Tendency of crystal orientation rotation toward stable {001} <100> during lateral crystal growth of Si thin film sandwiched by SiO₂

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Stable crystal orientation (CO) for lateral growth of Si thin film sandwiched by SiO₂ was evidenced to be only {001} in normal direction (ND {001}) and (100) ±5° in scanning direction (SD (100)). Crystal with ND{001} is quasi-stable when angle θ between inplane (110) and SD is among 15° ≤ θ ≤ 40° and is unstable when θ is < 15°. CO other than the stable CO will rotate spontaneously toward the stable CO, i.e. ND{001} with SD(100) ±5°. Most ND{001} crystal was ended by twinning before the CO come to the stable CO. The twinning was triggered by gas ejection or particles, so suppressing of these phenomena would be the key for increasing ND(001)/SD(100) crystal occupations. These results have been verified for crystal growth velocity among 0.04–45 mm s⁻¹. © 2021 The Author(s). Published on behalf of The Japan Society of Applied Physics by IOP Publishing Ltd

1. Introduction

Growth of high quality crystalline silicon thin film on amorphous materials such as quartz or glass substrate has long been a challenge for achieving both fully-depleted semiconductor on insulator transistors in integrated circuits¹,² and thin-film transistors (TFTs) for pixel driving circuit in active-matrix flat panel displays.³⁻⁹ With respect to the TFTs, importance of high mobility TFTs are increasing, because of demand for driving current-driven organic light emitting diode¹⁰,¹¹ or micro light emitting diode (µLED)¹²,¹³ In particular µLED display can provide very high brightness of up to 10⁷ cd m⁻²,¹⁴ current driving ability of TFTs must be correspondingly increased. Polycrystalline Si (poly-Si) film formed by excimer laser crystallization of amorphous Si (a-Si) film provide TFTs with a field effect electron mobility µ of >100 cm² V⁻¹ s⁻¹,³ however, because of its polycrystalline behavior, µ was lower and characteristic deviation was larger than these of single crystal Si. Alternatively, continuous wave laser crystallization (CLC)¹⁵⁻²⁰ became a promising candidate for realizing crystal Si film with high mobility because of continuous lateral grain growth. Problem of CLC was existence of long grain boundaries in parallel to laser scanning direction (SD) that will deteriorate off characteristics of transistors. Attempts to grow single crystal Si strip on SiO₂ in CLC have been performed in the early 1980s for SOI, by means of arranging laser beam shape such as donut-shape,¹⁵ twin Gaussian shape,¹⁶ and crescent shape.¹⁷ In those attempts, instable Ar gas laser was used, and furthermore, the beam dimension was as large as several ten microns with gentle laser beam edge so polycrystalline region existed at side of the single crystal strip. High-power ultraviolet (UV) or blue laser diode (LD) for CLC were proposed at around 2010²²,²⁵ in which we used line laser beam from a multimode UV-LD for CLC for the first time.²⁵ Then we proposed micro-chevron laser beam scanning (µCLBS) method²⁷ in which µCLBS was produced using one-sided Dove prism and a multimode UV-LD, by which single crystal Si strip free of random grain boundary (RGB) was realized. Then we clarified that twinning in the strip was growth twin and was generally originated at Si/SiO₂ interface, so improvement of the interface quality is essential for reducing twinning boundaries.²⁴ When TFTs were fabricated on a single strip, low characteristic deviation on subthreshold swing, threshold voltage, and off current achieved with a high field effect mobility (>500 cm² V⁻¹ s⁻¹).²⁹ However, because crystal orientation (CO) changed with crystal growth progressing along the Si strip, variability of field effect mobility of TFTs was as large as 34%. Therefore, control of CO in both normal direction (ND) and crystal growth direction [referred to as SD hereafter] become important for TFTs. As for the CO control, Sasaki and Kuroki found CLC at near threshold power density for lateral growth result in Si film preferentially {001} textured in ND (referred to as ND{001} hereafter) at first,²⁹,³⁰ however, the mechanism was not clarified and there still exist grains with other orientations. In µCLBS method, we reveal CO rotation in pitch direction is owing to expansion rate difference of the Si film between bottom surface and top surface during solidification.³¹ After that, CO rotation caused by expansion rate difference was suppressed by capping the Si film with SiO₂ film thicker than 200 nm, due to symmetric SiO₂/Si/SiO₂ sandwiched structure. As a result, stable growth of ND{001} crystal have realized and length ratio of ND{001} strip have increased to 91% by further optimizing laser scanning speed and laser power.³² But it was further found that in non ND{001} crystals, CO was still rotating in a random directions at glance. Furthermore, no predominant CO in SD was found in the ND{001} strips. Although Sasaki et al. recently claimed that they have realized ND{001}/SD(100) crystalline Si film in CLC, but they did not show the process to achieve it and did not provide convincing quantitative results.³³ For further control of CO, it is important to clear the mechanism of ND{001} crystal formation. Generally, there can be three possible options for the formation of ND{001} crystals. 1. Nuclei for crystal growth were all ND{001} crystals. 2. Nuclei were with random CO and crystals grew from it changed their CO to ND{001} during growth. 3. Nuclei were with random CO and only ND{001} crystal survived after
competition (this will not take place because only one grain growth is permitted in $\mu$CLBS). In this paper, the above questions will be clarified.

2. Experimental methods

60 nm thick a-Si film capped with 200–300 nm thick SiO$_2$ film on quartz glass was used as a sample. The Si film and SiO$_2$ film were deposited respectively by low pressure chemical vapor deposition method and reactive pulse-DC sputtering method. 405 nm wavelength chevron laser beam with a dimension of 10 $\mu$m was scanned through the sample at a speed $v$ among 0.04–45 mm s$^{-1}$ at laser power $P$ among 70–250 mW. The Si strips were then evaluated by electron backscatter diffraction (EBSD) method after removal of surface SiO$_2$ capping film by 5% HF solution.

3. Results and discussion

3.1. CO of nuclei in uncapped Si film at near completely melting threshold

Figure 1 shows (a) CO map in ND, (b) CO map in SD, and (c) grain-boundary (GB) map, of $\mu$CLBS Si strip with $v = 10$ mm s$^{-1}$ and with $P$ of 192, 201, and 209 mW. Coordinate indicating ND, SD, and transverse direction (TD) was shown in all figures necessary henceforth. In Si strip with $P = 209$ mW, there is no RGB, indicating the Si film was above complete melting threshold so lateral crystal growth continued. This result are the same as our previous results in Refs. 24 and 28. CO was rotating with crystal growth in pitch direction because of expansion difference in top surface and rear surface of Si film at solidification. There is no preferential CO, and twinning took place frequently when $\{111\}$ come near parallel to film plane. With $P = 201$ mW, lateral growth grains break apart. The Si film must be under completely melting threshold so lateral growth was frequently replaced by new grain originated from unmolten Si at Si/SiO$_2$ interface as nuclei. The CO of these small lateral grains reflects that of nuclei. Figure 2 shows inverse pole figure (IPF) in ND, SD, and TD of all grains with $P = 201$ mW, in which blue dot indicates CO of each grains, and red circles indicates CO of each grains weighted by its grain size. From distribution of blue dots, CO of all grains were weakly preferentially $\{001\}$ in ND, but no preferential CO in SD or TD. The CO distribution of these grains directly reflect that of nuclei. Besides, from distribution of red circles, CO of grains grew larger were preferentially $\{001\}$ in ND, but no preferential CO in SD or TD. This suggest ND$\{001\}$ is a most preferable CO for crystal growth in Si thin film. In the next section, preferential CO of Si strip was further investigated in Si film capped with SiO$_2$ for preventing unwanted pitch rotation.

3.2. Stable and quasi-stable CO of Si crystal capped with SiO$_2$

Figure 3 shows CO map in both ND and SD of twelve 5 mm long Si strips ($v = 0.4$ mm s$^{-1}$ and $P = 142$ mW). The displacement $x$ in SD was indicated in the figure, in which the origin of $x$ was an origin of EBSD measurement but was not an origin of laser scanning. CO of the Si strips were preferentially ND$\{001\}$, with a length ratio of approximately 78%. On the other hand, it seems that SD did not have a preferred CO. IPF plots in ND, SD, and TD of all Si strips

![Fig. 1.](https://example.com/fig1.png)  
(Color online) EBSD CO map in ND, CO map in SD, and GB map, of Si strips with laser power of 192 mW, 201 mW, and 209 mW.

![Fig. 2.](https://example.com/fig2.png)  
(Color online) IPF of all grains from a 1 mm long Si crystal with $P = 201$ mW in ND, SD, and TD. Blue dot indicates CO of each grains, and red circles indicates CO of each grains weighted by its grain size.

![Fig. 3.](https://example.com/fig3.png)  
(Color online) EBSD CO map in both ND and SD of twelve 5 mm long Si strips. Blue dot indicates CO of each grains, and red circles indicates CO of each grains weighted by its grain size.
longer than 200 $\mu$m in Fig. 3 were shown in Fig. 4(a), in which strip areas having misorientation angle $\phi$ from ND{001} of $0^\circ$–$2^\circ$, $2^\circ$–$5^\circ$, $5^\circ$–$10^\circ$, and $>10^\circ$ were respectively shown by red, green, blue, and black colors. Area fraction of these areas were shown in Fig. 4(b). Area fraction was $66.2\%$ for $\phi \leq 2^\circ$, $19.8\%$ for $2^\circ < \phi \leq 5^\circ$, $5.9\%$ for $5^\circ < \phi \leq 10^\circ$, and $8.1\%$ for $\phi > 10^\circ$. Area with $\phi \leq 2^\circ$ was from the stably growing ND{001} crystals, and area with $2^\circ < \phi < 10^\circ$ was from side region of the ND{001} crystal or from crystals on the way to ND{001}. From SD IPF in Fig. 4(a), only when angle between inplane {110} and SD, $\theta$, was larger than $8^\circ$, ND{001} crystal be present, else be absent. This suggest ND

Fig. 3. (Color online) EBSD CO map in both ND and SD of capped Si with $v = 0.4$ mm s$^{-1}$. There are twelve 5 mm long strips.

Fig. 4. (Color online) (a) IPF of all Si strips with its length longer than 200 $\mu$m in ND, SD, and TD. Crystals having misorientation angle $\phi$ from ND{001} of $0^\circ$–$2^\circ$, $2^\circ$–$5^\circ$, $5^\circ$–$10^\circ$, and $>10^\circ$ were respectively shown by red, green, blue, and black colors. (b) Area fraction of these crystals.
[001] crystal with at least $\theta < 8^\circ$ is unstable CO. An ND [001] strip which was in quasi stable CO at first and then entered unstable CO was shown in Fig. 5 to demonstrate what will happen to the CO when crystal progressed. Figure 5(a) shows EBSD CO map in ND; (b) shows PFs of CO path when crystal progressed from point A1 to point A4 in (a); (c) shows point-to-origin misorientation angle from A1 to A4; (d) shows magnified {001} PF of CO path when crystal progressed from A2 to A3; (e) shows diamond-structure rhombicuboctahedron of crystals estimated from PFs at points A1, A2, and A3. Faces with red, blue, and green colors respectively indicate {001}, {111}, and {011}. This strip was found at a condition of $v = 10 \text{ mm s}^{-1}$, and a similar strip was also found in our published paper with $v = 45 \text{ mm s}^{-1}$. At point A1, $\theta$ was 21° [blue colored poles in Fig. 5(b)]. Among A1 to A3, CO was rotating counterclockwise around ND at a very slow rotation rate $R_r$ of 0.0068° $\mu\text{m}^{-1}$, and once $\theta$ come into 15° at near A3, CO start to rotate around the inplane $\langle 110 \rangle$ at a very fast $R_r$ of 0.92° $\mu\text{m}^{-1}$ until a $\langle 111 \rangle$ come to ND at A4 [Figs. 5(c) and 5(e)]. These results suggest ND[001] crystal with $40^\circ > \theta \geq 15^\circ$ is quasi stable CO and that with $\theta < 15^\circ$ is an unstable CO. Here we can define $15^\circ$ as a threshold angle, $\theta_{th}$, at which CO became unstable. In Fig. 4 with $v = 0.4 \text{ mm s}^{-1}$ $\theta_{th}$ was 8°, and in our previous result with $v = 45 \text{ mm s}^{-1}$ $\theta_{th}$ was 10°. $R_r$ of ND[001] strips with $\theta > 15^\circ$ in Fig. 3 were investigated in relation to $\theta$, and was shown in Fig. 6. In this figure $R_r$ was investigated in relation to $\theta$, and was shown in Fig. 6.
have only one axis rotation around ND and the direction was toward decreasing $\theta$, so when $\theta$ was positive, CO rotate in counterclockwise direction else in clockwise direction. When $\theta \geq 40^\circ$, which means CO was SD(100) within $\pm 5^\circ$, $R_r$ was 0, means CO in this condition was stable. $R_r$ are enlarging with decreasing $\theta$, and became maximum $0.0068 \mu\text{m}^{-1}$ at $\theta = 15^\circ$, then entered unstable CO. Besides, a continuous CO spiral motion around ND within radius of $\sim 1^\circ$ was found in strips with $\theta > \theta_{th}$, as shown in Fig. 5(d). The direction of spiral motion in Fig. 5(d) was clockwise, as was indicated by dotted arrow. The direction of spiral motion was always opposite to $R_r$. When CO entered $\theta_{th}$, radius of spiral motion increased, and then get into orbit in CO rotation around the inplane (110) to get away from ND(001) [point A3 to A4 in Fig. 5(e)]. The mechanism of the CO spiral motion was not cleared yet.

3.3. Twinning in ND(001) crystal

Figure 7 shows EBSD CO map and scanning electron microscopy (SEM) image ($70^\circ$ tilted) of a portion where ND(001) crystal ended. The ND(001) crystal was always ended at a $\Sigma$3-CSL boundary. $\Sigma$3-CSL boundaries in $\mu$CLBS were growth twin and was generally originated at the melt/solid interface on a {111} facet having small inclined angle with Si/SiO$_2$ interface, but in ND(001) crystal all {111} planes have an large inclined angle (54.7°) with Si/SiO$_2$ interface, so twinning ought to be hard to take place. As can be seen in SEM image of a point where the twinning took place, there was always a hole or a particle. The hole might be formed by gas ejection or by a particle, which in turn triggered twinning, so suppression of the phenomenon will be a key for increasing length fraction of ND(001) crystal. From Fig. 7, twinning of ND(001) crystal generated a ND
{221} crystal, and twinning of ND{221} generated ND{447} crystal. Pole figures (PF) of these crystals are shown in Fig. 8(a), in which ND{001} crystal was by green poles, ND{221} by blue poles, and ND{447} crystal by red poles. Based on the PFs, diamond-structure rhombicuboctahedrons were shown in Fig. 8(b). In Fig. 8(b), only the positive hemisphere was displayed, so a pole in negative SD in fact corresponds to a point-symmetric pole in the negative hemisphere at positive SD that face to the melt/solid interface.

From the above discussion, stable CO is only ND{001} with \( \theta \geq 40^\circ \) (SD(100) ± 5°), and quasi-stable CO is ND{001} with \( 40^\circ > \theta > 15^\circ \), and the quasi-stable CO will decrease its \( \theta \) very slowly and finally became unstable CO when \( \theta < 15^\circ \).

3.4. **CO rotation toward stable ND{001} strip**

From the above discussion, stable CO is only ND{001} with \( \theta \geq 40^\circ \) (SD(100) ± 5°), and quasi-stable CO is ND{001} with \( 40^\circ > \theta > 15^\circ \), and the quasi-stable CO will decrease its \( \theta \) very slowly and finally became unstable CO when \( \theta < 15^\circ \).
Besides, twice twinning also generate unstable CO of ND{447}. These unstable CO tends to vary its CO toward stable CO. We found there are two CO-rotation paths depending on the value of $\theta$. Diamond-structure rhombicuboctahedron of ND{447} crystal in Fig. 9(a) was used to explain this condition. When $\theta$ is larger than 15°, CO-rotation path follows path I, else path II.

Figure 10 shows a typical example of a Si crystal with $\theta = 34^\circ$ following path I to ND{001}. This portion can be found at a point indicated “A” in Fig. 3. (a) EBSD CO map in ND; (b) PFs of CO path when crystal progress from points A3 (blue color) to A4 (red color). This portion can be found at a point indicated “A” in Fig. 3.

Figure 11 shows another example of a Si crystal following path I with $\theta = 63^\circ$. (a) EBSD CO map in ND; (b) PF of CO path when crystal proceeded from points B1 (blue color) to B2(red color), and then further from B2(black color) to B3(magenta). This portion can be found at a point indicated “B” in Fig. 3.

Path I. As shown in Figs. 9(b) and 9(c), the CO at A3 rotated around the inplane $\langle 110 \rangle$ toward a direction for a $\langle 001 \rangle$ nearest to ND to come to ND, then the strip became ND{001} crystal at A4. The ND{001} crystal was with $\theta = 34^\circ$ [Fig. 10(b), red colored PFs], so the CO met quasi-stable CO condition. As discussed in Sect. 3.2, CO will rotate around ND in a direction decreasing $\theta$ in a very slow rate, and CO will enter unstable CO after $\theta$ became smaller than 15° if twinning does not take place in midstream. This CO rotation process was shown in Figs. 9(c) and 9(d).

Figure 11 shows another example of a Si crystal following path I with $\theta = 63^\circ$. (a) EBSD CO map in ND; (b) PF of CO path when crystal proceeded from points B1 (blue color) to B2(red color), and then further from B2(black color) to B3(magenta). This portion can be found at a point indicated “B” in Fig. 3. In this example CO maintained near ND{447} (purple colored strip in the EBSD map)
map) for about 0.8 mm in length. CO rotated around ND toward a direction decreasing $\theta$ at $R_c = 0.014$ $\mu$m$^{-1}$, and at the same time CO fluctuated within $\pm 5^\circ$ with crystal progress from B1 to B2. When $\theta$ decreased to $< 52^\circ$ at B2, CO immediately converged on ND{001} at B3. This result suggest inplane $\langle 110 \rangle$ with $\theta > 52^\circ$ does not work as axis for CO rotation.

Figure 12 shows another example of a Si crystal following path I with $\theta = 20^\circ$, in which (a) shows EBSD map in ND; (b) shows PFs of CO path when crystal progressed from point C3 to C4 in (a). This portion can be found at a point indicated “C” in Fig. 3. Points C1, C2, and C3 belong to a single grain, but contained...
dense strains and twins. Especially among C2 to C3, the CO was changing continuously but with a very large CO rotation rate of 6° μm⁻¹. CO at C3 was ND{318} with θ = 20°, so CO rotated around inplane 〈110〉 for a 〈001〉 nearest to ND to come to ND. At near ND{001}, CO spiral rotation in a direction opposite to decreasing θ was observed again, and CO finally converged on ND{001}. Such a CO spiral rotation just before converging on ND{001} was generally observed in crystals with θ₀ < θ < 22°.

Figure 13 shows example of a Si crystal following path II with θ = ~0°. (a) EBSD map in ND; (b) PFs of CO path when crystal progressed from point E1 (blue color) through point E2 (green color) to point E3 (red color) in (a); (c) diamond-structure rhombicuboctahedron of crystals at points E1, E2, and E3. This strip was with v = 2 mm s⁻¹, but a similar strip with worse symmetry can be found at a point indicated “E” in Fig. 3.

Fig. 14. (Color online) Another example following path II with θ = ~0°. (a) EBSD map in ND; (b) PFs of CO path when crystal progressed from point E1 through point E2 to point E3 in (a); (c) diamond-structure rhombicuboctahedron of crystals at points E1, E2, and E3. This strip was with v = 2 mm s⁻¹, but a similar strip with worse symmetry can be found at a point indicated “E” in Fig. 3.

Fig. 15. (Color online) (a) EBSD map and (b) diamond-structure rhombicuboctahedrons showing mechanism of CO iteration between ND{447} and ND {221} when θ < 15°. Although this strip was obtained at v = 10 mm s⁻¹, similar tendency can be found at a point indicated “F” in Fig. 3. This process will be repeated, until CO deviated from the above CO due to strains, twinning from a {111} other than the one near parallel to film plane, or twinning misfired.
D2 (blue color) to D3 (red color) in (a); (c) shows diamond-structure rhombicuboctahedron of crystals at points D2 and D3. This portion can be found at a point indicated “D” in Fig. 3. Again, a ND{447} crystal at point D1 was generated after twice twinning in initial ND{001} crystal, then a near ND{001} crystal formed at D2 after a strip with dense twins and strains following D1. The near ND{001} crystal at D2 have an inplane E10 with $\theta = 10^\circ$, so this condition coincide with condition for Path II in Fig. 9. CO rotated around the inplane E10 toward a direction for a $\{110\}$ nearest to ND to come to ND. In this example, unfortunately, twinning took place when $\{111\}$ came to near ND at point D3, so further tracking of the CO rotation disabled in this crystal. Alternatively we found another example with CO started from ND{111} SD{100}, as were shown in Fig. 14, in which (a) shows EBSD map in ND; (b) shows PFs of CO path when crystal progressed from point E1 (blue color) through point E2 (green color) to point E3 (red color) in (a); (c) shows diamond-structure rhombicuboctahedron of crystals at points E1, E2, and E3. This strip was with $v = 2 \text{ mm s}^{-1}$, but a similar strip with a worse symmetry can be found at a point indicated “E” in Fig. 3. When crystal progressed from E1 to E2, the CO rotate around the inplane $\{110\}$ toward a direction for a $\{110\}$ nearest to ND to come to ND [Fig. 14(c), E1 – E2]. After the $\{110\}$ had come to ND at point E2, CO started to rotate around the inplane transverse 001 toward a direction for a $\{00\}$ nearest to ND to come to ND. Then stable ND{001}SD{100} crystal generated. From the discussion so far, tendency of CO rotation depending on $\theta$ was summarized in Fig. 9. It showed that all CO will finally converged on ND{001}SD{100}.

### 3.5. CO iteration in ND{447} derivation strip

We have shown that the ND{447} derivation crystal contained dense twins and strains. This is because that ND{447} derivation crystal have $\{111\}$ near parallel to Si/SiO$_2$ interface and have six near inplane $\{111\}$, so twins and strains generally induced during crystal growth. Especially when the ND{447} derivation crystal was with the condition of $\theta < \theta_0$, CO iteration between ND{447} and ND{221} took place, and the defective ND{447} derivation strip become prolonged, as was shown EBSD ND CO map in Fig. 15(a). Although this strip was obtained at $v = 10 \text{ mm s}^{-1}$, similar tendency can be found at a point indicated “F” in Fig. 3. Occurrence probability of such a CO iteration increased when $v$ increased. The mechanism of CO iteration was clarified in Fig. 15(b). Because $\theta < \theta_0$, CO of ND{447} crystal rotate toward ND{221}. When CO became ND {221}, since it is a short lifetime CO, twinning will immediately took place on a $\{111\}$ facet having small inclined angle (15.8°) with Si/SiO$_2$ interface, and the ND{447} crystal generated. This process will be repeated until, CO deviated from the above CO due to strains, twinning from a $\{111\}$ other than the one near parallel to film plane, or twinning misfired.

### 4. Conclusions

Stable CO for lateral crystal growth of Si thin film sandwiched by SiO$_2$ was evidenced to be ND{001} with SD{100}$\pm 5^\circ$. ND {001} crystal is quasi-stable when angle $\theta$ is $15^\circ \leq \theta < 40^\circ$ and is unstable when $\theta < 15^\circ$. CO other than the stable CO will rotate spontaneously toward the stable CO, i.e. ND{001} with SD{100}$\pm 5^\circ$. It was shown that in quasi-stable ND{001} crystals, CO rotate around ND toward a direction decreasing $\theta$ at a very slow rate among 0.001 to 0.0007 $\mu\text{m}^{-1}$; while in unstable ND{001} crystals, CO rotate around inplane (110) nearest to SD at a fast rate of 0.2–1$^\circ$ $\mu\text{m}^{-1}$. All crystals other than stable CO will finally converged on ND{001} with SD {100}$\pm 5^\circ$, but generally crystal was ended by twinning before the CO became stable. The twinning was triggered by gas ejection or particles, so suppressing of these phenomenon would be the key for increasing ND{001}SD{100} crystal occupations. These results have been verified for crystal growth velocity among 0.04–45 $\mu\text{m s}^{-1}$.

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