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Orientation dependence of growth quality in strained-layer superlattices: A model-potential study of the Si-Ge system

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Growth quality of Si-Ge/Si superlattices in various directions is studied using an empirical-potential-based approach. Kuan and Iyer found, experimentally, that growth morphology is best in the [100] direction while twin formation has substantial effects on growth in other directions. We show that twin formation does not relax misfit strain but instead allows growth to switch over to energetically favorable directions. We discuss the relative stability of strained Si-Ge superlattices for different orientations and the applicability of the results to III-V semiconductor superlattices. We also discuss experimental observations of long-range ordering which occur only in thick relaxed Si-Ge layers grown in the [100] direction and not in thin strained-layer superlattices grown in the same direction.

In this paper, we study the growth quality of strained-layer superlattices in different directions by means of a model potential. There have been many useful studies of semiconductor systems using empirical potentials.[1-8] Ito, Khor, and Das Sarma[7,8] have developed a potential which has been applied successfully to the elemental semiconductors and their binary systems; in particular, they obtained good global fit to cohesive energies for various structures, elastic constants, equilibrium lattice parameters, interplanar distances, and excess energies of various Si-Ge superlattices. This clearly demonstrates the validity of using these potentials in the study of strain effects in binary systems.

In this work, we apply the potential to throw light on the results of recent experimental work on Si-Ge superlattice growth by Kuan and Iyer[9] (KI), which we summarize in Table I. They studied the growth morphology of thin strained-layer superlattices (SLS), comprising 18–20 layers of $Si_{0.5}Ge_{0.5}$ on Si substrates at 400 °C and 580 °C. Growth quality in the [100] direction is very good. There were undulations of small amplitude (2 nm) and long wavelength (100 nm) in the layers (attributed to strain relaxation[9]) of the superlattices grown at 580 °C which we will not address here. In contrast, superlattices grown in the [110] direction exhibit a high density of coherent twins at both temperatures. Twins nucleate at the Si substrate and twin planes are inclined at 35° and 90° to the growth surface. The twinned crystal is such that growth is actually in the [411] direction, which, KI note, may be a more favorable growth surface than [110]. Growth on the [111] surface shows the poorest morphology of the three directions. Twins are present and layers are of irregular thickness. There are few dislocations and KI suggest that twins can relax strain more effectively than dislocations.

TABLE I. Summary of experimental results of Kuan and Iyer (Ref. 9) on growth quality of thin SiGe strained-layer superlattices (SLS's) in different directions. No long-range order was seen in any of the SLS's.

Growth direction **G**	Twin formation	Angle of twin surface with **G**	Growth quality
[100]	None	· · ·	Good; layers flat and uniform at 400 °C; small undulations at 580 °C.
[110]	Twinning	35°, 90°	Smooth layers; growth occurs in the [411] direction.
[111]	High density of twins	· · ·	Poorest morphology; layers of irregular thickness.

KI also studied thick (500 nm), partially relaxed $Si_{0.5}Ge_{0.5}$ layers grown on Si. Layers grown in the [100] direction exhibit long-range order along the [111] direction of the RH2 type, where the stacking sequence is made up of alternating Si-Si and Ge-Ge double layers. Ordering is not observed in thick layers grown in other directions or in any of the thin superlattices grown in any direction. This ordering in thick relaxed [100] layers was also observed by LeGoues, Kesan, and Iyer[10] and the mechanism discussed by LeGoues *et al.*[11] It is not easy to see why this mechanism does not also apply to thin [100] superlattices.

We note that the number of layers grown for the thin superlattices is less than the critical thickness of 70–80 layers (100 Å),[12] before misfit dislocations become the main mechanism for strain relaxation. Below the critical thickness, coherent twins can affect growth quality seriously as seen in the work of KI. Twinning boundaries correspond to the [111] planes, the lattices on either side of the plane are in direct contact and atoms maintain the tetrahedral arrangement in the region[13] (without bond bending or bond stretching if there is no misfit strain). The question then is if twinning relaxes misfit strain. To study this, we determine the elastic strain energies of $Si_{0.5}Ge_{0.5}$/Si superlattices grown in different directions.

Equal numbers of Si and Ge atoms, randomly distributed in a box and initially strained in all directions to Si parameters, are allowed to move via molecular dynamics as the system is slowly relaxed in the growth direction. The temperature is steadily reduced and the final relaxation is by means of the steepest descent method. To simulate twinning, we insert two twin planes in the [111] direction, one in the middle of the box and the other at the end; this allows the use of periodic boundary conditions in the simulations. As above, the system is then relaxed in the growth direction while strain is maintained in the lateral directions.

In Table II, we display the results of our simulations. Twin directions are given by vectors which lie in the three planes formed by [111] and each of the three directions $[\bar{1}11]$, $[1\bar{1}1]$, and $[11\bar{1}]$; these vectors and the growth directions are equally inclined to [111]. Column 4 gives the strain energy of the superlattice without its twin and the entries are arranged in descending order of strain energy. Column 2 shows the corresponding direction of the twin and column 3 lists the angle that the growth direction makes with the twin surface. In Table III, we compare the strain energies obtained for superlattices together with their twins, with the average of the strain energies of the separate surfaces (from column 4 of Table II). The results of Table III show clearly that twins do not relax misfit strain. From Table II, the [001] surface is lowest in energy; its twin $[22\bar{1}]$ lies some distance up the scale. On the other hand, the [110] surfaces have twins at the same energy ($[\bar{1}10]$), and at a slightly lower energy ([114]); growth would tend to switch over to this latter surface from the [110], as was observed by KI. Finally, the [111] surface has the highest energy of all and its twin, $[11\bar{5}]$, lies considerably below it. These results are consistent with the observations of KI in Table I. The [001] surface has the best morphology; it is free of twinning. The [111] surface has the poorest morphology. Both [110] and [111] surfaces show a high density of twins and the angles indicated in column 3 of Table II are observed. While twinning does not relax misfit strain, it does permit growth to switch over to energetically favorable directions. This conclusion is correct even when the energy cost of twin nucleation and faulting at twin surfaces (~60 meV per surface atom[14]) are taken into consideration. Considering only the energetics of twinning and faulting, we estimate domain dimensions (in $[1\bar{1}0]$ cross sections) of [110] SLS's to be of the order of a few hundred angstroms, which is consistent with experimental results.[9] The results of Table II are consistent with harmonic elasticity calculations.[15] Wood and Zunger[15] have shown that the elastic energy of strained superlattices is given by

$$\Delta E(a_s,\mathbf{G})=\tfrac{9}{8}\,q(\mathbf{G})Ba_{\mathrm{eq}}(a_s-a_{\mathrm{eq}})^2\,, \qquad (1)$$

where B is the bulk modulus, and a_{eq} and a_s are the unstrained and substrate lattice parameters, respectively. In addition

$$q(\mathbf{G})=1-B/\{c_{11}+\gamma(\mathbf{G})[c_{44}-(c_{11}-c_{12})/2]\}$$

and

$$\gamma(\mathbf{G})=2[1-(n_1^4+n_2^4+n_3^4)]\,.$$

n_1, n_2, and n_3 are the direction cosines of the growth direction **G**. As $c_{44}-(c_{11}-c_{12})/2>0$ for Si and Ge, the order of the strain energy ΔE is given by $\gamma(\mathbf{G})$, which is seen in the last column of Table I. Using average values of elastic constants and bulk moduli[7] in (1), we get ΔE to be 17.53 and 14.14 meV/atom for $\mathbf{G}=[111]$ and [001], respectively, which agree well with the figures of column 3, Table II.

TABLE II. Strain energies of superlattices in different growth directions.

Growth direction **G**	Twin directions	Angle with twin surface	Strain energy (meV/atom)	γ
[111]	$[11\bar{5}]$	109.5°	18.36	1.333
$[77\bar{8}]$	[114]	74.2°	18.35	1.322
$[22\bar{1}]$	$[00\bar{1}]$	54.7°	18.19	1.185
[110]	[114], $[\bar{1}10]$	35.3°, 90°	17.97	1.000
$[11\bar{4}]$	[114]	74.2°	17.39	0.412
[114]	[110], $[11\bar{4}]$, $[77\bar{8}]$	35.3°, 74.2°	17.38	0.412
$[11\bar{5}]$	[111]	109.5°	17.26	0.280
$[00\bar{1}]$	$[22\bar{1}]$	54.7°	16.95	0.0

TABLE III. A comparison of strain energies of superlattice-twin combinations and the average of the strain energies of the component superlattices.

Superlattice twin	Strain energy of combination (meV/atom)	Average energy of separate surfaces
$[00\bar{1}]$-$[22\bar{1}]$	17.54	17.57
[110]-[114]	17.64	17.67
[111]-$[11\bar{5}]$	17.76	17.80

We now consider the long-range (RH2) ordering seen in thick partially relaxed Si-Ge layers grown on [100] surfaces, but not observed in the thin strained-layer superlattices. Kelires and Tersoff[11] simulated the growth of SiGe alloys epitaxially, on Si substrates, taking into account the dimer reconstruction on the [100] surface. They observed segregation of the components at the surface as indicated in the first four layers of Fig. 1. All second-layer sites and third- and fourth-layer sites immediately below the dimers are under compressive stress, and so for energetic reasons, these sites are occupied by the smaller Si atoms. This tendency to segregate at the top four layers together with two assumptions (growth occurs by the motion of double steps and diffusion below the fourth layer is negligible) form the basis of the mechanism proposed by LeGoues *et al.*[11] for the RH2 ordering along [111] planes. They found it energetically favorable (~40 meV per surface atom) for the ordering to persist along a certain [111] plane rather than zigzagging about this direction. However, RH2 ordering is not observed in thin superlattices grown in the [100] direction, where this mechanism should also apply.

We apply our potential to the study of this problem. As π bonding plays an important role in the reconstructed [100] surface, we take it into account in our simulations.

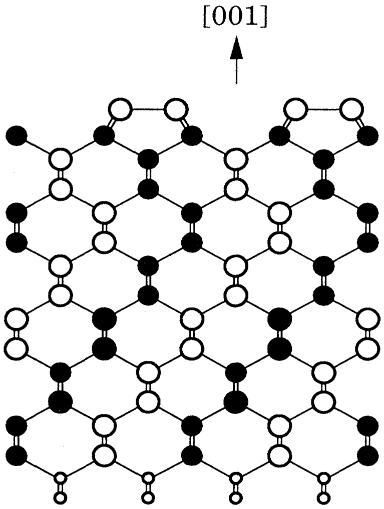

FIG. 1. [110] cross section of the [001] 2×1 dimerized surface. Open circles are Ge atoms. The arrangement is the most favorable one energetically, for the top four layers. Here we show a zigzag configuration. Small circles represent the rest of the system.

Total-energy calculations of Payne *et al.*[16] give the reconstruction energy of the Si [100] surface as 1.01 eV per surface atom and the dimer bond length as 2.23 Å, thus showing its π-bonding character. The surface is under tensile stress of 0.7 eV per 1×1 cell in the direction along the dimer and a compressive stress of 1.9 eV per 1×1 cell perpendicular to it. We modify the potential for surface atoms *only* to reflect the π-bonding character of the dimer bond; this bond is strengthened by 1.35 times (about the strength of a double bond), and the bond-bending constant is increased by a factor of 1.2 (reflecting the flatter geometry at the dimer required by π bonding). This gives a reconstruction energy of 0.98 eV per surface atom, a dimer bond length of 2.28 Å, and surface stresses of 0.7 eV and -1.6 eV per 1×1 cell along and perpendicular to the dimer, respectively. The results reported below are not sensitive to the numerical factors used above.

As a comparison, Kelires and Tersoff[11] obtained a difference in elastic energy of ~50 meV/atom, for the configuration comprising the first four layers of Fig. 1, when Ge atoms are substituted for Si in the second layer while we get 50 meV/atom (unrelaxed) and a 26 meV/atom (relaxed). We also obtain this configuration as being the most energetically favorable when we consider the energy of the top four layers only. In Table IV we show results obtained with 26-layer slab geometries; computations with a 14-layer geometry give essentially the same results. Column 1 gives the energy difference between an RH2 arrangement for the [100] surface and the corresponding zigzag configuration in Fig. 1 (where the top four layers have reversed registry) when the substrate is stressed to Si parameters and when the substrate is relaxed to the parameters of the SiGe alloy. This small difference is not surprising, since the two configurations actually differ only in the sixth layer, where the strain relaxations are already quite small. Our results indicate that the growth mechanism described above is, by itself, not adequate to enforce RH2 ordering along the [111] direction and that the relaxed character of the thick SiGe samples is not essential for ordering. This seems to be consistent with the experimental observation that thin [100] superlattices do not exhibit ordering. The energy differences for a D_B step[17] with RH2 ordering and zigzagged as above are shown in column 2; a double-layer step does not impose ordering. In Fig. 2 we show a D_B step with a D_A kink. The rebondings at the step edges are those found by Chadi[17] to be the most energetically favorable. The presence of a kink puts constraints in the formation of the upper step dimers. If a further layer is grown on the top layer of the configuration in Fig. 2, these constraints will force the dimers of this new layer into RH2 registry. This may be a possible extra condition needed to enforce long-range ordering, which may become observable only after a good number of layers have been grown and ordered domains have become large enough.

TABLE IV. Energy difference, in meV per 1×1 surface cell, between RH2 atomic arrangements and the corresponding zigzag configurations.

	[100] surface	[100] with D_B step
Unstressed	-1.5	-1.7
Stressed	-0.95	-1.3

In conclusion, we have shown that in the growth of thin strained SiGe alloy superlattices, twin surfaces do not relax misfit strain, but permit an easy path for growth to switch over to energetically more favorable twin directions. As a result, growth is best in the [100] direction and poorest in the [111] direction. Our calculations are consistent with the predictions of classical elasticity theory. These results should be applicable to some III-V semiconductor superlattices. We also considered the occurrence of RH2 ordering which appears only in thick relaxed SiGe alloy layers grown on [100] surfaces, and not in the thin strained-layer superlattices. Our results indicate that the mechanism that has been proposed recently for this ordering[11] may need to be supplemented by further conditions.

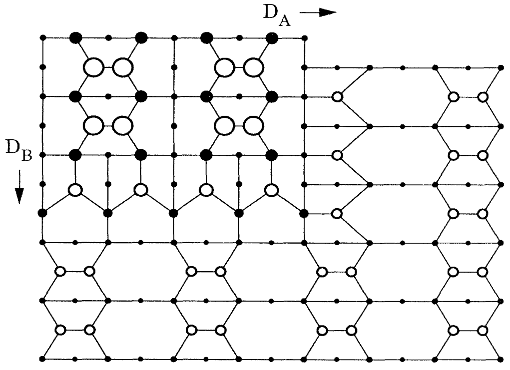

FIG. 2. Surface view of double steps D_B and D_A, in a kink arrangement. Open circles are atoms with dangling bonds.

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