In-situ Observation of Melt Growth Process of Bi (111) Thin Films by Transmission Electron Microscopy

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In-situ electron microscopy of the melt growth of Bi (111) films of purity 99.9 and 99.9999% was performed. The film was partially melted and regrown in the presence and in the absence of a small temperature oscillation at a cooling rate of $8.3 \times 10^{-3}$ to $2.7 \times 10^{-1}$ K/s, and the dynamic behavior of growing interface and crystal defect was observed.

The growing interface was smoothly curved more frequently than faceted in any growth condition. When a small temperature oscillation was imposed on cooling, the curved interface showed a back-and-forth advancement, while the faceted interface moved stepwise or steadily without back-melting. When the oscillation was absent, both the interfaces moved nearly steadily. The average velocities of the curved interface and the faceted one increased linearly with an increase of the cooling rate, though the former velocity was slightly larger than the latter one.

During the melt growth and subsequent cooling, short dislocations, dislocation loops, circular voids, lineage defects and triangular defects were introduced into the films. The density of short dislocations, the length of lineage defects and the sum of densities of dislocation loops and circular voids were large in the impure film, respectively.

From these results together with previous results on the (111) films of purity 99.9% and the (100) films of purity 99.9 and 99.9999%, some information concerning the effect of film orientation on the micromorphology of growing interface and the formation of crystal defect during the melt growth of Bi films was presented.

(Received October 12, 1992)

Keywords: crystal growth, solid-liquid interface, facet, crystal defect, dislocation, vacancy cluster, electron microscopy, bismuth, thin film

I. Introduction

Glicksman and Vold first observed in-situ the melting and solidification processes of Bi (purity 99.9999% (5N)) polycrystalline films by means of a transmission electron microscopy (TEM). They found that a facet which might be composed of sub-microscopic (100) faces appeared in a growing interface in a (111) oriented grain and that a dislocation array which formed a low angle tilt boundary was created near a solid-liquid interface during the melt growth.

Afterward, two of the present authors (Watanabe and Sugawara) and coworkers performed in-situ TEM observation of the melt growth process of Bi (111) thin films of purity 3N and (100) thin films of purity 3N and 6N, which were prepared by thinning bulk single crystals. The films were partially melted and regrown by cooling at an average rate of $8.3 \times 10^{-3}$ to $2.7 \times 10^{-1}$ K/s in the former films, and at an average rate of $8.3 \times 10^{-3}$ K/s in the latter ones.

The solid-liquid interface in the (111) film was always concave toward the melt and its curvature became large when the cooling rate was decreased, while the growing interface in the (100) film was sometimes faceted. It was inferred that the facet would correspond to the (100), (111) and (111) planes. The curved interface displayed a back-and-forth advancement at the slow cooling rate, a stepwise advancement at the intermediate cooling rate and a steady advancement at the high cooling rate. It was found that the oscillatory interface motion was resulted from a small oscillation of external temperatures. On the other hand, the faceted interface showed a stepwise or steady advancement without back-melting at any cooling rate.

Various crystal defects were introduced during the melt growth and/or subsequent cooling process. In the (111) films, long and short dislocations, dislocation loops, lineage defects probably composed of a dislocation array, circular voids and triangular defects were formed. In the (100) films, the crystal defects other than the long dislocations and the triangular defects were formed, though the defect densities were different from those in the (111) films. It is to be added that the densities of short dislocations and lineage defects were high in impure materials.

As an extension of previous study, we have now examined the features of solid-liquid interface and crystal defect formation in Bi (111) films of purity 3N and 6N which were grown from the melt in the presence or absence of a small temperature oscillation, respectively. In this paper, the results of observations are given including the previous results and are discussed in relation to the purity, the orientation and the cooling condition of Bi films.

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‡ In this paper Miller indices of a crystal plane of Bi are denoted by a face-centered rhombohedral notation.
II. Experimental

Foil specimens were prepared similarly as in our previous experiments\(^{(3)-(5)}\). Bi single crystals of purity 3N and 6N were grown by the Czochralski method and the orientation of the (111) plane was determined by the light figure method\(^{(6)}\). After slices 1.5 mm thick were cut out along the (111) plane by the use of a spark-erosion machine, they were thinned to 0.2 mm thick by chemical polishing, cut into polygonal discs about 3 mm in width and then thinned by a jet polishing and a final electropolishing. For the jet polishing, the electrolyte used was concentrated hydrochloric acid + methanol + ethylene glycol mono-butyl ether (10:39:1 in volume ratio) and the applied voltage was 7–8 V, and for the final polishing concentrated hydrochloric acid + methanol + ethylene glycol mono-butyl ether (5:19:1 in volume ratio, 269–278 K) and 7–13 V. Finally, carbon was deposited in a vacuum on both faces of the polished films in order to prevent vaporization and de-wetting of the molten Bi during melting in an electron microscope.

The (111) thin film thus prepared was fixed into a specimen holder and then mounted on a heating stage in a transmission electron microscope (JEOL, JEM-200A). The holder was provided with a PR thermocouple and a tungsten-wire heater. In order to control the specimen temperature, two types of controller were employed, as shown schematically in Fig. 1. One type indicated by dotted lines was the same as that used in our previous experiments\(^{(3)-(5)}\). Direct current was supplied to the heater from the stabilized power unit through the servo-resistor, which was regulated by a PID controller of on-and-off type (Chino Works, Ltd., NA-862) so that the heating temperature may follow a given pattern by a programmer (Chino Works, Ltd., UH-4N). This system will be called the old-type controller in the following. A typical example of temperature-time record at the programmed cooling rate of \(8.3 \times 10^{-3} \text{ K/s}\) is shown in Fig. 2(a). It can be seen that the cooling curve contains a small temperature oscillation, of which amplitude ranges from 0.04 to 0.06 K and periodic time varies from 2.1 to 2.3 s. In Bi (111) films grown under the same cooling condition\(^{(3)-(5)}\), the amplitude of oscillation was amounted to 0.01 – 0.03 K, and in the (100) films\(^{(3)}\) 0.01 – 0.06 K, though the periodic time in both films was the same as that in the (111) films. It must be noted that the small temperature oscillation gave rise to a back-and-forth advancement of curved solid-liquid interface during the melt-growth.

The other controller is shown with solid lines in Fig. 1. Direct current was supplied to the heater through an SCR and a rectifier, and the temperature was controlled according to a given pattern by a combination of a PID controller of current input type (Chino Works, Ltd., NA-863) and a digital programmer (Chino Works, Ltd., DP-30). This system will be called the new-type controller here. As shown in Fig. 2(b), the temperature-time curve at the programmed cooling rate of \(8.3 \times 10^{-3} \text{ K/s}\) is free from a small temperature oscillation, but it is de-

\(^{†}\) In the previous report\(^{(3)-(5)}\), we gave the incorrect values of 0.1 – 0.3 K through careless mistakes.
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III. Experimental Results and Discussion

1. Morphology and velocity of growing interface

Two typical TEM images of solid-liquid interfaces appeared during the melt growth of Bi (111) films are shown in Fig. 3, in which dark areas denoted by L correspond to the liquid and light areas denoted by S represent the solid. The curved interface (Fig. 3(a)) was concave toward the melt, corresponding to the isotherm of the growing system. On the other hand, the faceted interface (Fig. 3(b)) was composed of two facets which were joined by a small curved surface. From the orientation analysis of the selected area diffraction pattern, it was confirmed that the facet traces lay in [101] and [110] directions, respectively.

In Table 1, the results of observations on the morphology of growing interface in the (111) films are given together with the previous results on (111) \(^{[19]} \) and (100) \(^{[20]} \) films. The curved interface occurred frequently compared with the faceted interface in the (111) films similarly as in (100) and (111) films. The occurrence of curved interface became remarkable in the order of (111), (100) and (111) films, all interfaces being the curved ones in the last film (see Table 1; controller: old-type, cooling rate: \(3 \times 10^{-2} \text{K/s} \)). A comparison of different cooling rates of 6N (111) films showed that the curved interface developed more frequently at higher cooling rates. In the (111) films, the faceted interface occurred more often in the high purity material than in the low purity one (see Table 1, controller: old-type and new-type, cooling rate: \(1.7 \times 10^{-2}, 6.7 \times 10^{-2} \), or \(2.7 \times 10^{-1} \text{K/s} \), and bright-field images of a growing interface and a crystal defect were observed and photographed. If necessary, the interfacial process was recorded on a video-tape in order to determine the interface velocity using a TV imaging system. A selected area diffraction pattern taken from a fixed field showed that the thin film crystallized in the same orientation as that before melting. The observations were carried out with twenty-one to thirty-seven specimens in each growing condition.

Fig. 2 Temperature-time record at the programmed cooling rate of \(8.3 \times 10^{-3} \text{K/s} \). (a) Oscillatory cooling curve displayed by the old-type temperature controller and (b) smooth curve by the new-type one.

Fig. 3 TEM images of (a) a curved interface and (b) a faceted interface observed during the melt growth of Bi (111) films (purity 3N).
Table 1: Results of observation on the solid-liquid interface morphology of the melt-grown Bi films.

<table>
<thead>
<tr>
<th>Orientation</th>
<th>Purity</th>
<th>Controller</th>
<th>Rate (K/s)</th>
<th>Number of tested specimens</th>
<th>Number of occurred specimens</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Curved interface</td>
<td>Faceted interface</td>
</tr>
<tr>
<td>(111)</td>
<td>3N</td>
<td>old-type</td>
<td>8.3 x 10^{-3}</td>
<td>37</td>
<td>23</td>
</tr>
<tr>
<td></td>
<td>6N</td>
<td></td>
<td></td>
<td>30</td>
<td>17</td>
</tr>
<tr>
<td></td>
<td>3N</td>
<td>new-type</td>
<td>8.3 x 10^{-3}</td>
<td>21</td>
<td>17</td>
</tr>
<tr>
<td></td>
<td>6N</td>
<td></td>
<td>1.7 x 10^{-2}</td>
<td>30</td>
<td>16</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>6.7 x 10^{-2}</td>
<td>22</td>
<td>13</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2.7 x 10^{-1}</td>
<td>31</td>
<td>28</td>
</tr>
<tr>
<td>(111)</td>
<td>3N</td>
<td>old-type</td>
<td>8.3 x 10^{-3}</td>
<td>30</td>
<td>30</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1.7 x 10^{-3}</td>
<td>30</td>
<td>30</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>6.7 x 10^{-2}</td>
<td>30</td>
<td>30</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2.7 x 10^{-1}</td>
<td>30</td>
<td>30</td>
</tr>
<tr>
<td>(100)</td>
<td>3N</td>
<td>old-type</td>
<td>8.3 x 10^{-3}</td>
<td>22</td>
<td>18</td>
</tr>
<tr>
<td></td>
<td>6N</td>
<td></td>
<td></td>
<td>22</td>
<td>21</td>
</tr>
</tbody>
</table>

8.3 x 10^{-3} K/s). In the (100) films, however, the reverse relation seemed to be maintained. Moreover, it was noticed that the faceting was more remarkable in the low purity films grown in the presence of a small temperature oscillation than in those grown in the absence of the oscillation, though the faceted interface developed equally in the high purity ones grown under both cooling conditions (see Table 1; (111), controller: old-type and new-type, cooling rate: 8.3 x 10^{-3} K/s).

The morphology of growing interface can be predicted from Jackson’s parameter \( \alpha \) which specifies the atomic roughness of solid-liquid interface. If \( \alpha > 2 \), the solid-liquid interface would be atomically smooth and hence the growing interface would be faceted. If \( \alpha < 2 \), the interface would always be atomically rough and it would be curved following the isotherm of the solid-liquid system. Non-metallic materials may assume principally the former interface and metallic materials may possess the latter one. In Bi of which general properties fall between non-metallic and metallic classifications, the value of \( \alpha \) is 2.39 for the (111) face and 1.59 for \{111\}, \{100\}, \{112\}, \{121\} and \{122\} faces, respectively\( ^8 \). It has been pointed out that the Jackson’s criterion is not strictly valid for materials with \( 3 > \alpha > 2 \)\( ^8 \). Indeed, Bi showed a non-faceted growth typical of metals for which \( \alpha < 2 \) at high supercooling (cooling rate) and it displayed a faceted growth typical of non-metallic behavior for which \( \alpha > 3 \) at low supercooling (cooling rate)\( ^9 \). The dependency of interface morphology on the cooling rate can be explained from the above semi-metallic behavior of Bi.

Wagner and Brown\( ^9 \) observed that the \{100\} faces of low \( \alpha \) (1.59) are developed preferentially rather than the (111) face of the highest \( \alpha \) (2.39) in a bulk Bi crystal grown at moderate supercooling. Glicksman and Vold\( ^{10,12} \) reported that a facet appeared in a (111) oriented grain of polycrystal Bi film might be composed of a sub-microscopic (100) faces. These facts indicate the preference of (100) faces for faceting in melt-grown Bi. It is most probable that a facet in a growing interface would correspond to a low-index crystal face which lies nearly perpendicular to the growing direction. Taking account of the orientation of facet trace on the film plane, it was inferred that the \{100\} and \{111\} planes would compose a facet in the (111) films. In the (100) films, one more (111) plane would develop as a facet\( ^9 \).

When the (111) films were grown in the presence of the small temperature oscillation (controller: old-type, cooling rate: 8.3 x 10^{-3} K/s), the curved interface advanced back-and-forth with the periodicity of 2.2 s in response to the oscillation. Under the same cooling condition, the faceted interface advanced stepwise or steadily without back-melting. On the other hand, when the (111) film was grown without the temperature oscillation (controller: new-type), both of the curved interface and the faceted one showed the steady advancement at any cooling rate.

The dependence of average interface velocity on cooling rates is shown in Fig. 4. The data were obtained from video-images of 6N purity (111) films grown using the new-type controller. When the cooling rate increased from 8.3 x 10^{-3} to 2.7 x 10^{-1} K/s, the velocity of curved interface changed linearly from 0.05 to 0.53 \( \mu \)m/s and that of faceted interface from 0.02 to 0.38 \( \mu \)m/s, respectively. According to the kinetic theory of melt growth\( ^{10} \), the velocity of the curved interface would be controlled by the continuous growth mechanism, and that of faceted interface would be controlled by the lateral growth mechanism. The fundamental process for the continuous growth is the random attachment of atoms to natural steps on the atomically rough (non-singular) surface. On the other hand, two processes are generally fundamental for the lateral growth: the formation of a two-dimensional crystal nucleus on the atomically smooth (singular) surface and its lateral spreading over the crystal surface due to the attachment of atoms. It can be confidently expected that the rate of lateral growth is much smaller than that of continuous growth at a given undercooling because of the nucleation difficulty, the
It has been expected that when the (111) films are grown in the presence of a small temperature oscillation, extra dislocations would be introduced by a layer type of solute segregation due to the periodic change of growth rate\(^{(14)}\). However, no effect of temperature oscillation on the dislocation formation was observed (see Table 2; controller: old-type and new-type, cooling rate: \(8.3 \times 10^{-3} \text{ K/s}\)). The overall density of short dislocations varied very little with the cooling rate in the (111) films similarly as in the (111) films. It was found that the overall density of short dislocations became large in the order of (111), (100) and (111) films (see Table 2; controller: old-type, cooling rate: \(8.3 \times 10^{-3} \text{ K/s}\)). This would be ascribed to the anisotropy of the solute distribution coefficient.

2) Dislocation loop and circular void

A typical TEM image of dislocation loop observed in the (111) films is shown in Fig. 6. It is a common knowledge that the dislocation loop can be produced by a condensation of vacancies into disk void and its subsequent collapse\(^{(15)}\). Previously, we observed in the (111) films\(^{(7)}\) that some circular voids changed into dislocation loops during cooling after the melt growth. Figure 7 shows a TEM image of circular voids appeared in the melt-grown (111) films. The sum of densities of dislocation loops and circular voids was higher in 3N (111) films than in 6N (111) films grown under the same cooling condition (see Table 2; controller: old-type and new-type, cooling rate: \(8.3 \times 10^{-3} \text{ K/s}\)), while the purity dependency was not clear in the (100) films. We could not obtain any knowledge about the effects of cooling rate on the formation of dislocation loop and circular void in the (111) films, since these defects were not observed at any cooling rate in 6N specimens tested. However, it was found in 3N (111) films that the dislocation loops and circular voids tended to increase with an increase of cooling rate (see Table 2; controller: old-type). This may be attributed to an increase of the amount of quenched-in vacancies at high cooling rate or growth rate\(^{(13,17)}\). The sum of densities of these two defects was higher in the (111) films grown in the presence of the small temperature oscillation than in those grown without temperature oscillation (see Table 2; controller: old-type and new-type, cooling rate: \(8.3 \times 10^{-3} \text{ K/s}\)).

The formation of dislocation loops and circular voids depended also on the film orientation. Total amount of these two defects was found to be large in the order of (100), (111) and (111) (see Table 2; controller: old-type, cooling rate: \(8.3 \times 10^{-3} \text{ K/s}\)).

3) Lineage defect

Figure 8 shows a TEM image of lineage defect formed in the melt-grown (111) films. The lineage defect was introduced in a manner similar to those in the (111) and (100) films\(^{(7,15)}\). This defect was usually nucleated at a small depression of growing interface which occurred suddenly by a partial delay of the interface advance and then elongated into the grown part. Exceptionally, a short lineage defect was formed in a last solidified part of
Table 2  Average density and size of crystal defects formed in the melt-grown Bi films.

<table>
<thead>
<tr>
<th>Orientation</th>
<th>Purity</th>
<th>Controller</th>
<th>Rate ( /K, s^{-1})</th>
<th>Short dislocation</th>
<th>Dislocation loop</th>
<th>Circular void</th>
<th>Lineage defect</th>
<th>Triangular defect</th>
<th>Remark</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Oblique to films</td>
<td>Density /(10^9) m(^{-2})</td>
<td>Size /(\mu m)</td>
<td>Normal to films</td>
<td>Density /(10^9) m(^{-2})</td>
<td>Size /(\mu m)</td>
</tr>
<tr>
<td>(111)</td>
<td>3N</td>
<td>old-type</td>
<td>(8.3 \times 10^{-3})</td>
<td>2.2 0.17</td>
<td>5.0</td>
<td>25 0.04</td>
<td>4.6 0.05</td>
<td>6.4</td>
<td>7.1 0.05</td>
</tr>
<tr>
<td></td>
<td>6N</td>
<td></td>
<td>(8.3 \times 10^{-3})</td>
<td>1.5 0.18</td>
<td>1.1</td>
<td>4.6 0.02</td>
<td>4.6 0.04</td>
<td>5.4</td>
<td>21 0.05</td>
</tr>
<tr>
<td>(11̅1)</td>
<td>3N</td>
<td>new-type</td>
<td>(8.3 \times 10^{-3})</td>
<td>3.6 0.10</td>
<td>3.6</td>
<td>0.60 0.17</td>
<td>4.2 0.12</td>
<td>14</td>
<td>not observed</td>
</tr>
<tr>
<td></td>
<td>6N</td>
<td></td>
<td>(8.3 \times 10^{-3})</td>
<td>1.9 0.10</td>
<td>2.2</td>
<td>not observed</td>
<td>not observed</td>
<td>3.6</td>
<td>not observed</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>(1.7 \times 10^{-2})</td>
<td>0.3 0.51</td>
<td>0.52</td>
<td>not observed</td>
<td>not observed</td>
<td>1.5</td>
<td>not observed</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>(6.7 \times 10^{-2})</td>
<td>not observed</td>
<td>6.5</td>
<td>not observed</td>
<td>not observed</td>
<td>2.7 (10^{-1})</td>
<td>not observed</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td>(2.7 \times 10^{-1})</td>
<td>not observed</td>
<td>2.3</td>
<td>not observed</td>
<td>not observed</td>
<td>previous study</td>
<td>not observed</td>
</tr>
<tr>
<td>(100)</td>
<td>3N</td>
<td>old-type</td>
<td>(8.3 \times 10^{-3})</td>
<td>25 0.12</td>
<td>—</td>
<td>13 0.06</td>
<td>6.6 0.05</td>
<td>1.3</td>
<td>not observed</td>
</tr>
<tr>
<td></td>
<td>6N</td>
<td></td>
<td>(1.7 \times 10^{-2})</td>
<td>19 0.08</td>
<td>—</td>
<td>14 0.05</td>
<td>32 0.04</td>
<td>4.0</td>
<td>9.3 0.12</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>(6.7 \times 10^{-2})</td>
<td>2.8 0.07</td>
<td>—</td>
<td>24 0.03</td>
<td>33 0.02</td>
<td>5.0</td>
<td>2.8 0.02</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>(2.7 \times 10^{-1})</td>
<td>8.0 0.09</td>
<td>—</td>
<td>10 0.04</td>
<td>29 0.03</td>
<td>4.8</td>
<td>0.2 0.02</td>
</tr>
</tbody>
</table>

*In 3N (11̅1) films grown at the cooling rate of \(1.7 \times 10^{-2}\) K/s using the old-type controller, the long dislocation more than 2 \(\mu m\) in length was formed with an average density of \(6.1 \times 10^9\) m\(^{-2}\) in addition to the above-listed crystal defects.

Fig. 5  TEM images of short dislocations (indicated by arrows) formed during the melt growth of Bi (111) films (purity 3N). (a) Dislocations intersecting the film obliquely and (b) dislocations intersecting the film vertically. The sides of a triangle inserted correspond to \(\langle 110\rangle\) directions.
The lineage defect was longer in 3N films than in 6N films of both (111) and (100) orientations (see Table 2; controller: old-type, cooling rate: $8.3 \times 10^{-3} \text{ K/s}$). It can be concluded that the lineage defect may originate from the chemical inhomogeneity introduced during the melt growth\(^{(11)-(13)}\). The length of lineage defect in 6N (111) films seemed to decrease with an increase of cooling rate (see Table 2; controller: new-type), while the situation was reversed in 3N (111) films (see Table 2; controller: old-type). From a comparison of different orientations of 3N films, it can be seen that the formation of lineage became remarkable in the order of (111), (111)\(\approx\) (100) (see Table 2; controller: old-type, cooling rate: $8.3 \times 10^{-3} \text{ K/s}$). As to the effect of small temperature oscillation on the lineage formation, we could not obtain any definite conclusion. As discussed in the previous report\(^{(14)}\), the experimental results can not be explained fully from the impurity mechanisms\(^{(11)-(13)}\) so far proposed for the lineage formation.

### (4) Triangular defect

A TEM image of triangular defects observed in the (111) films is shown in Fig. 9. Each edge of the triangle lies in a \(\langle 1\overline{1}0\rangle\) direction. The triangular defects appeared just after the melt growth and grew in size during subsequent cooling. Similar defects were also formed in (111) films\(^{(14)}\) under some growth conditions, but not in the (100) films\(^{(15)}\). The effects of the cooling condition and purity of materials were not clarified from the experimental data.

In a vapor-deposited Bi film, Fedorenko \textit{et al.}\(^{(16)}\) observed triangular voids which contained a stacking fault surrounded by partial dislocations. They suggested that these defects were formed by the condensation of vacancies into a disk on the (111) plane and its subsequent collapse. However, it is doubtful that the triangular defects observed in the melt-grown Bi are vacancy clusters, since the maximum concentration of vacancies to be quenched-in\(^{(19)}\) is too low to produce vacancy clusters of dislocation loop and circular defect if the triangular defect is added as a vacancy void, as described...
in the previous report\textsuperscript{\text((34))}. The origin of the triangular defects is unknown at present.

IV. Summary

The dynamic process of melt growth of Bi (111) thin films was observed in-situ by an electron microscopy. The thin films of purity 3N and 6N were partially melted and regrown at an average cooling rate of $8.3 \times 10^{-3}$ to $2.7 \times 10^{-1}$ K/s in a transmission electron microscope.

The growing interface was concave toward the melt in more than half of the specimens in any specified condition and it was faceted in the remainder. The faceted interface was composed of two or more $\{100\}$ or $\{111\}$ facets which were connected by a curved surface with each other. The curved interface occurred more often when the material was impure and the cooling rate was high. The impure material exhibited a tendency to increase the probability of faceting when a small temperature oscillation was imposed during the melt growth.

During the melt growth in the presence of the small temperature oscillation, the curved interface advanced with a back-and-forth oscillation, while the faceted interface advanced stepwise or steadily without back-melting. On the other hand, both the interfaces moved nearly steadily when the temperature oscillation was absent.

The average interface velocity increased as the cooling rate increased. The velocity of faceted interface was slightly lower than that of curved one.

The crystal defects introduced in the grown films were the short dislocations inclined obliquely and vertically to the film plane, the dislocation loop, the circular void, the lineage defect which is composed of a dislocation array and the triangular defect.

The density of short dislocations and the length of lineage defects were large in the impure material, indicating that extra dislocations would be introduced due to segregation of impurity atoms. The dislocation loop and circular void which may be caused by a condensation of quenched-in vacancies were formed more frequently than the other crystal defects. The sum of densities of these defects was also high in impure material. The amounts of these four crystal defects varied with the cooling rate. The triangular defects produced just after the melt growth grew in size during subsequent cooling. The effect of growth conditions on the formation of this defect could not be clarified.

Acknowledgment

The authors wish to thank Dr. An-Pang Tsai and Mr. Yoshio Bizen for the useful help in the performance of experiment. This work was partially supported by a Grant-in-Aid for Scientific Research from the Ministry of Education, Science and Culture, Japan.

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