# Computation of Strained Epitaxial Growth in Three Dimensions by Kinetic Monte Carlo 

Giovanni Russo* Peter Smereka ${ }^{\dagger}$

November 2, 2018


#### Abstract

A numerical method for computation of heteroepitaxial growth in the presence of strain is presented. The model used is based on a solid-on-solid model with a cubic lattice. Elastic effects are incorporated using a ball and spring type model. The growing film is evolved using Kinetic Monte Carlo (KMC) and it is assumed that the film is in mechanical equilibrium. The strain field in the substrate is computed by an exact solution which is efficiently evaluated using the fast Fourier transform. The strain field in the growing film is computed directly. The resulting coupled system is solved iteratively using the conjugate gradient method. Finally we introduce various approximations in the implementation of KMC to improve the computation speed. Numerical results show that layer-by-layer growth is unstable if the misfit is large enough resulting in the formation of three dimensional islands.


## 1 Introduction

Epitaxial growth is the process where crystals are grown by the deposition of atoms in a vacuum. Typically the deposition rate is small and the crystal is grown, loosely speaking, one layer at a time. In this paper, we consider the computation of strained epitaxial growth when the strain arises because the natural lattice spacing of the substrate and the deposited material are different. This difference is called the mismatch. Heteroepitaxial growth is experimentally observed to grow in the following growth modes:

1. Frank-Van der Meer growth: crystal surface remains fairly flat, growth occurs in the layer-by-layer fashion.
2. Volmer-Weber growth: three dimensional islands form on the substrate without a wetting layer.
3. Stranski-Krastanov growth: the film grows in a layer-by- layer fashion for a few layers, and then Volmer-Weber growth begins. This results in three dimensional islands on top of a wetting layer.

[^0]

Figure 1: Germanium on Silicon- Due to elastic interaction. The bottom configuration has less energy than the top one

The type of growth mode depends on many parameters, an important one is the mismatch. In many cases, when the mismatch is high, one finds Volmer-Weber growth and when the mismatch is small layer-by-layer growth is observed. For intermediate values of the mismatch Stranski-Krastanov growth is often seen. For an overview see, for example, Ref. [21, 14].

In homoepitaxy, the effects of strain are usually very small and quite often ignored in the many models. In general, the morphology of a growing film by homoepitaxy is reasonably well understood. It is known that in some cases a homoepitaxially grown film can undergo an instability resulting in mound formation. Typically these phenomena are due to kinetic effects, for example a step-edge barrier [2, 6, 7, or enhanced edge diffusion [15] can cause mound formation.

However, when a species of atoms grows on a substrate of a different species, formation of 3D islands is observed in many situations. It is generally believed in many cases (for example for the growth of Germanium on Silicon) that this is a thermodynamical effect. In particular, the elastic energy stored in a strained flat interface is greater than when there are three dimensional islands. This is due to the fact that in the latter case the atoms have more opportunity to relax (see Figure (1). However, the surface energy of three dimensional islands is greater than that of a flat interface. This implies that the morphology of heteroepitaxially grown films is determined by the interplay between elastic energy which is a bulk effect and surface energy which arises from broken bonds.

### 1.1 Modeling Elastic Effects

Elastic effects in thin films can be studied with fully continuum models or Burton-Cabrera-Frank [5 type models that consider elastic effects between steps. In this paper, we shall consider a fully discrete model which is evolved in time using a kinetic Monte

Carlo method. Naturally, such an approach has the disadvantage of not being able to simulate on large length scales. However, it offers the advantage that nanoscale physical effects such as island shape fluctuations and nucleation are naturally incorporated. One of the first, if not the first, model in this direction is due to Orr et al. [13]. They accounted for the elastic interactions using a ball and spring model, which takes into account nearest neighbor and next nearest neighbor interactions. This was combined with a solid-on-solid type model which was then used with KMC to simulate a growing heteroepitaxial film in $1+1$ dimensions. If the misfit was below a critical value, the film grew in a layer-by-layer fashion. On the other hand, if the misfit was above the same critical value, then the film was observed to grow in the Volmer-Weber fashion. Later Lam, Lee and Sander [8] provided a more efficient implementation of this model, which allowed them to perform simulations using parameter values that were more physically reasonable and compute for larger domains. This work has been recently been extended to three dimensions [9].

Ratsch et al. [18] studied three dimensional heteroepitaxy, however they did not take explicitly into account the harmonic forces between atoms, but rather they used an approximate treatment [17] based on the Frenkel-Kontorova model. The model was used to investigate the island size distribution in heteroepitaxial growth [16].

Off lattice KMC simulations of heteroepitaxial growth in $1+1$ dimensions where presented in a series of papers [10, 3, 11. In these computations the forces between atoms were modelled using Lennard-Jones interactions. The misfit is easily incorporated by changing parameters in the potential. One advantage of this approach is that dislocations are naturally included, which is not the case with the ball and spring model. These simulations also demonstrate that if the misfit is sufficiently large, layer-by-layer growth is unstable and mounds form.

A more sophisticated discrete elastic model was introduced by Schindler et al. 20. This model is based on a discrete form of the continuum elasticity equations. The approach presented here could be used to solve their model as well.

## 2 Model description and Kinetic Monte Carlo

The model we shall use is a three dimensional version of the model proposed in Refs. [13, 8]. For the convenience of the reader we shall now describe this model. To fix ideas we shall assume that the deposited atoms are Germanium and the substrate is composed of Silicon. The atoms occupy sites arranged on a simple cubic lattice with no over hanging atoms allowed. This means that the height of the surface is a function of the substrate location. We assume that atoms bond with their nearest and next nearest neighbors. Each atom can be linked to its six nearest neighbors located at a distance $a$, and to its twelve next neighbors located at a distance $a \sqrt{2}$. For example, an atom on a flat plane surface orthogonal to one of the coordinate axis will have five bonds with nearest neighbors, and eight bonds with next nearest neighbors, while an atom sitting on top of that same flat surface will have five bonds (one with a nearest neighbor and four with next near neighbors). We shall assume the chemical energy associated to all these bonds is the same. The total chemical bond energy associated to each atom is therefore $E_{b}=-\gamma N_{b}$, where $N_{b}$ is the number of bonds of each atom, and $\gamma$ the energy associated to each bond.

The elastic effects in this model are taken into account by assuming that the bonds
will act like a spring between the atoms. We will use $a_{s}$ and $a_{g}$ to denote the lattice spacing between Silicon and Germanium atoms respectively. We shall denote respectively by $k_{L}$ and $k_{D}$ the spring constants corresponding to longitudinal (nearest neighbor) and diagonal (next nearest neighbor) bonds. For ease of exposition, we shall assume that both Silicon and Germanium have the same spring constants. Since $a_{g} \neq a_{s}$ mechanical, force will arises (the calculation of which is described in detail below). In many systems, the time taken for sound waves to propagate across the sample is much smaller than the time scales associated to any growth process. Therefore we assume that our mass-spring system is always in mechanical equilibrium.

Each atom, $p$, of the system will hop with a rate $\Gamma_{m}$ [] given by

$$
\begin{equation*}
\Gamma_{m}=R_{0} \exp \left(\frac{-\Delta E}{k_{B} T}\right) \tag{1}
\end{equation*}
$$

where

$$
\begin{equation*}
\Delta E=E(\text { without atom } p)-E(\text { with atom } p) \tag{2}
\end{equation*}
$$

is the change in energy of the entire system when atom $p$ is completely removed. $R_{0}$ is the attempt frequency, $k_{B}$ is the Boltzmann factor, and $T$ is the lattice temperature. Since the chemical bonds are purely local, then we can write (2) as

$$
\Delta E=n_{b} \gamma-\Delta E_{\text {elas }}
$$

where $n_{b}$ is the number of chemical bonds of the atom, $\gamma$ is the energy associated to the chemical bond, and

$$
\begin{equation*}
\Delta E_{\text {elas }}=E_{\text {elas }}(\text { with atom } p)-E_{\text {elas }}(\text { without atom } p) \tag{3}
\end{equation*}
$$

We note that $\Delta E_{\text {elas }}$ is always nonnegative and when combined with (2) implies that elastic effects will always increase the hopping rate.

We shall evolve the model in time by the use of kinetic Monte Carlo (KMC). The basic KMC method can be described as follows.

1. Pick an atom at random on the surface, uniformly among all surface atoms
2. Compute its number of bonds
3. Compute the contribution of the elastic energy associated to the atom
4. Pick a random number, $r$, uniformly distributed in $[0,1]$.
5. If $r<\exp \left(-\left(n_{b} \gamma-\Delta E_{\text {elas }}-K\right) / k_{B} T\right)$ then move the atom, according to a random direction, chosen uniformly among all possible directions

The constant $K$ is computed in such a way that the numerator $n_{b} \gamma-\Delta E_{\text {elas }}-K$ that appears in the exponential is always non negative. This sets the fastest rate in the problem to unity. The method described is based on rejection. It is well known that if the number of possible event types is small (as is the case for this model when there are no elastic effects) then rejection-free Kinetic Monte Carlo can provide a much more efficient algorithm.

While this model is idealized, it nevertheless captures the essential physicals effects of heteroepitaxial growth, such as adatom diffusion, nucleation, surface diffusion, long range elastic interaction. In addition, since the model is evolved in time using kinetic Monte Carlo, it naturally captures effects associated with fluctuations.


Figure 2: The reference configuration is obtained by compressing the Germanium atoms to have the same horizontal spacing as the Silicon atoms. The vertical spacing is chosen so that Germanium atoms are in equilibrium.

## 3 Elastic computations

The main difficulty in the implementation of this model is the computation of the strain field. In this section we shall outline our approach for solving this problem. For the basic set up we follow Lam et al. [8], as is described in next section. However, our numerical implementation is different from the one used in [8]. One important feature of our work is that we provide an exact solution for the elastic displacement in the substrate which is efficiently evaluated using fast Fourier transforms.

### 3.1 The reference configuration

The reference configuration we choose consists of a periodic array of complete layers of Germanium atoms on top of a periodic array of Silicon. The Germanium atoms are compressed so that their horizontal lattice spacing matches that of the Silicon atoms, see Figure 3. The vertical lattice spacing, $a_{L}$, is chosen so that the resulting system is in mechanical equilibrium. We will now describe the computation of $a_{L}$ in two dimensions. It is useful to introduce the following dimensionless quantity

$$
\epsilon=\frac{a_{g}-a_{s}}{a_{g}} .
$$

which is denoted as the mismatch. Typical values of $\epsilon$ range from -0.05 to 0.05 . For example the mismatch for Germanium on a Silicon substrate is 0.04 . In order to deduce the atom displacement with respect to the reference configuration we need to compute the forces experienced by an atom due to each of its neighbors. Elementary considerations allow to compute that, to first order in the ratio $\epsilon$, one has
$\vec{F}_{1}=F_{H}\binom{1}{0}, \vec{F}_{2}=F_{D V}\binom{1}{1}, \vec{F}_{3}=F_{V}\binom{0}{-1}, \vec{F}_{4}=F_{D V}\binom{-1}{1}, \vec{F}_{5}=-\vec{F}_{1}$,
where $F_{V}=k_{L}\left(a_{L}-a_{s}\right), F_{H}=k_{L}\left(a_{g}-a_{s}\right)$, and $F_{D V}=k_{D}\left(2 a_{g}-a_{L}-a_{s}\right) / 2$.
The value of $a_{L}$ is determined by requiring that these five forces sum to zero for atoms in the reference configuration. By symmetry, the forces in the $x$ direction sum to


Figure 3: The reference configuration is obtained by compressing the Germanium atoms to have the same horizontal spacing as the Silicon atoms. The vertical spacing is chosen so that complete layers of Germanium are in equilibrium. $\vec{b}$ is the net force on an atom due to the compression. In principle $\vec{b}$ can be nonzero for any Germanium or Silicon atoms in the top row, if the top layer is not complete
zero. On the other hand balancing the $z$ components of the force one has $2 F_{D V}=F_{V}$ which implies

$$
k_{D}\left(2 a_{g}-a_{L}-a_{s}\right)+k_{L}\left(a_{g}-a_{L}\right)=0,
$$

which gives the following expression for $a_{L}$

$$
a_{L}=a_{g}\left(1+\epsilon \frac{k_{D}}{k_{L}+k_{D}}\right) .
$$

A similar argument can be applied to the three dimensional lattice. In this case each atom can interact with six nearest neighbors, located at a distance $a$, and twelve diagonal next to nearest neighbors, located at distance $a \sqrt{2}$. The interaction with the 8 corner neighbors, located at a distance $a \sqrt{3}$, are neglected.

As in the two dimensional case, we shall denote by $k_{L}$ and $k_{D}$ the two spring constants corresponding to the interaction between nearest neighbors and diagonal neighbors. Each bulk atom is surrounded by 18 neighbors, (six longitudinal and twelve diagonal). We denote by $\vec{F}_{i j k}$ the contribution of the force on a given atom due to the presence of its neighbor in the direction $(i, j, k)$. For example, the 3D equivalent of force $\vec{F}_{3}$ of Figure 3 would be $\vec{F}_{0,0,-1}$.

The six forces aligned along the coordinate axis have the expression

$$
\vec{F}_{i j k}=\left(\begin{array}{c}
-i F_{H}  \tag{4}\\
-j F_{H} \\
k F_{V}
\end{array}\right), \quad \text { with } \quad i, j, k \in\{-1,0,1\}, \quad \text { where } \quad|i|+|j|+|k|=1 .
$$

$F_{V}$ and $F_{H}$ are given above. The twelve diagonal forces are given by

$$
\begin{align*}
& \vec{F}_{i 0 k}=-\left(\begin{array}{c}
i F_{D V} \\
0 \\
k F_{D V}
\end{array}\right), \quad \text { with } \quad i, k \in\{-1,1\},  \tag{5}\\
& \vec{F}_{0 j k}=-\left(\begin{array}{c}
0 \\
j F_{D V} \\
k F_{D V}
\end{array}\right), \quad \text { with } \quad i, k \in\{-1,1\}, \tag{6}
\end{align*}
$$

and

$$
\vec{F}_{i j 0}=-\left(\begin{array}{c}
i F_{D H}  \tag{7}\\
j F_{D H} \\
0
\end{array}\right), \quad \text { with } \quad i, j \in\{-1,1\}
$$

$F_{D V}$ is given above and $F_{D H}=k_{D}\left(a_{g}-a_{s}\right)$. It is convenient to set $\vec{F}_{i j k}=0$ if $|i|+|j|+|k|=3$.

As in the two dimensional case these forces must sum to zero in the reference configuration. The $x$ and $y$ components will vanish by symmetry. The forces in the $z$ direction vanish if

$$
F_{V}+4 F_{D V}=0
$$

which implies

$$
2 k_{D}\left(2 a_{g}-a_{L}-a_{s}\right)+k_{L}\left(a_{g}-a_{L}\right)=0 .
$$

This gives the following expression for $a_{L}$,

$$
\begin{equation*}
a_{L}=a_{g}\left(1+\epsilon \frac{2 k_{D}}{k_{L}+2 k_{D}}\right) . \tag{8}
\end{equation*}
$$

### 3.2 Computation of the interaction

Let us denote the displacement, with respect to the reference configuration, of an atom at site $(\ell, j, k)$ by the vector $\left(u_{\ell j k}, v_{\ell j k}, w_{\ell j k}\right)$ and the force experienced by this atom as $\left(f_{\ell j k}, g_{\ell j}, h_{\ell j k}\right)$. This force will arise from the interaction of the atom with its nearest neighbors and next nearest neighbors. For example the $x$ component of the force is given by

$$
\begin{aligned}
f_{\ell j k} & =k_{L}\left(\left[u_{\ell+1 j k}-u_{\ell j k}\right]+\left[u_{\ell-1 j k}-u_{\ell j k}\right]\right) \\
& +\frac{k_{D}}{2}\left(\left[u_{\ell+1 j k+1}-u_{\ell j k}\right]+\left[u_{\ell-1 j k+1}-u_{\ell j k}\right]\right) \\
& +\frac{k_{D}}{2}\left(\left[u_{\ell+1 j k-1}-u_{\ell j k}\right]+\left[u_{\ell-1 j k-1}-u_{\ell j k}\right]\right) \\
& +\frac{k_{D}}{2}\left(\left[u_{\ell+1 j+1 k}-u_{\ell j k}\right]+\left[u_{\ell-1 j+1 k}-u_{\ell j k}\right]\right) \\
& +\frac{k_{D}}{2}\left(\left[u_{\ell+1 j-1 k}-u_{\ell j k}\right]+\left[u_{\ell-1 j-1 k}-u_{\ell j k}\right]\right) \\
& +\frac{k_{D}}{2}\left(\left[v_{\ell+1 j+1 k}-v_{\ell j k}\right]+\left[v_{\ell-1 j-1 k}-v_{\ell j k}\right]\right) \\
& -\frac{k_{D}}{2}\left(\left[v_{\ell+1 j-1 k}-v_{\ell j k}\right]+\left[v_{\ell-1 j+1 k}-v_{\ell j k}\right]\right)
\end{aligned}
$$

$$
\begin{align*}
& +\frac{k_{D}}{2}\left(\left[w_{\ell+1 j k+1}-w_{\ell j k}\right]+\left[w_{\ell-1 j k-1}-w_{\ell j k}\right]\right) \\
& -\frac{k_{D}}{2}\left(\left[w_{\ell+1 j k-1}-w_{\ell j k}\right]+\left[w_{\ell-1 j k+1}-u_{\ell j k}\right]\right) \\
& +\sum_{(m, n, q) \in \operatorname{neigh}(\ell, j, k)} \vec{F}_{m n q} \cdot \mathbf{e}_{x} \tag{9}
\end{align*}
$$

where $\vec{F}_{m n q}$ are given by (4][7]. Each term in square brackets and each $\vec{F}_{m n q}$ represents the interaction of an atom at site $(\ell, j, k)$ with potential nearest and next nearest neighbors. If no such neighbor exists then the term should not be included.

Suppose we have $N$ atoms and we denote the relative displacement of the $p$ th atom by $\vec{u}_{p}$ and let $\vec{F}_{p}$ denote the force it experiences. We also let $\vec{b}_{p}$ denote the sum of all forces given by (4.7), acting on the atom when its position is the reference configuration. Next we define the following vectors in $\mathbb{R}^{3 N}, \mathbf{u}=\left(\vec{u}_{1}, \ldots, \vec{u}_{N}\right)^{T}, \mathbf{b}$ and $\mathbf{F}$ are similarly defined. Then we can write

$$
\mathbf{F}=\mathcal{A} \mathbf{u}+\mathbf{b}
$$

We remark that for atoms that are completely surrounded by other 18 atoms, or atoms that are on a horizontal surface, the corresponding $\vec{b}$ is zero, since all the forces acting on them sum up to zero, which is consistent to the fact that a rectangular box of atoms in the reference configuration is in equilibrium. As a consequence, the vector $\mathbf{b}$ has nonzero elements only for atoms at the surface. The matrix vector product, $\mathcal{A} \mathbf{u}$ can be deduced from (9) and similar relations for $g_{\ell j k}$ and $h_{\ell j k}$.

The equilibrium position of atoms in a given configuration is obtained by setting $\mathbf{F}=0$, i.e. by solving the large linear system

$$
\mathcal{A} \mathbf{u}+\mathbf{b}=0 .
$$

### 3.3 Contribution of the substrate

It is known that elastic interactions can be very long ranged. For example the elastic interaction between two island behaves like $d^{-2}$ where $d$ is the distance between the island centers [19. This indicates that elastic interaction can penetrate deep into the substrate. On the other hand, the interaction range is certainly much shorter than the thickness of the substrate. For this reason it is prudent to consider the substrate to be semi-infinite in the $z$-direction. To reduce boundary effects we consider periodic boundary conditions in both the $x$ and $y$ directions. In this section we shall derive a formula that expresses the force on the surface atoms of the substrate completely in terms of their displacement.

The surface of the substrate corresponds to $k=0$, and the atoms of the bulk substrate will be indexed using negative $k$ values. In the substrate ( $k \leq-1$ ) all atoms have a complete set of neighbors, consequently we can write the force, in component form, on the atom at site $(\ell, j, k), k \leq-1$, as

$$
\begin{aligned}
f_{\ell j k}= & k_{L}\left(u_{\ell+1 j k}-2 u_{\ell j k}+u_{\ell-1 j k}\right) \\
& +\frac{k_{D}}{2}\left(u_{\ell+1 j k+1}+u_{\ell-1 j k+1}+u_{\ell+1 j k-1}+u_{\ell-1 j k-1}\right. \\
& \left.+u_{\ell+1 j+1 k}+u_{\ell-1 j+1 k}+u_{\ell+1 j-1 k}+u_{\ell-1 j-1 k}-8 u_{\ell j k}\right)
\end{aligned}
$$

$$
\begin{align*}
& +\frac{k_{D}}{2}\left(v_{\ell+1 j+1 k}+v_{\ell-1 j-1 k}-v_{\ell+1 j-1 k}-v_{\ell-1 j+1 k}\right) \\
& +\frac{k_{D}}{2}\left(w_{\ell+1 j k+1}+w_{\ell-1 j k-1}-w_{\ell+1 j k-1}-w_{\ell-1 j k+1}\right) \tag{10}
\end{align*}
$$

$$
\begin{align*}
g_{\ell j k}= & k_{L}\left(v_{\ell j+1 k}-2 v_{\ell j k}+v_{\ell j-1 k}\right) \\
& +\frac{k_{D}}{2}\left(v_{\ell j+1 k+1}+v_{\ell j-1 k+1}+v_{\ell j+1 k-1}+v_{\ell j-1 k-1}\right. \\
& \left.+v_{\ell+1 j+1 k}+v_{\ell-1 j+1 k}+v_{\ell+1 j-1 k}+v_{\ell-1 j-1 k}-8 v_{\ell j k}\right) \\
& +\frac{k_{D}}{2}\left(u_{\ell+1 j+1 k}+u_{\ell-1 j-1 k}-u_{\ell+1 j-1 k}-u_{\ell-1 j+1 k}\right) \\
& +\frac{k_{D}}{2}\left(w_{\ell j+1 k+1}+w_{\ell j-1 k-1}-w_{\ell j+1 k-1}-w_{\ell j-1 k+1}\right),  \tag{11}\\
h_{\ell j k}= & k_{L}\left(w_{\ell j k+1}-2 w_{\ell j k}+w_{\ell j k-1}\right) \\
& +\frac{k_{D}}{2}\left(w_{\ell j+1 k+1}+w_{\ell j+1 k-1}+w_{\ell j-1 k+1}+w_{\ell j-1 k-1}\right. \\
& \left.+w_{\ell+1 j k+1}+w_{\ell-1 j k+1}+w_{\ell+1 j k-1}+w_{\ell-1 j k-1}-8 w_{\ell j k}\right) \\
& +\frac{k_{D}}{2}\left(u_{\ell+1 j k+1}+u_{\ell-1 j k-1}-u_{\ell+1 j k-1}-u_{\ell-1 j k+1}\right) \\
& +\frac{k_{D}}{2}\left(v_{\ell j+1 k+1}+v_{\ell j-1 k-1}-v_{\ell j+1 k-1}-v_{\ell j-1 k+1}\right) . \tag{12}
\end{align*}
$$

At the surface of the substrate $(k=0)$, one has a slightly different expression, because there are no atoms on top, namely:

$$
\begin{align*}
f_{\ell j 0}= & k_{L}\left(u_{\ell+1, j, 0}-2 u_{\ell, j, 0}+u_{\ell-1, j, 0}\right) \\
& +\frac{k_{D}}{2}\left(u_{\ell+1, j,-1}+u_{\ell-1, j,-1}+u_{\ell+1, j+1,0}+u_{\ell-1, j+1,0}\right. \\
& \left.+u_{\ell+1, j-1,0}+u_{\ell-1, j-1,0}-6 u_{\ell, j, 0}\right) \\
& +\frac{k_{D}}{2}\left(v_{\ell+1, j+1,0}+v_{\ell-1, j-1,0}-v_{\ell+1, j-1,0}-v_{\ell-1, j+1,0}\right) \\
& +\frac{k_{D}}{2}\left(w_{\ell-1, j,-1}-w_{\ell+1, j,-1}\right)  \tag{13}\\
g_{\ell j 0}= & k_{L}\left(v_{\ell, j+1,0}-2 v_{\ell, j, 0}+v_{\ell, j-1,0}\right) \\
& +\frac{k_{D}}{2}\left(v_{\ell+1, j+1,0}+v_{\ell-1, j+1,0}+v_{\ell+1, j-1,0}+v_{\ell-1, j-1,0}\right. \\
& \left.+v_{\ell, j+1,-1}+v_{\ell, j-1,-1}-6 v_{\ell, j, 0}\right) \\
& +\frac{k_{D}}{2}\left(u_{\ell+1, j+1,0}+u_{\ell-1, j-1,0}-u_{\ell+1, j-1,0}-u_{\ell-1, j+1,0}\right) \\
& +\frac{k_{D}}{2}\left(w_{\ell, j-1,-1}-w_{\ell, j+1,-1}\right)  \tag{14}\\
h_{\ell j 0}= & k_{L}\left(w_{\ell, j,-1}-w_{\ell, j, 0}\right) \\
& +\frac{k_{D}}{2}\left(w_{\ell+1, j,-1}+w_{\ell-1, j,-1}+w_{\ell, j+1,-1}+w_{\ell, j-1,-1}-4 w_{\ell, j, 0}\right)
\end{align*}
$$

$$
\begin{align*}
& +\frac{k_{D}}{2}\left(u_{\ell-1, j,-1}-u_{\ell+1, j,-1}\right) \\
& +\frac{k_{D}}{2}\left(v_{\ell, j-1,-1}-v_{\ell, j+1,-1}\right) \tag{15}
\end{align*}
$$

Let us now consider a Fourier expansion of the displacement in the $x$ and $y$ direction. The generic Fourier mode will take the form

$$
\begin{align*}
u_{\ell j k} & =\hat{u}_{k}(\xi, \eta) e^{i(\ell \xi+j \eta)} \\
v_{\ell j k} & =\hat{v}_{k}(\xi, \eta) e^{i(\ell \xi+j \eta)}  \tag{16}\\
w_{\ell j k} & =\hat{w}_{k}(\xi, \eta) e^{i(\ell \xi+j \eta)} .
\end{align*}
$$

By inserting this Fourier expansion in the expression of the surface force (13 (15) one obtains the relations

$$
\begin{align*}
\hat{f}_{0}= & 2 k_{L} \hat{u}_{0}(\cos \xi-1)+k_{D}\left[\hat{u}_{-1} \cos \xi+\hat{u}_{0}(2 \cos \xi \cos \eta-3)\right. \\
& \left.-2 \hat{v}_{0} \sin \xi \sin \eta-i \hat{w}_{-1} \sin \xi\right], \\
\hat{g}_{0}= & 2 k_{L} \hat{v}_{0}(\cos \eta-1)+k_{D}\left[\hat{v}_{-1} \cos \eta+\hat{v}_{0}(2 \cos \xi \cos \eta-3)\right. \\
& \left.-2 \hat{u}_{0} \sin \xi \sin \eta-i \hat{w}_{-1} \sin \eta\right],  \tag{17}\\
\hat{h}_{0}= & k_{L}\left(\hat{w}_{-1}-\hat{w}_{0}\right)+k_{D}\left[\hat{w}_{-1}(\cos \xi+\cos \eta)-2 \hat{w}_{0}\right] \\
& \left.-i\left(\hat{u}_{-1} \sin \xi+\hat{v}_{-1} \sin \eta\right)\right] .
\end{align*}
$$

In the relation above we have suppressed the dependence of all Fourier modes on $(\xi, \eta)$.
Eq.(17) gives a relation between the Fourier modes of the force and the Fourier modes of the displacement. However, our goal is to express $\left(\hat{f}_{0}, \hat{g}_{0}, \hat{h}_{0}\right)$ in terms of $\left(\hat{u}_{0}, \hat{v}_{0}, \hat{w}_{0}\right)$. Once this is done then the force field at the surface can be computed from its Fourier modes by inverse discrete Fourier transform. In order to accomplish our goal, we need to express $\hat{u}_{-1}, \hat{v}_{-1}, \hat{w}_{-1}$ in terms of $\hat{u}_{0}, \hat{v}_{0}, \hat{w}_{0}$, and substitute their expression into (17). This can be done as follows. First, let us insert the Fourier expansion (16) into (10 12) obtaining:

$$
\begin{align*}
\hat{f}_{k}= & 2 k_{L} \hat{u}_{k}(\cos \xi-1)+k_{D}\left[\left(\hat{u}_{k+1}+\hat{u}_{k-1}\right) \cos \xi+\hat{u}_{k}(2 \cos \eta \cos \xi-4)\right] \\
& +i k_{D}\left(\hat{w}_{k+1}-\hat{w}_{k-1}\right) \sin \xi-2 k_{D} \hat{v}_{k} \sin \xi \sin \eta \\
\hat{g}_{k}= & 2 k_{L} \hat{v}_{k}(\cos \eta-1)+k_{D}\left[\left(\hat{v}_{k+1}+\hat{v}_{k-1}\right) \cos \eta+\hat{v}_{k}(2 \cos \eta \cos \xi-4)\right]  \tag{18}\\
& +i k_{D}\left(\hat{w}_{k+1}-\hat{w}_{k-1}\right) \sin \eta-2 k_{D} \hat{u}_{k} \sin \xi \sin \eta \\
\hat{h}_{k}= & k_{L}\left(\hat{w}_{k+1}-2 \hat{w}_{k}+\hat{w}_{k-1}\right) \\
& +k_{D}\left[\left(\hat{w}_{k+1}+\hat{w}_{k-1}\right)(\cos \xi+\cos \eta)-4 \hat{w}_{k}\right] \\
& +i k_{D}\left[\left(\hat{u}_{k+1}-\hat{u}_{k-1}\right) \sin \xi+\left(\hat{v}_{k+1}-\hat{v}_{k-1}\right) \sin \eta\right]
\end{align*}
$$

The discrete equations given by (18) are solved using the following substitution

$$
\begin{equation*}
\hat{u}_{k}=\hat{u} \alpha^{k}, \quad \hat{v}_{k}=\hat{v} \alpha^{k}, \quad \hat{w}_{k}=\quad \hat{w} \alpha^{k} \tag{19}
\end{equation*}
$$

where we look for solutions with $|\alpha|>1$, since we expect the Fourier modes to decay as $k \rightarrow-\infty$. Inserting this ansatz into the expression (18) of the Fourier modes of the force acting on the inner points of the substrate, one obtains:

$$
\left(\begin{array}{c}
\hat{f}  \tag{20}\\
\hat{g} \\
\hat{h}
\end{array}\right)=\Omega(\alpha)\left(\begin{array}{c}
\hat{u} \\
\hat{v} \\
\hat{w}
\end{array}\right)
$$

where the entries of the matrix $\Omega$ are given by

$$
\begin{aligned}
\omega_{11} & =2 k_{L}(\cos \xi-1) \alpha+k_{D}\left[\cos \xi\left(1+\alpha^{2}\right)+2(\cos \eta \cos \xi-2) \alpha\right] \\
\omega_{22} & =2 k_{L}(\cos \eta-1) \alpha+k_{D}\left[\cos \eta\left(1+\alpha^{2}\right)+2(\cos \eta \cos \xi-2) \alpha\right] \\
\omega_{33} & =k_{L}\left(\alpha^{2}-2 \alpha+1\right)+k_{D}\left[\left(\alpha^{2}+1\right)(\cos \xi+\cos \eta)-4\right] \\
\omega_{12} & =\omega_{21}=-2 \alpha k_{D} \sin \xi \sin \eta \\
\omega_{13} & =\omega_{31}=i k_{D}\left(\alpha^{2}-1\right) \sin \xi \\
\omega_{23} & =\omega_{32}=i k_{D}\left(\alpha^{2}-1\right) \sin \eta
\end{aligned}
$$

Note that matrix $\Omega$ is symmetric, but not self-adjoint.
Since all forces in the bulk have to be zero (all such atoms are in mechanical equilibrium), then one has

$$
\Omega\left(\begin{array}{c}
\hat{u}  \tag{21}\\
\hat{v} \\
\hat{w}
\end{array}\right)=0
$$

This homogeneous system has nontrivial solutions only if

$$
\begin{equation*}
P(\alpha) \equiv \operatorname{det}(\Omega)=0 \tag{22}
\end{equation*}
$$

This relation results in an algebraic equation for the values of $\alpha$. The polynomial $P(\alpha)$ is a six degree polynomial, therefore it admits, in general, six roots. Note that, because of the structure of the matrix $\Omega$, matrix $\alpha^{2} \Omega(1 / \alpha)$ is equal to $\Omega(\alpha)$ with $\omega_{13}=\omega_{31}$ and $\omega_{23}=\omega_{32}$ of opposite sign. This does not change the expression of the determinant, and therefore if $\tilde{\alpha} \neq 0$ is a root, then also $1 / \tilde{\alpha}$ is a root. This means that the number of roots $\tilde{\alpha}$ such that $|\tilde{\alpha}|>1$ is equal to the number of (nonzero) roots $\tilde{\alpha}$ such that $|\tilde{\alpha}|<1$. The roots that are of interest for us are the ones that decay as $k \rightarrow \infty$, i.e. $\tilde{\alpha}$ $:|\tilde{\alpha}|>1$.

### 3.3.1 Eigenvector computation

In this subsection we describe a general procedure for the computation of the eigenvalues and eigenvectors, that works also in the case of multiple eigenvalues.

The goal is to solve the problem given by (21), which we write as

$$
\begin{equation*}
\Omega r=0 \tag{23}
\end{equation*}
$$

where $r$ is a three-component vector (we drop the arrow on top), and to find independent eigenvectors even if some eigenvalues coincide. First compute the eigenvalues by solving the algebraic equation (22). Consider the three eigenvalues $\alpha_{\ell}$ such that
$\left|\alpha_{\ell}\right|>1, \ell=1, \ldots, 3$. If they are all distinct, then the three eigenvectors corresponding to them will be independent. If two of them are coincident, let us say $\alpha_{2}=\alpha_{3}$, then one has to find two independent eigenvectors corresponding to the coincident eigenvalues.

A unified treatment of the problem is obtained by the use of the singular value decomposition (SVD) of matrix $\Omega$. The procedure works as follows. First compute $\alpha_{1}, \alpha_{2}, \alpha_{3}$. If they are distinct, for each of them compute $\Omega_{\ell}=\Omega\left(\alpha_{\ell}\right), \ell=1, \ldots, 3$. Perform the SVD of $\Omega_{\ell}: \Omega_{\ell}=U \Sigma V^{\dagger}$, where $U$ and $V$ are unitary matrices (i.e. $U U^{\dagger}=I, V V^{\dagger}=I$ ), and $\Sigma$ is a diagonal matrix containing the singular values of $\Omega_{\ell}$. Taking into account that $U$ is non singular, problem (23) reads

$$
\Sigma V^{\dagger} r=0
$$

Since $\Omega_{\ell}$ is singular, then $\Sigma=\operatorname{diag}\left(\sigma_{1}, \sigma_{2}, 0\right)$, therefore one has

$$
\sigma_{1}\left(V^{\dagger} r\right)_{1}=0, \quad \sigma_{2}\left(V^{\dagger} r\right)_{2}=0, \quad\left(V^{\dagger} r\right)_{3}=\text { arbitrary }
$$

Let us choose $\left(V^{\dagger} r\right)_{3}=1$.
Assuming $\sigma_{2} \neq 0$, i.e. that the matrix $\Sigma_{\ell}$ has rank 2, then one has

$$
V^{\dagger} r=\left(\begin{array}{l}
0 \\
0 \\
1
\end{array}\right)
$$

therefore

$$
r=V\left(\begin{array}{l}
0 \\
0 \\
1
\end{array}\right)
$$

i.e. $r$ is the third column of $V$.

If two roots are coincident, say $\alpha_{1}, \alpha_{2}=\alpha_{3}$, then first compute the eigenvector $r_{1}$ using the procedure above applied to matrix $\Omega\left(\alpha_{1}\right)$. For the computation of the other eigenvectors there are two possibilities: either the rank of the matrix $\Omega_{2}=\Omega_{3}$ is 1 , i.e. $\quad \sigma_{2}=0$, or the rank of the matrix is 2 , i.e. $\sigma_{2} \neq 0$. However, the latter case never happened in all our computations, and we conjecture it can never happen for our problem. Therefore we assume that $\sigma_{2}=0$. Repeating the procedure above, one finds that $r_{2}$ and $r_{3}$ can be computed, respectively, as the second and third column of the matrix $V$.

### 3.3.2 Surface Force Formula

We denote by $\vec{r}_{\ell}$ a solution of the system

$$
\Omega\left(\alpha_{\ell}\right) \vec{r}=0,
$$

with $\ell=1,2,3$. Then we can decompose the vector $\left(\hat{u}_{0}, \hat{v}_{0}, \hat{w}_{0}\right)^{T}$ on the basis of the eigenvectors, i.e.

$$
\left(\begin{array}{c}
\hat{u}_{0} \\
\hat{v}_{0} \\
\hat{w}_{0}
\end{array}\right)=c_{1} \vec{r}_{1}+c_{2} \vec{r}_{2}+c_{3} \vec{r}_{3} .
$$

Once the constants $c_{1}, c_{2}, c_{3}$ are computed, one can write

$$
\left(\begin{array}{c}
\hat{u}_{-1} \\
\hat{v}_{-1} \\
\hat{w}_{-1}
\end{array}\right)=\frac{c_{1}}{\alpha_{1}} \vec{r}_{1}+\frac{c_{2}}{\alpha_{2}} \vec{r}_{2}+\frac{c_{3}}{\alpha_{3}} \vec{r}_{3} .
$$

The two relations allow to express $\hat{u}_{-1}, \hat{v}_{-1}, \hat{w}_{-1}$ in terms of $\hat{u}_{0}, \hat{v}_{0}, \hat{w}_{0}$. Once this is done, one can substitute this expression into (17) and obtain the final relation between $\left(\hat{f}_{0}, \hat{g}_{0}, \hat{h}_{0}\right)$ and $\left(\hat{u}_{0}, \hat{v}_{0}, \hat{w}_{0}\right)$. This relation has to be computed for all Fourier modes $(\xi, \eta)$. Periodicity implies that $\xi=2 \pi m / M, \eta=2 \pi n / M, m, n=1, \ldots, M$.

The complete algorithm for the computation of $\vec{f}_{\ell j 0}$ from $\vec{u}_{\ell j 0}$ can be summarized as follows.

## Computation of $\vec{f}_{\ell j 0}$ from $\vec{u}_{\ell j 0}$

0. Preprocessing. Given $M$, for each mode $\left(m_{1}, m_{2}\right), m_{1}, m_{2}=1, \ldots, M$, solve the eigenvalue problem (21), and store the eigenvalues and eigenvectors.
1. Given $\vec{u}_{\ell j 0}$, perform the discrete Fourier transform in $\ell$ and $j$ and compute all Fourier modes $\hat{u}_{0}, \hat{v}_{0}, \hat{w}_{0}$.
2. For each mode, compute $\hat{u}_{-1}, \hat{v}_{-1}, \hat{w}_{-1}$ using pre-computed values of eigenvalues and eigenvectors.
3. Compute the Fourier modes of the force $\hat{f}, \hat{g}, \hat{h}$, using Eq.(17)
4. Compute the force by inverse discrete Fourier transform

All discrete Fourier transforms can be efficiently computed by FFT algorithms in $O\left(M^{2} \log M\right)$ operations. In all our calculations we used the FFTW package developed at MIT [1].

### 3.4 Elastic Displacement Computation

Let us assume that we have deposited $N$ atoms on a substrate of size $M \times M$. Let us use $\mathbf{u}_{g} \in \mathbb{R}^{3 N}$ to denote the relative displacement of the Germanium atoms on the substrate. We use $\mathbf{u}_{s} \in \mathbb{R}^{3 M^{2}}$ to denote the relative displacement of the top layer of atoms on the substrate. Then the equilibrium position of the particles can be obtained by solving the following linear system

$$
\mathbf{F} \equiv\binom{\mathbf{F}_{s}}{\mathbf{F}_{g}}=\left(\begin{array}{cc}
S & B  \tag{24}\\
B^{T} & A
\end{array}\right)\binom{\mathbf{u}_{s}}{\mathbf{u}_{g}}+\binom{\mathbf{b}_{s}}{\mathbf{b}_{g}}=0
$$

The matrices appearing in the system have the following meaning. The forces acting on the $M^{2}$ Silicon atoms on the surface of the substrate are

$$
\mathbf{F}_{s}=S \mathbf{u}_{s}+B \mathbf{u}_{g}+\mathbf{b}_{s}
$$

Here $S \mathbf{u}_{s}$ is the force on the atoms at the surface of the substrate due to all the (Silicon) atoms in the substrate. This is efficiently computed using the results from the previous section. $B \mathbf{u}_{g}$ is the force on the substrate surface due to the Germanium atoms, and $\mathbf{b}_{s}$ is the sum of the forces given by (4] 7). The force acting on the $N$ Germanium atoms on the substrate is given by

$$
\mathbf{F}_{g}=B^{T} \mathbf{u}_{s}+A \mathbf{u}_{g}+\mathbf{b}_{g}
$$

where $A \mathbf{u}_{g}$ are the forces that arise from the interactions between the Germanium atoms, $B^{T} \mathbf{u}_{s}$ is the force on the Germanium atoms due to the top layer of Silicon atoms, and $\mathbf{b}_{g}$ is the sum of the forces given by (47).

We observe that the matrix

$$
\left(\begin{array}{cc}
S & B  \tag{25}\\
B^{T} & A
\end{array}\right)
$$

is a symmetric negative semi-definite matrix; it has 3 zero eigenvalues, corresponding to the free translation in the 3 directions of the coordinate axis. The system is clearly invariant for translation along the directions parallel to the substrate. It is also invariant along the direction orthogonal to the substrate, because the substrate is considered semi-infinite. This can be understood by the following argument. For a substrate of a finite thickness, let us say of $N_{L}$ layers, a unit displacement in the direction orthogonal to the substrate will produce an elastic force per unit atom equal to

$$
F=\frac{1}{N_{L}}\left(k_{L}+2 k_{D}\right)
$$

which vanishes as $N_{L} \rightarrow \infty$. Therefore no resistance is opposed to any translation.
Notice that the matrix $B^{T}$ is the transpose of matrix $B \in \mathbb{R}^{3 M^{2} \times 3 N} . A$ is a $3 N \times 3 N$ matrix. $A$ and $B$ are sparse and the matrix vector products are efficiently evaluated using expressions similar to (9). System (24) can be solved by an iterative scheme for large, sparse linear systems, making use of the symmetry and definiteness of the coefficient matrix. Here we shall use just a simple conjugate gradient method, leaving the search for a more efficient method to future investigations.

## 4 Evaluation of the elastic energy

Once the strain field is determined, the elastic energy is computed as follows. The energy associated to the bonds is given by

$$
E_{\text {elas }}=E_{\mathrm{Ge}-\mathrm{Ge}}+E_{\mathrm{Ge}-\mathrm{Si}}+E_{\mathrm{Si}-\mathrm{Si}},
$$

where $E_{\mathrm{Si}-\mathrm{Si}}$ is the energy due to the interaction between the Silicon atoms. The other terms are analogously defined. One has

$$
\begin{equation*}
E_{\mathrm{Si}-\mathrm{Si}}=\sum_{\mathrm{Si}-\mathrm{Si} \text { bonds }} \frac{1}{2} k_{\text {bond }}\left(\ell_{\mathrm{bond}}\right)^{2} . \tag{26}
\end{equation*}
$$

where $k_{\text {bond }}$ is either $k_{L}$ or $k_{D}$ depending on whether the bond is longitudinal or diagonal. $\ell_{\text {bond }}$ is the amount the bond has been stretched from the equilibrium configuration. This can be written in terms of the displacement field as

$$
\begin{equation*}
E_{\mathrm{Si}-\mathrm{Si}}=-\frac{1}{2} \mathbf{u}_{\mathrm{Si}}^{T} A_{\mathrm{Si}} \mathbf{u}_{\mathrm{Si}} \tag{27}
\end{equation*}
$$

where we denote by $A_{\mathrm{Si}}$ the (infinite dimensional) matrix that provides the force on all Silicon atoms as a function of the position of the Silicon atoms.

The energy due to the interaction of the Germanium atoms can be written as

$$
\begin{equation*}
E_{\mathrm{Ge}-\mathrm{Ge}}+E_{\mathrm{Ge}-\mathrm{Si}}=\sum_{\text {all Ge bonds }} \frac{1}{2} k_{\mathrm{bond}}\left(\ell_{\mathrm{bond}}\right)^{2}, \tag{28}
\end{equation*}
$$

where $k_{\text {bond }}$ is as in (26) but here $\ell_{\text {bond }}$ represents the amount the Germanium bonds have been stretched from their original equilibrium configuration (as opposed to the reference configuration). This can be written as

$$
\begin{equation*}
E_{\mathrm{Ge}-\mathrm{Ge}}+E_{\mathrm{Ge}-\mathrm{Si}}=\frac{1}{2} \sum_{\substack{\text { all atoms } \\ \text { bonded to } \\ \text { a Ge atom }}}^{\sum e_{\ell j k}} \tag{29}
\end{equation*}
$$

where $e_{\ell j k}$ is the total elastic energy stored in all the bonds associated to the atom located at site $(\ell, j, k)$. The factor $\frac{1}{2}$ accounts for the double counting of the summation. We can write

$$
e_{\ell j k}=e_{\ell j k}^{x}+e_{\ell j k}^{y}+e_{\ell j k}^{z}
$$

with

$$
\begin{aligned}
e_{\ell j k}^{x} & =\frac{k_{L}}{2}\left(\left[u_{\ell+1 j k}-u_{\ell j k}+d_{x}\right]^{2}+\left[u_{\ell-1 j k}-u_{\ell j k}-d_{x}\right]^{2}\right) \\
& +\frac{k_{D}}{2}\left(\left[u_{\ell+1 j k+1}-u_{\ell j k}+d_{x}\right]^{2}+\left[u_{\ell-1 j k+1}-u_{\ell j k}-d_{x}\right]^{2}\right) \\
& +\frac{k_{D}}{2}\left(\left[u_{\ell+1 j k-1}-u_{\ell j k}+d_{x}\right]^{2}+\left[u_{\ell-1 j k-1}-u_{\ell j k}-d_{x}\right]^{2}\right) \\
& +\frac{k_{D}}{2}\left(\left[u_{\ell+1 j+1 k}-u_{\ell j k}+d_{x}\right]^{2}+\left[u_{\ell-1 j+1 k}-u_{\ell j k}-d_{x}\right]^{2}\right) \\
& +\frac{k_{D}}{2}\left(\left[u_{\ell+1 j-1 k}-u_{\ell j k}+d_{x}\right]^{2}+\left[u_{\ell-1 j-1 k}-u_{\ell j k}-d_{x}\right]^{2}\right)
\end{aligned}
$$

where $d_{x}=a_{s}-a_{g}$. In the above expression, each term in square brackets represents the contribution to the elastic energy by a pair of atoms. If no such pair exists then the term is not included. One can derive analogous expressions for $e_{\ell j k}^{y}$ and $e_{\ell j k}^{z}$ where $d_{y}=d_{x}$ and $d_{z}=a_{L}-a_{g}$.

To compute the total elastic energy, the sum in Eq. (28) is computed directly, while the sum in Eq. (26), which contains infinitely many terms, can be computed by the following argument. First, let us distinguish among the surface and the bulk atoms. Let us denote by $-\mathbf{F}_{\mathrm{Si}}$ the force acting on Silicon due to the presence of the Germanium. They take the form

$$
\mathbf{F}_{\mathrm{Si}}=\binom{\mathbf{F}_{s}}{0}
$$

where the dimension of the vector $\mathbf{F}_{s}$ is equal to the dimension of the surface of the substrate, while 0 represent the force acting on rest of all the infinite atoms of the bulk.

At equilibrium, the net force acting on all Silicon atoms is zero, therefore we may write

$$
-\mathbf{F}_{\mathrm{Si}}+A_{\mathrm{Si}}\binom{\mathbf{u}_{s}}{\mathbf{u}_{\mathrm{bulk}}}=0
$$

Using the above relations one obtains

$$
E_{\mathrm{Si}-\mathrm{Si}}=-\frac{1}{2}\binom{\mathbf{u}_{s}}{\mathbf{u}_{\mathrm{bulk}}}^{T}\binom{\mathbf{F}_{s}}{0}=-\frac{1}{2} \mathbf{u}_{s}^{T} \mathbf{F}_{s}
$$

We remark here that $\mathbf{F}_{s}$ is the same surface force computed in the previous section using the discrete Fourier transform.

## 5 Time stepping approximations

The method outlined in Section 22 is impractically slow for the following reasons

1. For each attempted hop, a complete elastic computation has to be performed. Since most attempts are rejected, most of the time would be spent performing elastic computations that are never used.
2. Even without elastic effects, the simple rejection-based KMC described here is very slow. A more effective technique would be to use rejection-free KMC, in which all possible events are sampled according to their probability [4, 12]. However, with the inclusion of elastic effects, the implementation of rejection-free KMC is not so straightforward.
Here we shall outline various approximations of the model which lead to a much faster code, without significantly compromising the physical fidelity.

As mentioned above, in order to know the rate at which an atom might hop we must compute the change in elastic energy of the entire system with and without that atom present. We make the following approximation, we assume that the change in the elastic energy is due to the energy in the bonds that directly connect that atom. Therefore we have

$$
\begin{equation*}
\Delta E_{\text {elas }} \approx \sum_{\substack{\text { bonds to } \\ \text { atom } p}} \frac{k_{\mathrm{bond}}}{2}\left(\ell_{\mathrm{bond}}\right)^{2}=e_{\ell j k} \tag{30}
\end{equation*}
$$

where $(\ell, j, k)$ is the site of the $p$ th atom. The advantage of this approach is that a new equilibrium configuration has to be computed only if the move is accepted. Another approximation involves updating the displacement field after a given number $J$ of hops. Furthermore, one can verify that the change in elastic energy for atoms which are lightly bonded $\left(N_{b} \leq 5\right)$ is very small and consequently we assume it to be zero.

Finally, in order to reduce the number of rejections we separate the lightly bonded $\left(N_{b} \leq 5\right)$ and the more strongly bonded $\left(N_{b}>5\right)$ atoms in our implementation of kinetic Monte Carlo. This is done as follows. We take $Q$ steps where we update the lightly bonded atoms and then take one step where the strongly bonded atoms are allowed to move. The accepted rate for the strongly bonded atoms is increased by a factor $Q$.

The above discussion can be conveniently summarized as the following algorithm.

## ALGORITHM

1. Choose a site at random among all $M \times M$ sites (only atoms on the surface are allowed to move)
2. If there is Germanium atom present compute $N_{b}$ (number of bonds)
3. Let it hop without any elastic computation if $N_{b} \leq 5$
4. If $N_{b}>5$ ignore the atom
5. Repeat Steps 1 to $4 M^{2}$ times
6. Repeat Steps 1 to $5 Q$ times
7. Let the system come to mechanical equilibrium (perform an elastic solve).
8. Choose a site at random
9. If there is Germanium atom present compute $N_{b}$
10. If $N_{b} \leq 5$ do nothing
11. If $N_{b}>5$ then compute a random number, $r \in U[0,1]$
12. If $r<Q \exp \left(-\gamma\left(N_{b}-5\right)+\Delta E_{s}\right)$ perform a hop
13. After $J$ hops update the displacement field
14. Repeat Steps 9 to $13 M^{2}$ times.
15. We have now advanced $Q$ steps

## 6 Numerical results

At the present time the algorithm is to too slow to perform computations for realistic values for the parameter values and see effects due to elastic strain. For example, quantum dots observed in experiments are on the order of 20 nm . This would suggest that we should be computing on domains on the order of $1000 \times 1000$. At the present time the largest domain for which we can simulate in a reasonable time is $64 \times 64$. Since elastic phenomena are a bulk effect, then we must increase the spring constants to be unphysically large in order to observe significant elastic interaction. In our simulations we chose $k_{L}=500$ and $k_{D}=250$. These values are approximately 10 times larger than physical values. In addition we chose $F=10^{-5}$. Since the hopping rate for an adatom is unity then it follows that the diffusion coefficient is $D=\frac{1}{4}$. Therefore in our simulations we have $D / F=2.5 \times 10^{4}$. From an experimental point of view this is small. Realistic values for $D / F$ are typically larger than $10^{5}$. We have chosen high deposition rate to reduce the simulation time. We have taken $\gamma=2$. Finally we have used $Q=5$ and $J=8$. Numerical experiments revealed taking smaller values for $Q$ and $J$ did not change the answer appreciably.

In Figures 4, 5] and 6, we present computations using the parameter values discussed above but we allow the misfit to vary. Figure 4 shows the growth for $\epsilon=0.02$. Here one observes layer-by-layer growth. Figure 5 presents the results when $\epsilon=0.04$. One observes that in the initial stages of growth the morophology is similar to layer-bylayer growth but three dimensional islands form by nucleation type events and by the formation of trenches [22. The result for the case $\epsilon=0.06$ are shown in Figure 6. In this situation three dimensional islands form very quickly by nucleation.

In both cases, $\epsilon=0.04$ and $\epsilon=0.06$ we observe Volmer-Weber growth. Simulations were performed over a wide range of parameter values and they always revealed a sharp transition between layer-by-layer growth and Volmer-Weber growth; Stranski-Krastanov growth was not observed. Our results are consistent with previous simulations in this regard.

## 7 Summary

A numerical method for the computation of heteroepitaxial growth in the presence of strain using kinetic Monte Carlo has been presented. A solid-on-solid model is used and the elastic effects are incorporated using a linear ball and spring model. It is


Figure 4: Heteroepitaxial simulations with $\epsilon=0.02$, all other parameter values are given in the text. (a) 0.5 monolayers, (b) 1.5 monolayers, (c) 2.5 monolayers, and (d) 3.5 monolayers. The number of gray levels is equal to the maximum height. Note: higer resolution versions of the figures can be found at www.math.lsa.umich.edu/~psmereka


Figure 5: Heteroepitaxial simulations with $\epsilon=0.04$, all other parameter values are given in the text. (a) 0.5 monolayers, (b) 1.5 monolayers, (c) 2.5 monolayers, and (d) 3.5 monolayers. The number of gray levels is equal to the maximum height.


Figure 6: Heteroepitaxial simulations with $\epsilon=0.06$, all other parameter values are given in the text. (a) 0.5 monolayers, (b) 1.5 monolayers, (c) 2.5 monolayers, and (d) 3.5 monolayers. The number of gray levels is equal to the maximum height.
assumed that the film is mechanical equilibrium. The strain field in the substrate is computed by an exact solution which is efficiently evaluated using the fast Fourier transform. The strain field in the growing film is computed directly. The resulting coupled system is solved iteratively using the conjugate gradient method. Finally we introduce various approximations in the implementation of the KMC to improve the computation speed. Numerical results show that layer-by-layer growth is unstable if the misfit is large enough resulting in the formation of three dimensional islands. Our results are in agreement with previous studies [3, 10, 11, [8, 13, 18].

Currently, we are in the process of solving the elastic equations for the deposited atoms using multigrid and then coupling the multigrid solver to the exact solution in the substrate. In addition we plan to extend the model to allow for the deposition of several different atomistic species.

## Acknowledgments

The authors thank Len Sander for many useful conversations. The authors also thank Russel Caflisch and Christian Ratsch for helpful remarks; in particular for comments that ultimately lead to the approximation given by Eq. (30). PS was supported by NSF grants DMS-0207402 and DMS-0244419. GR was supported by a grant from the Michigan Center for Theoretical Physics and grant from the Italian Government (PRIN project 2003, prot.n.2003011441_004).

## References

[1] The FFTW package was developed at MIT by Matteo Frigo and Steven G. Johnson. Informations can be found at the site http://www.fftw.org/.
[2] J.G. Amar and F. Family, Mound Formation, Coarsening and Instabilities in Epitaxial Growth, Surface Rev. and Lett. 5 851-864 (1998).
[3] M. Biehl, M. Ahr, W. Kinzel, and F. Much, Kinetic Monte Carlo simulations of heteroepitaxial growth, Thin Solid Films, 428 52-55 (2003).
[4] A.B. Bortz, M.K. Kalos, and J.L. Lebowitz, New algorithm for Monte-Carlo simulation of Ising spin systems, J. Comput. Phys. 17 10-18 (1975).
[5] W.K. Burton, N. Cabrera, and F.C. Frank, The growth of crystals and the equilibrium structure of their surfaces, Trans. R. Soc. London Ser. A 243, 299 (1951).
[6] M.D. Johnson, C. Orme, A.W. Hunt, D. Graff, J. Sudijono, L.M. Sander, B.G. Orr, Stable and unstable growth in molecular beam epitaxy, Phys. Rev. Lett. 72 116 (1994).
[7] J. Krug, P. Politi, and Michely, Island nucleation in the presence of step-edge barriers: Theory and applications, Phys. Rev. $B 6114037$ (2000).
[8] C.H. Lam, C.K. Lee, and L.M. Sander, Competing Roughening Mechanisms in Strained Heteroepitaxy: A Fast Kinetic Monte Carlo Study, Phys. Rev. Lett. 89 16102 (1-4) (2002).
[9] M. T. Lung, C-H Lam, and L. M. Sander, preprint
[10] F. Much, M. Ahr, M. Biehl, and W.Kinzel, A Kinetic Monte Carlo method for the simulation of heteroepitaxial growth, Comput. Phys. Commun. 147 (2002) 226-229.
[11] F. Much and M. Biehl, Simulation of wetting-layer and island formation in heteroepitaxial growth, Europhysics Lett. 63 (2003) 14-20.
[12] M.E.J. Newman and G.T. Barkema, Monte Carlo Methods in Statistical Physics, Oxford University Press, New York, (1999).
[13] B.G. Orr, D.A. Kessler, C.W. Snyder, and L.M. Sander, A model for straininduced roughening and coherent island growth, Europhysics Lett. 19 33-38 (1992).
[14] P. Politi, G. Grenet, A. Marty, A. Ponchet, and J. Villain Instabilities in crystal growth by atomic or molecular beams Phys. Rep. 324271 (2000).
[15] C. Ratsch, M.C. Wheeler, and M.F. Gyure, Roughening due to edge diffusion for irreversible aggregation Phys. Rev. B62 12636 (2000)
[16] C. Ratsch, A. Zangwill, and P. Šmilauer, Scaling of heteroepitaxial island sizes. Surface Science Letters 314 L937-L942 (1994).
[17] C. Ratsch and A. Zangwill, Equilibrium theory of the Stranski-Krastanov epitaxial morphology, Surface Science 293 123-131 (1993).
[18] C.Ratsch, P.S̆milauer, D.D. Vvedensky, A. Zangwill, Mechanism for Coherent Island Formation during Heteroepitaxy, J. Phys. I France 6 575-581 (1996).
[19] J.M. Rickman and D.J. Srolovitz, Defect interactions on solid surfaces, Surface Science 284 211-221 (1993).
[20] A.C. Schindler, M.F. Gyure, G.D. Simms, D.D. Vvedensky, R.E. Caflisch, C. Connell, and E. Lou, Theory of strain relaxation in heteroepitaxial systems, Phys. Rev. B. 67075316 (2003).
[21] V.A. Shchukin and D. Bimberg, Spontaneous ordering of nanostructure on crystal surfaces Rev. Mod. Phys. 711125 (1999).
[22] J. Tersoff and F.K. LeGoues, Competing relaxation mechanisms in strained layers, Phys. Rev. Lett. 723570 (1994).


[^0]:    *Dipartimento di Matematica e Informatica, Università di Catania, Catania, Italy
    ${ }^{\dagger}$ Department of Mathematics, University of Michigan and the Michigan Center for Theoretical Physics, Ann Arbor, MI 48109-1109

