Atomistic simulation study of misfit strain relaxation mechanisms in heteroepitaxial islands

Avinash M. Dongare and Leonid V. Zhigilei

Department of Materials Science and Engineering, University of Virginia

116 Engineer's Way, Charlottesville, VA 22904-4745

ABSTRACT

The mechanisms of the misfit strain relaxation in heteroepitaxial islands are investigated in two-dimensional molecular dynamics simulations. Stress distributions are analyzed for coherent and dislocated islands. Thermally-activated nucleation of misfit dislocations upon annealing at an elevated temperature and their motion from the edges of the islands towards the positions corresponding to the maximum strain relief is observed and related to the corresponding decrease of the total strain energy of the system. Differences between the predictions of the energy balance and force balance criteria for the appearance of misfit dislocations is discussed. Simulations of an island located at different distances form the edge of a mesa indicate that the energy of the system decreases sharply as the island position shifts toward the edge. These results suggest that there may be a region near the edge of a mesa where nucleation and growth of ordered arrays of islands is favored.

INTRODUCTION

The effect of the quantum confinement of charge carriers within zero-dimensional "quantum dots" has a promise of a remarkable advancement of electronic and optoelectronic devices. Realization of this promise hinges on the development of efficient methods for generation of ordered arrays of small, tens of nm in size, clusters or islands. One of the methods of quantum dot generation is lattice mismatched heteroepitaxial growth [1,2]. The electronic structure of a coherently strained island formed by heteroepitaxial growth is sensitive to the stress distribution in and around the island. Moreover, when the island size exceeds a certain critical size, the high misfit strain in the growing island can be released through the nucleation of misfit dislocations, which can also affect the electronic properties. Understanding the stress distributions and the mechanisms of stress relaxation are, therefore, important for the advancement of nanoscale electronic devices.

While there have been significant theoretical and computational efforts aimed at analysis of stress/strain distributions in and around coherently strained heteroepitaxial islands, e.g. [3,4,5,6], the relaxation of the lattice mismatch stresses related to the misfit dislocations is much less studied. It is difficult to include the description of the processes of dislocation nucleation and migration into continuum models, whereas the scale of the atomistic simulations is not sufficient to study these processes. Experimentally, the transition from a coherently growing strained island to the one in which misfit dislocations partially relieve the strain energy has been observed via high resolution electron microscopy [7,8]. It has been found that mismatch dislocations typically nucleate at the corners of the island, the regions of the maximum strain concentration. The critical dimensions for the appearance of misfit dislocations are commonly discussed based on either energy balance models, when energies of coherently strained and plastically relaxed islands are compared, or force balance models, when dislocation nucleation is considered as a limiting step in the relaxation process [3,9,10]. In this paper we use a two-dimensional atomistic model of a lattice mismatched island to compare the predictions of the two approaches and to

investigate the role of the energy barrier for dislocation nucleation and the effect of the dislocations on the stress distribution in and around the island.

In addition to the requirement of the size uniformity of the nano-islands, most of the potential electronic applications require the ability to control the spatial distribution of the islands in one-, two- or three-dimensional devices, such as single-electron transistors and memory cells. Several methods have been proposed to achieve ordered arrangement of the quantum dots, including growth on substrates with dislocation networks [11] or stacking of successive layers of buried heteroepitaxial islands [12]. One of the most promising approaches to induce self-organization of heteroepitaxial islands is the self-assembly on patterned substrates [13,14,15]. In particular, Kamins and Williams [13] demonstrated one-dimensional ordering of Ge islands along the edges of lithographically prepared Si mesas. The ordering of self-assembled Ge islands has been attributed either to the kinetics of island nucleation in the presence of the edges of the mesa serving as barriers to surface diffusion or to a higher compliance of the substrate material near the edges allowing for the decrease of the mismatch strain energy. In this work we investigate the later possibility by performing a series of simulations of heteroepitaxial islands located on a plateau of a mesa. The total strain energy is calculated for islands located at different distances from the edge of the mesa and the spatial extent of the effect of the edge on the strain energy relief is analyzed.

COMPUTATIONAL METHOD

Large-scale two-dimensional (2D) molecular dynamics (MD) simulations are used in this work to investigate the mechanisms of the misfit strain relaxation in heteroepitaxial islands and, in particular, the effect of the misfit dislocations and surface topography on stress distribution and energy of the islands. While 2D simulations do not allow for a direct quantitative comparison with experimental data, they are capable of revealing a general physical picture of the mechanisms of the misfit strain relaxation and can provide a basis for interpretation of experimental results [3,16,17].

Atomistic representation of the substrate in the simulations consists of 57142 atoms initially arranged in a close-packed 2D triangular lattice with dimensions of 80 nm in the direction parallel to the surface and 35 nm in the direction normal to the surface. Periodic boundary conditions are applied in the direction parallel to the substrate and the bottom layer of the substrate is maintained to be rigid. The size of the substrate is chosen to be sufficiently large to avoid the effect of the boundary conditions on the simulation results. Epitaxial islands located on the substrate have different height, from 1.2 nm to 10 nm, and contact length with the substrate of 20 nm. To study the effect of the plateau edge on strain relaxation in a system of an island located on a mesa, as in experiment by Kamins and Williams [13], a system consisting of a substrate, a mesa, and an epitaxial island is used. The mesa has a base of 80 nm, a plateau of 68 nm, and a height of 20 nm. The height of the island is 3 nm and contact length with the plateau is 20 nm. The energy of the system is calculated for different locations of the island with respect to the edge of the plateau. Calculations of stress distributions and energies of any atomic configuration are preceded by complete relaxation and quenching of the system to 0 K. The quenching is done by MD with atom-by-atom damping method, when the velocity of any atom drops to zero at a time when it reaches maximum value. The time of the relaxation/quenching is chosen so that no further changes in the total energy of the system can be achieved by further relaxation.

The interatomic interaction is described by the Lennard-Jones potential with parameters chosen to roughly reproduce the bond energies and lattice parameters in Si and Ge for atoms in the substrate and the island respectively. The lattice cross-species parameters are fixed using the Lorentz-Berthelot rules [18]. The lattice mismatch in the simulations is defined as $f = 2(a_{ss}-a_{ii})/(a_{ss}+a_{ii}) = 4.17$ % where $a_{ss} = 2.35$ Å and $a_{ii} = 2.45$ Å are the equilibrium inter-atomic distances in the substrate and island materials respectively.

The elements of the tensor of atomic-level stresses are calculated as

$$\sigma_{\alpha\beta}(i) = -\frac{1}{\Omega_0}\left[\frac{1}{2}\sum_j F_{ij}^{\alpha} r_{ij}^{\beta} + M_i v_i^{\alpha} v_i^{\beta}\right]$$

where α and β label the Cartesian components, Ω_0 is the atomic volume, F_{ij} is the force on atom i due to atom j, M_i is the mass of atom i, and v_i is the velocity of atom i. The atomically resolved hydrostatic pressure is then calculated as $p(i) = -\text{Tr}[\sigma(i)]/2$. Positive values of pressure correspond to compression; negative values correspond to tensile stresses.

RESULTS AND DISCUSSION

a) *Nucleation of Misfit Dislocations*

Distributions of the hydrostatic pressure shown in the left part of Fig. 1 are calculated for systems that are relaxed/quenched directly from the initial state. Due to the lattice mismatch there is an abrupt transition at the interface from compression in the island to expansion in the substrate. The tensile stresses propagate deeper into the substrate with increasing height of the island. At the same time, the compressive stress relaxation in taller islands is more pronounced due to the ability of the island to expand in the lateral direction. As a result, the maximum compressive stresses inside the island are observed in the case of the thinnest island, whereas the maximum tensile stresses in the substrate are observed for the largest island. The expansion of the island also creates very strong compression in the substrate just below the island edges. The areas near the edges of the island are also the ones where a high intensity of the shear stresses is observed and where misfit dislocations nucleate upon annealing, as discussed below.

While no misfit dislocations appear during the relaxation and quenching of the initial system, nucleation of dislocations can be induced by annealing of the system at an elevated temperature, Fig. 1, right frames. The annealing temperature helps to overcome the nucleation barrier and the number of dislocations depends on the size of the island. Two dislocations appear when configuration with the 10 nm island is annealed at 750 K, leading to the decrease of the total energy of the system in a quenched, 0 K, state by approximately 56 eV. An appearance of two dislocations can be also induced in a configuration with the 6 nm island by annealing at 1000 K with an associated decrease in the total energy of the system by 58.9 eV. However, only one dislocation appears when the same system is annealed at 750 K, leading to a decrease in energy by 38 eV. For the 3 nm island, only one dislocation appears when annealed at 1000 K with a decrease in the total energy by 33.5 eV. No dislocation nucleation is observed in the case of the smallest 1.5 nm island after annealing at 1000 K. In all cases the misfit dislocations nucleate at the island edges and then slide along the interface to settle down at minimum energy positions. Annealing at a high temperature helps the system to overcome the energy barrier required to nucleate the dislocations at the island edges.
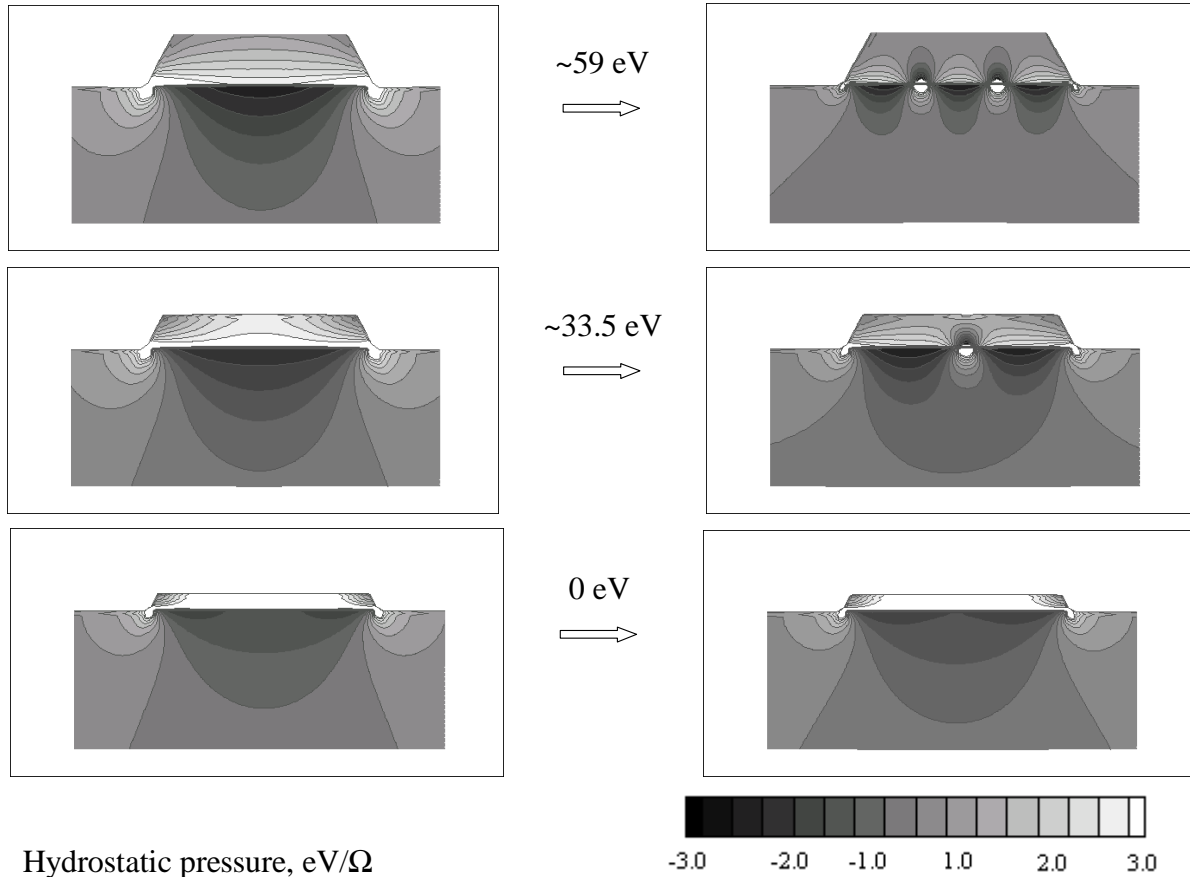
Figure 1. Contour plots of hydrostatic pressure in and around the island before (left) and after annealing at 1000 K (right) for different island sizes. Only part of the substrate is shown. Dislocations that appear during the relaxation or annealing of the initial configuration are identified by construction of a Burgers circuit.

In order to compare strain energies in systems with islands of different sizes, the interfacial energy, E_{IF}, i.e. the elastic energy due to the presence of the misfit interface, is calculated as $E_{IF} = E_T - (E_{ISL} + E_{SUB})$, where E_T is the total energy of the system, and E_{ISL} and E_{SUB} are the energies of the island and the substrate independent of each other [10]. The interface energy is calculated for systems with three different island sizes shown in Fig. 1. Interfacial energies for systems without dislocations, with one dislocation, and with two dislocations are plotted as a function of the height of the island in Fig. 2. Systems that cannot be created by thermally activated nucleation of dislocation(s) during annealing at temperatures up to 1000 K were created by manual introduction of dislocation(s). Either nucleation or manual introduction of the dislocations results in the relaxation of the mismatch stresses, Fig. 1, and associated decrease in the total strain energy of the system, Fig. 2. For islands of all sizes considered the energy decrease is the largest when two dislocations are created. Even in a system with a 1.5 nm island (lowest frames in Fig. 1), where no dislocations have appeared during annealing at 1000 K, introduction of dislocations leads to a substantial decrease of the interfacial energy. These results illustrate the gap between the energy balance and force balance criteria for the appearance of misfit dislocations and point to the importance of the kinetics of dislocation nucleation.

b) *Edge Effect*

In order to investigate the origin of the observed self-organization of heteroepitaxial islands along the edges of mesoscale topographical surface features [13,14,15], we have performed a series of simulations of an island on a mesa. Stress distributions and the total strain energy are calculated for islands located at different distances from the edge of a mesa, Fig. 3. Presence of the edge reduces constraints on the substrate relaxation, facilitating expansion of the substrate (larger areas of tensile stresses in the lower frame of Fig. 3) and partial relaxation of the compressive stresses in the island. The dependence of the total energy of the system on the island position relatively to the edge of the mesa shows the decrease of the strain energy of the system as the island approaches the edge.

Figure 2. Variation of interfacial energy with height of the island for a coherent island-substrate interface and interfaces with one and two misfit dislocations.

This energy dependence can be related to the experimental observation by Kamins and Williams [13] of the preferential nucleation and alignment of several rows of Ge islands along the edge of a mesa. In the simulations, the decrease in the strain energy is relatively small when the island is moved from the center of the plateau by 140 nm, but becomes much steeper as the island position is further shifted towards the edge of the mesa. These results suggest that the decrease of the strain energy near the edge of a mesa may be the reason for the formation of an ordered layer of islands, observed experimentally. The results of the simulations obtained for a system with a smaller thickness of the substrate are in a good quantitative agreement with the results shown in Fig. 3. Further three-dimensional computational investigations are needed to obtain quantitative information on the spatial extent of the low-energy region near the edge of a mesa and its relation to the size/shape of the mesa and the islands.
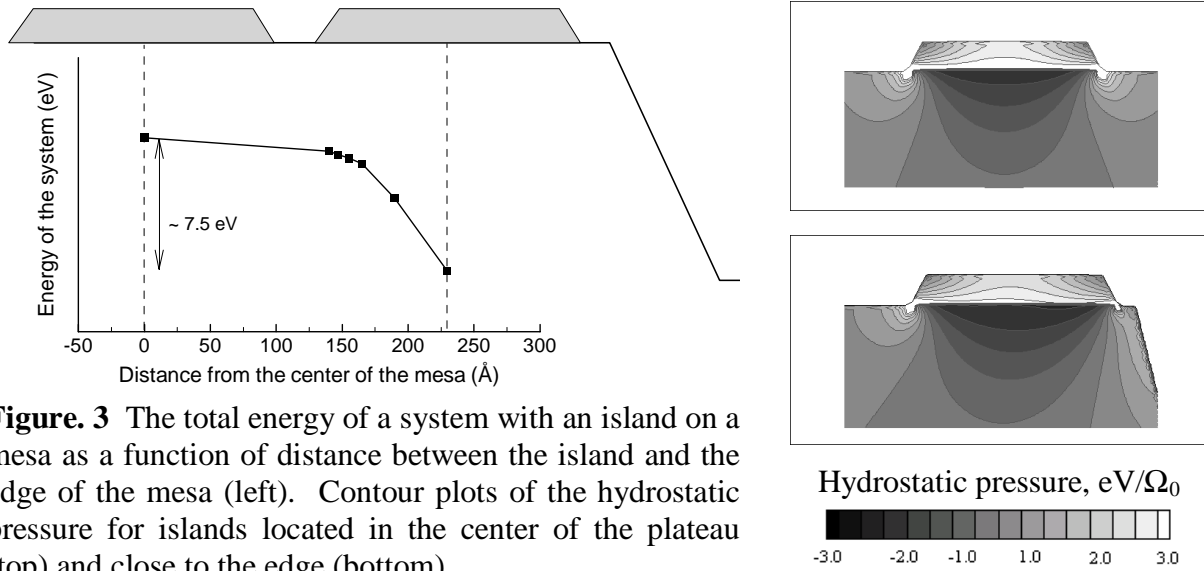

Figure. 3 The total energy of a system with an island on a mesa as a function of distance between the island and the edge of the mesa (left). Contour plots of the hydrostatic pressure for islands located in the center of the plateau (top) and close to the edge (bottom).
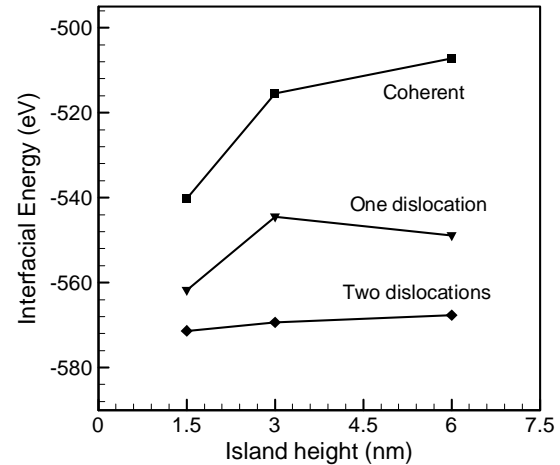
Hydrostatic pressure, eV/Ω_0

SUMMARY

Distribution of atomic-level stresses and the mechanisms of the misfit strain relaxation in heteroepitaxial islands are investigated in a series of 2D MD simulations. Stress concentration near the edges of the islands is observed to induce nucleation of misfit dislocations upon annealing at an elevated temperature. The dislocations move from the edges of the islands and end up in the positions corresponding to the maximum strain relief. A significant energy decrease is also observed as a result of a manual introduction of misfit dislocations in systems where the nucleation barrier cannot be overcome by a thermal fluctuation, pointing to a significant gap between the energy balance and force balance criteria for the appearance of misfit dislocations. Simulations of an island located at different distances from the edge of a mesa indicate that the energy of the system decreases sharply as the island position shifts toward the edge. These results suggest that there may be a low energy region near the edge of a mesa where nucleation and growth of an ordered layer of islands is favored.

REFERENCES

[1] D. Bimberg, M. Grundmann, N.N. Ledentsov, *Quantum Dot Heterostructures* (John Wiley & Sons, Chichester, 1998).

[2] *Germanium silicon: Physics and materials*, edited by R. Hull and J. C. Bean (Academic Press, San Diego, 1999).

[3] H. T. Johnson and L. B. Freund, *J. Appl. Phys.* **81**, 6081 (1997).

[4] X. Su, R.K. Kalia, A. Nakano, P. Vashishta, A. Madhukar, *Appl. Phys. Lett.* **79**, 4577 (2001).

[5] M.A. Makeev and A. Madhukar, *Appl. Pys. Lett.* **81**, 3789 (2002).

[6] P. Raiteri, F. Valentinotti, and L Miglio, *Appl. Surf. Sci.* **188**, 4 (2002).

[7] S. Guha, A. Madhukar and K.C. Rajkumar, *Appl. Phys. Lett.* **57**, 2110 (1990).

[8] Y. Chen, X. W. Lin, Z. Liliental-Weber, and J. Washburn, *Appl. Phys. Lett.* **68**, 111 (1996).

[9] V. Gopal, A. L. Vasiliev, and E. P. Kvam, *Phil. Mag. A* **81**, 2481 (2001).

[10] W. Wunderlich, M. Fujimoto, H. Ohsato, *Thin Solid Films* **375**, 9 (2000).

[11] S.Y. Shiryaev, F. Jensen, J.L. Hansen, J. W. Petersen, A.N. Larsen, *Phys. Rev. Lett.* **78**, 503 (1997).

[12] J. Tersoff, C. Teichert, and M.G. Lagally, *Phys. Rev. Lett.* **76**, 1675 (1996).

[13] T.I. Kamins and R.S. Williams, *Appl. Phys. Lett.* **71**, 1201 (1997).

[14] G. Jin, J. Wan, Y.H. Luo, J.L. Liu, and K.L. Wang, *J. Cryst. Growth* **227-228**, 1100 (2001).

[15] Q. Gong, R. Notzel, H.P. Schonherr, K.H. Ploog, *Physica E* **13**, 1176 (2002).

[16] A. Kuronen, K. Kaski, L.F. Perondi and J. Rintala, *Europhys. Lett.* **55**, 19 (2001).

[17] L. Dong, J. Schnitker, R.W. Smith, and D.J. Srolovitz, *J. Appl. Phys.* **83**, 217 (1998).

[18] J. S. Rowlinson, *Liquid and liquid mixtures* (Butterworth Scientific, London, 1982).