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Analysis of the processes of silicon epitaxial lateral overgrowth in Ar ambient gas

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Liquid phase epitaxy (LPE) and, in particular, epitaxial lateral overgrowth (ELO) is an attractive method of thin film deposition, owing to the simplicity of its technology and a reduced growth temperature. In this paper, we present recent results of the ELO of silicon layers carried out by means of LPE using Ar as an ambient gas, without any addition of hydrogen, potentially explosive gas, which makes this deposition technique a very safe process. The aim of the this work focused on the silicon ELO growth on partially masked substrates was to determine optimal conditions of growth resulting in ELO layers of the maximum aspect ratio and minimum defect density. Data presented herein clearly show that the epitaxial layers characterized by the maximum value of the aspect ratio can be obtained by application of the 0.25 °C/min cooling rate. Noteworthy is the fact that in the same conditions the defect density achieves the minimum value of 1.07×10^4 cm^{-2}, which is the amount smaller by the factor of 10 than the defect density of Si substrates used (1.7×10^5 cm^{-2}). It confirms the ELO technique as a promising tool for the fabrication of low-defect density silicon layers of good morphology.

Key words: *liquid phase epitaxy; silicon thin films; crystal morphology*

1. Introduction

Attention of scientists has been focused in recent years on thin film materials technologies, as they seem to be promising alternatives to bulk silicon materials reducing the cost of photovoltaic (PV) modules production. Due to this fact, an increased activity has taken place using liquid phase epitaxy (LPE) and, in particular, epitaxial lateral overgrowth (ELO), to prepare silicon thin film materials for solar cells applications. LPE is an attractive method of the thin film deposition processes, owing to the simplicity of its technology and a reduced growth temperature (below the melting point of silicon).

ELO is a method of epitaxial growth on a partially masked substrate. Prior to the growth, the substrate is covered with a thin masking film of insulator and patterned by

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means of a standard photolithography technique forming a suitable set of silicon open windows on the sample area. Afterwards, the silicon epilayer is deposited on the surface of the masked substrate (Fig. 1). The growth of Si occurs selectively – it starts in the seeding windows and proceeds in two directions: normal to the surface, and also spreads over the parts of the insulating layer [1]. In favourable conditions, the adjacent stripes of Si tend to coalesce forming a continuous epilayer. One of the advantages of such an approach is that the thin masking film prevents the propagation of defects present in the Si substrate into the ELO layer, thus the ELO process is a promising tool in reducing defect density of epitaxial silicon layers.

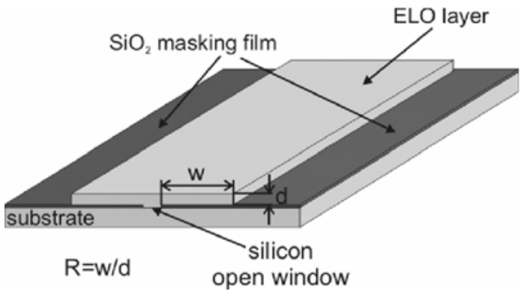

Fig 1. Schematic cross section of the ELO layer: the width *w* and thickness *d*, as well as the aspect ratio *R* of the ELO layer are defined herein

Generally, LPE is a promising technique in reducing the costs of the PV modules not only because of low-cost exploitation of the process but also because of the simplicity of the apparatus design. However, usually when it comes to the LPE process, a continuously flowing H_2 is used as a process gas, hence the safety level of the process decreases while there is a rise in the costs. A few attempts to avoid the H_2 application have been made, such as growth in a vacuum [2] or the use of Ar/H_2 mixture as ambient gas [3]. This paper reports on the results of the epitaxial lateral overgrowth of Si carried out by means of LPE using Ar as an ambient gas, without any additions of hydrogen. Elimination of a potentially explosive gas makes this technique a very safe process.

2. Experimental

The growth of Si epilayers was conducted by means of LPE with a horizontal sliding graphite boat system, with application of Ar pure gas. Si thin layers were grown on silicon substrates (111) oriented and partially masked with an SiO_2 film (0.1 μm thick) obtained by oxidization of Si wafers in dry O_2 for 1 hour at 1000 °C, and patterned by conventional photolithography technique, forming a suitable set of openings for seeding windows. Sample dimensions were 10×10 mm^2. As the result, a set of parallel lines of silicon seeding areas 50 μm wide with 100 μm SiO_2 spacing, oriented along $<0\bar{1}1>$ direction were formed.

We chose (111) oriented Si substrates as it is known [4] that the aspect ratio of Si films grown on (111) Si, defined as the width of the laterally overgrown Si to the thickness of Si film (see Fig. 1) is higher than in the case of (100) surface orientation, even though from the atomistic point of view the (100) surface offers more nucleation sites.

Prior to the growth, both substrate and the source Si were cleaned with organic solutions using acetone and trichloroethylene, then chemically etched in H_2SO_4:H_2O_2, 2:1 solution and dipped in 4% HF. To prepare the melt we used tin (5 *N*) as the solvent, which was etched in 36% HCl. In addition, small amounts (0.3 wt. % approx.) of Al (5 *N*) were incorporated into the growth solution, in order to remove native oxides formed on the silicon open window surfaces. After 1 hour of saturation at 920 °C, the growth was performed with various cooling rates: 0.25 °C/min, 0.5 °C/min, 1.0 °C/min with a fixed value of the supersaturation.

After the process of growth, the substrate with Si thin epilayers and the solution were disconnected, cooled to the room temperature and cleaned with aqua regia (HNO_3:HCl, 1:3) in order to remove any residual remains of the solution. The silicon thin layers grown on Si substrates masked with SiO_2 were examined using a scanning electron microscope (SEM).

3. Results and discussion

The experiments carried out, followed by the measurements of the width and thickness of the silicon ELO layers, resulted in the dependences of the parameters characterizing the ELO layers on the cooling rate at constant supersaturation. The values of particular parameters are the average values obtained after a series of experiments.

As shown in Fig. 2, the width of the ELO layer decreases with the increase of the cooling rate at constant supersaturation. This finding can be explained by consideration of the length of the growth period applied in each case. According to the data reported in previous works [5, 6], the value of width of the ELO layers depends on the time of growth at constant supersaturation, i.e. it increases in time until a plateau is achieved. At the first 2 hours of growth this dependence is approximately linear. Thus, as a high value of the cooling rate at a constant supersaturation means that the time of growth is shorter, it becomes clear why ELO layers obtained during the growth at lower values of cooling rates (here 0.25 °C/min) achieve higher values of width than in the case of 1.0 °C/min cooling rate, for example. In the cases investigated in this work, the difference between these two extreme widths reaches 20 μm, as the growth of ELO layer at 0.25 °C/min results in the widths equal 50 μm on average, while the width of ELO layer grown at 1.0 °C/min is ca. 30 μm.

A slightly different behaviour can be observed in the changes of the thickness of the ELO layers on the cooling rate. As the data show, the thickness of the ELO layer is maximum reaching 38 μm when the 0.5°C/min cooling rate is applied during the growth, while the values in the two other cases are smaller by 3–5 μm, the differences being within the measurement error. That is why it is worthy to consider another parameter characterizing the ELO layers, i.e. the aspect ratio.

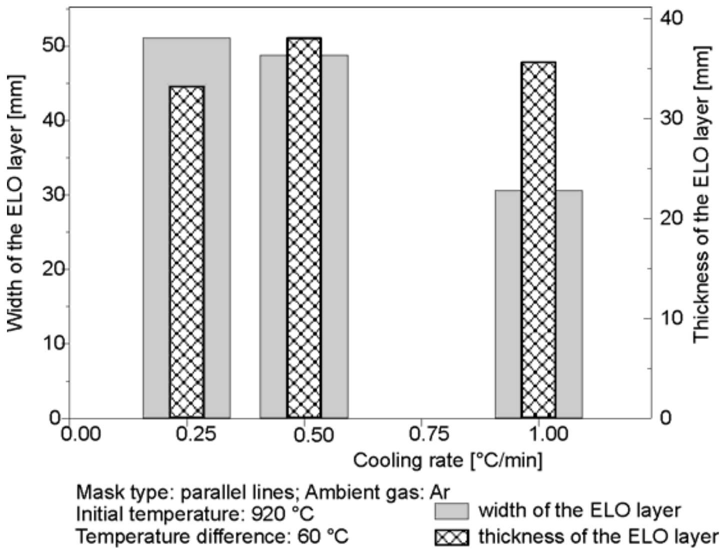

Fig. 2. Dependences of the width and thickness of the ELO layers on the cooling rate at a constant supersaturation

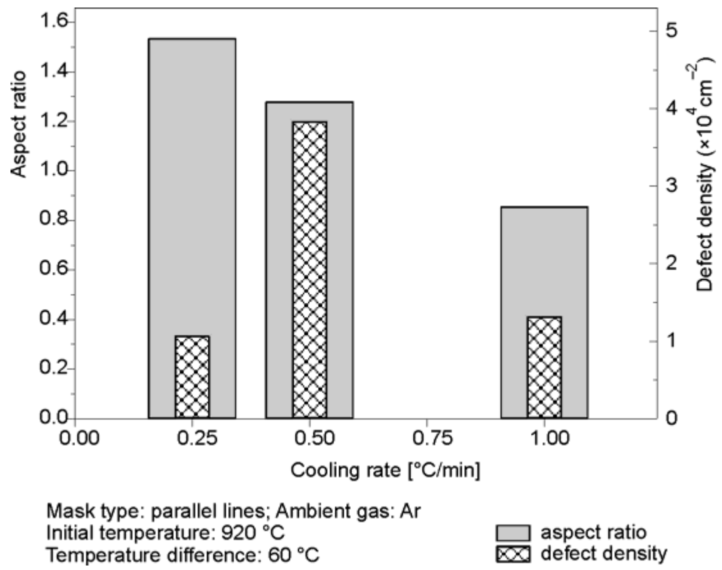

Fig. 3. Dependences of the aspect ratio and defect density on the cooling rate at a constant supersaturation

The dependences of the aspect ratio and of the defect density on the cooling rate at a constant supersaturation are presented in Fig. 3. The defect densities of silicon ELO layers as well as the Si substrates used were determined using the Secco etching method [7].

The aim of the this work, focused on the silicon ELO growth on partially masked substrates, was to determine the optimal conditions of growth resulting in the ELO layers of the maximum aspect ratio value and the minimum defect density. The data presented in Fig. 3 clearly show that the epitaxial layers grown in the conditions specified above, characterized by the maximum value of the aspect ratio can be obtained by application of the 0.25 °C/min cooling rate. Noteworthy is the fact that in the same conditions the defect density achieves the minimum value of 1.07×10^4 cm^{-2}, which is the amount smaller by one order of magnitude than the defect density of Si substrates used (1.7×10^5 cm^{-2}). It confirms the ELO technique as a promising tool for the fabrication of low-defect density silicon layers of good morphology.

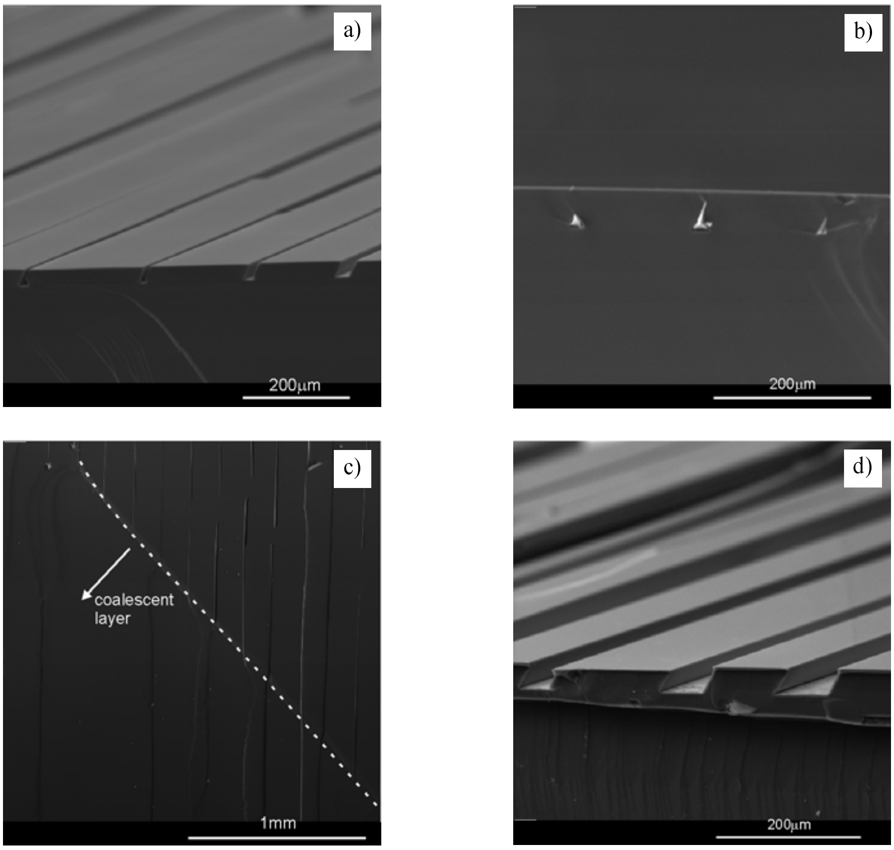

Fig. 4. The view of the epitaxial silicon layers grown with the application of various cooling rates: a) 0.25°C/min, b, c) 0.5°C/min and d) 1.0°C/min at constant supersaturation

The morphology of epitaxial layers is an important indicator of the crystal quality. It is essential to obtain planar, mirror-like surfaces, free of any surface imperfections or inclusions. The results of the investigations presented here show that the cooling rate is an important parameter influencing the layer morphology. Figure 4 presents the view of the epitaxially grown Si layers obtained in the experiments carried out under three different cooling rates.

The layers grown at 0.25 °C/min cooling rate are thinner than those obtained at 1.0 °C/min, but of the equal thickness of the neighbouring stripes (Fig. 4a), which is not observed in the epilayers grown at 1.0 °C/min cooling rate (Fig. 4d). The rise in the value of the cooling rate manifests itself also in the presence of tin inclusions observed in the epitaxial layers (Fig. 4b) which are the leftovers of the growth solution jammed between the adjoining stripes. These inclusions are distributed throughout the epitaxial layer and not just confined to the interface between epilayer and the substrate.

In some cases, the leftovers of the growth solution were present on the sample surface which served as a source of Si atoms being able to build in the crystal forming the upper parts of the ELO layer, leading to a continuous coalescent epilayer. The clear border line on the left-down of which the fully coalescent layer is visible can be seen in Fig. 4c.

4. Conclusions

The results of investigation presented here demonstrate that the ELO technique is a suitable method enabling one to obtain silicon thin layers without any use of H_2, with Ar as an ambient gas. The procedure reduces the costs simultaneously making the ELO a very safe process. The kinetics of growth as well as the morphology of the epitaxially overgrown layers have been studied. Data presented here show that the widths of the ELO layers strongly depend on the cooling rate at a constant supersaturation. A slightly different behaviour is observed in the case of the thickness of the epilayer, which stays at the same level in the range of the measurement error value. The results show that the epitaxial overgrowth resulting in the epilayers characterized by the maximum aspect ratio value and minimum defect density at the same time is possible. Epitaxial lateral overgrowth technique has been confirmed as a promising tool and suitable method for the fabrication of low-defect density silicon layers of good morphology.

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