A systematic study and optimization of parameters affecting grain size and surface roughness in excimer laser annealed polysilicon thin films

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A thorough investigation of polysilicon films crystallized from amorphous silicon using a XeCl excimer laser has been completed. Emphasis was placed on the development of large crystals, good uniformity, and low surface roughness, with an eye towards improving throughput. This was accomplished through use of SiO$_2$ barrier layers, multiple passes, and careful consideration of annealing ambient. Thermal modeling was performed to help interpret the observed trends and aid in optimization. By controlling these process parameters, samples were produced with a high degree of uniformity and two-dimensional structural ordering, with grains nearly identical in size and stacked in rows and columns. However, a biproduct of the ordering was the creation of large topographical features at the intersection points of multiple grain boundaries, which lead to surface roughness in the range of 12 nm. By applying more shots to the same area, films with maximum grain sizes on the order of 3 μm and reduced surface roughness (6 nm) were realized. © 1997 American Institute of Physics. [S0021-8979(97)08121-8]

I. INTRODUCTION

There is an increasing interest in the use of polycrystalline silicon (poly-Si) thin film transistors (TFTs) for application in the circuitry of active-matrix addressed flat panel displays (FPDs). Poly-Si films used in the fabrication of TFTs have been traditionally obtained by solid-phase crystallization (SPC) of as-deposited amorphous silicon (a-Si) films, at relatively low crystallization temperatures. However, crystals made in this manner have a high density of intragrain defects that tend to increase the threshold voltage and decrease the field effect mobility of the TFT, resulting in poor transfer characteristics.

Excimer laser annealing (ELA) has been actively investigated as an alternative method for the crystallization of a-Si films. The major advantages of this technique are the formation of polysilicon grains with excellent structural quality and the ability to process selected areas. This alleviates the aforementioned problems of SPC and allows a-Si:H and polysilicon devices to be produced on the same substrate.

The most common problem that plagues ELA is the narrow process window associated with the development of a large and uniform grain size. Moreover, the surface roughness inherent to this process is also troublesome. Currently, equipment manufacturers promote ELA at elevated temperatures of uniformity and two-dimensional structural ordering, with grains nearly identical in size and stacked in rows and columns. However, a biproduct of the ordering was the creation of large topographical features at the intersection points of multiple grain boundaries, which lead to surface roughness in the range of 12 nm. By applying more shots to the same area, films with maximum grain sizes on the order of 3 μm and reduced surface roughness (6 nm) were realized. © 1997 American Institute of Physics. [S0021-8979(97)08121-8]

II. EXPERIMENT

Using plasma enhanced chemical vapor deposition (PECVD), 50 nm thick layers of a-Si were deposited, at a temperature of 390 °C and a corresponding deposition rate of 60 nm/min, on 1737 Corning glass substrates. If a barrier layer was needed, SiO$_2$ was deposited using PECVD, at a temperature of 390 °C and a deposition rate of 120 nm/min, on the glass prior to silicon deposition. Samples were then preheated to 450 °C for 2 h in a conventional oven under nitrogen flow to dehydrogenate the deposited silicon film. Otherwise, during laser annealing, the film would ablate...
from explosive hydrogen evolution, due to the high hydrogen content of PECVD deposited $\alpha$-Si (around 10%).

The laser processing took place in a custom made stainless steel chamber with a quartz window on top. The system has a 1 mTorr base pressure and a valve which can be used to bleed in purified gases. A hot stage sample holder is able to heat samples up to 500 °C, but for this set of experiments, all laser processing was done at 400 °C or at room temperature (RT). The processing chamber sits on an $x-y$, motor driven stage with 0.1 μm step resolution and variable speed control. The amorphous silicon was crystallized by XeCl excimer laser pulses (308 nm), with a 25 ns pulse duration, from a Lambda Physik laser. The beam was passed through a homogenizer and focused down to a spot size of 5 mm×6 mm.

Samples were placed in the chamber and scanned perpendicularly to the leading beam edge, with a 96% overlap from pulse to pulse, for a variety of different conditions. The repetition rate of the laser was maintained at 35 Hz. After the annealing was completed, transmission electron microscopy (TEM) was used to determine crystalline quality and size, and atomic force microscopy (AFM) was used to study the effect of different annealing conditions on surface roughness. The size of the measured area for AFM was 10 μm×10 μm. For a few samples, secondary ion mass spectroscopy (SIMS) was employed to examine if impurities were leaching into the silicon from the glass substrate.

III. RESULTS AND DISCUSSION

It is well known that large grains can enhance the performance of polysilicon TFTs. The melt duration of the molten film and the number of pulses on a given area are primarily responsible for allowing large grains to form in laser annealed polysilicon films. One method to affect the melt duration is to heat the substrate. Another is to change the cooling rate or thermal conductivity of the surrounding medium. This can be accomplished from below the silicon by using barrier layers or above by changing the gaseous ambient. Though multiple pulses increase the average grain size in most cases, they can create substrate damage which may serve to breakup large grains. These three variables (barrier layers, ambient, multiple passes) were studied carefully to see their overall effect on the quality of laser annealed polysilicon films.

A. Barrier layer

When high temperature processes are needed to crystallize amorphous silicon films, quartz substrates must be used because typical strain temperatures for glass are around 600 °C. However, quartz substrates are expensive. In terms of applications toward large screen FPDs, the thermal constraints imposed by the substrate are even more severe. During excimer laser annealing of amorphous silicon, the majority of the photon energy is absorbed within the first 20 nm. This is a consequence of the short laser pulse width (15–30 ns typically) and high absorption coefficient of silicon in this part of the spectrum ($2 \times 10^6$ cm$^{-1}$ ). Once the energy is absorbed, it spreads through the film and substrate by thermal conduction. As a result of the high temperatures reached in the film, the Si/glass interface can experience temperatures up to 1700 °C for very short durations. At these temperatures, glass may deform and even melt, thus influencing the nucleation and recrystallization process in the top silicon layer. To alleviate these problems, a SiO$_2$ barrier layer can be added. In this manner, the temperature at the surface of the substrate can be moderated. A question, however, exists as to what are the effects of the barrier layer on the structural characteristics of the annealed silicon film. To address this question, we studied in detail the characteristics of excimer laser annealed polysilicon films with barrier layers ranging from 100 nm to 400 nm, over a wide range of energy densities, and compared them to samples made without a barrier. We found that barriers provide a simple but effective way to improve grain size and reduce contamination from the glass.

The results from our barrier study are shown in Fig. 1. The two most noticeable features from the plot are an increase in grain size and an increase in the energy density at which the maximum grain size is achieved for thicker barriers. Both of these results can be explained by using a simple heat transfer argument and relating this to the crystal growth mechanism as described by Im$^{11}$ et al. It has been reported that the thermal conductivity for deposited SiO$_2$ is lower than glass over the entire temperature range of interest.$^{12}$ As a result, the addition of a barrier layer leads to slower cooling and hence a longer melt duration for energy densities which can cause liquefaction of the top silicon layer. Figure 2 illustrates the result of a rapid cool as opposed to a slower one. On the left are nucleation sites at the silicon-SiO$_2$/glass interface which have survived the partial melting of the silicon film. With a slow cool, the grains grow until their boundaries impinge resulting in a monomodal grain size distribution and a large average grain size. For a fast cool, the grains grow until the unsolidified material becomes supercooled, whereby homogenous nucleation can occur. This results in a bimodal

![FIG. 1. Average grain size as a function of laser energy density with SiO$_2$ barrier thickness as a parameter (substrate temperature=400 °C).](image-url)
grain size distribution, where a few large grains are surrounded by an abundance of much smaller grains, and thus a smaller average grain size. Experimentally, as higher energy densities were used, fewer nucleation sites remained and so longer grain growth periods were needed to eliminate homogeneous nucleation. This was accomplished by using progressively thicker barriers, which resulted in larger grains, as indicated in Fig. 1.

Computer simulations were performed to verify the relationship between the melt duration, barrier thickness, and grain size. A one-dimensional solution to Fick’s thermal diffusion Eq. (1) was obtained using a commercial partial differential equation solver which employs a finite element method with weighted residuals:

$$\frac{\partial q}{\partial t} = k \frac{\partial^2 T}{\partial z^2} + S.$$  

In this equation, $q$ is the energy per unit volume, $T$ is the temperature, $S$ is the source term, $k$ is the thermal conductivity, $t$ is the time variable, and $z$ is the position variable. More specifically,

$$q = (\rho)^*(c_p)^*(T - T_{\text{ref}}) + (H_f),$$  

and

$$S = (1 - R)^*(E/\tau)^*\alpha^* \exp^{-\alpha z^*} u(t - \tau),$$  

where

- $\rho$ is the material density,
- $c_p$ is the heat capacity,
- $T_{\text{ref}}$ is a reference temperature ($=298$ K),
- $E$ is the laser energy density,
- $H_f$ is the melt latent heat,
- $R$ is the surface reflectance,
- $\tau$ is the pulse duration,
- $\alpha$ is the absorption coefficient of $a$-Si at 308 nm,
- $u(t - \tau)$ is a time dependent step function which turns the pulse off at $t > \tau$.

Some of the parameters used in the calculation are shown in Table I. A substrate temperature of 400 °C was assumed and, for faster convergence, the thermal properties were made to be temperature invariant. From these simulations, the melt duration as a function of barrier thickness was determined and is plotted in Fig. 3 along with the maximum grain size. This figure makes it apparent that, as suggested above, thicker barriers allow longer melt durations, which, in turn, create larger grains.

Another benefit of using a barrier layer is the reduction in impurities segregating into the silicon from the glass. From SIMS analysis, we determined the concentrations of Al, Na, and K at the polysilicon/SiO$_2$ interface as a function of barrier layer thickness. These results are shown in Fig. 4. The particular glass used was Corning 1737, which is an alumina-silicate, with low alkaline concentration. It can be seen that the amount of aluminum at the interface declined markedly from no barrier to 100 nm and changed very little for thicker barriers. Sodium and potassium concentrations at the interface declined until the barrier was 200 nm thick, and

<table>
<thead>
<tr>
<th>Material</th>
<th>Melting point (K)</th>
<th>Specific heat (cal/g K)</th>
<th>Density (g/cm$^3$)</th>
<th>Therm. conduct. (cal/cm s K)</th>
<th>Latent heat (cal/cm$^3$)</th>
<th>Absorption at 308 nm (cm$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a$-Si</td>
<td>1273</td>
<td>0.1673</td>
<td>2.33</td>
<td>0.3583</td>
<td>693</td>
<td>$1.8 \times 10^{-6}$</td>
</tr>
<tr>
<td>Glass</td>
<td>1113</td>
<td>0.1309</td>
<td>2.76</td>
<td>0.012</td>
<td>2.0</td>
<td>$2.0 \times 10^{-3}$</td>
</tr>
<tr>
<td>SiO$_2$</td>
<td>1993</td>
<td>0.1309</td>
<td>2.76</td>
<td>0.00956</td>
<td>2.0</td>
<td>$2.0 \times 10^{-3}$</td>
</tr>
</tbody>
</table>
changed very little as the barrier thickness was increased to 400 nm. We concluded from these results that a 200 nm barrier would be sufficient to maximize the impurity blocking effect of the barrier layer.

B. Annealing ambient

The choice of annealing ambient can significantly affect the grain growth characteristics of laser annealed polysilicon films. The presence of gas in the annealing chamber cools the sample surface directly, which can change both the melt duration and the energy density threshold where melting occurs. As with the barrier layer study, and for the same reasons, a faster cooling rate limits the maximum achievable grain size. The ambients we choose to study were nitrogen, helium, argon, and rough vacuum. Air was not selected because our previous work,13 as shown in Fig. 5, and work done by others14 suggests that oxygen is detrimental to surface roughness. Results from the ambient investigation are shown in Fig. 6, where for this experiment, a 200 nm barrier was employed. Figure 7 shows the relationship between the maximum grain size from Fig. 6 and the thermal conductivity \( k \) of the ambients, assuming \( k=0 \) for vacuum. This figure emphasizes the importance of heat transfer in limiting grain size.

It can also be seen from Fig. 6 that large grain growth initiated at a lower energy density for samples made in either nitrogen, helium, or argon as opposed to vacuum. This may have been due to a delay in the onset of liquefaction during the pulse duration, from the cooling effect of the gas. This delay could have prevented the increased reflectivity of the liquid phase to an extent that more energy of each pulse was absorbed, the effect being a shift of the curves depicting grain size versus energy density toward lower energy densities.

Another source of concern when an ambient other than vacuum was used is uniformity. Especially with helium, it became difficult to create films composed entirely of large grains because small grains formed homogeneously could be found under almost any condition, creating regions with vastly different structural characteristics. The problem was less severe with nitrogen and argon and could be avoided entirely with a careful selection of energy density.
C. Multiple passes

The effect of multiple passes is shown in Fig. 8. Other authors have reported similar results in regards to the effect of the number of pulses for a given area on grain size.15,16 From the plot, most noticeably, multiple passes lead to an increase in the average grain size. When using a lower energy density (260 mJ/cm²) with no barrier and substrate heating, the average grain size first increased then saturated. When a higher energy density (290 mJ/cm²) was used, the average grain size increased initially and then decreased with more passes. This decrease in average grain size was likely due to the kind of surface roughening described by Anderson et al.17 That is, damage to the glass interface generated nucleation sites which reduced the average grain size. This is not to say that this mechanism of grain size reduction was the source of small grains in single passes at high energy densities. On the contrary, two step experiments were conducted where small grains were created by an initial scan at a high energy density (around 400 mJ/cm²) then a second scan was made at a lower energy density (around 310 mJ/cm²) and large grains were recovered suggests otherwise. But interface roughening can create enough nucleation sites to limit large grains which would form if a smooth interface were maintained. From TEM images, it appeared that some of the largest crystals which formed after the eighth pass were broken up after 16 passes. When the same conditions were applied to a sample with a 200 nm barrier, a decrease in grain size did not occur from 8 to 16 passes. The higher melting point of SiO₂ makes it more resistant to thermal damage. Hence, it provided a thermal buffer between the silicon and glass, leading to a lesser likelihood of damage induced nucleation sites, and as a result, large grains remained intact after the sixteenth pass.

Figure 8 also shows the results of using a multiple scan process on a sample annealed at RT. For comparison purposes, a higher energy density was used so that the grain size after the first scan would be approximately the same as the sample crystallized at 400 °C using 290 mJ/cm². In this case, the grain size continually increased as more passes were completed. The lack of substrate heating caused the average grain size to increase at a slower rate, but after the sixteenth pass, the transformation to a film composed entirely of large grains was nearly finalized. With this technique, and the use of a barrier layer, we were able to increase the average grain size to upwards of 650 nm without substrate heating.
From inspection of TEM images, it can also be seen that multiple passes can enhance two-dimensional ordering in laser annealed polysilicon films, as shown in Figs. 9(a)–9(c). After one scan, the grains were more or less randomly located, but after two scans, the grains in a significant portion of the film became organized into well-defined perpendicular rows and columns. At this point, uniformity was excellent, and coupled with the ordering effect, the film took on a honeycomb appearance. With more passes, the grains grew, but not necessarily uniformly. Rather, a few grains became much larger than the rest. After eight and 16 passes, the grains had all grown large, some on the order of several microns, and they appeared entirely different from grains formed initially, being less regularly shaped. The ordering was still quite noticeable, though the relationship between ordering and the grain size was less obvious.

It may be that this ordering phenomenon is related to the so-called laser induced periodic surface structure or LIPSS. Other authors have produced LIPSS using a similar process, and this conjecture has support in that the wavelength of the ordering is approximately equal to the wavelength of the incident radiation (308 nm), as one would expect for

<table>
<thead>
<tr>
<th>Number of scans</th>
<th>1</th>
<th>2</th>
<th>8</th>
</tr>
</thead>
<tbody>
<tr>
<td>rms surface roughness (nm)</td>
<td>12.8</td>
<td>12.3</td>
<td>5.7</td>
</tr>
</tbody>
</table>

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![AFM images of polysilicon scanned at a laser fluence of 305 mJ/cm² with a 200 nm barrier](image-url)
LIPSS.\textsuperscript{14} The question arises though as to why the ordering occurred in two dimensions, and not one, as is typical for LIPSS. It is important to note that this effect only occurred over a narrow range of energy densities. It also occurred under certain conditions in both argon and nitrogen and strongly in samples annealed at RT.

To see the effect of multiple passes on surface roughness, AFM measurements were performed. The results are shown in Table II. The most striking feature here is that the surface roughness dropped dramatically from the fourth pass to the eighth. In many cases, surface roughness increases as the average grain size increases,\textsuperscript{19} but, as our results indicate, this is not always so. What is more important in relating surface roughness, as measured with AFM, to film characteristics, as seen with TEM, is the particular grain structure itself. Four AFM images are shown in Figs. 10(a)–10(d). Figures 10(a) and 10(b) are a planar and three-dimensional view taken from a sample, with a 200 nm barrier, scanned twice (2$\times$) at 290 mJ/cm$^2$ with a substrate temperature of 400 °C. Figures 10(c) and 10(d) are likewise taken from a sample scanned eight times (8$\times$). Though the grain size was smaller in the 2$\times$ sample, the roughness was certainly greater. The major cause of this increased roughness was not the grain boundaries, but rather the intersection of grain boundaries. The regularity of the structure in the 2$\times$ sample created an abundance of points where many grain boundaries converged. These vertices were also where the largest surface features were located, as seen in Fig. 10(b). In the 8$\times$ sample, the grains were more irregular and there were fewer points where multiple grain boundaries intersected, so the surface roughness was less. An explanation of the relationship between grain boundaries and surface roughness was given by Fork et al.\textsuperscript{18} In the 2$\times$ sample, at the points where several melt fronts would have been converging upon solidification, the liquid silicon was squeezed from several directions by the expanding, solidified silicon into a sharply peaked bump. At a typical grain boundary, the force on the melted area comes from only two directions, so the magnitude of the feature is less.

IV. CONCLUSION

Several methods have been studied in an attempt to produce high quality laser annealed polysilicon films and to improve throughput. SiO$_2$ barrier layers allowed the formation of larger grains by slowing the cooling rate. Barriers also provided an effective means of blocking contaminant segregation from the glass. Laser annealing in a nitrogen or argon ambient allowed the formation of films with qualities comparable to vacuum. Uniformity was generally good and despite smaller grains, the films were in many cases indistinguishable from samples made in vacuum. Helium, on the other hand, did not produce high quality samples. Almost without exception, the films were nonuniform, as there were unavoidably regions where large grains began to grow, but were halted by the rapid cooling. Multiple passes created ordered structures and increased grain size at the same time. However, due to the points where many grain boundaries converged in the ordered films, the roughness was quite high. This continued until very large grain growth was initiated, whereupon the roughness was dramatically reduced. Multiple passes, especially when a barrier layer was employed, proved to be a viable technique to produce large grains without substrate heating.

\begin{itemize}
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\item[]\textsuperscript{11}J. S. Im, H. J. Kim, and M. O. Thompson, Appl. Phys. Lett. 63, 1969 (1993).
\end{itemize}