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# A Study of Nucleation and Grain Growth in Silicon Implanted a-Silicon Films

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The solid-phase-recrystallization process was investigated for Si implanted amorphous silicon films. This process consists of nucleation and grain growth. The recrystallization process had two stages. The nucleation of grains was dominant at the first stage. Both nucleation and grain growth occurred at the second stage. The activation energies for nucleation and grain growth were determined by a new statistical approach, using the number of grains and the distribution of grain size. The obtained activation energies for nucleation and grain growth were 5.9 eV and 2.8 eV, respectively. The rate-determining step of the grain growth seemed to be similar to that of lateral solid phase epitaxy.

### 1. INTRODUCTION

Recently, thin film transistors (TFTs) are fabricated in solid-phase-recrystallized polycrystalline silicon films in order to reduce their size and to increase their speed in such devices as SRAMs and LCDs<sup>1,2)</sup>. The grain size strongly affects the device characteristics<sup>3)</sup>. It is important to control the grain size in order to use these devices. Grain size is determined by the nucleation rate and grain-growth rate in amorphous silicon (a-Si) films. Understanding the mechanism of nucleation and grain growth is necessary to control the grain size. Nucleation and grain growth should be clearly distinguished for precise analysis. However, these phenomena had not been well investigated, because obtained experimental results are a combination of nucleation and grain growth.

This paper discusses the results of investigating the solid-phase-recrystallization process by separating this

process into nucleation and grain growth, by using a new statistical approach. The number of grains was counted exactly during the early regrowth stage, and the distribution of grain size was measured during the growth stage by TEM observation. The nucleation rate was determined directly. The growth rate of a grain was derived by a newly proposed model for the growth stage. The activation energies for both nucleation and grain growth were determined. The mechanism of grain growth is discussed with obtained results.

#### 2. EXPERIMENT

2000 Å-polycrystalline silicon films were deposited on silicon wafers covered with thermally grown  $SiO_2$  layers. Amorphous silicon films were fabricated by siliconion-implantation into the deposited layer. The silicon-ion-implantation condition was  $2.5x10^{15}$ cm<sup>-2</sup> at 50 keV and  $5.4x10^{15}$ cm<sup>-2</sup> at 120 keV. The amorphized films were recrystallized by isothermal furnace annealing in the temperature range from 600 °C to 640 °C in a nitrogen ambient.

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Fig.1 TEM photographs of films annealed at 610 °C for (a)12 h, (b)16 h and (c)20 h

The recrystallization process was investigated by TEM observation. The number of grains and the area of each grain were measured for various annealing times. These values were determined by planer TEM photographs, such as Figs. 1 (a)-(c). Defects in the grains were precisely observed by high-resolution TEM (400 keV).

#### 3. RESULTS

Figure 2 shows the annealing time dependence of the crystalline fraction at various annealing temperatures. This figure shows that the recrystallization process can be divided into two stages. During the first stage (I), crystallization hardly proceeds. During the second stage (II), the crystalline fraction increases very rapidly.

The number of grains was directly counted by TEM observation. The result is shown in Fig.3 as a function of annealing time. At first, nucleation of a grain was hardly observed. Then, the nucleation rate became constant.

The authors deduced the nucleation rate by derivating the number of grains. Figure 4 shows the annealing time dependence of the nucleation rate at various annealing temperatures. During stage (I), the nucleation rate increased as the annealing time increased. The energy relaxation time of nucleation ( $\mathcal{T}$ ), which was defined from the slope of the solid line in Fig.4, is also presented in this figure.

The incubation time  $(t_0)$  was determined as the intersecting point of the extrapolation of the straight part of the lines and the axis of the annealing time in Fig.3. Figure 5 shows the annealing temperature dependence of the incubation time. This Arrhenius plot has a slope of 3.9 eV.

The steady-state nucleation rate is derived from the slope of the straight part of the lines in Fig.3. Figure 6 shows the effect of annealing temperature on the steady-state nucleation rate, and its activation energy was 5.9 eV.

Figure 7 shows the distribution of the grain area, determined by TEM photographs (Fig.1), for films annealed for 12, 16 and 20 hours at 610 °C. The solid lines show the theoretical curves calculated by the newly



Fig.2 Crystalline fraction vs. annealing time





proposed model, in which the activation energy of nucleation (5.9 eV) was considered, as will be described in the following section. Figure 8 shows the annealing temperature dependence of the growth rate of a grain, which was deduced using this model. The activation energy of grain growth was 2.8 eV.

## 4. DISCUSSION

To calculate the theoretical curves shown in Fig.7, the annealing-time dependence of the nucleation rate was considered, which is clearly observed in Fig.4. It had been assumed in most of the studies that the nucleation rate until the incubation time  $(\tau_0)$  was zero and then became constant<sup>4,5</sup>). In this study, its annealing-time dependence was approximated as a power solution, and the theoretical distribution of the area of a grain was expressed as

$$f(S) = \frac{\gamma}{2\sqrt{\pi S} v_g} (t - \frac{\sqrt{S}}{\sqrt{\pi} v_g})^3 \quad (1)$$

where grains were assumed to grow in a twodimensional (cylindrical) way. It is obvious that the experimental results and the theoretical curves agree well. The increase in the number of small grains cannot be explained, if the annealing-time dependence of the nucleation rate is not considered. Thus, this method of numerical







Fig.7 Distribution of grain area

solution is inevitable for the precise determination of the growth rate  $(v_g)$  and other parameters.

The experimentally obtained activation energies are related to the important thermodynamic parameters of nucleation and grain growth in a-Si. The activation free energy for the formation of a cluster  $(\Delta G_n^*)$ , having a critical size above which the cluster can grow and below which it can vanish, and the free-energy difference between the amorphous phase and crystalline phase ( $\Delta G'$ ) were calculated as follows:

 $\Delta G_n^* = E(\gamma_n) - E(\tau_0)$ (2) and

$$\Delta G' = -2(E(v_g) - E_d),$$
 (3)

where  $E(\gamma_n)$  is the activation energy of the steady-state nucleation rate,  $E(\mathcal{I}_0)$  is the slope of the incubation time on an Arrhenius plot,  $E(v_g)$  is the activation energy of the growth rate, and  $E_d$  is the activation energy of self-diffusion in a-Si.  $E_d$  is written as  $E_d = E(\mathcal{I}_0) + kT$ .



Fig.6 Steady-state nucleation rate vs. annealing time



Using these equations,  $\varDelta \texttt{G}^{\ *}_n$  and  $\varDelta \texttt{G}^{\prime}$  can be derived as

 $\Delta G_n^* = 2.0 eV$ 

 $\Delta G' = 2.0 eV.$ 

(5)

(4)

Iverson and Reif<sup>5)</sup> had derived these values by a similar experimental method. They found  $\Delta G_n^*$  to be 2.4 eV and  $\Delta G'$  to be -0.1 eV. In the authors' experiment, the distribution of grain size during recrystallization was precisely investigated for the first time and the growth rate of a grain was able to be derived directly by TEM observation.

The growth rate was much lower than that of lateral solid phase epitaxy (L-SPE), while the derived activation energy of grain growth, 2.8 eV, was almost the same as that of L-SPE<sup>6,7)</sup>. These results suggest that the mechanism of grain growth is not essentially different from that of L-SPE but that the migration rate from the amorphous phase to the crystalline phase is much lower. Figure 9 shows a TEM image of a typical grain and the diffraction pattern at the center of the grain. Every grain had twins in its center as shown by the diffraction pattern, and it grew preferentially along the twin plane. This suggests that the twin-edges are the main growth point. The activation energy of grain-growth seems to be mainly based on growth at the twin. A probable atomic model



Fig.9 TEM image of typical grain and diffraction pattern at center of grain



Fig.10 Atomic model of twin-edges

for the twin-edges is shown in Fig.10, and the white arrow indicates the growth point. This point has a structural advantage for grain growth, such as bond bending relaxation or a high probability of bonding. The mechanism for twin-growth is supported by these atomic structure effects.

#### 5. CONCLUSION

Solid-phase-recrystallization of a-Si films was investigated by considering that the recrystallization process had two stages. During the first stage, grain nucleation was dominant. During the second stage, both nucleation and grain growth occurred. The activation energies for nucleation and grain growth were determined by a new statistical approach, considering the number of nucleated grains and the distribution of grain size. The obtained activation energy for nucleation and grain growth were 3.9 eV and 2.8 eV, respectively. The rate-determining step of grain growth seemed to be similar to that of lateral solid phase epitaxy.

#### REFERENCES

- T. Yamanaka et al.; IEDM Technical Digest, p.48, 1988.
- A. Miura et al.; IEEE Trans. Electron Devices, <u>EDL-36</u> (1989) 351.
  T. Katoh; IEEE Trans. Electron Devices,
- 3) T. Katoh; IEEE Trans. Electron Devices, EDL-35 (1988) 923.
- 4) K. Zellama et al.; J. Appl. Phys. <u>50</u> (1979) 6995.
- 5) R. B. Iverson and R. Reif; J. Appl. Phys. <u>62</u> (1987) 1675.
- 6) H. Yamamoto et al.; Jpn. J. Appl. Phys. 25 (1986) 667.
- 7) M.Tabe and Y. Kunii; Mat. Res. Soc. Symp. Proc. <u>35</u> (1985) 695.