E_d)$ is the monomer diffusivity with a diffusion barrier E_d . For all supercritical clusters combined, the monomer addition rate is then given by

$$U_x = \sigma_x D n_1 n_x + F a_x n_x, \quad (6)$$

where σ_x is the average capture number for all supercritical islands, and a_x is the average substrate surface area covered by stable clusters, so that $a_x n_x$ represents the total time-dependent surface coverage by stable clusters. Finally, assuming that all supercritical islands are three-dimensional hemispheres gives,

$$U_c \approx \frac{1}{2} \sum_k n_x^2 \frac{d}{dt} (\pi (2r_x)^2), \quad (7)$$

where r_x is the average radius for all stable clusters.

The balances in Eqs. (2-4) may be solved for the cluster size distribution; see refs. [45, 46] for details. For the extremely incomplete condensation regime that was found to describe the experiments in ref. [19], the saturation (i.e., maximum) number density of stable islands is given by [44]

$$n_{x,\max} \sim (F)^{\frac{2i}{3}} \exp\left(\frac{E}{k_B T}\right), \quad (8)$$

where $E = \frac{2}{3} [E_i + (i+1)E_a - E_d]$. Importantly, the value of $n_{x,\max}$ can be straightforwardly measured in experiments and has been used to deduce activation energies for adsorption and surface diffusion, as well as critical cluster binding energies in a variety of systems [56-59]. Below, we use this quantity to establish direct connections between simulation and experiment.

3. METHODOLOGY

3.1 General Considerations

All simulations were performed using an empirical interatomic potential model for the ternary Si-Ge-O system based on the Tersoff framework [60]. In ref. [20], we combined the well-established Tersoff parameters for the binary Si-Ge system [60] with a recent parameterization for the Si-O binary system [61]. Using a single fitting parameter that represented the Ge-O interaction strength, the ternary potential model was found to reproduce well a wide range of structural and thermodynamic properties for bulk a-SiO₂, the Si-SiO₂ interface, and Ge adatom binding on a-SiO₂ surfaces [20]. All molecular dynamics simulations were performed using the LAMMPS software package [62]. The simulations were conducted in the constant pressure/constant temperature (NPT) ensemble unless otherwise specified, and Nose-Hoover thermostats and barostats [63] were applied to control the temperature and pressure of the system, respectively. A time step of 1 fs was applied in all cases. Static relaxations were performed using the conjugate gradient algorithm in the LAMMPS software.

3.2 Preparation of a-SiO₂ Surfaces

Bulk a-SiO₂ was prepared using a melt-quench sequence [20]. First, a cubic β -cristobalite SiO₂ lattice consisting of 4096 SiO₂ units with dimensions 5.8×5.8×5.8 nm³ was created with periodic boundary conditions applied on all sides of the system. The SiO₂ lattice was melted at 5000K for 5 ns, followed by rapid cooling to 0K at a linear cooling rate of 1×10¹² K/s. The quality of the resulting a-SiO₂ has been discussed in detail in our previous study [20] and overall exhibits good agreement with bulk density, radial distribution function, and bond angle distributions predicted by *ab initio* calculations and experiments.

Following the formation of bulk a-SiO₂, periodic boundary conditions in the *z*-direction were removed and the slab sliced in two along the *xy*-plane to create an a-SiO₂ slab with dimensions approximately 6×6×3 nm³. Note that the sliced slab is still periodic in the *xy*-plane. In all subsequent deposition simulations, the bottom 1 nm of the a-SiO₂ slab was fixed to emulate a bulk-like environment for the 2 nm-thick active layer above. In each case, before Ge deposition was initiated, the temperature in the active layer was increased at a linear rate of 1×10¹² K/s to the deposition temperature, which ranged from 1800K to 2300K. The temperature was then held constant until the system potential energy equilibrated; the equilibration period was typically 30 ns long. The annealing process was not found to strongly impact the final surface roughness: the root-mean-square surface roughness increased with annealing temperature from about 0.08 nm at 2000K to about 0.11 nm at 2300K. Moreover, the small diffusion distance of Ge on a-SiO₂ (see Section 4.1) would further mitigate any impact of varying surface roughness with temperature. Following equilibration, the a-SiO₂ slab was replicated four times in the *x* and *y* directions, resulting in a slab with dimensions 24×24×3 nm³. The replicated surface finally was equilibrated at constant temperature for a further 0.2 ns. Note that for all deposition simulations, the top *z*-boundary of the simulation domain was placed at least 5 nm higher than the highest point of the a-SiO₂ surface to prevent any bias in the Ge deposition due to boundary effects.

3.3 Ge Deposition Simulations

Simulations of Ge deposition on the a-SiO₂ surface were performed in the constant volume/constant temperature (NVT) ensemble. A 2D schematic representation of the deposition simulation system is shown in Figure 1. Ge atoms were introduced into the region above the slab at a constant rate to simulate deposition at a prescribed flux ranging from 5×10^{21} to 6.9×10^{24} atoms/cm²s. The x and y coordinates of each inserted atom were chosen randomly, while the z -coordinate was set to be 5 Å above the maximum height of all atoms within a 5 Å radius along the xy -plane. All inserted atoms were initialized with downward velocity corresponding to the deposition temperature.

Figure 1. Schematic diagram of the system. Ge atoms are represented by the green particles. The fixed and active SiO₂ layers (1 nm and 2 nm in thickness, respectively) are denoted in the figure. The arrow indicates a Ge atom that was newly added to the system that is 5 Å above the highest atom within a 5 Å radius along the xy plane. This atom is given a downward velocity moving towards the a-SiO₂ surface.

Following each atomic insertion, the system was evolved with MD until the addition of the next Ge atom. An island was defined as a group of interacting Ge particles consisting of two or more Ge monomers with inter-particle distances being less than the Ge-Ge potential cutoff distance (3.1 Å). A Ge

particle was defined as adsorbed if its distance to the nearest Si or O atom is within the respective Ge-Si or Ge-O potential cutoff distances (2.9462Å and 2.49Å, respectively).

Ge atoms desorbing from the a-SiO₂ surface require special attention. Under some conditions, namely when the deposition rate is large, desorbed atoms rising from the surface may collide and bind with downward-moving atoms to form aggregates in the vapor phase. These aggregates represent an artifact that subsequently impacts the effective deposition flux to the surface. In order to address this issue, checks were performed before every Ge atom addition to remove any desorbed Ge atoms from the system. Desorbed atoms were identified as Ge atoms with an upward-directed (+z direction) velocity component and zero potential energy.

4. RESULTS

4.1 Single Ge Adatoms on a-SiO₂

Measurements of Ge adatom residence time and displacement distributions on the a-SiO₂ surface were made by depositing a single Ge atom onto the surface and then tracking its trajectory. The displacement, X_a , of the Ge atom on the surface was defined as the distance between the positions at which the particle adsorbs (potential energy first becomes nonzero) and desorbs (potential energy becomes zero). The residence time, τ_a , correspondingly was defined as the duration of the adsorption time. For each sample, the simulation was terminated either when the Ge atom desorbed or when the simulation time exceeded 50 ps. The single-atom simulation described here was repeated at least $O(10^4)$ times at both 1800K and 2300K to obtain converged distributions for X_a .

Our previous characterization of the a-SiO₂ surface [20] showed that the surface is highly heterogeneous in terms of Ge binding strength, and is comprised of favorable binding sites interspersed with non-binding or even repulsive patches. The presence of unfavorable binding regions in between the binding sites leads to extremely short adatom residence time and surface displacement distributions. As shown in Figure 2, more than 90% of Ge adatoms leave the surface after diffusing less than 3Å at both

temperatures. The corresponding residence time for these displacements is less than 1 ps. In fact, the mean displacement before desorption for a Ge adatom on the surface is less than $1\text{\AA}$, which is on the order of the atomic vibrational amplitude, further emphasizing that adatoms are essentially immobile under the simulations conditions considered here.

Figure 2. Ge atom displacement probability distributions on amorphous SiO_2 surface at 1800K (blue circles) and 2300K (red diamonds).

4.2 Ge Deposition and Island Nucleation

Ge deposition runs were carried out for deposition fluxes ranging from 5×10^{21} atoms/cm²s to 6.9×10^{24} atoms/cm²s at temperatures between 1800K and 2300K. Figure 3 shows four top-down snapshots of the a-SiO₂ surface, all taken when the area density of Ge atoms brought down to the surface, N_{add} , had reached 6.6×10^{14} atoms/cm²; the four snapshots correspond to different deposition fluxes and temperatures (see Figure 3 caption). While the resulting Ge islands are always randomly distributed and amorphous in structure, their areal density and sizes are strongly impacted by the deposition rate and substrate temperature. Higher temperatures clearly destabilize islands and lead to lower densities of smaller islands, while increasing the deposition rate leads to a higher density of larger islands.

Figure 3. System configurations at $N_{add} = 6.6 \times 10^{14}$ atoms/cm² for different substrate temperatures and deposition fluxes: (a) 2000K, 2.76×10^{22} atoms/cm²s, (b) 2200K, 2.76×10^{22} atoms/cm²s, (c) 2000K, 1.38×10^{24} atoms/cm²s, (d) 2200K, 1.38×10^{24} atoms/cm²s. Red atoms: O. Blue atoms: Si. Green atoms: Ge. Renderings generated using the OVITO visualization software [64].

The stable surface island density, n_x , is shown in Figure 4 as a function of substrate temperature at several deposition fluxes for $N_{add} = 3.47 \times 10^{14}$ atoms/cm². Here, and in all subsequent discussion, we assume that the critical island size, i , is less than 2 and all clusters are included in the evaluation of n_x . In Section 4.3, we self-consistently validate this assumption. At every flux, the island density decreases slowly as the temperature is increased from 1800K to about 2000K and then decreases more rapidly as the temperature is further increased. Over the entire flux range considered, and for all temperatures, higher fluxes generally correspond to higher island densities. These trends are in qualitative agreement with the experimental observations by Leonhardt *et al.* [19].

Figure 4. Surface island density, n_x , as a function of temperature at different deposition fluxes at $N_{add} = 3.47 \times 10^{14}$ atoms/cm². Blue circles: $F = 4.14 \times 10^{24}$ atoms/cm²s. Cyan left triangles: $F = 1.38 \times 10^{24}$ atoms/cm²s. Green diamonds: $F = 6.9 \times 10^{23}$ atoms/cm²s. Dark green squares: $F = 3.45 \times 10^{23}$ atoms/cm²s. Brown circles: $F = 1.38 \times 10^{23}$ atoms/cm²s. Orange diamonds: $F = 6.9 \times 10^{22}$ atoms/cm²s. Dark brown gradients: $F = 2.76 \times 10^{22}$ atoms/cm²s. Pink right triangles: $F = 1.38 \times 10^{22}$ atoms/cm²s. Red deltas: $F = 5 \times 10^{21}$ atoms/cm²s.

The evolution of Ge monomer density, n_1 , the total island density, n_x , and the total amount of Ge condensed on the surface, N_{cond} , as a function of N_{add} at 2100K are shown in Figure 5 for high ($F = 4.14 \times 10^{24}$ atoms/cm²s), medium ($F = 6.9 \times 10^{23}$ atoms/cm²s), and low ($F = 2.76 \times 10^{22}$ atoms/cm²s) deposition fluxes. There are several processes dictating the evolution of n_1 and n_x , including (1) monomer formation on the surface by deposition, (2) monomer desorption from the surface, and (3) island nucleation and growth as Ge atoms from the deposition flux impinge onto monomers or islands already present on the surface. During the initial stages of deposition when the surface coverage is very low, n_1 evolves according to a balance between monomer deposition and monomer desorption, and no islands are present at this point, i.e., $n_1 = N_{cond}$. As islands begin to nucleate by impingement of surface

monomers with arriving Ge atoms, n_1 begins to deviate from N_{cond} and quickly reaches a peak value, at which the island nucleation rate becomes the same as the rate of n_1 growth by the balance between deposition and desorption. As the rate of island nucleation subsides and the island density, n_x , reaches saturation, n_1 evolves largely independently of n_x , a consequence of the fact that island growth proceeds by direct impingement from the vapor, rather than by surface diffusion of monomers to the islands. In other words, n_1 depends only on the relative arrival and desorption rates and the fraction of the surface area that is not occupied by islands. Finally, for all fluxes considered here, the island density reaches a saturation value, at which point no new islands are formed and existing islands are growing (and therefore increasing N_{cond}).

Figure 5. Island distribution evolution as a function of N_{add} (and therefore implicitly as a function of time) during deposition at 2100K at deposition fluxes of (a) $F = 4.14 \times 10^{24}$ atoms/cm²s, (b) $F = 6.9 \times 10^{23}$ atoms/cm²s, and (c) $F = 2.76 \times 10^{22}$ atoms/cm²s. Data shown includes the total density of Ge condensed on the surface (N_{cond} - orange squares), the density of surface monomers (n_1 - purple diamonds) and the total density of stable clusters (n_x - green circles).

Shown in Figure 6 is the evolution of the *integral condensation coefficient*, $\alpha \equiv N_{cond}/N_{add}$, as a function of the number of deposited atoms per unit area at 2200K and various deposition fluxes [65, 66]. Note that the number of deposited atoms is linearly related to deposition time but by a different factor at each flux. For times shorter than the desorption timescale, the integral condensation coefficient is expected to be roughly constant at a value that is dictated by the fraction of surface that is binding. For larger times, desorption from binding sites acts to lower α as seen in Figure 6. Finally, as the island coverage fraction increases, α reaches a minimum (e.g., at $N_{add} \sim 5 \times 10^{14} \text{ cm}^{-2}$ for the highest three fluxes considered in Figure 6) and eventually begins to rise because adsorbed Ge islands always represent favorable binding environments for incoming Ge atoms. The impact of deposition flux is strong across all three regimes. Higher fluxes lead to higher initial α and a stronger impact by island coverage at later times.

Figure 6. Integral condensation coefficient, α , as a function of the number of Ge added to the system per unit area, N_{add} , at 2200K and different fluxes. Green diamonds: $F = 6.9 \times 10^{23} \text{ atoms/cm}^2\text{s}$, dark green squares: $F = 3.45 \times 10^{23} \text{ atoms/cm}^2\text{s}$, brown circles: $F = 1.38 \times 10^{23} \text{ atoms/cm}^2\text{s}$, dark brown gradients: $F = 2.76 \times 10^{22} \text{ atoms/cm}^2\text{s}$, red deltas: $F = 5 \times 10^{21} \text{ atoms/cm}^2\text{s}$.

The integral condensation coefficient provides information about the deposition regime; complete condensation corresponds to $\alpha = 1$, while extremely incomplete condensation implies that $\alpha \ll 1$. Leonhardt *et al.* [19] concluded that Ge deposition on a-SiO₂ occurs in the extremely incomplete condensation regime. The values of α in Figure 6, particularly at very early time and very high fluxes, are as high as 0.4, which is not necessarily indicative of extremely incomplete condensation. To further characterize the deposition regime in the simulations, we compared the distribution of inter-island distances, X_{island} , with the adatom diffusion distance, X_a , at different deposition fluxes. Inter-island distances were computed on the basis of island centers-of-mass. Islands were approximated to be circular in the xy -plane projection and the radius of each island circle was taken to be the distance between the center-of-mass and the furthest atom in the island. The distance between two islands was then calculated as the xy -projected distance between the edges of the two circles. Islands were defined as “neighbors” if the line connecting their centers did not cross any areas projected by other islands. Since many Ge islands on the surface deviate strongly from circularity, the circular approximation represents an upper-bound for island radius and the approximated inter-island distances represent lower-bounds.

Shown in Figure 7(a) is the mean inter-island distance, $\bar{X}_{island}$, as a function of N_{add} for the same three deposition fluxes considered in Figure 5. The value of $\bar{X}_{island}$ decreases as islands nucleate on the surface, then levels off when n_x reaches saturation density. The distance between islands decreases with increasing deposition flux due to the increasing island density, with the lowest value on the order of 10 Å. Taking into account the adatom displacement distribution shown previously in Figure 2 and the probability distributions of X_{island} at the saturation island density found for each deposition flux [Figure 7(b)], the probability of island growth due to adatom migration on the surface is below 1% at high deposition flux, and less than 0.1% for the medium and low flux cases. Because the inter-island distances used here represent lower-bound estimates for the actual island separations on the surface, these probabilities represent upper-bounds for the actual probability of island growth by surface adatom capture.

The low probability of island growth due to surface capture confirms that the system is in the extremely incomplete condensation regime at all deposition fluxes considered in our simulations. Moreover, since adatom displacement does not change significantly with temperature (at least over the range considered here), the condensation regime is confirmed to remain extremely incomplete at all temperatures sampled in this work.

Figure 7. (a) Mean inter-island distance as a function of Ge atoms brought down to the surface per unit area at different deposition fluxes and 2100K. Dark brown gradients: $F = 2.76 \times 10^{22}$ atoms/cm²s, green diamonds: $F = 6.9 \times 10^{23}$ atoms/cm²s, blue circles: $F = 4.14 \times 10^{24}$ atoms/cm²s. (b) Probability distributions of the inter-island distance when n_x reaches the saturation island density for the three cases shown in (a).

Dark brown gradients: $F = 2.76 \times 10^{22}$ atoms/cm²s at $N_{add} = 9 \times 10^{14}$ atoms/cm², green diamonds: $F = 6.9 \times 10^{23}$ atoms/cm²s at $N_{add} = 6 \times 10^{14}$ atoms/cm², blue circles: $F = 4.14 \times 10^{24}$ atoms/cm²s at $N_{add} = 7 \times 10^{14}$ atoms/cm².

4.3 Scaling Analysis and Quantitative Comparison with Experiment

While the experimentally observed condensation regime is qualitatively reproduced in simulations, quantitative measures of the deposition system are needed to make more meaningful connections between the computational model and the experiments. As mentioned previously in Section 2, the saturation island density, $n_{x,max}$, may be directly measured in experiments. Recall that in Eq. (8), $n_{x,max}$ was found to scale with temperature and the deposition flux according to

$$n_{x,max} \sim (F)^{\frac{2i}{3}} \exp\left(\frac{E}{k_B T}\right),$$

where $E = \frac{2}{3}(E_i + (i+1)E_a - E_d)$ in the extremely incomplete condensation regime. Thus, by measuring the saturation island density as a function of temperature, the slope, E/k_B , may be used to deduce the energy barriers relevant for island nucleation. For example, in Ref. [19], saturation island density measurements were used to compute the diffusion activation energy for Ge on a-SiO₂.

Shown in Figure 8 are Arrhenius plots of saturation island density versus temperature at several different fluxes ranging from 5×10^{21} atoms/cm²s to 6.9×10^{24} atoms/cm²s. The slope, E/k_B , changes abruptly at 2000K for the three deposition fluxes at which a wide range of temperatures were considered (see Figure 8). The change of slope separating the saturation island density into “high” and “low” temperature regimes is in excellent qualitative agreement with similar experimental observations reported by Leonhardt et al. [19]. Establishing a quantitative connection between the simulation and experimental temperature scales is, however, somewhat challenging. For instance, the Tersoff potential is known to

significantly overestimate the melting temperatures of Si and Ge ($T_m \sim 2550\text{K}$ for both), and the usual approach is to introduce a scaling factor, e.g., $T_{m,\text{sim}}/T_{m,\text{exp}}$, when comparing simulation and experimental results. However, in the present case it is not clear whether the melting temperature of Si or Ge, or the glass transition temperature of a-SiO₂ should be used to establish a scaling factor for the temperature. Given these ambiguities, we refrain from making a direct comparison to the experimental transition temperature reported in ref. [19], which was around 773K.

Figure 8. Saturation island density, $n_{x,\text{max}}$, as a function of inverse temperature. Purple diamonds: $F = 6.90 \times 10^{24}$ atoms/cm²s. Blue circles: $F = 4.14 \times 10^{24}$ atoms/cm²s. Cyan left triangles: $F = 1.38 \times 10^{24}$ atoms/cm²s. Dark green squares: $F = 3.45 \times 10^{23}$ atoms/cm²s. Orange diamonds: $F = 6.9 \times 10^{22}$ atoms/cm²s. Dark brown gradients: $F = 2.76 \times 10^{22}$ atoms/cm²s. Pink right triangles: $F = 1.38 \times 10^{22}$ atoms/cm²s. Red deltas: $F = 5 \times 10^{21}$ atoms/cm²s. Solid lines are linear regression fits in the high temperature regime; dashed lines are linear regression fits in the low temperature regime.

Although the relationship between the Tersoff and experimental temperature scales is unclear, the temperature *dependence* of the saturation island density may be analyzed more directly. It is immediately obvious that the slopes extracted from both the high and low temperature regimes in Figure 8 are strong

functions of deposition flux. In the low temperature regime, E/k_B increases from $\sim 1600 \pm 100\text{K}$ at 1.38×10^{24} atoms/cm²s to $6100 \pm 900\text{K}$ at 6.9×10^{22} atom/cm²s, while at high temperature E/k_B rises from $12800 \pm 900\text{K}$ at 1.38×10^{24} atoms/cm²s to $34000 \pm 6000\text{K}$ at 6.9×10^{22} atom/cm²s. Here and in Figure 9, the error bars represent the standard error of the linear regression slope. On the other hand, the analysis in ref. [19] suggests that these values are independent of the deposition flux, and are about 5030K and 40400K in the low and high temperature regimes, respectively.

In the following, we focus principally on the high temperature regime ($T \geq 2000\text{K}$), for which we have data over a much larger range of fluxes. Shown in Figure 9 is a plot of E/k_B as a function of flux for the high temperature regime. E/k_B rises monotonically as the deposition flux decreases, eventually plateauing at a value that is tantalizingly close to the experimentally reported value (horizontal line); the onset of the plateau appears around a deposition flux of 7×10^{22} atoms/cm²s. While further decreases in deposition flux would have been useful to consider, the slowest deposition run corresponded to introducing one Ge atom every 34.5 ps, and required an MD simulation of 173 ns for a system containing 10^5 atoms, corresponding approximately to 48,000 CPU hours on a modern supercomputer.

The existence of a plateau in E/k_B for $F \leq O(10^{22})$ atoms/cm²s, along with the excellent prediction of the final value, strongly indicates that for these fluxes, the system has reached a “slow deposition” regime that extends all the way to experimentally realistic fluxes. In other words, for fluxes below $O(10^{22})$ atoms/cm²s, the deposition timescale is slower than other important timescales that govern island rearrangement (for example) or desorption. Recall that surface diffusion does not appear to be relevant for this system due to the presence of extremely incomplete condensation conditions and therefore the timescales associated with inter-island diffusion are not likely to be important.

Bearing the above considerations in mind, and assuming that monomer equilibration on the surface is the rate limiting process that competes with the deposition rate, we may then estimate the maximum flux at which the system is still in the “slow deposition” regime. Recalling that the average

residence time (between adsorption and desorption) for single Ge monomers was found to be on the order of 10 ps, and requiring that the deposition rate be lower than the desorption rate, gives a maximum desorption flux that is $O(10^{22})$ atoms/cm²s for the 24 nm by 24 nm a-SiO₂ surface used in this study. This is in excellent agreement with the flux at which the plateau is observed in Figure 9!

Figure 9. Arrhenius slope for saturation island density in the high temperature regime as a function of deposition flux. The solid line represents the (flux independent) value obtained in experiments by Leonhardt *et al.* [19].

Finally, we return to the issue of critical island size, which has been assumed to be smaller than 2 throughout the preceding analysis; i.e., all islands, including dimers, have been included in the evaluation of n_x . According to Eq. (8), which applies when the deposition is in the extremely incomplete condensation regime, the saturation island density is related to the deposition flux as $n_{x,\max} \sim F^{2i/3}$. Shown in Figure 10 are log-log plots of the saturation island density as a function of deposition flux at several different temperatures – the slopes of these curves therefore correspond to the quantity $2i/3$. As expected, the slopes, and therefore the critical sizes, are not constant across the entire flux range. However, we do expect that the slope for each temperature reaches a constant value once the slow deposition regime is attained over the last 3 or 4 data points – although this is difficult to ascertain given

the scatter in the data and the minimum flux that can be accessed. Nonetheless, the slope across the 3-4 points that correspond to the slowest deposition fluxes at each temperature corresponds to critical sizes in the range 1-2. Importantly, for higher deposition fluxes or at lower temperatures, the critical size estimate is even lower. Collectively, these results self-consistently validate the assumption that the critical size is less than 2. The critical size was also estimated in the experimental analysis in ref. [19] and found to be in the range 2-3 in the “high” temperature regime (and 1-2 in the “low” temperature regime), again in excellent agreement with the present simulation results.

Figure 10. Log-log plot of saturation island density versus deposition flux at different temperatures. Lines represent linear regression fits. Red squares: 2000K. Green circles: 2050K. Blue diamonds: 2100K. Orange deltas: 2150K.

5. CONCLUSIONS

The nucleation of Ge islands on amorphous SiO₂ during Ge deposition was investigated with direct MD simulations based on a Tersoff-type empirical interatomic potential model parameterized for the ternary Ge-Si-O system. In accord with prior experimental analysis, the deposition physics, which we probed across a wide range of fluxes and temperatures, were found to be described well by the so-called

extremely incomplete condensation regime whereby island growth proceeds by direct impingement from the vapor phase rather than diffusional transport on the surface. These conditions exist because of the nature of the a-SiO₂ surface, which we have previously shown [20] to present a spatially heterogeneous binding energy distribution to Ge atoms. More specifically, the surface exhibits localized binding sites of varying strengths isolated from each other by regions of unfavorable binding, thereby leading to very limited Ge monomer mobility on the a-SiO₂ surface and a rather low average barrier for desorption.

Two major conclusions may be drawn from our study. First, and perhaps most importantly, we provide evidence that it is possible to simulate atomic deposition, using straightforward, direct MD simulations, at experimentally meaningful conditions. The timescale restrictions associated with MD simulations of atomic deposition are well-established in the literature and typically require that deposition rates be many orders-of-magnitude higher than experimentally realistic rates. However, by sampling deposition fluxes over a wide enough range (nearly 3 orders-of-magnitude), we found that it is possible to (just) reach a regime where key measures of the deposition process no longer depend on the flux, enabling us to make detailed, quantitative comparisons to experimental measurements. Of course, this may not always be possible in all materials systems, but it does provide strong evidence that realistic deposition rates do not necessarily have to be achieved in atomistic simulations in order to make quantitative predictions.

Second, and somewhat surprisingly given the complexity of the ternary Si-Ge-O system, we find that predictions of various measures, namely the critical island size and the quantitative temperature dependence, are in excellent agreement with values obtained from experiments. The connection between the simulations and experimental observations of much larger islands formed under much slower deposition conditions was established using a rate equation-based scaling analysis. The rate equation framework was first used to identify the deposition regime under which the experiments were performed, and then to systematically steer the simulations towards it by altering the temperature and deposition flux. Once this was established, the rate equation framework was used to identify quantities that could be compared quantitatively – even though the simulation length, time, and temperature scales are all

different than the experimental ones. Collectively, our findings confirm that the empirical potential model for the Si-Ge-O system used here and in our previous study is well-suited for the atomistic study of Ge-on-Si selective epitaxial growth and related systems.

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