Materials Science-Poland, 30(4), 2012, pp. 414-418
http://www.materialsscience.pwr.wroc.pl/
DOI: 10.2478/s13536-012-0042-z

Finite element method simulation of interface evolution during epitaxial growth*

S. GULKOWSKI[1]†, J.M. OLCHOWIK[1], K. CIESLAK[1], P.P. MOSKVIN[2]

[1] Lublin University of Technology, Nadbystrzycka 38, 20-618 Lublin, Poland

[2] Zhitomir State Technological University, 103 Chernyakhovskogo, vul. 10005 Zhitomir, Ukraine

Epitaxial Lateral Overgrowth (ELO) is a method of epitaxial growth on a partially masked substrate. It can be a promising method for photovoltaic applications due to a possibility of producing thin and high quality silicon substrates. Since the mask prevents propagation of the substrate dislocations to the laterally overgrown parts of the ELO layer they are characterized by a lower dislocation density than the substrate. It means that it is possible to fabricate good quality solar cells on a poor quality Si substrate. The main goal of the research is to obtain a higher growth rate in the lateral direction than in the direction normal to the substrate. The epilayer growth kinetics depends on many technological factors, basically the growth temperature, the cooling rate, the solvent and the mask filling factor. For this reason the best way to achieve the goal is a computational analysis of the epitaxial layer growth process. This work presents a two-dimensional computational study of such a process of growth for different technological conditions. The computational model is based on the assumption of pure diffusion control growth.

Keywords: *epitaxial lateral overgrowth, liquid phase epitaxy, computer simulation*

1. Introduction

In the Epitaxial Lateral Overgrowth (ELO) technique the amorphous mask is deposited on a silicon substrate and a narrow window is created in the mask by means of a standard photolithography process. Epitaxial growth begins inside the window and then proceeds over the mask with a different growth rate which depends on the technological conditions of the experimental process [1–7]. The Liquid Phase Epitaxy (LPE) method, which offers high crystal quality with cost-effective and simple apparatus, may be used to produce epitaxialy overgrown layers. The most common method is the horizontal sliding graphite boat system in which a saturated solution is placed over the growth substrate and the temperature of the system is lowered with a constant cooling rate. The initial temperature of the system is set depending on the chosen solvent. Detailed descriptions of the sliding boat LPE method can be found, for example, in [8].

The purpose of using a dielectric mask deposited on the growth substrate is to stop the propagation of dislocations from the substrate. This is the major point of the ELO method as it reduces the defect density in the new layer. The substrate defect filtration in the ELO method was demonstrated in [9], where the layer was deposited on a buffer substrate with a very high density of etch pits ($\sim 10^8$ cm^{-2}). It was shown that the dislocations present in the substrate propagated only to the very narrow area above the opened window (seed). The rest of the layer was nearly dislocation free. A defect density analysis of the ELO Si layers growth on a Si substrate was shown in [10]. A comparison of the quality of the Si layers obtained by the standard LPE and ELO methods was presented. The defect density for the LPE layers was about 10^4 cm^{-2}, whereas this value was measured as 10^3 cm^{-2} in the ELO method. It proves efficient defect filtering in the Epitaxial Lateral Overgrowth method. A sufficient ELO procedure requires a high difference between the lateral and normal growth rate (high aspect ratio).

*This paper was presented at the Conference Functional and Nanostructured Materials, FNMA 11, 6–9 September 2011, Szczecin, Poland

†E-mail: s.gulkowski@pollub.pl

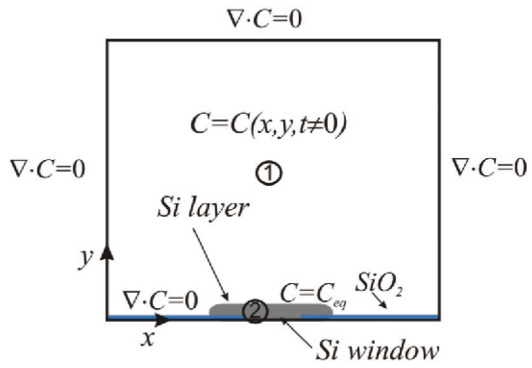

Fig. 1. Schematic view of the computational domain used in the simulations. The growth cell consists of two main parts: the solution and the epitaxial layer region. The moving interface of the layer is therefore the border between these two regions.

Unfortunately, finding an optimized set of technological parameters involves numerous experiments. To reduce the number of experiments – numerical calculations can be carried out. This paper presents a two-dimensional computational study of the epilayer interface evolution during growth from a solution in the ELO method. As distinct from the well known approaches to the ELO growth phenomena described, for example, in [11–13], our model is based on the assumption that growth is controlled only by the diffusion of Si in the solution. It means that a thermodynamic equilibrium and a high ratio of the surface reaction exist on the boundary surface. The interface position is obtained from the growth rates calculated on the basis of the concentration gradients in the normal direction to the interface. A triangle adaptive mesh was used to improve the solute concentration profiles near the grown layer border.

2. Mathematical model

A 2D computational domain for ELO growth is shown in Fig. 1. As can be seen, it consists of two main parts: the first part is the solution area which keeps information about the concentration of the solute in the solution. The second part is the moving interface grid represented by a set of connected points which change their position in time according to the calculated growth rates. Information about the location of the moving points, on the one hand, and the concentration field in the vicinity of the interface, on the other hand, must be passed between these two parts of the domain.

The set of equations for this problem can be written as follow:

$$\frac{\partial C(\xi,t)}{\partial t} = D_{Si}\nabla^2 C(\xi,t)\,, \tag{1}$$

$$D_{Si}\left.\frac{\partial C}{\partial n}\right|_L = \nu\cdot(1-C_{eq})\,, \tag{2}$$

$$C(\xi,0) = C_0(T_0)\,, \tag{3}$$

$$C(\xi,t) = C_{eq}(T)\,, \tag{4}$$

$$\left.\frac{\partial C}{\partial x}\right|_{\substack{x=0\\x=N_x}} = 0\,, \quad \left.\frac{\partial C}{\partial y}\right|_{y=N_x} \tag{5}$$

where $C(\xi,t)$ is the mass fraction of the solute and D is the diffusion coefficient of the solute. C_{eq} is the equilibrium concentration obtained from the phase diagram, and ν_n is the interface growth velocity. The solute concentration field in the liquid phase is obtained by solving the transport Equation (1) in absence of convection. This assumption is valid in the horizontal sliding method of growth in which the substrate is placed under the solution. Although the convective mass transfer appears after a short time just after the motion in the sliding process, it disappears very fast and it does not have any influence on the layer growth which is controlled by the diffusion only [14]. The initial concentration, C_0, for all the elements is set as the equilibrium concentration at the starting temperature, T_0, according to the formula (3) and it is taken from the phase diagram of the Si–Sn solution. The phase diagram is obtained from [8]. During the growth process the temperature, T, decreases with time with a constant cooling rate, hence, the equilibrium concentration is calculated in each time step of the simulation. For the two vertical walls as well as for the area between the solution and the oxide mask no flux Neumann boundary conditions (5) were used (Fig. 1). The mass balance condition (2) was set for the interface.

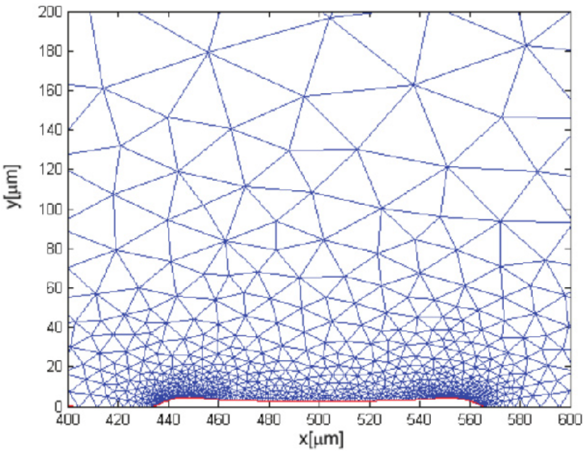

Fig. 2. Improvement of concentration field calculations in vicinity of grown layer with the use of adaptive mesh grid.

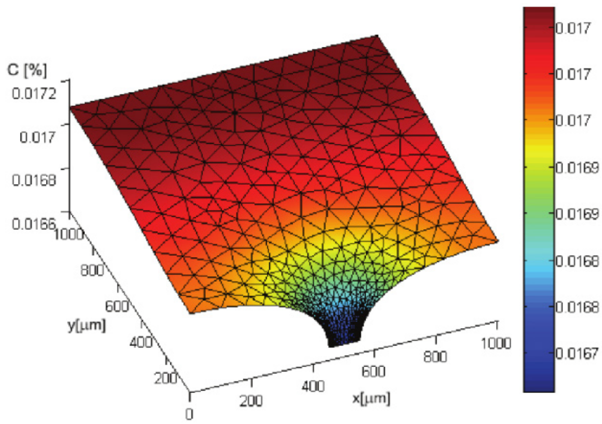

Fig. 3. Surface concentration of Si solute in Si–Sn solution after 1500 s of ELO growth.

From this condition the gradients of concentration in the normal direction to the local curvature of the interface and the resulting growth rate can be determined. After the growth rate calculation, each point of the interface is moved to a new location according to the advection relation.

The finite element method was used to solve the equations given above. As the growth rate v_n is determined from the gradient of concentration, it is very important to calculate the concentration field near the layer very precisely. For this reason, the adaptive mesh grid near the edge of the layer was used (Fig. 2). As the interface moves with time it changes the geometry of the computational domain. In each time step a new grid is generated and information of the concentration is passed to the grid. It should be pointed out that in the presented method the adaptive mesh evolves with time as well as the interface. The computational procedure consist of the following main steps:

1. Set initial values of the numerical process.
2. Generate the mesh for the given geometry of the domain.
3. Set the initial concentration profile for every node of the mesh.
4. Decrease the temperature with the given cooling rate.
5. Set the equilibrium concentration on the interface on the basis of the phase diagram.
6. Calculate the concentration profile from Equation (1).
7. Calculate the gradients, growth rate and new position of the interface.
8. Update the geometry of the domain, mesh and return to the next step.

3. Results and discussion

The computer simulations were performed for the Si ELO layer grown by LPE on an Si partially masked substrate. 100 μm windows in a domain which was 1 mm wide were used. The initial temperature of the system was set as 920 °C, 960 °C and 1000 °C. The computations were performed for different cooling rates which were as follows: 0.1°C/min, 0.5°C/min, 1.0°C/min. The diffusion coefficient was taken after [15] as $D = 3.0 \cdot 10^{-5}$ cm^2/s.

Fig. 3 shows the Si concentration surfaces in the Si–Sn solution with time. The calculations start from the solute equilibrium concentration at the initial temperature for the whole domain. The decreasing temperature in time leads to epitaxial growth and, in consequence, to a decrease in the concentration of the solute species in the open window area. The difference in the concentration of Si in the Si–Sn solution causes the flux of the solute to move towards the growing interface.

The concentration gradients calculated at the edge of the layer are much higher than those calcu-

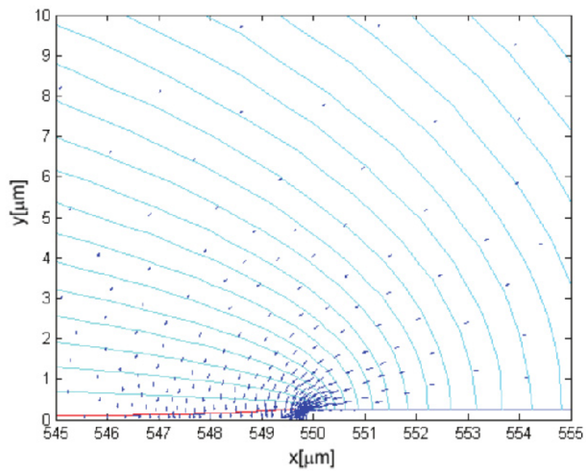

Fig. 4. Flux of Si towards growing layer in 30 s of simulation. An early stage of the interface position can be also seen.

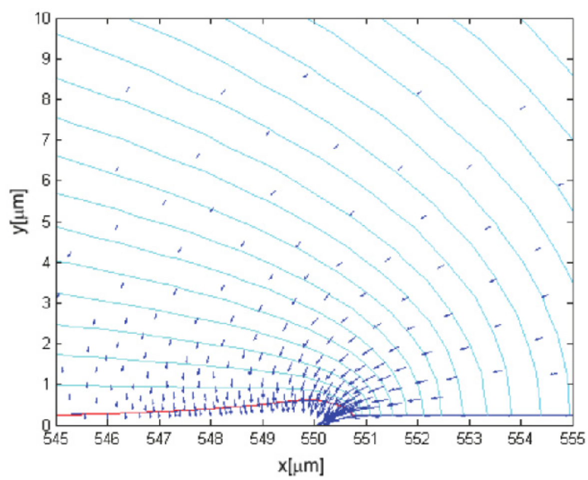

Fig. 5. Flux of Si towards interface after few minutes of growth. The epitaxial layer interface is created due to growth.

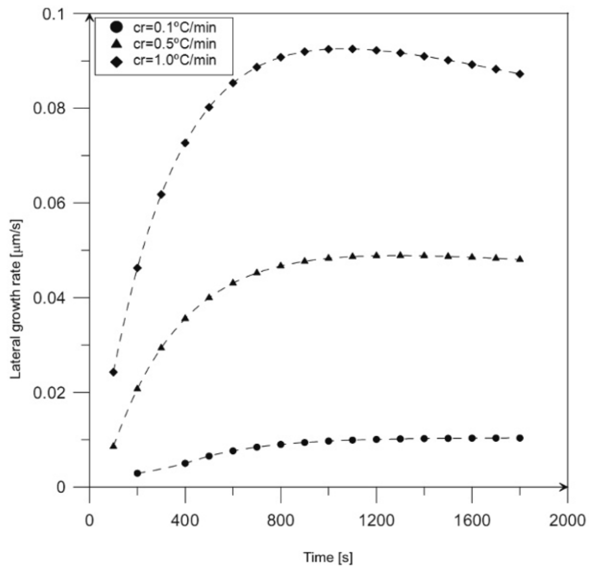

Fig. 6. Comparison of lateral growth rates for different cooling rates.

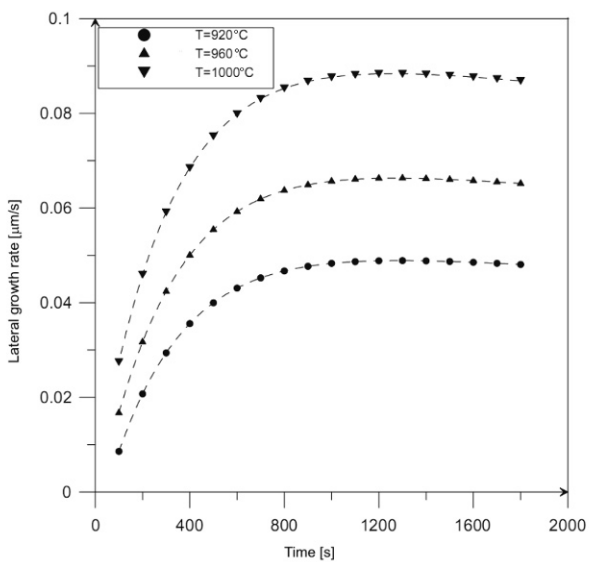

Fig. 7. Comparison of lateral growth rates for different initial system temperature.

lated for the planar regions of the layer. It can be seen in Fig. 4 and Fig. 5. The flux of the solute in the solution is marked by arrows. The arrow length is proportional to the concentration gradient at a given point. The number of arrows is connected to the number of mesh triangles. It should be pointed out that the difference is obtained due to the mask presence on the growing substrate. As there is no flux boundary condition for the mask, all the Si species move from the mask area towards the grown layer region. It leads to a higher concentration gradient at the edges of the layer and to faster growth in this region. Thus, the geometry of the system can be the reason for the difference in the growth rate in the lateral and normal direction.

The simulation results of the epitaxial layer growth for different cooling rates and initial temperature are shown in Fig. 6 and Fig. 7. As can be seen, the growth rate is rising for the first 800 s and becomes constant after that time. It is caused by the finite size of the domain. It is visible especially in the third plot of Fig. 6 where the cooling rate is 1.0. After reaching the maximum, the growth rate is decreasing due to a lower solute concentration in the whole system domain.

Figs. 6 and 7 show also the dependence of the lateral growth rate on the growth temperature and the cooling rate. Increasing one of the parameters results in an increase in the growth rate. This is a desirable effect for growing thin and wide epitaxial layers on a partially masked substrate. The experimental results of the epitaxial growth of Si on a Si substrate presented in [16] confirm that the layer width (as the growth rate is in the lateral direction) increases with an increase in the value of the cooling rate. It leads to a higher aspect ratio which was also presented in the above mentioned paper.

4. Conclusions

A two dimensional computer simulation of the ELO layer growth was performed by the LPE method on the basis of the assumption of the diffusion limited growth. The Finite Element Method with an adaptive mesh grid was used to obtain high precision of the concentration gradient calculations near the interface. The growth rate of the interface was obtained from the concentration gradient in the normal direction in the vicinity of the surface. The interface evolution during the growth was investigated and the results of the calculations were shown. The lateral growth rate dependence on the initial temperature of the system and the cooling rate observed in the numerical calculations as well as the experimental work leads to the conclusion that the diffusion-limited model could be applicable to the description of the epitaxial layer growth on a masked substrate.

References

[1] LIU Y.C., ZYTKIEWICZ Z.R., DOST S., *J. Crystal Growth*, 265 (2004), 241.

[2] NISHINAGA T., *Crystal Properties Prep.*, 31 (1991), 92.

[3] KHENNER M., BRAUN R.J., MAUK M.G., *J. Crystal Growth*, 235 (2002), 425.

[4] KHENNER M., BRAUN R.J., MAUK M.G., *J. Crystal Growth*, 241 (2002), 330.

[5] ZYTKIEWICZ Z.R., *J. Crystal Growth*, 172 (1996), 259.

[6] ZYTKIEWICZ Z.R., *Cryst. Res. Technol.*, 34 (1999), 573.

[7] DOST S., LENT B., *Single Crystal Growth of Semiconductors from Metallic Solutions*, Elsevier 2007.

[8] CAPPER P., MAUK M., LIQUID PHASE EPITAXY OF ELECTRONIC, OPTICAL AND OPTOELECTRONIC MATERIALS, John Willey & Sons, 2007.

[9] ZYTKIEWICZ Z.R., *Thin Solid Films*, 412 (2002), 64.

[10] KRAIEM J., FAVE A., KAMIŃSKI A., LEMITI M., JOZWIK I., OLCHOWIK J.M., Proc. 19th EPSEC, 7–11 June 2004, Paris, France, p. 1158.

[11] YAN Z., NISHINAGA S., NISHINAGA T., *J. Crystal Growth*, 209 (2000), 1.

[12] LIU Y.C., ZYTKIEWICZ Z.R, DOST S., *J. Crystal Growth*, 275 (2005), e953–e957.

[13] KIMURA M., DJILALI N., DOST S., KANAI H., TANAKA A., SUKEGAWA T., *J. Crystal Growth*, 167 (1996), 516.

[14] CHEN L.J., CHEN L.C., WAN C., LIEN C., *The Chemical Engineering Journal and the Biochemical Engineering Journal*, 62 (1996), 43.

[15] KIMURA M., DJILALI N., DOST S., *J. Crystal Growth*, 143 (1994), 334.

[16] JOZWIK I., OLCHOWIK J.M., Proc. 21st EPSEC, 4–8 September 2006, Dresden, Germany, (CD) p. 1005.

Received 2012-07-07
Accepted 2012-08-27