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Self Organized Grain Growth Larger than 1 μm through Pulsed-Laser-Induced Melting of Silicon Films

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Large-grain growth (>1 μm) was observed through pulsed-laser-induced melting of silicon films. When silicon islands with a width of 10 μm and a thickness of 70 nm formed on a quartz substrate were melted completely by irradiation of a XeCl excimer laser with an energy of 360 mJ/cm^2, the shape of the film was changed into a globular shape. The width shrank to 2.4 μm and the film thickness increased to 270 nm near the edge. Large grains with a size of 1.2 μm were grown and grains were lined two deep in the globular islands. The large-grain growth occurs along a temperature gradient in the lateral direction, which is caused during the change of the film shape.

KEYWORDS: thin film transistor, lateral grain growth, interface-controlled growth, homogeneous solidification

Polycrystalline silicon with large grains (>1 μm) is an attractive material for fabricating polycrystalline thin-film transistors (poly-Si TFTs) with a large carrier mobility. A few methods have been proposed to form poly-Si films with large grains.[1,2] However, there is still a problem of controlling the grain size and shape. Large but random grains can cause large variations of carrier mobility and threshold voltage in poly-Si TFTs because the number of grain boundaries in the channel region cannot be controlled. Moreover, poly-Si TFTs must be fabricated at a low processing temperature (<400°C) if they are applied to devices on a substrate with no heat resistance such as glass. We reports a mode of pulsed-laser-induced melting followed by regrowth of silicon films, which provides a crystalline grain larger than 1 μm at room temperature ambient. The possibility of controlling the grain size and shape is also discussed.

Polycrystalline silicon films with a thickness of 70 nm were deposited using low-pressure chemical vapor deposition (LPCVD) on quartz substrates. Silicon islands with a width of 10 μm and a length of 40 μm were then defined. The samples were irradiated with XeCl excimer laser pulses with a pulse of 30 ns in full width at half-maximum and a beam size of 5 mm × 5 mm at room temperature in a vacuum chamber. The silicon islands were melted by irradiation with an energy density of 360 mJ/cm^2 and 340 mJ/cm^2. A change of shape into a globular shape was observed. The widths of the islands were reduced to 2.4 μm and 7 μm with the irradiation energies of 360 and 340 mJ/cm^2, respectively. The thicknesses were increased to 270 nm and 180 nm, which were maximal near the edge, for the irradiation energies of 360 and 340 mJ/cm^2. Electron channelling measurement was used to investigate the crystalline state of the laser-annealed films. Figure 1 shows photographs of an electron channelling image. Crystalline grains with a size of about 1.2 μm $\times$ 0.5 μm were observed. The large grains were formed over the entire islands, and they were lined two deep along the length of the silicon island for the 360 mJ/cm^2 irradiation case. On the other hand, large grains were oberved in the edge regions and they were lined along the length of the silicon island for the 340 mJ/cm^2 irradiation case. Fine grains were formed in the middle regions, and their grain size was smaller than the detection limit of the electron channeling measurement.

We have already demonstrated two solidification modes of molten silicon induced by XeCl excimer laser pulses formed on quartz substrate for non patterned silicon films.[3–6] They are interface-controlled growth and homogeneous solidification. In the interface-controlled growth, silicon films are melted and a liquid-solid interface is formed in the films by irradiation with energy above the surface melting threshold of 160 mJ/cm^2. After irradiation, the temperature decreases and the interface moves toward the surface through heat diffusion into the substrate. The speed of interface mortion is governed by heat diffusion into the quartz substrate; it was experimentally determined as 0.6 m/s using transient conductance for 20-nm-thick silicon films.[6] Solidification therefore occurs along the direction of interface movement in the direction vertical to the surface. Crystalline grains can grow spherically from nucleation sites located near the Si/quartz interface in the initial stage of melting. The grain growth is limited by the reduction of the melt region in the vertical direction by the rate of 0.6 m/s. We previously determined that non patterned 30-nm-thick silicon films are crystallized with grain size smaller than 60 nm through melting with a duration of 80 ns at the surface, induced by irradiation with an energy of 270 mJ/cm^2. Crystalline grains were distributed randomly. On the other hand, homogeneous solidification occurs when silicon films are melted completely by irradiation with higher energy. When the temperature gradient in the liquid silicon is reduced to lower than 1×10^5 K/cm during long melting, solidification occurs homogeneously throughout the film through the isothermal supercooling state. The solidification occurs much more rapidly than the inerface controlled growth.[6] This solidification mode can therefore lead to silicon films in the amorphous state. Newly solidified amorphous state is reheated with recalescence caused by latent heat which is released during rapid solidification. Crystallization of fine grains is induced by recalescence when the film is thicker than 24 nm because the latent heat energy per unit area increases as film thickness increases.[5]

In the present laser-induced melting of silicon islands, the shrinkage of islands was observed. This

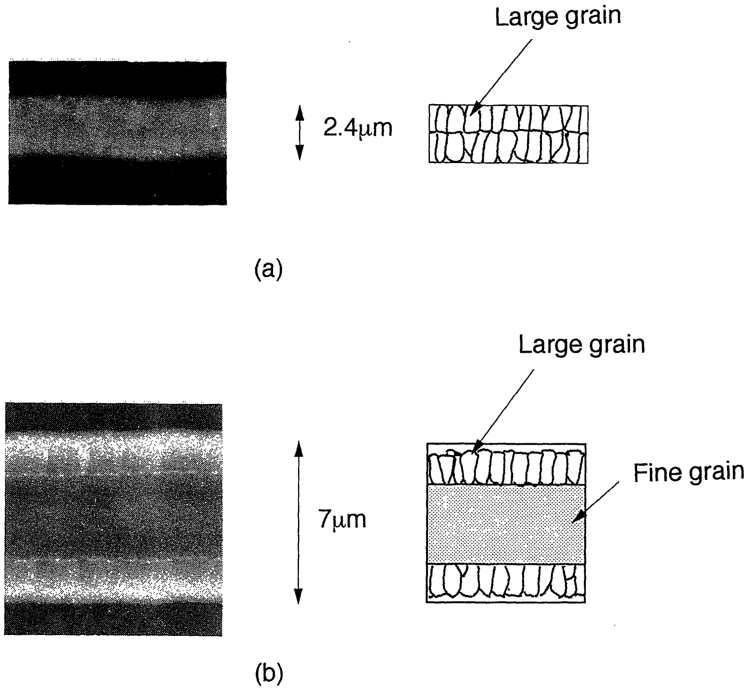

Fig. 1. Photographs of electron channelling image of laser-annealed silicon islands with laser energy of 360 mJ/cm^2 (a) and 340 mJ/cm^2 (b). Schematic distributions of grain boundaries are also given.

result shows that the silicon islands was melted completely. However, the silicon islands were not amorphized, in contrast to non patterned silicon film case. Large-grain growth occurred on the islands, as shown in Fig. 1. These results indicate that another solidification mode can occur with shrinkage of islands. When silicon islands formed on quartz substrates are melted completely by irradiation, the films undergo a change of shape into a globular shape due to surface tension. Because the area shrinks from the edges and the thickness increases at edge regions, heat diffusion is reduced and thermal energy is piled up in liquid silicon at edge regions during melting, as shown by the illustration for the 360 mJ/cm^2 irradiation case in Fig. 2. The increase of the thickenss of molten silicon therefore lowers the cooling rate and lengthens the melt duration. On the other hand, the degree of heat diffusion in the middle region of the island is almost the same as that of non patterned silicon films formed on quartz substrates.

Figure 3 shows the calculated cooling rate at the Si/quartz interface, if the solidification point is assumed to be 1412°C, as a function of the thickness of molten silicon with different melt durations using the one-dimensional heat diffusion equation.[7,8] The cooling rate decreases as the thickness increases for each melt duration. Heat flow calculation gave the melt duration of 70-nm-thick films as 140 ns for an energy of 360 mJ/cm^2, and the cooling rate is 6.5×10^9 K/s at the solidification point. If the silicon films change into a globular shape and the thickness increases to 270 nm immediately after the silicon film is melted completely, the melt duration is increased to 400 ns and the cooling rate is reduced to 2×10^9 K/s. The temperature at the edge region therefore decreases more slowly than that at the middle region, and a temperature gradient is formed in the lateral direction. When the temperature gradient is larger than 1×10^5 K/cm, interface-controlled growth in the lateral direction can occur, as discussed above. On the other hand, the temperature gradient in the vertical direction is lower than 1×10^5 K/cm because the irradiation energy is higher than the required energy of homogeneous solidification for non patterned silicon films. The present crystallization of silicon islands can therefore be interpreted as interface-controlled growth in the lateral direction from the middle region toward the edge as can be seen in Fig. 2. The grain growth should occur spherically from crystalline nucleation sites in the initial stage, but it is governed by the interface movement in the lateral direction. Figure 1 shows that the grain length of 1.2 μm and the position of grains in the direction of grain length were defined well. This result is evidence that the crystallization occurs along the thermal gradient in the lateral direction. The change of shape of liquid silicon on quartz substrates, which induces the temperature gradient in the lateral direction, is a self-organized phenomenon. The 360 mJ/cm^2 irradiation melted the silicon islands long enough to cause the shrinkage and to crystallize silicon islands in the lateral direction over the entire islands with the initial width of 10 μm. However, for the 340 mJ/cm^2-irradiation case, the melt duration was shorter so that the shrinkage of islands and crystallization in the lateral direction occurred in only the edge region, and fine grains were formed in the middle region through homogeneous solidification because the temperature gradient was lower than 1×10^5 K/cm in both of lateral and vertical directions in the

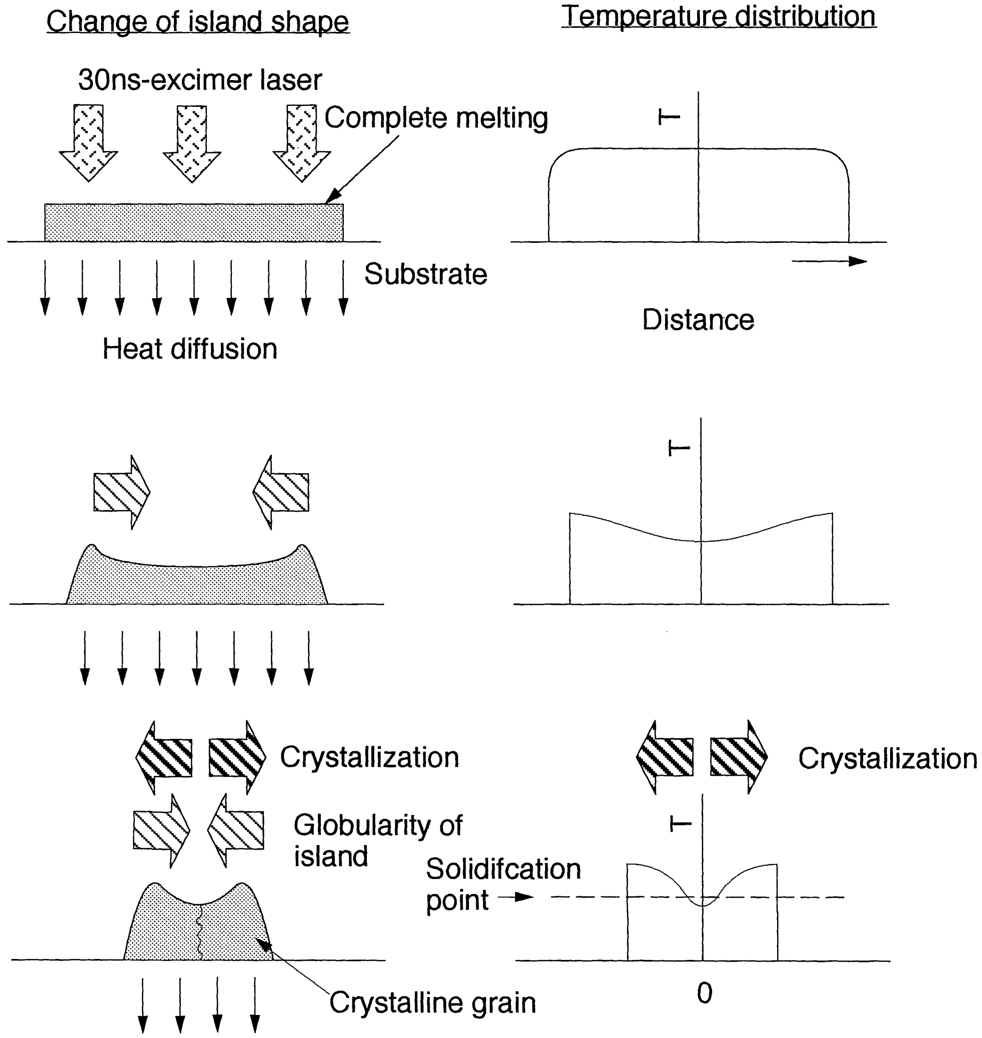

Fig. 2. Illustration of change of island into a globular shape and evolution of temperature gradient for the 360 mJ/cm^2 irradiation case.

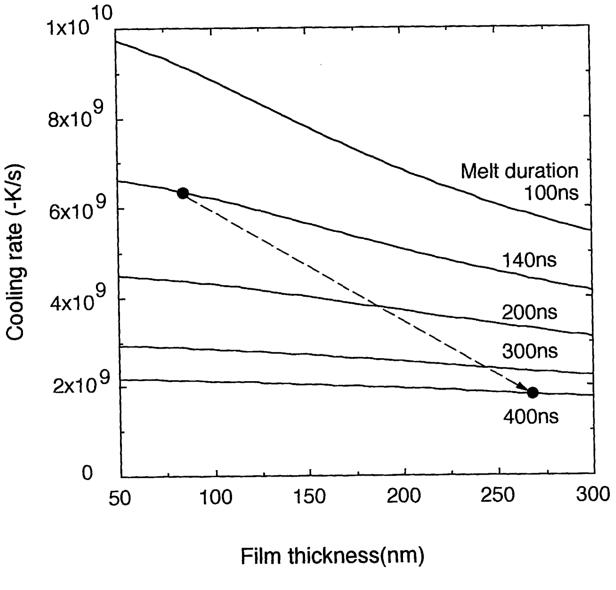

Fig. 3. Calculated cooling rate as a function of film thickness at solidification point of 1412°C with different melt durations. Solid marks represent cooling rates at the solidification point for 70-nm-thick non patterned silicon film melted by irradiation with 360 mJ/cm^2, and for 270-nm-thick silicon film if the film changed immediately to the globular shape.

middle region. The control of size and position of grains is an advantage in fabrication of electron devices such as thin film transistors, by controlling grain boundary in a channel region. The grain size in the other direction is about 0.5 μm, although it was not controlled well. The length (40 μm) of the islands was much longer than the degree of shrinkage of the islands. The degree of temperature gradient in this lateral direction was therefore too small to cause the crystallization along this direction. Random nucleation consequently occurred in this direction.

In summary, large-grain growth (>1 μm) was observed through pulsed laser-induced melting of silicon film. When silicon islands with a width of 10 μm and a thickness of 70 nm formed on a quartz substrate were melted completely with irradiation of a pulsed XeCl excimer laser with an energy of 360 mJ/cm^2, the shape of the film was changed into a globular shape. The width shrank to 2.4 μm and the film thickness increased to 270 nm near the edge. Large grains with a size of 1.2 μm were grown, and grains were lined two deep on the globular islands. The large-grain growth occurs along a temperature gradient in the lateral direction, which is caused during the change of the film shape. A temperature gradient occurs in the lateral direction during the shrinkage of silicon islands because thickness increases near the edge and thermal energy becomes larger than that of the middle region. Crystallization occurs along the temperature gradient from the middle to the edge. The present process has an advantage on controlling grain size because a temperature gradient is induced in the lateral direction during melting.

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