Self-assembly of steps and vacancy lines during the early stages of Ge/Si(001) heteroepitaxy

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The wetting layer formed during the early stages of Ge/Si(001) growth has been found in recent experiments to undergo a roughening process, where the $S_A$ surface steps affect the spatial organization of vacancy lines (VLs) by increasing (stretching) or decreasing (squeezing) their average spacing. Using a combination of atomistic simulations and elastic theory of surface defects, we have computed the interaction energy of the $S_A$ steps and VLs for each of the observed defect configurations. We find that the repulsive $S_A$-VL interactions lead to an increase in the spacing of the VLs in the “stretch” arrangement, but do not significantly affect the VL spacing in the “squeeze” configuration, providing an explanation for the observed correlations in the wetting layer roughness. © 2005 American Institute of Physics. [DOI: 10.1063/1.2147720]

The strained-layer Ge/Si(001) system shows a remarkable variety of structural and morphological changes depending on growth conditions and Ge coverage. The system undergoes transformations from the 2×1 structure in the absence of Ge, to the 2×N reconstructed wetting layer, to the formation of hut and dome quantum dots at Ge coverages larger than about three monolayers (ML). While the formation of pyramid and dome clusters has been actively studied in the last two decades, in comparison, the initial roughening of the wetting layer that occurs just before the formation of pyramids has remained virtually unexplored. The scanning tunneling microscopy (STM) experiments of Sutter et al. have recently provided important insights on the initial roughening of the Ge film using microscopy techniques capable of achieving a remarkable degree of detail and image statistics over wide areas of the film surface. Their work shows that the interactions between the $S_A$ steps and the vacancy lines (VLs) drive the roughening of the Ge film through the formation of ordered stripe patterns.

Motivated by these elegant experiments, we have investigated the energetics of the observed patterns, using both atomistic simulations and elastic theory of surface defects. The aim of our calculations is to determine the interactions between the $S_A$ steps and VLs and to study the role of these interactions in the spatial ordering of these line defects. Our simulations predict that the repulsive $S_A$-VL interactions decay with an inverse-distance law, in agreement with the behavior expected for lines of monopoles interacting with dipoles through their elastic fields. Furthermore, we find that these interactions modify the mean distance between the VLs, depending on the way the $S_A$ steps and VLs are distributed on the surface.

The structural models that correspond to the stretch and the squeeze arrangements are illustrated in Fig. 1. In the former case, an island of width $d$ bounded by $S_A$ steps is sandwiched between VLs, which leads to an increase (or stretching) in the mean spacing of the VLs. In contrast, the two VLs sandwiched between the similar islands are observed to be “squeezed” by the $S_A$ steps in the latter case. Using the Tersoff potential for Si-Ge, we have performed relaxations for 90-layer-thick computational cells with 2-ML or 3-ML Ge coverage and for periods $D=Nd$ ($2<N<4$), where $a=3.84$ Å is the lattice constant of the Si(001) surface. From the total energy of the slab $E$, we compute the surface energy $\gamma=(E-n_{Si}\mu_{Si}-n_{Ge}\mu_{Ge})/A$ as the excess energy per area $A=2aD$, where $n_{Ge}$, $\mu_{Ge}$ ($n_{Si}$, $\mu_{Si}$) are the number of atoms and the chemical potential of Ge (Si), respectively. The overall surface energy $\gamma$ can be written as

$$\gamma = \gamma_u \frac{d}{D} + \gamma_l \frac{D - d}{D} + \frac{\lambda}{D},$$

where the surface energy $\gamma_u$ ($\gamma_l$) of the upper (lower) terrace can be obtained from separate calculations for defect-free Ge/Si(001) surfaces and $\lambda$ (subsequently referred to as $\lambda_u$ or $\lambda_l$ for the two cases in Fig. 1) includes the formation energies of $S_A$ steps and VLs and their interactions.

Before presenting the results for the interactions between $S_A$ steps and VLs, it is useful to separately consider the energetics of periodic arrays of each of these line defects. Since the VLs can be described as elastic-force dipoles in the plane of the surface, they interact with each other with a repulsive inverse-square distance law. The energy per unit length of an array of VLs with the spacing $D$ can then be written as

$$\lambda_{VL}(D) = A_{VL}^{0} + A_{VL}^{1} \frac{n_{VL}^{2}}{(6D^{2})},$$

where $A_{VL}^{0}$ is the formation energy of a VL and the second term is the interaction energy of the uniform array, where $A_{VL}^{1}$ is the strength of the dipolar interaction. The up- and down-$S_A$ steps on the sides of an island, on the other hand, behave like force monopoles due to the discontinuity of surface stress at the island edges. Following the notations...

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in Eq. (2), the energy of a periodic array of such islands of width $d$ and period $D$ is given by\textsuperscript{9,10}

$$
\lambda_{SA}(d,D) = 2\Lambda^0_{SA} - \Lambda^1_{SA} \log \left( \frac{D}{\pi c} \sin \left( \frac{\pi d}{D} \right) \right),
$$

(3)

where $c$ is a half an atomic scale cutoff introduced to regularize the logarithmic divergence.\textsuperscript{11} The parameters $\Lambda^0$ and $\Lambda^1$ in Eqs. (2) and (3) obtained from atomistic simulations as described in Refs. 6 and 10 are given in Table I.

We now consider the stretch configuration [Fig. 1(a)], for which the defect energy can be written as

$$
\lambda_{a}(d,D) = \lambda_{VL} + \lambda_{SA} + \lambda_{int}^{VL}.
$$

(4)

where $\lambda_{VL}$ and $\lambda_{SA}$ are given by Eqs. (2) and (3), respectively, and the third term denotes the interaction between the monopoles at the $S_A$ steps and the dipoles at the VLs. We have calculated this term from atomistic relaxations of the stretch structure [Fig. 1(a)] and plotted it in Fig. 2(a) as a function of the VL spacing $D$, for fixed island width $d=4a$.

In order to extract the strength of the $S_A$-VL interaction from this data, we note that according to elasticity theory, a single $S_A$ step interacts with a single VL with a power law that decays as the inverse of the distance between them. Using $\Lambda^1_{VL}$ to denote the strength of this monopole-dipole interaction, the contributions corresponding to the steps and VLs in the unit cell of the stretch configuration and its periodic images can be summed analytically to

$$
\lambda_{int}^{VL} = \Lambda^1_{VL} \frac{2\pi}{D} \sin \left( \frac{\pi d}{2D} \right).
$$

(5)

It can be seen from Fig. 2(a) that this functional form provides an excellent fit to the results of the simulations. Furthermore, the coefficient $\Lambda^1_{VL}=254.7$ meV obtained from the fitting procedure is very close to the geometric average $\sqrt{\Lambda^1_{VL} \Lambda^1_{SA}}=247.4$ meV that is expected\textsuperscript{12} for the interaction of a monopole with a dipole. These observations confirm that

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{(Color online) Surface unit cells for the stretch (a) and squeeze (b) configurations reported in Ref. 3. The Ge film is shown in black, and the Si substrate in gray. The island size is $d$ and the distance between an $S_A$ step and a VL is $p$. Configuration (a) requires another parameter $q$ to denote the distance between the two VLs of the unit cell. The periods $D$ along the [110] direction are $D=2p+d$ and $D=2p+d+q$ for models (a) and (b), respectively. The periodic length of the cells in the [110] direction is $2a$ ($a=3.84\,\text{Å}$). The horizontal arrows schematically show the force monopoles and dipoles at the steps and VLs, respectively.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig2.png}
\caption{(Color online) Interaction energy of $S_A$ steps and VLs obtained from atomistic simulations in the stretch configuration [(a), triangles up] and in the squeeze pattern [(b), triangles down], along with the fitting curves given by Eq. (5) and Eq. (8), respectively. The dashed curves show the interaction energy between VLs [refer to Eqs. (2) and (7)] in the two configurations. The island size is $d=4a$ in both cases, and the $S_A$-VL distance in case (b) is $p=2.5a$.}
\end{figure}

\begin{table}[h]
\centering
\caption{Defect formation energies $\Lambda^0$ and interaction strength parameters $\Lambda^1$ computed using the Tersoff interatomic potential (see Ref. 4).}
\begin{tabular}{|c|c|c|}
\hline
Defect & $\Lambda^0$ (meV/Å) & $\Lambda^1$ (meV/Å) \\
\hline
VL & $-133.934 \pm 0.201$ & $13434 \pm 89$ \\
$S_A$ & $-15.56 \pm 0.11$ & $4556 \pm 0.076$ \\
$S_A$-VL & $254.7 \pm 1.077$ & \\
\hline
\end{tabular}
\end{table}
spacings between VLs is determined by minimizing the defect energy per period of the defect array, which is \( \lambda_{\text{VL}}/D \) [refer to Eq. (2)]. As shown in Ref. 6, a competition between the negative formation energy of VLs and their repulsive interactions leads to a mean spacing \( D = 6a \), which compares favorably with the experimental value of \( (8 \pm 0.9)a \). To see how this spacing is altered in the two structures in Fig. 1, we have plotted \( \lambda_{a,b}/D \) [from Eq. (4) and Eq. (6)] as functions of the spacing between the VLs in Fig. 3, choosing a typical island width of \( d = 4a \). While the island width \( d \) and the VL-VL separation \( D \) determine the \( S_{\text{VL}} \)-VL spacing \( ((D - d)/2) \) in the stretch configuration, this spacing \( (p) \) is an independent parameter for the squeeze structures. We have found, however, that the defect energy per period \( \lambda_{a,b}/D \) is minimum at the \( S_{\text{VL}} \)-VL separation of \( p = 2.5a \) for island widths in the range \( 2a \leq d \leq 8a \). This result is in agreement with the observations of Sutter et al., who find that \( S_{\text{VL}} \)-VL spacings of 1.5\( a \) and 2.5\( a \) occur much more frequently than any other \( S_{\text{VL}} \)-VL separation. In what follows, we discuss the variation of the defect energy \( \lambda/D \) as a function of VL-VL distance.

A comparative analysis of the VL-VL spacing between stretch, free and squeeze configurations reveals a remarkable consistency with experimental findings. As seen in Fig. 3, the optimum VL-VL distance in the stretch case is \( D = 10a \), which is larger than the spacing of free VLs (Ref. 6) by four Si(001) lattice constants. A similar increase in the VL spacing has been observed by Sutter et al., who report separations of \( (8.0 \pm 0.9)a \) and \( (11.9 \pm 1.7)a \) for the free and stretch configurations, respectively. For squeeze structures with \( d = 4a \) and \( p = 2.5a \), we find nearly the same optimum VL-VL distance \( (q = 6a) \) as in the case of free VLs. This finding is also consistent with the STM observations, which show only small change in the VL-VL distance from \( (8.0 \pm 0.9)a \) (free) to \( (7.1 \pm 0.9)a \) (squeeze). The close agreement of the computational and analytical results with experiments suggests that the empirical potential used in our work is able to capture the key features of the interactions of line defects on Ge/Si(001).

The pronounced difference between the statistics of VL separations in the stretch and squeeze configurations can be understood by considering the relative contributions of the monopole-dipole and dipole-dipole interactions to the surface energy of each configuration. In the stretch case, the \( S_{\text{VL}} \)-VL (monopole-dipole) interactions are stronger than the VL-VL interactions, as seen in Fig. 2(a). Therefore, the optimum spacing of \( 6a \) determined for a uniform array of VLs (see Ref. 6) will be increased significantly by the presence of islands between the VLs [Fig. 1(a)]. In the squeeze case [Fig. 2(b)], since the islands are located between pairs of VLs, the VL-VL repulsion dominates the \( S_{\text{VL}} \)-VL interaction. Indeed, the monopole-dipole terms are found to have little influence on the VL-VL distance in the squeeze configuration.

In summary, we have studied the energetics of \( S_{\text{VL}} \)-VL stretch and squeeze patterns using a linear elasticity based approach combined with atomistic simulations. Our study demonstrates the existence of an effective repulsion between steps and VLs, which has been identified as a key factor in the initial roughening of the Ge/Si(001) system. Our analysis also predicts optimal separations for the VLs, in agreement with recent microscopy experiments. While the experimental distributions of VL spacings contain information on thermal fluctuations, these effects are not included in the present work, but will be addressed in the future.