Numerical simulation of pulsed-laser induced lateral growth of pc-Si from amorphous layers

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Abstract

The mechanisms of lateral growth of pc-Si induced by the irradiation of an a-Si thin film by a ns-pulse laser moving with constant velocity are studied in the paper. Pulsed-laser induced phase transformations in a-Si are modeled using two basic types of approach. The first of them assumes a direct transition between a-Si and 1-Si without any intermediate phases, the other approach is based on the assumption that melting of a thin surface layer is immediately followed by resolidification in the form of pc-Si, which initiates explosive crystallization. In a practical application of the computational model, the lateral growth of pc-Si induced by a CuBr laser is simulated. The temperature field, the positions and velocities of the phase interfaces, the reflectivity of the probe laser beam and the length of the laterally crystallized region are calculated in dependence on the pulsed-laser energy density and on the type of the physical model.

1 Introduction

Pulsed-laser induced phase transformations in amorphous silicon (a-Si) thin films have been a subject of intensive studies, particularly in the last two decades. Currently, phase transition mechanisms during the initial heating phase are described using two basic types of models. The first of them assumes a direct transition between a-Si and liquid Si (l-Si) (see [1–4]) without any intermediate phases, i.e., only l-Si/a-Si interface appears in the
The second type of model is based on the assumption that melting of a thin surface layer is immediately followed by resolidification in the form of polycrystalline silicon (pc-Si), which initiates the explosive crystallization (XC). The XC process consists in the fact that a thin liquid layer is propagating fast through the amorphous material due to the difference between the latent heat of crystallization of pc-Si and the latent heat of melting of a-Si [5–8]. With further increase of surface temperature, the newly formed pc-Si layer is melted from the surface, and the phase change mechanism is similar to the case of melting of a monocrystalline layer. In this type of model, we generally have three phase interfaces, the first of them is the l-Si/pc-Si front, the other two are the pc-Si/l-Si and l-Si/a-Si interfaces on both sides of the buried molten layer propagating into the a-Si layer at XC.

Similar mechanisms can also be applied to phase transitions in the lateral direction (perpendicular to the laser beam). However, this type of phase change processes has not been studied so frequently as those in the direction of the laser beam, and if so, then only under special conditions. For instance, Andrä et al. [9,10] studied the XC process in the lateral growth of Si in such an experimental situation when the substrate was heated to higher temperatures (~ 1200 K) by a free-running ms-pulse laser, and a second laser then increased locally the temperature above the melting point of a-Si.

In this paper, we analyze the mechanisms of lateral growth of pc-Si induced by irradiation of an a-Si layer by a ns-pulse laser moving with a constant velocity.

2 Models of lateral growth of pc-Si

We consider the following experimental situation: a thin a-Si layer (typically, 10-500 nm) is deposited on 400 μm thick Corning glass substrate. The sample, which is moving with a constant speed v = 200 μm/s, i.e., 20 nm/pulse, is irradiated by a CuBr laser (511 and 578 nm, 50 ns FWHM, laser spot 200 μm in diameter) with a f = 10 kHz repetition rate. The aim of this treatment is to obtain large-grain (LG) pc-Si in large areas.

Owing to the experimental situation (ns-pulse laser), we can employ a thermal model for the description of laser interactions with the specimen. Contrary to the most often modeled experimental setups with both sample and laser motionless, where a 1-D approximation is commonly used because the dimension of the spot is several orders of magnitude larger than the layer thickness, here we have generally a 3-D problem. However, solving a 3-D problem with several moving boundaries would significantly exceed the capacity of commonly accessible computers, and therefore some simplifications need to be done.

The main idea of our simplification is a conversion of the general 3-D case into a set of 1-D solutions. As the main concern of experimentalists
is to know about the crystallization process in the direction of the sample motion, we will mainly concentrate ourselves on solving this problem.

The idea of the solution technique is strongly influenced by a choice of the model of a-Si phase transformations in the initial heating phase. We will employ both the 1-D models mentioned in the Introduction, namely the model with explosive crystallization (XC model) and the model with direct a-Si \(\rightarrow\) l-Si phase transition (AL model).

These models are described as follows:

**A. The XC model (see [11])**

Assuming one-dimensional heat conduction to be the dominant mode of energy transfer, we can write the energy balance in the form

\[
\rho_i c_i \frac{\partial T_i}{\partial t} = \frac{\partial}{\partial x} \left( K_i \frac{\partial T_i}{\partial x} \right) + S_i(x, t), \quad i = l, pc, a, s,
\]

where \(\rho\) is the density, \(c\) the specific heat, \(T\) the temperature, \(K\) the thermal conductivity, the indices \(pc, a, l\) mean the polycrystalline, amorphous and liquid phases, respectively, and the index \(s\) means the substrate.

The volume heating term \(S_i\) arising due to the laser irradiation can be expressed in the form

\[
S_i(x, t) = (1 - R(t))\alpha(x)I_0(t)\exp(- \int_{Z_0(t)}^x \alpha(\eta)d\eta),
\]

where \(I_0(t)\) is the power density of the pulse (power per unit area), \(\alpha\) the optical absorption coefficient, \(Z_0(t)\) is the position of the liquid/vapor interface (the surface of the sample). The reflectivity \(R = R(t)\) is calculated using the theory of absorbing optically nonhomogeneous media (see [12]).

All phase changes are considered to be nonequilibrium, which follows from the rapidity of the process. Therefore, at every phase interface we have a heat balance condition and a kinetic condition. In modeling the XC process, we utilize the fact that the explosively propagating liquid layer is very thin, and replace it by a discontinuity surface (XC front) where the difference between the latent heat of crystallization of pc-Si and the latent heat of melting of a-Si, \(L_{xc} = L_{pc} - L_a\) is released.

The heat balance condition at the liquid/vapor interface can be expressed as follows:

\[
\rho_i L_v \frac{dZ_0}{dt} = K_l \left. \left( \frac{\partial T_l}{\partial x} \right) \right|_{x=Z_0(t)} - \varepsilon \sigma_{SB} (T_{Z_0}^4 - T_e^4),
\]

where \(\varepsilon\) is the emissivity from the liquid surface, \(\sigma_{SB}\) the Stefan-Boltzmann constant, \(T_{Z_0}\) the temperature of the liquid/vapor interface, \(L_v\) the latent heat of evaporation, \(T_e\) the temperature of the surroundings, i.e., for \(x \to -\infty\).
For evaporation into vacuum, the kinetic condition reads (see [13])

$$\frac{dZ_0}{dt} = \frac{1}{\rho_l} \sqrt{\frac{M}{2\pi R_g}} T_{Z_0}^{C-0.5} \cdot 10^{-\frac{A+B}{T_{Z_0}}}$$

(4)

where $T_{Z_0}$ is the absolute temperature, $M$ the molar mass, $R_g$ the universal gas constant, and $A, B, C$ are material parameters.

On the pc-Si/l-Si interface we have the heat balance condition

$$\rho_{pc}L_{pc} \frac{dZ_1}{dt} = K_{pc} \left( \frac{\partial T_{pc}}{\partial x} \right)_{x=Z_1(t)_+} - K_{l} \left( \frac{\partial T_{l}}{\partial x} \right)_{x=Z_1(t)_-}$$

(5)

where $L_{pc}$ is the latent heat of melting of pc-Si, $Z_1$ the position of the pc-Si/l-Si interface, and the kinetic condition can be expressed schematically in the form

$$\frac{dZ_1}{dt} = f_1(T_{Z_1})$$

(6)

where $f_1(T_{Z_1})$ is the pc-Si/l-Si interface response function.

Finally, on the XC front we can write the conditions

$$\rho_aL_{XC} \frac{dZ_2}{dt} = K_{pc} \left( \frac{\partial T_{pc}}{\partial x} \right)_{x=Z_2(t)_-} - K_a \left( \frac{\partial T_a}{\partial x} \right)_{x=Z_2(t)_+}$$

(7)

$$\frac{dZ_2}{dt} = f_2(T_{Z_2})$$

(8)

where $Z_2$ is the position of the XC front, and $f_2(T_{Z_2})$ the XC front response function. The velocity $dZ_2/dt$ of the XC front is assumed to be always positive because the character of XC described before excludes the reverse transition from pc-Si to a-Si in this way.

The other boundary conditions and initial conditions have the form

$$\frac{\partial T}{\partial x}(D, t) = 0$$

(9)

$$T(x, 0) = T_0(x), \quad x \in [0, D]$$

(10)

$$Z_0(0) = Z_{00}, \quad Z_1(0) = Z_{10}, \quad Z_2(0) = Z_{20},$$

(11)

where $D$ is the thickness of the sample, $D = A + B$, $A$ is the thickness of the a-Si layer, and $B$ the thickness of the Corning glass plate.

**B. The AL model**

In the heating phase it is supposed that the surface temperature increase is so fast that XC does not appear. Therefore, we have no pc-Si layer, and only the liquid/vapor and l-Si/a-Si interfaces are modeled. Eqs. (1)–(4) of the XC model remain unchanged, and we have only two relations for the l-Si/a-Si interface instead of Eqs. (5)–(8). The heat balance condition reads

$$\rho_aL_a \frac{dZ_1}{dt} = K_a \left( \frac{\partial T_a}{\partial x} \right)_{x=Z_1(t)_+} - K_{l} \left( \frac{\partial T_{l}}{\partial x} \right)_{x=Z_1(t)_-}$$

(12)
where $L_a$ is the latent heat of melting of a-Si, $Z_1$ the position of the l-Si/a-Si interface, and the kinetic condition can be expressed as

$$\frac{dZ_1}{dt} = f_3(T_{Z_1}),$$

(13)

where $f_3(T_{Z_1})$ is the l-Si/a-Si interface response function.

The other initial and boundary conditions are the same as for the XC model (except the last condition of (11), because there is no $Z_2$ interface here).

In the solidification phase ($dZ_1/dt < 0$) it is assumed that after the thickness of the newly solidified layer, $\Delta Z_1 = Z_{1,\text{max}} - Z_1(t)$ ($Z_{1,\text{max}}$ is the maximum thickness of the molten layer), exceeds certain critical value (typically, $\Delta Z_1 \sim 1$ nm), the crystallization process from the interface is initiated. a pc-Si layer appears and instead of Eqs. (12)–(13) we have

$$\rho_{pc} L_{pc} \frac{dZ_1}{dt} = K_{pc} \left( \frac{\partial T_{pc}}{\partial x} \right)_{x=Z_1(t)} - K_1 \left( \frac{\partial T_1}{\partial x} \right)_{x=Z_1(t)}$$

(14)

$$\frac{dZ_1}{dt} = f_1(T_{Z_1}).$$

(15)

Using the XC model, the lateral growth process can work without the pulse energy density being changed during the sample movement only in the case that the surface is melted not only in the first shot but also repeatedly during the subsequent shots, and the crystallization proceeds in the lateral direction this way (Mechanism A). Mathematically this can be described by the following algorithm:

1. The temperature field and other characteristic parameters of the process, such as the positions and velocities of the phase boundaries, are calculated in the direction of the laser beam using a 1-D model.

2. The temperature field determined in the first step is employed in the form of Dirichlet boundary conditions for calculating the pc-Si growth in the area outside the laser spot, in the direction perpendicular to the laser beam. Here, a 1-D problem with radial symmetry is solved.

This calculation scheme is analogous also for further laser shots. The temperature fields and positions of phase boundaries at the time $t_{\text{fin}} = 1/f = 10^5$ ns from the previous shot are taken as initial conditions for the next shot, and the calculations as in the first and second step of the above algorithm are performed again.

Applying the AL model, we have one more possibility in how to achieve the lateral growth of pc-Si. We choose the laser energy density in the way that the surface of the sample at the laser spot is melted in the first shot only, and in the subsequent shots, only that part of the laser spot which was originally formed by a-Si is melted, while the pc-Si part remains solid due to the higher thermal conductivity of pc-Si compared to a-Si.

The mechanism of lateral growth (Mechanism B) consists then in the fact that when l-Si appears in the place where the a-Si part of the laser spot
was, it is severely undercooled with respect to the pc-Si part, and therefore a very fast crystallization from the interface is started.

The computational scheme is a little more complicated than in the previous case and is as follows:

1. The temperature field and positions and velocities of the phase boundaries at the first shot are calculated in the direction of laser beam using the 1-D AL model.

2. a) In the second shot, the temperature field in the direction of the laser beam for both pc-Si and a-Si parts of the laser spot is calculated (and possibly also the positions and velocities of the phase interfaces, if the surface is melted), as if both parts were thermally insulated from each other.

b) Using the temperature fields in the pc-Si and a-Si (l-Si) parts, a lumped model for calculating the positions and velocities of the pc-Si/l-Si interface in the lateral direction (i.e., the lateral growth of pc-Si) is proposed:

We assume the relation between the temperature and velocity of the pc-Si/l-Si interface is in the form ([14])

\[ v(T) = \frac{D}{\Lambda^2 f_A l} \cdot \exp \left( -\frac{\Delta S}{k_B} \right) \cdot \left[ 1 - \exp \left( -\frac{\Delta \mu}{k_B T} \right) \right], \]  

where \( D \) is the diffusion coefficient in the liquid, \( \Lambda \) is the mean free path in the liquid, so that \( D/\Lambda^2 \) is the atomic jump frequency, \( f_A \) is the active site fraction, \( l \) is the thickness of the layer of the liquid atoms that are sufficiently close to the crystal so that they can reach crystal lattice sites with a single jump, \( \Delta S \) is the entropy of fusion, \( \Delta \mu \) is the change of chemical potential during the phase change, and \( k_B \) is the Boltzmann constant. Then, we can calculate the amount of thermal energy \( \Delta Q_S \) released at the pc-Si/l-Si interface during the time interval \( \Delta t \) as follows:

\[ \Delta Q_S = \rho_{pc} L_{pc} v(T) \Delta t \cdot S, \]  

where \( S \) is the interface area.

In the lumped model approximation working with integral balances only, we assume that both the pc-Si and the l-Si parts have constant space temperature distributions and that the thermal energy \( Q_S \) is spent on heating up the cooler of the pc-Si and l-Si parts of the spot, i.e.,

\[ \Delta Q_S = \Delta Q_i = \rho_i c_i S d_{c,i} \Delta T, \]  

where the index \( i \) means either pc or l.

Using (17) and (18), we can calculate the temperature increase \( \Delta T \) during the time interval \( \Delta t \) as

\[ \Delta T = \frac{\rho_{pc} L_{pc} v(T) \Delta t}{\rho_i c_i d_{c,i}}, \]  

where again the index \( i \) is either pc or l.
The only quantity which has to be specified in Eq. (19) is the characteristic length $d_{c,i}$. If the heated region is pc-Si, we can approximate $d_{c,pc}$ as the thermal length,

$$d_{c,pc} = \sqrt{\frac{K_{pc}}{\rho_{pc} c_{pc}}} \cdot t_c,$$

(20)

where $t_c$ is the duration of the surface melt in the former a-Si part of the laser spot calculated in 2.a).

The same treatment cannot be employed if the l-Si region is heated because the length of the l-Si part in the lateral direction, $d_l$, is usually much lower than the thermal length. Therefore, we choose $d_l$ as the characteristic length $d_{c,l}$.

However, the calculation of $d_l$ is more complicated than the determination of the thermal length because $d_l$ generally consists of two parts. One of them, $d_{c,l1}$, is equal to the sample shift between two laser shots, the second, $d_{c,l2}$, is the molten area outside the spot. Therefore, we employ the temperature field determined in the a-Si part of the spot in the second shot (point 2.a)) as a Dirichlet boundary condition for calculating the position and velocity of the l-Si/a-Si interface outside the laser spot, in the direction perpendicular to the laser beam, using the 1-D AL model with radial symmetry, and choose the calculated maximum length of the molten layer as $d_{c,l2}$. The computational scheme in points 2.a), 2.b) is then repeated also for the subsequent shots.

It should be noted that for the AL model, the mechanism of lateral growth with melting the full spot area in the first pulse only is not the only possible one. The mechanism shown before for the XC model (Mechanism A), when the spot area is repeatedly melted, can also be applied for the AL model.

### 3 Computational results

The mathematical models of lateral growth of pc-Si were solved numerically using the Galerkin finite element method, and the computer implementation was done.

The main aim of the numerical experiments performed was to analyze the applicability of the proposed mechanisms of lateral growth and particularly to determine the application limits concerning the laser energy density $E$ and the a-Si layer thickness $A$.

Fig. 1a shows the application limits of the Mechanism B of lateral growth for $A = 240$ nm. Apparently, the appropriate conditions begin at $E = 0.09$ Jcm$^{-2}$ when melting of a-Si starts, and are terminated at $E = 0.185$ Jcm$^{-2}$ when melting of the newly formed polycrystalline layer in the second shot appears. The corresponding lengths of the laterally crystallized regions on the sample surface, calculated using the lumped model, which are presented in Fig. 2a, are well above the laser shift of 20 nm between two consecutive shots and depend very weakly on the laser energy
Figure 1: Maximum thickness of the molten layer as a function of the laser energy density for two consecutive shots and $A = 240$ nm calculated in the direction of the laser beam using a) the AL model, b) the XC model.

Figure 2: Length of the laterally crystallized region on the sample surface as a function of the laser energy density, calculated for the second laser shot using a) the lumped model with the temperature fields obtained for $A = 240$ nm by the AL model, b) Mechanism A and the XC model.
Figure 3: Comparison of maximum molten layer thickness as a function of laser energy density in the first shot, calculated by the XC and AL models for $A = 240$ nm and in the direction of the laser beam.

Figure 4: Maximum thickness of the explosively crystallized layer as a function of the energy density for the XC model, $A = 240$ nm, first laser shot.

density in the interval $E \in [0.09 \text{ Jcm}^{-2}, 0.185 \text{ Jcm}^{-2}]$.

The situation might change dramatically when the thickness of the a-Si layer is varied. Our calculations revealed that, for instance, for $A = 60$ nm the Mechanism B cannot work any more because the melting thresholds for the first and the second shots are almost the same.

The reason for these variations was found to be in dramatic changes of the reflectivity of the incident CuBr laser beam between $A = 60$ nm and $A = 240$ nm. The final values of the reflectivity after the first laser shot differed quite a lot; while for 240 nm they were $\sim 0.30$, for 60 nm only $\sim 0.07$. Therefore, in the second shot more energy could penetrate into the 60 nm layer than into the 240 nm layer, and heating of the surface was faster.

Figs. 1b, 2b show the application limits of the Mechanism A of the lateral growth, demonstrated on the XC model. Basically, the process can start when the surface is melted in the second shot, i.e., for $E \geq 0.20 \text{ Jcm}^{-2}$. If we require a homogeneous spot area after the laser shift, i.e., the length of the laterally crystallized region in the second and further shots higher than 20 nm, then we must have $E > 0.33 \text{ Jcm}^{-2}$.

As follows from the character of the Mechanism A, there is no other upper limit for its practical application than the evaporation threshold. Moreover, the process cannot be negatively influenced by the changes of reflectivity in the second shot as it was in the Mechanism B.
Fig. 3 illustrates the differences between the XC and AL models using the comparison of maximum molten layer thickness. The melting thresholds are very similar, because XC increases the temperatures near to the surface very fast due to the latent heat release so that the surface melts soon after being transformed to pc-Si. However, for higher energy densities the differences in the maximum melt depth are higher because the thermal conductivity of pc-Si is several times higher than that of a-Si. The delay in the XC-model curve is due to the fact that in the corresponding range of energy densities, a fast increase in the maximum thickness of the explosively crystallized layer appears, as it is demonstrated in Fig. 4, and therefore the influence of a higher thermal conductivity of pc-Si is more pronounced.

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