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Analysis of the Features of Meniscus Profile Curves during Growth of Base-Faceted Sapphire Ribbons

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Abstract—The dependence of the step parameters on the surface of base-faceted sapphire ribbons on the angle of deviation of crystallographic orientation has been measured. A model of step formation on the lateral ribbon surface is proposed. Hysteretic character of the dependence of the lateral ribbon surface inclination on the meniscus angle at the triple point is shown. The shapes of the lateral surfaces are calculated and the range of variation in the meniscus angle is determined.

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INTRODUCTION

Growth of single-crystal sapphire ribbons with 90° orientation (the ribbon plane coincides with crystallographic basal plane) by the Stepanov method makes it possible to obtain atomic-smooth ribbon surface due to the emergence of the basal face on the ribbon surface; i.e., base-faceted (BF) ribbons can be grown. It is difficult to implement this in practice because (i) the crystallographic and geometric ribbon planes should strictly coincide and (ii) the ribbon thickness must remain constant. The first condition can be satisfied by exact orientation of the seed crystal with respect to the growth direction and the shaper edges. The second condition is related to the parameters of melt liquid column at the crystallization front and stability of the capillary and thermal growth conditions. It was shown in [1] that, aligning the seed accurate to several arcmins (using laser beam reflection from the seed crystal faces), one can obtain specular areas on the ribbon surface several square centimeters in size. However, the ribbon surface contained growth steps. The vertical steps are formed on the edges due to the decrease in the ribbon thickness. The horizontal ones are related to both the residual misorientation and a small variation in the crystallization front height during growth.

If the basal crystallographic plane of a BF ribbon deviates from the growth direction by a small angle during growth, strictly periodic steps are observed on the lateral ribbon surface, whose size depends on the inclination of the basal plane with respect to vertical [2, 3]. Each step consists of two parts: a singular (1000) face and a nonsingular rounded surface. A more detailed analysis of this surface with an optical microscope [4] shows that it is also composed of micrometer-sized steps formed by the same face.

The surface areas between faceted steps were assumed to be isotropic in [2] and their shape was approximated by an exponential function. This approximation was shown to give good correspondence with the experimental results. However, the choice of this approximation was not justified. It was shown in [3] that the structure of the growth steps is different on the opposite sides of BF ribbon, and this discrepancy is due to the difference between the growth angles and meniscus shape on the opposite sides of BF ribbon with inclined orientation. Voronkov [5–7] has theoretically considered the problems of face formation during crystal growth from melt and explained the nature of faceting of germanium and silicon semiconductor crystals. The surface areas between the faces were assumed to be nonsingular.

The macroscopic effects of capillary shaping are considered in this study. Hence, we also assume this surface to be "isotropic", bearing in mind that, in contrast to classical rough surface, it consists of microscopic faceted steps. The mechanism of formation of such a fine structure on this surface is currently unclear and needs separate discussion.

On the whole, the nature of face formation during capillary shaping of grown sapphire crystals has been studied insufficiently. In this paper, we report the results of experimental investigation of the surface of BF sapphire ribbons at different angles of misorientation, analysis of the change in the angular relations between the meniscus inclination and the growing crystal lateral surface at the triple point, numerical simulation of the melt meniscus shape during BF sapphire ribbon growth, and calculation of the crystal lateral surface.

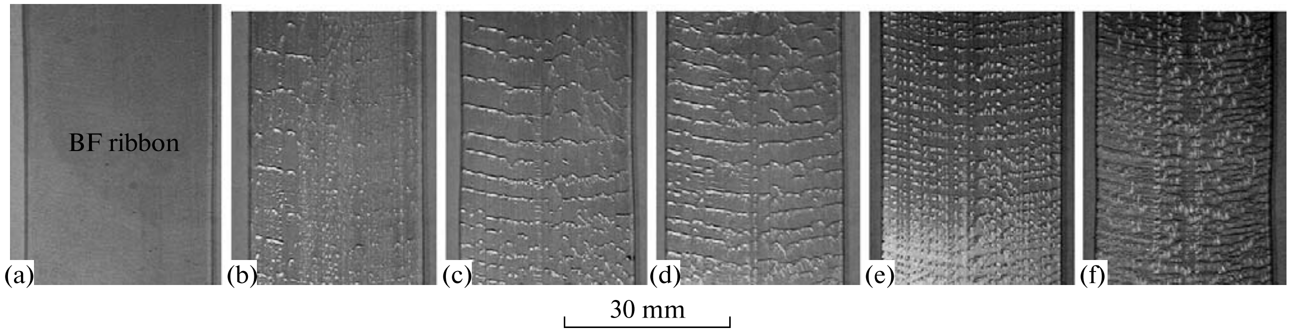

Fig. 1. Surface photographs of BF ribbons grown with a specified basal plane inclination with respect to the ribbon surface. The growth direction and normals to the basal plane and ribbon surface lie in the same plane. The angles of inclination are (a) 0°, (b) 0.35°, (c) 1.1°, (d) 1.7°, (e) 2.7°, and (f) 5°.

GROWTH OF BF RIBBONS WITH INCLINED ORIENTATION

To study the surface structure of BF ribbons, the latter were grown with a specified inclination of the normal basal face with respect to the ribbon surface. In this case, the growth direction and the normals to the basal face and ribbon surface lie in the same plane. The ribbons were grown from molybdenum shaper and crucible in a graphite resistive heat zone with a rate of 7 mm/min. The exact orientation was provided by applying seed crystals with basal (reference) areas of faces and laser-oriented seeds directly in the growth chamber. The orientation error did not exceed several arcmin. The constant ribbon thickness was provided by the shaper design (bent upper edges along the ribbon, according to the curvature of the thermal field isotherms in the growth zone) and high process stabilization with visual monitoring of the ribbon width during growth. Photographs of the ribbon surface for angles of inclination of 0°, 0.35°, 1.1°, 1.7°, 2.7°, and 5° are shown in Fig. 1. It can be seen that the ribbon surface with a face inclined toward the melt consists of periodically repeating specular (faceted) areas, separated by transition regions, reproducing the crystallization front shape. The surface of the opposite ribbon side also contains steps with a smaller period. Figure 2a schematically shows the longitudinal ribbon cross section, and Fig. 2b demonstrates the measured dependence of the step period on the angle of face inclination with respect to the growth direction. A decrease in the angle of inclination leads to a more rapid increase in the size of the faceted areas in comparison with the isotropic surface areas.

MODEL OF FORMATION OF STEP BF RIBBON SURFACE

Obviously, the surface formation mechanism periodically changes during base-faceted ribbon growth: the faceting stage passes to the stage of isotropic surface growth and vice versa. Any significant meniscus vibrations are not observed. Let us assume that the crystallization front height remains constant during growth and that melt inleakage on the face is absent. We assume also that the growth angle ε_0 is constant during isotropic surface growth and the angle of lateral surface inclination $\varphi_S = \varphi_L - \varepsilon_0$, where φ_L is the meniscus angle at the triple point. At the faceting stage the angle φ_S is constant and equal to the seed misorientation angle with respect to the growth direction (α). In this case, the angle φ_L may take any values and is determined from the condition of meniscus pinning at the shaper edge and the constancy of meniscus height during growth.

Let us consider the change in the inclination angles of the tangents to the crystal lateral surface (φ_S) and to the meniscus (φ_L) at the triple point. When a face begins to grow on the crystal lateral surface (Fig. 3a), the growth angle ceases to be constant, as a result of which the angle φ_S stepwise decreases with a change in sign to opposite (transition from point *1* to point *2* in the diagram of Fig. 3b). During the face growth, the angle φ_S does not change (segment 2→3). With a decrease in the crystal thickness (due to the face growth), the angle φ_L increases. When it reaches the critical value φ_L^{max}, a stepwise transition from point *3* to point *4* occurs, which corresponds to the transition from the faceting stage to isotropic stage (segment 4→1).

Thus, the dependence $\varphi_S(\varphi_L)$ contains a hysteresis (has different character at different growth stages), and there are some critical values of the angle φ_L (φ_L^{min} and φ_L^{max}); when reaching these values, the stage of isotropic growth changes with faceting and vice versa. The possibility of violating the condition of growth angle constancy at crystal faceting was indicated by Voronkov: he reported the dependences $\varphi_S(\varphi_L)$ in [4], which also contain discontinuities and areas of constant angle φ_S with the angle φ_L varying in some range.

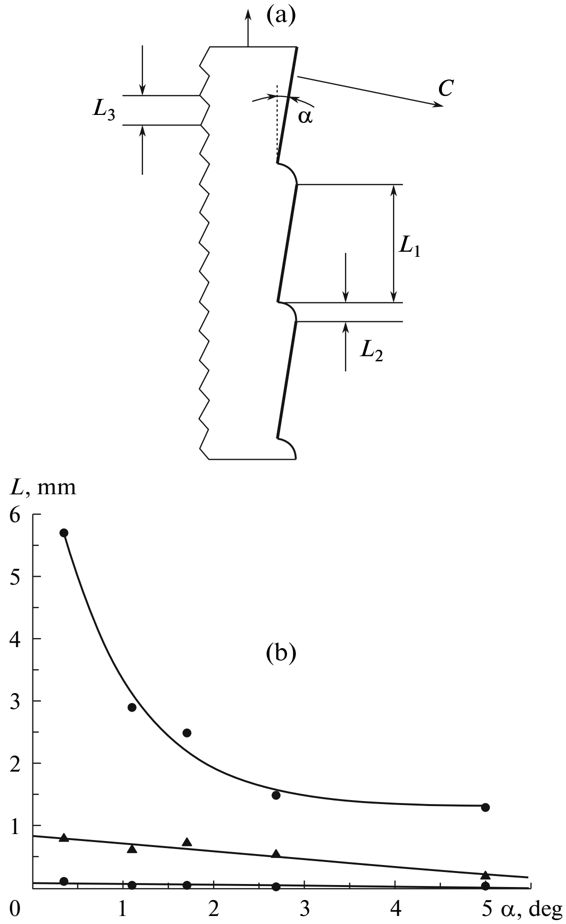

Fig. 2. (a) Schematic of the longitudinal cross section of BF ribbon at a small deviation of crystallographic orientation and (b) the dependence of the step period on the angle of inclination. The upper curve is the length of the faces inclined toward the melt (L_1), the intermediate curve is the length of the extension portions (L_2), and the lower curve is the step period at the ribbon rear side (L_3).

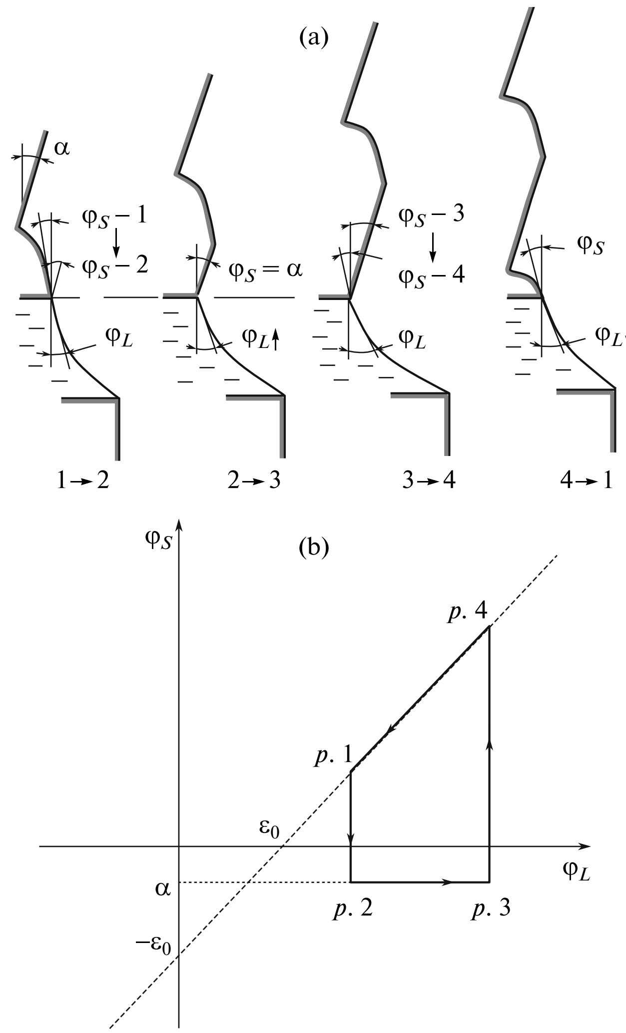

Fig. 3. (a) Schematic diagram of the step formation on the lateral side of base-faceted ribbon and (b) the hysteretic dependence of the angle of inclination of the crystal lateral surface (φ_S) on the meniscus angle at the triple point (φ_L) for the growth of base-faceted ribbon; ε_0 is the equilibrium growth angle and α is the angle of crystal misorientation.

CALCULATION OF PROFILE CURVE

The meniscus profile curves were calculated by solving numerically the Laplace capillary equation, written in the dimensionless form [8]:

$$z'' \pm 2(d - z)[1 + (z')^2]^{3/2} = 0. \quad (1)$$

A profile curve is given by the dependence $z(x)$, and the linear sizes are expressed in terms of the capillary constant $a = (2\sigma/\rho_L g)^{1/2}$, where σ is the surface tension coefficient of the melt, ρ_L is the melt density, g is the acceleration of gravity, and d is the dimensionless melt pressure at the shaper cut ($z = 0$). The pressure unit is the pressure of melt column as high as one capillary constant a.

The first boundary condition $z(0) = 0$ corresponds the meniscus pinning at the shaper edge. The second boundary condition can reflect the constancy of either growth angle ($z'(x_c) = \varepsilon - \pi/2$) or meniscus height ($z(x_c) = H$, where H is the crystallization front height).

To find the critical (minimum and maximum) angles $\varphi_L^{\min}$ and $\varphi_L^{\max}$, we used the experimental data on the lengths of faceting and isotropic growth areas on BF ribbons with different orientations.

It was established that within the proposed model the shape of the crystal lateral surface can be described for many various combinations of the crystallization front height and the horizontal distance between the working shaper edge and the three-phase line. These sets of values for different misorientation angles are shown in Fig. 4. The horizontal distance B in this figure is the maximum distance along the horizontal axis between the three-phase line and the shaper edge during growth (because the model assumes that the three-phase line constantly oscillates along the horizontal at a constant height H above the shaper). The depen-

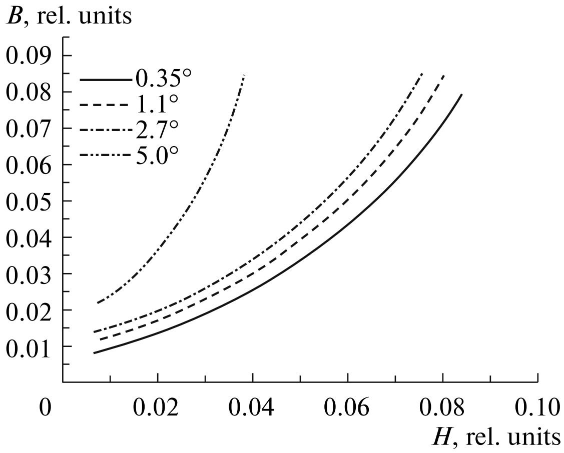

Fig. 4. Dependence of the horizontal shaper–crystal distance B on the meniscus height H at different crystal misorientation angles ("right" side).

dences of the critical angles φ_L^{min} and φ_L^{max} on the shaper–crystal distance for BF ribbons with different misorientations were calculated in the same way (Fig. 5).

Figure 6 shows also the calculated profile of the lateral surface of base-faceted ribbon with a misorientation angle of 1.1° and the change in the angle φ_L along the ribbon length. The Z axis in this figure coincides with the crystal growth direction, and the X axis is directed outward of the crystal (the linear size unit is the capillary constant, equal to 6 mm for sapphire). The faceting range corresponds to Z values from −0.48 to 0, and the isotropic growth (expansion) range corresponds to Z from 0 to 0.10.

As was established in [3, 9], the growth of base-faceted sapphire ribbons is accompanied by the growing ribbon shift with respect to the shaper axial plane, which is related to the deviation of the BF ribbon orientation from 90° direction. In addition, the sizes of steps formed as a result of faceting on the opposite ribbon sides differ by several tens of times. For example, for a ribbon with the misorientation angle $\alpha = 1.1°$, the average step period is 3.5 mm for the right side (where faceting leads to crystal narrowing) and 0.061 mm for the left side (where faceting results in crystal expansion).

It was fairly difficult to determine the ribbon shift by direct observation. Such a shift was estimated in [3] by growing a special bicrystal ribbon with two halves misoriented by 1.1° with respect to the opposite sides. Then, having measured the shift of the two ribbon halves and knowing the shaper thickness, one can estimate the growing crystal position with respect to the shaper. The following data were reported in [3]: at a ribbon thickness of 1.2 mm and a distance between the shaper edges of 1.72 mm the width of the gaps between the ribbon surface and shaper at both ribbon sides is 0.195 and 0.325 mm; the larger value corresponds to the right ribbon side, characterized by facial narrowing. These data can be used to estimate indirectly the model adequacy by calculating independently the meniscus height for both the right and left ribbon sides. For the right side, according to the dependence

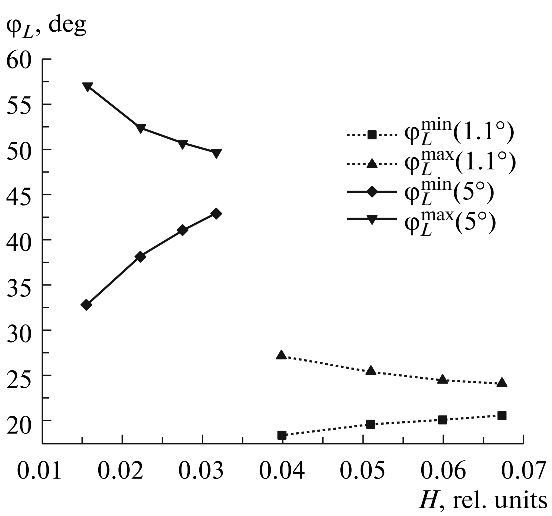

Fig. 5. Dependences of the critical angles φ_L^{min} and φ_L^{max} on the meniscus height H for base-faceted ribbons with different misorientation angles α.

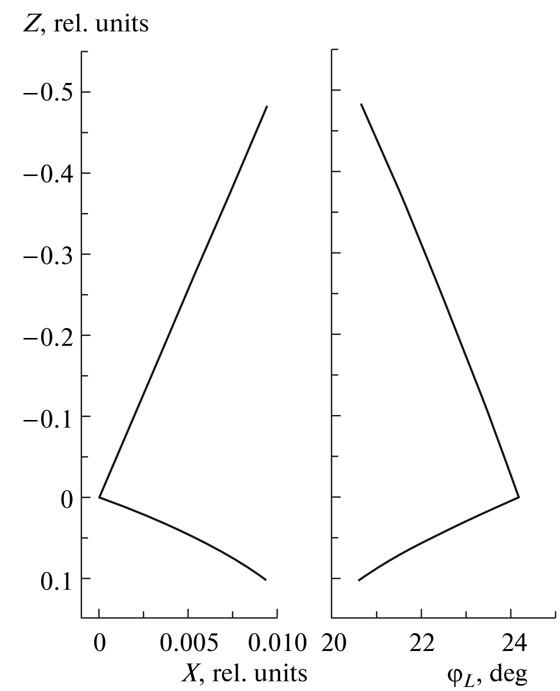

Fig. 6. Calculated profile of the lateral surface of base-faceted ribbon $X(Z)$ and the change in the angle φ_L along the ribbon length (misorientation angle 1.1°).

in Fig. 4, we have $H \approx 0.41$ mm. The calculations for the left side were performed assuming that the growth angle remains constant and obtained as a result $H \approx 0.40$ mm. In our opinion, the close values obtained are in agreement with the assumption about flat crystallization front.

CONCLUSIONS

Thus, our experiments on the growth of sapphire BF ribbons with different misorientations from 90° show that the surface energy anisotropy affects significantly the formation of the ribbon surface structure.

A model of step formation on the BF ribbon lateral surface is proposed, according to which the growth angle is constant only during transient region growth between the two regions of crystal facial narrowing, whereas during the growth of faceted regions the shape of the meniscus profile curve is determined by only the meniscus height and the current value of crystal thickness (position of the three-phase line). The meniscus height is assumed to be constant for all growth stages.

Based on measuring the sizes of the steps formed on the lateral surfaces of differently oriented ribbons, the model was parameterized, the shapes of grown ribbon lateral surfaces were calculated, and the range of variation in the meniscus angles at the triple point was found.

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