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Peiching Ling
(Ph.D. Thesis)

January 1983

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A MODEL OF AMORPHOUS SILICON LAYER REGROWTH

Peiching Ling
(Ph.D. Thesis)

Lawrence Berkeley Laboratory
University of California
Berkeley, California

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A MODEL OF AMORPHOUS SILICON LAYER REGROWTH

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ABSTRACT

A new model which combines the heterogenous nucleation and growth process for the mechanism of regrowth of amorphous silicon, in contact with a single crystal substrate, is presented. The model explains the effects of substrate orientation and impurity concentration on the kinetics of regrowth and also on the formation of twins during regrowth. The mechanism of redistribution of impurities and the formation of secondary defects during crystall regrowth are also discussed.

A comparison of experimental results with the predictions of the model is also included.
1. INTRODUCTION

The regrowth of amorphous Si layers has several well known characteristics: (1) the substrate orientation affects the regrowth kinetics; (2) impurities have an effect on regrowth kinetics; (3) impurity redistribution and supersaturation can occur due to regrowth; (4) secondary defects are formed during crystal regrowth.

Although several models have been proposed to explain the orientation effects or the impurity effects separately, there is no single theory or model which can explain all these characteristics, together. An attempt will be made here to develop a model which incorporates both heterogenous nucleation of growth steps and their migration and which explains all aspects of the regrowth of amorphous layers. A comparison of the results obtained in this study with recently published experimental findings will be made.

1.1. Amorphous Layer Formation

Amorphous silicon layers can be formed by several techniques such as vapor deposition, sputtering and ion implantation. However, unless stringent surface cleaning precautions are taken and depositions are carried out in ultra-high vacuum, the presence of surface oxidation or impurity layers on the crystal substrate prevents direct contact between the amorphous layers and substrate. This interface layer between the amorphous and single crystal regions blocks epitaxial growth during elevated temperature annealing. Ion implantation is used in this study since it allows precise impurity control and precludes the possibility of the existance of an oxide and/or contamination at the original crystalline amorphous interface. Also the regrowth of the
amorphous layers created by ion implantation is of practical importance to the semiconductor industry.

A first ion implantation step with self ions is used to create an amorphous layer free of impurity. A second implantation step can then be used to introduce a known amount of impurities into the specimen, if impurity effects are being investigated. If the implantation dose is above a critical level, an amorphous layer is formed. The thickness of this layer is of the order of $2R_p$, where $R_p$ is the average projected range of the incident ions.\(^4\)

There are several variables that affect the ion dose required to form an amorphous layer. The ion species is critical\(^5\) since heavy ions cause amorphitization more readily. The ion accelerating voltage\(^6\) is not of major importance in determining the critical dose. Care must be taken to prevent substrate heating during ion implantation. This is because a dislocation network can form in the transition region between the amorphous and crystalline material at elevated temperatures.\(^7,8,9\) In some cases, buried amorphous layers instead of total amorphitization are found.\(^10,11\) These problems can be avoided by maintaining the substrate near liquid nitrogen temperatures and by maintaining low beam current density.\(^12\)

1.2. The Kinetics of the Regrowth Process

(a) The Orientation Effect. The regrowth of epitaxial Si from implanted-amorphous layers is strongly dependent on the orientation of the underlying substrate\(^13\) as shown in Fig. 1. The regrowth rate for a $<100>$ sample is about three times that for $<110>$ samples and
about 25 times the initial regrowth rate for <111> samples. The regrowth rate for <111> oriented Si samples is nonlinear in annealing time. The slopes of Arrhenius plots of the regrowth rates show an activation energy of 2.35 eV.

It was found that twins are the major defects in regrown <111> samples. L. Csepregi et al. tried to use twin formation to explain the nonlinear dependence of the growth rate in <111> samples. Twins in a <111> oriented sample imply the presence of a <111> plane inclined to the surface. The axial direction in the twin normal to the surface is a <511> direction. Since the growth rate in the <511> direction is much faster than that in the <111> direction, one might expect that the average growth rate in regions containing twins would be increased. There are two difficulties with this model:

(i). Although twin <111> corresponds to <511> of the matrix, the twin-amorphous interface still tends to develop as a <111> facet so the growth rate should be unaffected.

(ii). If the twin <111> growth rate were the same as that along the <511> direction, the growth rate at a region containing twins should be extremely rapid compared to the initial growth rate. The ratio of growth along <511> to that along <111> is about 20. From experimental data however, we know that the ratio of initial and final <111> regrowth rate is just about 2.5 times. This nonlinear effect will be discussed later. (6.1)

There are several kinds of models to explain the orientation effect, e.g. (1) geometric model (2) stress relaxation model (3) surface reconstruction model. Here we discuss the model proposed by R. Drosd and J. Washburn.
The model is based on the ability of an atom leaving the amorphous phase to find a stable bonding site at the crystal surface. To do this, the incoming atom must complete two of its four possible bonds with the established crystal. On the <100> silicon surface a single atom may successfully attach itself to the crystal; but on a <110> surface, a pair of atoms is required; and a cluster of three is needed on a <111> face. The schematic drawing in Fig. 2 is a [110] projection of a diamond cubic lattice where the critical size nucleus on each crystal face is shown. On the <111> face it is seen that there are two positions in which the group of three atoms can attach. In the one labeled "correct" the crystal has been continued, defect free. The nucleus labeled "incorrect" would start the growth of a twin. Since the atoms in the twin-oriented nucleus make only second nearest neighbor mistakes such nuclei are expected to occur frequently. Stacking mistakes on the <100> or <110> surfaces would involve first nearest neighbor mistakes and hence are unlikely to occur.

This model explains the major features of amorphous layer recrystallization. However, it cannot explain the impurity effect on the regrowth rate and the observed activation energy.

(b) The Impurity Effect. The presence of impurities can have a pronounced influence on regrowth kinetics.\textsuperscript{18,19} Table 1 lists some representative values of the regrowth rate for various impurities. It was shown that the presence of B, P, and As increase the regrowth rate, but C, O, and Ar reduce it. The apparent activation energies for growth of impurity implanted layers were found to be quite similar.
Table 1. Some representative values of the regrowth rates for various impurities

<table>
<thead>
<tr>
<th>Impurity</th>
<th>Concentration (atoms cm$^{-3}$) x 10$^{-20}$</th>
<th>Regrowth rate, Å min$^{-1}$</th>
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<tr>
<td>$^{11}\text{B}$</td>
<td>2.5</td>
<td>1028$^a$</td>
</tr>
<tr>
<td>$^{12}\text{C}$</td>
<td>1.8</td>
<td>55</td>
</tr>
<tr>
<td>$^{14}\text{N}$</td>
<td>2.5</td>
<td>7.7</td>
</tr>
<tr>
<td>$^{16}\text{O}$</td>
<td>2.4</td>
<td>9.0</td>
</tr>
<tr>
<td>$^{20}\text{Ne}$</td>
<td>2.8</td>
<td>3.5</td>
</tr>
<tr>
<td>$^{28}\text{Si}$</td>
<td>---</td>
<td>85</td>
</tr>
<tr>
<td>$^{31}\text{P}$</td>
<td>2.5</td>
<td>527$^a$</td>
</tr>
<tr>
<td>$^{40}\text{Ar}$</td>
<td>1.5</td>
<td>2.4</td>
</tr>
<tr>
<td>$^{74}\text{Ge}$</td>
<td>2.0</td>
<td>87</td>
</tr>
<tr>
<td>$^{75}\text{As}$</td>
<td>2.0</td>
<td>480$^a$</td>
</tr>
</tbody>
</table>

$^a$Extrapolated value from 500°C. (Data taken from Ref. 3)
Data on growth rate vs. $1/T$ are plotted in Fig. 3 for different impurities. Of the species represented, B produces the strongest increase, and O the strongest decrease in growth rate. Although the data are taken over relatively narrow temperature ranges the slope of the plot for B is somewhat less than, and that of the O data and somewhat greater than, the slope for impurity free Si.

Recently, Suni et al., found that compensated layers with equal concentrations of B and P or B and As showed a strong decrease in the regrowth rate whereas for the layers with overlapping As and P profiles no compensation was found.

The Turnbull model of diffusion-like growth process cannot explain this result. In the Turnbull model the growth rate $U$ can be expressed as

$$U = \lambda \nu \exp(-\Delta g_a/kT)(1-\exp(-|\Delta g|/kT))$$

(see Section 2.3).

Here

- $\lambda$ is the atom jump distance
- $\nu$ is the atomic vibration frequency
- $\Delta g_a$ is the activation Gibbs free energy
- $\Delta g$ is the Gibbs free energy difference between the two phases per atom.

According to this relationship, at low temperature $U$ is approximately independent of $\Delta g$ (impurity concentration). From experimental data however, we know that at low temperature the growth rate is dependent on impurity concentration and the lower the temperature the larger the difference.
P. Ling and J. Washburn\textsuperscript{23} have proposed that all impurities accelerate the regrowth rate when present in a concentration below their solubility limit in crystalline Si. Conversely, all impurities will decrease the regrowth rate when present in a concentration above their solid solubility limit, though this is only a qualitative model. There are no other theories or models that can completely explain the impurity effect during regrowth.

1.3. The Supersaturation, Redistribution Effect and Secondary Defect Formation during Regrowth

Substitutional impurity concentrations in excess of the solid solubility have often been observed in regrown Si. Examples are Al, Sb, Ga, Ba and As.\textsuperscript{24,25,26} The redistribution of implanted species in regrown layers has been investigated quite extensively.\textsuperscript{24,27,28} The results are summarized below:

1. annealing at low temperature (\textdegree550°C) of amorphous Si layers obtained by implantation of species such as In, Sb, and Pb on \textlangle 100\rangle, \textlangle 110\rangle, and \textlangle 111\rangle samples leads to epitaxial regrowth. Redistribution of these implanted species was not observed. However redistribution of Al was found even at 480°C.

2. Direct annealing of these samples at high temperature can cause major redistribution, and in some cases significant loss of implanted species is observed in \textlangle 111\rangle samples. For \textlangle 100\rangle and \textlangle 110\rangle samples, epitaxial regrowth occurs with varying degrees of minor redistribution and loss of implanted species depending on the implantation energy and species.
Usually annealed ion-implanted silicon has one or two layers of secondary defects. The defects include interstitial loops, precipitates, twins and, in some cases polycrystalline Si.\textsuperscript{29,30,31,32} Annealing of the initial point defects and amorphous clusters produced by the ion implantation explains the formation of the deeper buried layer of secondary defects.
2. A MODEL OF AMORPHOUS LAYER REGROWTH

2.1. Assumptions

In this section, a thermodynamical model of amorphous layer regrowth based on heterogenous nucleation and migration of growth steps will be discussed. It is assumed that

(1) The nuclei are disc shaped and one or two atomic layers thick at the amorphous-crystalline interface. The number of atomic layers will be discussed in 3.1.

(2) There exists an elastic stress to balance the surface tension and pressure inside and outside the nucleus. The strain energy is negligible.

(3) New nuclei can form on the top of expanding nuclei. This is equivalent to saying that the total nucleation rate is constant all the time during the regrowth process, since the total interface area for nucleation is constant.

(4) The impurity concentration does not change during regrowth and the one-dimensional migration rate of the growth steps is constant.

(5) The regrowth rate \( R \) is defined as \( R = a/t^* \), where \( a \) is the disc thickness, \( t^* \) is the time at which the area swept by growth steps is equal to the substrate area.

Based on these assumptions, the regrowth occurs as follows: after nucleation all the disc shaped growth steps at the amorphous-crystalline interface will migrate in a direction parallel to the faces of the disc (expansion). When they grow large enough, these growth steps meet each other and form a complete (crystalline) layer of atoms over
the entire interface. The amorphous-crystalline interface has advanced into the amorphous region by a distance equal to the height of the growth steps and the regrowth rate is $a/t^*$. 

2.2. The Nucleation Rate of Growth Steps

Consider the formation of the disc shaped nuclei on the amorphous-crystalline interface. It is assumed that the nuclei are disc shaped, but this model (with suitable modifications) is applicable to any shape. When a nucleus forms, a new interface will be created around its edge.

Let: $g_a$ be the Gibbs free energy per atom for the amorphous phase
$g_c$ be the Gibbs free energy per atom for the crystalline phase at the same pressure and temperature as the amorphous phase
$\Delta g$ is then the free energy change per atom of the regrown Si, $g_c - g_a$.

If the elastic strain energy is neglected, the total free energy change $\Delta G$ in making a disc nucleus, of radius $r$ and thickness $'a'$ (as shown in Fig. 4), is given by

$$\Delta G = \pi r^2 a \rho \Delta g + 2 \pi r a \sigma$$

(2.1)

and $\rho$ is the number of atoms per unit volume in the crystalline phase. $\sigma$ is the surface energy of the surface around the edge of the nucleus.

$\Delta G$ has the maximum value $\Delta G^*$ at $r = r^*$. It is possible to compute $r^*$ by taking the derivative of $\Delta G$ with respect to $r$ and setting it equal to zero.

$$2 \pi r^* a \rho \Delta g + 2 \pi a \sigma = 0$$

$$r^* = - \sigma / \rho \Delta g$$
and then

\[ \Delta G^* = -\pi a^2 / \rho \Delta g. \] (2.2)

The number \( N_c \) of critical nuclei per unit area with radius \( r^* \) is given by \( N_c = N_S \exp(-\Delta G^*/K T) \). \( N_S \) is the number of atoms per unit area.

The rate of nucleation \( I \), that is, the number of discs going over the energy hump \( \Delta G^* \) per unit time and area, is given by multiplying the number of critical nuclei by the rate of growth of the disc. Neglecting the reverse reaction, this can be expressed as:\(^{21}\) (see Section 2.3)

\[ I = N_c S^* v \exp(-\Delta g_a / K T) \]

Also,

\[ I = S^* v N_S \exp(-\Delta G^*/K T \exp(-\Delta g_a / K T) \] (2.3)

\( S^* \): the number of atoms facing the critical nuclei in the amorphous phase

\( v \): the number of times per second an atom tries to jump the barrier

\( \Delta g_a \): the activation energy for an atom to cross the interface between the amorphous and crystalline phase.

2.3 The Growth Step Migration Rate

The derivation which follows is given by Turnbull.\(^{21,22}\) Referring to Fig. 5, \( \Delta g_a \) is the activation Gibbs free energy for one silicon atom to leave the amorphous phase, cross the interface, and attach itself to the growth step. \( \Delta g \) is as defined before. The thickness of one atomic layer is defined as \( \lambda \). That is, if one atom from the amorphous phase jumps across the interface to the crystalline phase, the
crystal grows at this point by $\lambda$. If $S$ atoms in the amorphous phase cross the interface, the rate of transfer of silicon from the amorphous phase to the crystalline phase is $S\exp(-\Delta_g a)/KT$. The rate of transfer of Si from the crystalline phase to the amorphous phase is $S\exp(-\Delta_g a + |\Delta g|)/KT$. The net transfer of atoms from amorphous phase to the crystalline phase per unit time is therefore $S\exp(-\Delta_g a)/KT [1-\exp(-|\Delta g|)/KT]$.

If $S$ atoms are transferred to the crystalline phase, the crystalline phase grows one layer. The radius growth rate, $U$, of a disc shaped growth step is equal to the product of $\lambda$ and the number of atoms transferred per unit time divided by $S$.

$$U = \lambda S\exp(-\Delta_g a)/KT [1-\exp(-|\Delta g|)/KT].$$ (2.4)

2.4 Regrowth Rate. Combined Nucleation and Growth at Constant Temperature.

Suppose that a particular growth disc nucleated at a time $t'$. The radius of the disc at time $t$ where $t > t'$ is given by $U(t-t')$, then the surface of this particular disc area $A_i$ will be given by the following expression.

$$A_i = \pi U^2(t-t')^2.$$ (2.5)

The total number of nuclei forming at time integral $dt$ around $t'$ can be expressed as $IA_0 dt$. Where $I$ is the nucleation rate per unit area, and $A_0$ is the total amorphous-crystalline interface. Since we assumed that new nuclei can form on the top of growing discs, $IA_0$ is independent of time and then the total area $A$ of all discs at time $t$ can be expressed as
$$A = \int_{t' = 0}^{t' = t} SIA_0 dt'.$$

From Eq. 2.6, we can find the time $t^*$ at which the total area of all growing discs is the same as the total area of the amorphous-crystalline interface. This is

$$A_0 = \int_{t' = 0}^{t' = t^*} A_1 I A_0 dt'.$$

$$A_0 = \int_{t' = 0}^{t' = t^*} \pi U^2 (t^* - t')^2 I A_0 dt.$$

$$A_0 = \frac{\pi U^2 t^*^3}{3} I A_0$$

$$\frac{1}{t^*} = \left( \frac{\pi}{3} U^2 I \right)^{\frac{1}{3}}$$

then one can calculate the regrowth rate $R = \frac{a}{t^*}$.

$$R = a \left( \frac{\pi}{3} U^2 I \right)^{\frac{1}{3}}$$

in substituting for $I$ and $U$ into this equation, one obtains

$$R = a \left( \frac{\pi}{3} N_{s^*} \lambda^2 S^* \right)^{\frac{1}{3}} \exp(-\Delta G^* / 3K T \exp(-\Delta g_a / K T)) \left( 1 - \exp(-|\Delta g| / K T) \right) .$$

For $N_{s^*} \lambda^2 - 1$ one can make a low temperature approximation

$$R = a v \left( \frac{\pi}{3} S^* \right)^{\frac{1}{3}} \exp(-\Delta G^* / 3K T \exp(-\Delta g_a / K T) .$$

(2.8)
3. MODEL PREDICTIONS

The regrowth rate at low temperature can be expressed as

\[ R = a v \left( \frac{\pi S^*}{3} \right)^{1/3} \exp[-\Delta G^*/3kT] \exp[-\Delta g_a/kT] \]

where

\[ \Delta G^* = -\pi a \sigma^2/\rho \Delta g. \]

Usually \( \Delta g_a \) is not sensitive to substrate orientation and impurity concentration if the impurity concentration is low. Although 'a' (plane distance) is different in different orientations, it results in only a small difference in regrowth rate. The main difference arises from \( \exp[-\Delta G^*/3kT] \). Thus the main factor which influences the orientation effect is the difference in nucleation rates for different orientations. Similarly the prime reason for the occurrence of the impurity effect is the difference in nucleation rate with differing impurities. The growth (migration) rate of the disc shaped nuclei is nearly the same in all cases.

3.1. The Orientation Effect

The orientation effect can be deduced from the model shown above and some geometrical facts, as shown below.

The chemical bond between silicon atoms is a covalent bond. At the interface between amorphous and crystalline silicon, each atom in the crystalline surface has incomplete or distorted bonds toward the amorphous phase. When the regrowth beings, single atomic layer embryos are formed at the interface. The increase in the number of incompletely or distorted bonds associated with each embryo will cause an increase
of surface energy. Therefore the greater the number of incomplete or distorted bonds, the smaller the regrowth rate.

(a) \(<100>\) Orientation. If we examine the lattice of silicon along a \(<100>\) direction we can see that each atom in a \({100}\) atomic layer has four nearest neighbors, two in the layer below and two above, which form four first order covalent bonds around the atom as shown in Fig. 6a. When an embryo is formed, each atom which is not at the edge of the embryo has the same energy condition (bonding condition) as the original interface. However, at the edge of this embryo, each atom has two incomplete first neighbor bonds and some incompleted or distorted second neighbor bonds. These incomplete second neighbor bonds cause an increase in the surface energy around the embryo. Such changes of energy are smaller than the changes caused by incomplete or distorted first neighbor bonds.

(b) \(<110>\) Orientation. Each atom in a \({110}\) atomic layer has four first neighbor covalent bonds, two of them in the same atomic layer, one above and the last one below this layer, as shown in Fig. 6b. Since atoms on the top surface of the embryo have the same energy condition as the original amorphous-crystalline interface, it isn't necessary to consider the atoms which are not at the edge of an embryo. At the edge of an embryo, each atom has zero, or one or two more incomplete or distorted first neighbor covalent bonds than atoms inside the embryo. The increase in the number of incomplete or distorted first neighbor bonds causes the surface energy to increase, and such variation of surface energy around this edge of the embryo is much larger than the variation of surface energy of \(<100>\) oriented silicon. This
is the reason that the regrowth rate of <110> oriented silicon is smaller than <100> oriented silicon.

(c) <111> Orientation. The first neighbor bond structure of an atom in a {111} atomic layer is shown in Fig. 6c, three bonds are on one side of this layer, and the last one is on the other side. It is reasonable to assume that all the atoms at the crystalline surface of the original amorphous-crystalline interface have only one incomplete or distorted first order covalent bond along a direction toward the amorphous phase.

When regrowth begins, a silicon atom in the amorphous surface returns to a suitable site to help form an atomic layer of crystalline phase. But after such process, there will be three incompleted or distorted first neighbor bonds for each atom on the newly formed crystalline phase, these incompleted bonds cause a great increase in surface energy. Hence, the <111> substrate cannot form a single atomic layer embryo (nucleus) and the stable nucleus should have two atomic layers as discussed below. Therefore, the number of nuclei in a <111> oriented wafer must be much smaller than in <110> and <100> oriented wafers. Furthermore, the regrowth rate of <111> wafers is the smallest among these three differently oriented wafers.

When three atoms (which are nearest neighbors to each other in {111} plane) have returned to their suitable crystalline sites, a fourth atom will attach on the top of these three atoms to reduce the surface energy. Therefore for <111> oriented silicon, it is energetically favorable that each nucleus has two atomic layers. The top layer has the same bonding condition as the original amorphous-crystalline
interface, and the atoms inside the nucleus have, in all, four nearest neighbors. All the atoms around the edge and on the bottom layer of the nucleus have one more incomplete or distorted bond more than the original crystalline interface atoms.

3.2. The Impurity Effect

(a) The Kinetics Effect. First it is assumed that the Gibbs free energy per atom of the amorphous phase is higher than that of the crystalline phase if the amount of impurity is lower than the maximum solubility, $C_m$, in this crystalline phase. At a critical concentration, $C^*$, the Gibbs free energy is the same in both phases as shown in Fig. 7. Assuming that Gibbs free energy curves are parabolic, the difference $\Delta g = g_c - g_\alpha$ is also parabolic and has a minimum value around $C_m$. As the amount of impurity increases, $\Delta g$ will first decrease (i.e., become more negative), reach its minimum value, and then increase (i.e., become less negative) and approach zero. From Eq. (2.2) $\Delta G^* = -\pi a^2 / \rho \Delta g$ will decrease with a decrease in $\Delta g$ and increase with an increase in $\Delta g$; $\Delta g$ is negative. This tells us that the driving force for regrowth should first increase with all kinds of impurities if $\exp(-\Delta G^*/kT)$ is the rate controlling factor and would be maximum when $\Delta g$ is minimum, (at an impurity concentration around $C_m$.) The regrowth rate might then decrease with further impurity increase and approach zero at $C^*$. If the amount of impurity in the amorphous phase is higher than $C^*$, the amorphous-crystalline phase transformation cannot occur directly. In this situation, either a precipitate would first or there would be no phase transformation.
(b) **Supersaturation effect.** When the impurity concentration is larger than \( C_m \) and smaller than \( C^* \), the Gibbs free energy of the amorphous phase is still larger than that of the crystalline phase, so the amorphous phase will initially transform to the crystalline phase although the crystalline phase is not stable; this is a metastable phase. Since the amorphous-crystal-line transformation is short range rearrangement, this metastable crystalline phase will decompose to two phases [crystalline phase with maximum solubility and impurity precipitate] when there is sufficient thermal energy. This is consistent with published experimental data.\(^{24,25,26}\)

### 3.3 The Activation Energy of Crystal Regrowth

The activation Gibbs free energy can be expressed as \( \Delta E - T \Delta S \) where \( \Delta E \) is the internal energy and \( \Delta S \) is the entropy change during the transfer of an amorphous silicon atom to the crystalline phase. The regrowth rate can be rewritten as:

\[
R = a \nu \left( \frac{\pi}{3} S^* \right)^{\frac{1}{3}} \exp \left( \frac{\Delta S}{K} \right) \exp \left( \frac{-\Delta G^*}{3} - \frac{\Delta E}{K} \right) / KT \quad (3.1)
\]

From Eq. (3.1) it is seen that the activation energy for regrowth can be expressed as \( \Delta E + \Delta G^* / 3 \) and \( \Delta E \) is not sensitive to low concentration. From this it follows that the activation energy will increase with increasing \( \Delta G^* \). From the former discussion, the growth rate will decrease with increasing \( \Delta G^* \). Hence it is concluded that the larger the growth rate, the smaller will be the activation energy and the smaller the growth rate, the larger will be the activation energy.
4. REDISTRIBUTION OF IMPURITY AND SECONDARY DEFECT FORMATION DUE TO REGROWTH

One of the assumptions of the model of the regrowth rate is that the impurity concentration does not change during regrowth. In discussing the redistribution of impurities, this assumption will be modified. Secondary defect formation is usually influenced by redistribution, and these will be discussed together below.

4.1 The Redistribution Effect

The problem of redistribution of impurities can be solved by using the moving boundary diffusion equation,

\[
\frac{\partial C_\alpha}{\partial t} = D_\alpha \frac{\partial^2 C_\alpha}{\partial x^2} + R \frac{\partial C_\alpha}{\partial x}
\]  

(4.1)

\[
\frac{\partial C_c}{\partial t} = D_c \frac{\partial^2 C_c}{\partial x^2} - R \frac{\partial C_c}{\partial x}
\]  

(4.2)

where

- \( R \) = regrowth rate
- \( D_\alpha, C_\alpha \) = diffusivity and impurity concentration in amorphous phase
- \( D_c, C_c \) = diffusivity and impurity concentration in crystalline phase.

Unfortunately, a lack of information about the thermodynamical properties at the amorphous-crystalline interface is a problem in solving Eqs. 1 and 2. For example, does the amorphous-crystalline interface reach equilibrium during the regrowth process? Does the ratio of impurity concentration between amorphous and crystalline phase at the
interface change with the variation of impurity concentration? In other words, the appropriate initial and boundary conditions of Eqs. (4.1) and (4.2) are not known. Even in this case, it is still possible to get some qualitative results from the comparison of the values between regrowth rate $R$ and diffusivity $D$.

(1) Let diffusivity $D$ be constant. During regrowth the time for which impurities stay in the amorphous phase is decided by the crystal regrowth rate. The lower the regrowth rate, the longer the time that impurities stay in the amorphous phase. The longer the time for the diffusion of impurities in the amorphous phase, the more redistribution of the impurities. Furthermore, if the concentration of impurities is higher than the maximum solubility concentration, the extra impurities may be carried by the moving interface if the growth rate is small.

(2) Let regrowth rate $R$ be constant. A larger diffusivity will cause more redistribution of impurities, and a smaller diffusivity will cause less redistribution.

From above arguments, it is possible to arrive at two conclusions.

(a) The impurities will tend to redistribute if $R/D$ is small. If the concentration of impurities is higher than the maximum solubility concentration, the extra impurities will be carried by the moving interface.

(b) The redistribution of impurities will be less if $R/D$ is large, and in this case, supersaturation in the regrown crystal can occur if the initial impurity concentration in the amorphous phase is higher than the maximum solubility.
Moveover, two predictions can be obtained from the above arguments.

(a) The impurities will be more redistributed in \(<111>\) orientation than in \(<100>\) orientation since the regrowth rate of \(<111>\) orientation is much slower than the regrowth rate of \(<100>\) orientation.

(b) As discussed in the former section, it is known that the regrowth rate reaches a maximum value if the concentration of impurities has a value around the maximum solubility concentration. This implies that the impurities will be less redistributed when the value of impurity concentration comes close to the maximum solubility concentration.

4.2 The Formation of Secondary Defects

Usually, regrown ion-implanted silicon has one or two secondary defect layers. The defects include interstitial dislocation loops, precipitates, twins, and in some cases polycrystalline silicon. The formation of twins will be discussed in 6.1. The defects of the deeper layer are interstitial dislocation loops.\(^{33}\) Annealing of the initial point defects and amorphous clusters around the amorphous-crystalline interface produced by the ion implantation explains the formation of interstitial dislocation loops.\(^{16,17}\)

If the impurity concentration is larger than the maximum solubility, the regrown amorphous silicon will form an upper layer of defects.

There are two cases of upper layer defects.

(1) If \(R/D\) is large, then the amorphous silicon transforms to crystalline silicon with supersaturated impurity. In this case, it is difficult to redistribute the impurity. The only way to compensate the extra impurity is to form crystalline defects to trap the impurity
e.g. formation of loops. After further annealing it will form precipitates. The formation and types of precipitate will depend on the impurity diffusivity in the crystalline phase and the formation energy of the precipitate e.g. strain energy, surface energy of the precipitates.

(2) If $R/D$ is small, the impurity will move with the interface. In this case the impurity segregates at the interface and should retard the regrowth rate, a precipitate then forms around the amorphous-crystalline interface. Later, polycrystalline silicon will nucleate around the precipitates. There is another possibility. The precipitates forms in the amorphous phase, if the diffusivity is very large (interstitial type), before the interface moves to the higher concentration region, and then polycrystalline silicon forms around the precipitates.
5. EXPERIMENTAL PROCEDURE

5.1 Ion Implantation Procedure and Starting Materials

Silicon wafers of n type (2-5 ohm-cm) were obtained from a commercial supplier (Monsanto). They were in the form of 3" discs of 0.017" thickness. Prior to implantation they were cleaned using the Piranet\textsuperscript{34} etch followed by boiling Trichloroethylene (T.C.E.) and a rinse in distilled water. The wafers were then attached to the ion implantation specimen holder using diffusion oil mixed with silver powder as paste. This was to ensure good thermal contact to the holder. Ion beam currents were maintained sufficiently low during implantation so that the sample remained near -180°C during the implantation. The implantation schedules listed in Table 2 consist of, first, a low temperature Si implant to form the amorphous layer and, second, implantation of the desired impurity species.

5.2 Annealing Procedure

The samples were annealed in a conventional tube furnace in an atmosphere of flowing nitrogen. A thermocouple placed beside the specimen allowed the temperature to be controlled within two degrees of the desired temperature.

5.3 RBS Measurement

To measure the thickness of the amorphous layer and the regrowth of the layer following thermal annealing, the channeling-backscattering techniques was used. The details of this techniques has been discussed by W. Chu et al.\textsuperscript{35} Channeling measurements were performed with 1.5 MeV $^4\text{He}^+$ at normal incidence with the detector mounted at 170° to the incident beam direction. The 1.5 MeV $^4\text{He}^+$ beam was accelerated in a Van de Graaff generator with an energy resolution of ± 500 eV.
Table 2. Implantation sequences employed to created amorphous layer on crystalline Si doped with O and Al

<table>
<thead>
<tr>
<th></th>
<th>Energy KeV</th>
<th>Dose cm⁻²</th>
<th>Energy KeV</th>
<th>Dose cm⁻²</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>100</td>
<td>1x10¹⁶</td>
<td>160⁺</td>
<td>60</td>
</tr>
<tr>
<td>B</td>
<td>100</td>
<td>1x10¹⁶</td>
<td>27Al⁺</td>
<td>90</td>
</tr>
<tr>
<td>C</td>
<td>100</td>
<td>1x10¹⁶</td>
<td>27Al⁺</td>
<td>90</td>
</tr>
</tbody>
</table>
6. RESULTS AND DISCUSSION

6.1 Comparison of Experimental Results with Predictions of the Model

(a) The Orientation Effect. L. Csepregi et al.\textsuperscript{13} published extensive results on the variation in regrowth rates of ion implanted amorphous silicon layers with different substrate orientations. They found that the \textit{<100>} growth direction is the fastest, with the \textit{<110>} being about 2.5 times slower and the \textit{<111>} about 25 times slower. The \textit{<111>} growth rate is nonlinear. The final growth rate is about three times the initial growth rate. Their experiment were repeated here with the same results. These are consistent with the predictions of the above model taking into account the different surface energies associated with the edge of the disc shaped growth step nucleus.

(b) The \textit{<111>} Nonlinear Growth Rate. The number of nuclei for \textit{<111>} substrate orientation is very small as discussed in 3.1. There is a large probability that the top surface of an expanding growth step nucleus becomes a new nucleation site. Such a process can lead to the formation of ledges. The growth form might be polygonal instead of disc shaped. The ledge surface should tend to have the lowest energy and the lowest energy interface is \textit{<111>} with one first neighbor incomplete\textsuperscript{36} bond. This faceting effect increases the amorphous–crystalline interface. The nucleation rate is proportional to the interface area. Hence the nucleation rate will be increased with increased faceting of the surface. This can explain the \textit{<111>} nonlinear regrowth rate for \textit{<111>} Si. For a \textit{<111>} wafer the maximum interface area would be three times the initial interface area. This
could explain why the final regrowth rate is about three times the initial growth rate. One thing should be mentioned here. In other orientations there can also be faceting. However in these cases it would lead to a decrease in crystal growth rates because the facet surfaces would still tend to be \{111\}.

(c) The Impurity Effect: Kinetics and Activation Energy.

Arrehenius plots of the regrowth rate for \(<100>\) Si implanted with Al and O with two different concentrations are shown in Fig. 8. This figure shows that the regrowth rate increases with increasing Al concentration when the Al concentration is below the maximum solubility concentration, and the activation energy of both are around 2.35 eV. It also shows that the regrowth rate decreases with increasing oxygen concentration when the concentration of oxygen concentration is higher the maximum solubility. The activation energy with \(1 \times 10^{20}/\text{cm}^3\) oxygen is 2.6 eV, while the activation energy with \(2 \times 10^{20}/\text{cm}^3\) oxygen is 2.8 eV.

These activation energy measurements are consistent with the predictions in Section 3.3, which suggest that a slower regrowth rate has a larger activation energy. The regrowth rate measurements are also consistent with the predictions in Section 3.2a.

In Section 3.2a, it was shown that all impurities should accelerate the regrowth rate when the concentration of impurities is below their solubility limit in crystalline Si. Conversely, all impurities should decrease the regrowth rate when the concentration is above the solubility limit. From this point of view, the data shown in Table 1, can
be rationalized; e.g., why B, P and As always enhance the regrowth rate and C, O, Ar decrease the regrowth rate. For B, P and As, the concentrations have been measured in the region just below or around the maximum solubility,\textsuperscript{37} therefore those elements have always been observed to increase the regrowth rate. For C, O, Ar, etc. the maximum solubility is very small and the effects of those elements on regrowth rate have been measured only in the regrowth of the maximum solubility limit. Therefore those elements have been always observed to decrease the regrowth rate.

Recently, the regrowth rates of $\langle 100 \rangle$ Si implanted with Sb, Te, and As\textsuperscript{38,39,40} have been observed to first increase with increasing the concentrations of Sb, Te and As, to a maximum value around their maximum solubility concentration, and then decrease with the increasing of the concentration.

The lack of knowledge of how $\Delta g$ changes specifically with different impurities makes it difficult to predict which elements will have the largest enhancement effect.

From the ternary phase diagrams it is possible to explain why the compensated layers with equal concentration of B and P or B and As show a strong decrease of the regrowth rate whereas for the layer with overlapping As and P profiles no compensation has been found.

(d) Impurity Redistribution Effect. Figure 9a (data taken from Ref. 26) shows the backscattering spectra for a $\langle 100 \rangle$ silicon sample implanted at $-190^\circ C$ with Sb\textsuperscript{+}, $10^{10}$ ions/cm\textsuperscript{2} at 150 keV, both as-implanted and after an anneal at 1000°C. Figure 9a shows the backscattering spectra for a $\langle 111 \rangle$ silicon with the same ion implanted and
annealing condition as Fig. 9a. These figures show that Sb redistributes more in <111> orientation than in <100> orientation. This is consistent with the model that a slow regrowth rate will lead to more redistribution.

Figure 10 shows (data taken from Ref. 28) the antimony redistribution effect with different concentrations at different annealing temperatures in <111> Si. Figure 10a has the lowest concentration and the least redistribution. Figure 10c has the highest concentration and the greatest redistribution effect. From the published data, it is found that the regrowth rate corresponding with Fig. 10a is maximum. From Section 3.2a, it was predicted that the regrowth rate corresponding to Fig. 10c should be slowest. This is consistent with the model that the slower regrowth rate will correspond to the greater redistribution effect.

6.2 The Formation of Twins During Regrowth

(a) <111> Substrate Case. The following model is a modification of one described by Drosd and Washburn to explain the formation of twins. From Section 3.1 it is known that the embryo for a {111} face should be two atomic layers thick. When three nearest neighbor atoms on {111} transfer to the crystalline phase then another atom will attach itself on the top of these three atoms. The position of the first three atoms will determine whether a twin is nucleated. From Fig. 11 it can be seen why and how twins form during crystal growth on the {111} plane. Suppose the first two atoms bond to the crystalline phase in a column at position 'a'. If a third atom is attached at position 'b', then the next atom will bond to the a and
b atoms at position 'd'. There will be no twin formation with this nucleus. On the other hand, if the third atom is located on atomic column 'c' instead of 'b', the fourth atom would attach at position 'e', and a twin will be formed. The choice of the first three atom attachments from the amorphous phase to the crystalline phase are random. That means it should be very easy to form a twin on a {111} plane. If the faceting effect is taken into account with the formation of a new nucleus on one of the facets, than the (111) regrown wafer will have not only (111) twins, but also other {111} twins and secondary twins. These 111 twins and secondary twins are commonly found in <111> regrown silicon. 41

(b) Other Orientation. Regrown silicon of other orientations has also been found to have twins. 38, 41 This can be explained as follows.

When ΔG* increases, the number of nuclei will decrease and facets may be formed as discussed previously. The facets produces a new 111 surface on which twins can form. There are several ways to decrease the number of nuclei.

(1) As discussed by Barret, et al. 4 the ΔG* has a minimum at intermediate temperature and at higher or lower temperatures ΔG* will increase. Therefore, the twin will be found at higher and lower temperature. For example, twins were observed on one occasion in <100> regrown silicon which had been annealed 13 days at 462°C. Unfortunately an attempt to reproduce this experiment failed.
In conventional furnace annealing at higher temperatures, twins cannot form since the heating rate is low. This means that the regrowth process was finished before the specimen reached the required temperature. By using fast heating, it is possible to form twins in other orientations. Recently, Sadana formed twins in <100> regrown silicon with heat-pulse heating at 700°C, 60 sec. 44

(2) The second method to increase $\Delta G^*$ is to decrease the magnitude of $\Delta g$, since $\Delta G^* = \pi \sigma^2 / \rho |\Delta g|$. This can be achieved by increasing the impurity to over the maximum solubility, as discussed in 3.2a. The twins have been found in <100> regrown silicon in which the impurity concentration is much higher than the maximum solubility. 38, 42

6.3 Suggestions for Further Work

(a) The Ratio of Growth Rates for Different Orientations.

It is known that the ratio of regrowth rate of <100> to <111> silicon is about 25, if there are no impurities. The ratio of the regrowth rate can be expressed as $R_{100}/R_{111} = C_1 \exp(-C_2/\rho |\Delta g|)$, where $C_1$ is a constant and $C_2 = \frac{\pi}{3\rho} [(\alpha \sigma^2)_{100} - (\alpha \sigma^2)_{111}]$. From Section 3.1, it is seen that $C_2$ is negative. It can be concluded therefore, that the larger the magnitude of $|\Delta g|$ the smaller the ratio. But from Section 3.2a, it was shown that larger $|\Delta g|$ will lead to a faster regrowth rate. Therefore the ratio $R_{100}/R_{111}$ of faster regrowth rate is smaller than that of slower regrowth rate. The only data available at present are for Si without impurities. Further investigation of the variation of this ratio with different regrowth rates should be done.
(b) Twin Formation in Other Orientations. More evidence is needed that twins can be formed during low temperature annealing in orientations other than $\langle 111 \rangle$ and also at high temperatures with fast heating, e.g. heat-pulse.
7. CONCLUSIONS

The proposed model which combines heterogenous nucleation of growth steps and their migration fits the known experimental results quite well. Several conclusions that follow from the model are listed below:

(1) The regrowth rate of <100> substrates is faster than that of <110> substrates, and the regrowth rate of <111> substrates are the least.

(2) All impurities accelerate the regrowth rate when present in concentrations below their solubility limit in crystalline Si. Conversely, all impurities will decrease the regrowth rate when present in concentrations above the solubility limit. Supersaturation of the impurity in the regrown crystal is possible if the diffusivity of the impurity in the amorphous phase is not very large.

(3) The faster the regrowth rate, the smaller will be the activation energy and the smaller will be the ratio of regrowth rates for <001> and <111> orientations.

(4) If R/D is large, the impurity finds it difficult to redistribute. Superstratation can occur with formation of upper layer defects if the impurity concentration is higher than the solubility limit.

(5) If R/D is small, the impurity redistributes. In this case it is possible to nucleate polycrystalline Si at precipitates that form in the amorphous phase.
I have profitted greatly from studying under the guidance of Professor J. Washburn. His attitude of conducting research aimed at solving a fundamental question, rather than being a "wandwering apparatus in search of a problem", is one which I will carry away.

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FIGURE CAPTIONS

Figure 1. (a) Regrowth rates vs time for Si implanted Si annealed at 550°C (data taken from Ref. 13).

(b) Regrowth rate vs $10^3 \frac{1}{T(k)}$ for different substrate orientations of Si (data taken from Ref. 13).

Figure 2. A schematic <110> projection of the diamond cubic lattice showing the minimum size of stable atomic clusters on the major crystal faces (data taken from Ref. 16).

Figure 3. Growth rate versus $10^3 /T(k)$ for <100> Si implanted with B, O and P ions at concentration levels near 0.5 atomic percent (data taken from Ref. 18-19).

Figure 4. The disc shaped nuclei on the amorphous-crystalline interface.

Figure 5. Free energy profile vs. between amorphous and crystalline phases. $\lambda$ is the distance between the amorphous and crystalline layers at the surface between them.

Figure 6. Atomic configuration on the (a) <100> (b) <110> (c) <111> planes.

Figure 7. Gibbs free energy per atom, g, vs concentration of I atoms for different phases.

Figure 8. Growth rate versus $10^3 /T(k)$ for <100> Si implanted with Al and O with two different concentrations.

Figure 9a. MeV$^4$He$^+$ backscattering spectra for Sb implanted <100> silicon, both as implanted and annealed at 1000°C. The aligned spectra were measured by channeling in the <111> minimum, the random spectra at 5° off the minimum.
Figure 9b. MeV $^4$He$^+$ backscattering spectra for Sb implanted <111> silicon, both as implanted and annealed at 1000°C. The aligned spectra were measured by channeling in the <100> minimum, the random spectra at 5° off the minimum (data taken from Ref. 27).

Figure 10. A plot of Pb distribution in Si after various annealing stages for 80 keV Pb implanted into Si at (a) $2.5 \times 10^{14}$ cm$^{-2}$, (b) $3 \times 10^{15}$ cm$^{-2}$ and (c) $1 \times 10^{16}$ cm$^{-2}$, showing pronounced outdiffusion effect at the higher doses (data taken from Ref. 28).

Figure 11. A schematic <110> projection of the diamond cubic lattice showing twin formation on 111 planes during regrowth.
Epitaxial regrowth of amorphous Si implanted Si \( T = 550^\circ C \)

![Graph](image-url)

**Fig. 1**
Fig. 4
Fig. 7
Fig. 8

Regrowth rate (Å/min) vs. $10^3/T$ ($^\circ$K)

- $4 \times 10^{18}$ Al/cm$^3$
- $4 \times 10^{17}$ Al/cm$^3$
- $1 \times 10^{20}$ O/cm$^3$
- $2 \times 10^{20}$ O/cm$^3$
Fig. 10
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