THE STRUCTURAL EVOLUTION OF LELY SEEDS DURING THE INITIAL STAGES OF SiC SUBLIMATION GROWTH

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ABSTRACT

The as-grown surfaces of 6H SiC seed crystals grown by the Lely method have been characterized by visible light microscopy and atomic force microscopy. The surfaces were then used to seed the sublimation growth of SiC at 2300°C, in 650 torr of Ar, with a gradient of 8 °C/cm. Repeated observations of the seed surface structure after predetermined intervals in the growth reactor demonstrated that transport starts at less than 2160 °C and that voids are nucleated by heterogeneous material on the original seed surface. Near these voids, the concentration of unit screw dislocations is increased and these defects migrate during growth. Screw dislocations of opposite sign act as step sources and build mesa structures around the heterogeneous material. After only 10 min at the growth temperature, micropipe/super screw dislocation complexes form in the centers of the mesas, possibly by the migration of unit dislocations to the voids.

INTRODUCTION

The physical vapor transport (PVT) growth method (a.k.a. seeded sublimation or modified-Lely growth) is currently accepted as the most practical way to produce large SiC monocrystals [1-3]. While wafers sliced from PVT grown monocrystalline boules are now used as device substrates, extended defects ultimately limit the yield of high power diodes [4]. The extended defects in typical wafers include low angle grain boundaries, second phase inclusions, polytypic inclusions, radial cracks, basal plane and threading dislocations, and micropipes [2]. Growth experiments conducted on nearly perfect seeds grown by the Lely method have demonstrated that the defects are not inherited from the seed, but form during growth.

The present paper describes a set of experiments aimed at understanding how extended defects are generated in PVT grown SiC. Prior to growth, the surface structures of Lely seeds were characterized by visible light microscopy (VLM) and atomic force microscopy (AFM). Structural characterization was then repeated after short treatments in the growth furnace. We have observed that threading screw dislocations nucleate at surface heterogeneities and that pairs of such defects act as sources for step loops; the continuous generation of step loops leads to the formation of mesa structures. Micropipes are frequently found in the centers of such mesas and a potential mechanism for their formation is described.

EXPERIMENTAL

The Lely seeds used in this study came from two sources. One set of seeds, with grown in pn junctions, were provided by D.L. Barrett. These crystals were grown by the Lely method [5] from Si and C powder, in flowing Ar, at 2550 °C. Al was incorporated in the charge material as a p-type dopant and N_2 was admitted to the growth ambient at later stages to produce an n-type layer. Additional Lely plates, nominally undoped, were obtained from Sterling Semiconductor. Prior to characterization or growth, a wet oxidation treatment (1100 ± 50° C, for 2-4 hours, in H_2O saturated O_2) was used to determine polarity and to remove surface impurities. The oxide was stripped in HF prior to growth, which was always carried out on the (0001) Si face and yielded the 6H polytype.

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The growth experiments were carried out using an apparatus similar to those described in the literature [6]. The seed and charge are enclosed in a graphite crucible that is surrounded by an inductively heated graphite susceptor. Both are surrounded by insulation and the assembly is contained in an Ar filled, water cooled quartz tube. The seed is mounted on the crucible lid and the charge (1-2 mm high purity Acheson powder, Elektroschmelzwerk Delfzijl) rests in the bottom of the crucible. Optical paths through the insulation allow the temperature at the top and the bottom of the crucible to be measured by separate two-color pyrometers.

In a typical growth run, the seed is "glued" to the crucible lid using caramelized sugar. To remove volatile components in the sugar, the mounted crystal/lid assembly is put in the furnace with an empty crucible and the system is pumped down to a pressure below $3x10^{-7}$ torr. The furnace is then heated in stages to about 1200 °C and held for 10 min or until the pressure is reduced below 1 x 10^{-4} torr. The charge is independently outgassed in a similar manner. During each growth run, the system is pumped to below $3x10^{-7}$ torr and then backfilled with 650 torr of ultrahigh purity (6.0) Ar . The crucible is then ramped to the growth temperature at a rate of 24°C/min. When the desired axial gradient is established, the pressure is reduced to increase the transport rate of Si and C from the charge to the seed. Because the objective of the experiments described here was to understand the initial stages of growth, the crystal was cooled at a rate of 80°C/min after a short (0-10 min) dwell at the growth temperature (approximately 2300 °C). In some cases, the heating was interrupted during the ramp.

RESULTS

The growth surfaces of Lely seed crystals had large flat regions covered with continuous step trains and a number of characteristic heterogeneities (see Fig. 1a). The typical heterogeneous features include second phase material and pits with circular or faceted hexagonal shapes. AFM images of the step trains show that atomically flat terraces, 1 to several microns wide, are separated risers that are several times the c lattice parameter (15 Å) in height. While surface/screw dislocation intersections have been found in the centers of the circular and faceted features, none have ever been imaged in the areas containing continuous step trains.

A second image of the same seed is shown after it was ramped to 2300 °C in 650 torr of Ar and held for 5 min with a gradient of 8 °C/cm across the crucible. Even during this very short treatment at a relatively high pressure of Ar, there is obvious growth on the surface of the crystal. Growth begins with the formation of mesas at the locations of surface heterogeneities. In Fig. 1b, it is obvious that a mesa has formed at each of the heterogeneities labeled with an arrow in Fig. 1a. Other crystal growth trials (not shown) were interrupted at earlier times or allowed to remain at the growth temperature for longer times. Based on observations from these runs, we conclude that the mesa structures first nucleate near the heterogeneities, then grow radially outward until they coalesce. An experiment that was interrupted at 2160 °C, during the heating ramp, showed that mesa structures had already formed on the seed.

AFM images show that in the center of each mesa, above the heterogeneous material, there is a pit in the surface layer (the black contrast in the center of Fig. 2a) and in the vicinity of this pit, there are numerous dislocations intersecting the surface. In any 50 μm x 50 μm image of the surface, at least one surface/screw dislocation intersection is observed. This places a lower limit on the dislocation concentration at 4 x 10^4/cm^2. Dislocations of opposite sign act as sources of step loops which expand outward during growth and create the mesa structures. A step fixed at two ends by dislocations of opposite sign expands outward during growth until line segments traveling toward one another annihilate, creating a complete loop and a shorter step fixed at the two defects [7]. As this process repeats, concentric step loops are ejected from the source. In Fig. 2, the steps are created on one side of the pit (labeled c) and different segments of the same step annihilate on the opposite side (labeled a) to form the complete loops that expand outward around the pit. Such sources are found on all of the mesas and another example is shown in Fig. 2b.

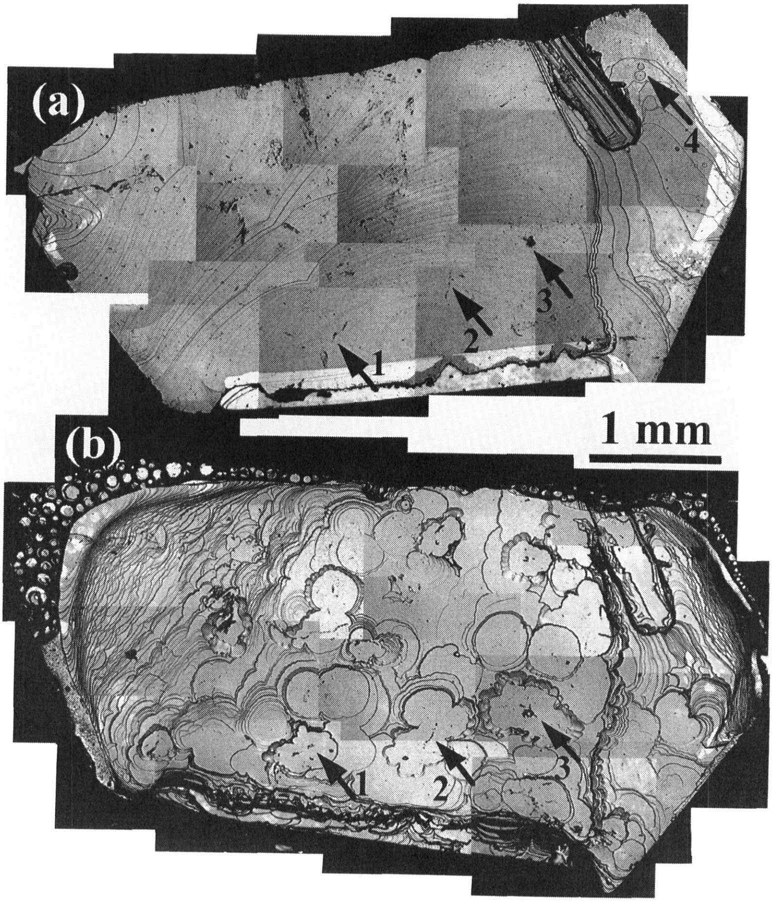

Figure 1. VLM images of the seed surface (a) before and (b) after growth for 5 min at 2300 °C in 650 torr of Ar. (a) Arrows 1, 2, and 3 point to examples of the heterogeneous surface material (black contrast). Arrow 4 points to examples of the circular and faceted pits. (b) Arrows 1, 2, and 3 point to mesa structures that formed above the heterogeneous material.

A second treatment of 5 min at 2300 °C resulted in expansion of the mesas and changes in the positions of the surface/screw dislocation intersections. For example, the AFM image in Fig. 3a shows the position where 5 dislocations with a unit screw component (b = 15 Å) intersect the surface after the first 5 min. of growth. Four of the dislocations are of the same sign and have clockwise rotating steps while the fifth has the opposite sign and rotates in the counter-clockwise direction. An AFM image of the same spot, after the second 5 min of growth, shows that the relative positions of the surface/dislocation intersection have changed (see Fig. 3b). It was also possible to identify the first micropipe/super screw dislocation complexes at this point in the growth. In each case, micropipes were found in the vicinity of the surface pits and an example is shown in Fig. 3c. While several types of cylindrical voids can be found in SiC, we reserve the term "micropipe" for the hollow cylinders with screw components of the Burgers vectors greater than or equal to 4 times the unit screw dislocation Burgers vector (b_0=15 Å). The hollow cylinder associated with these defects occurs to relieve the elastic strain at the dislocation's core [8-10]. The Burgers vector of the micropipe shown in Fig. 3c is $9b_0$.

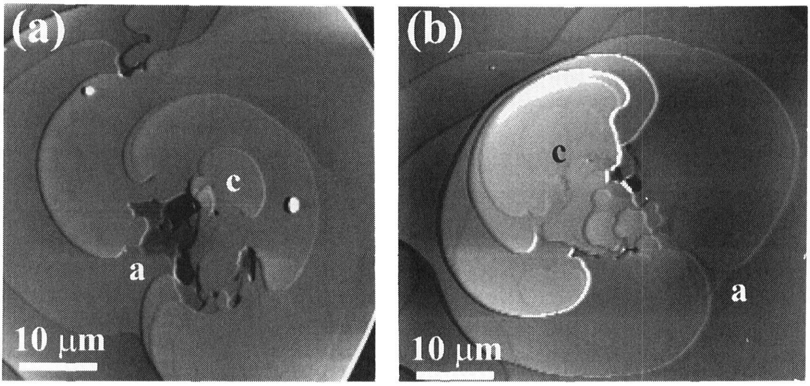

Figure 2. AFM images showing step sources on the tops of mesas after 5 min. at 2300 °C in 650 torr of Ar. The black-to-white vertical contrast is (a) 80 Å and (b) 125 Å. The steps in these images range in height from $2b_0$ Å (central loop) to $12b_0$ Å. See text for further descriptions.

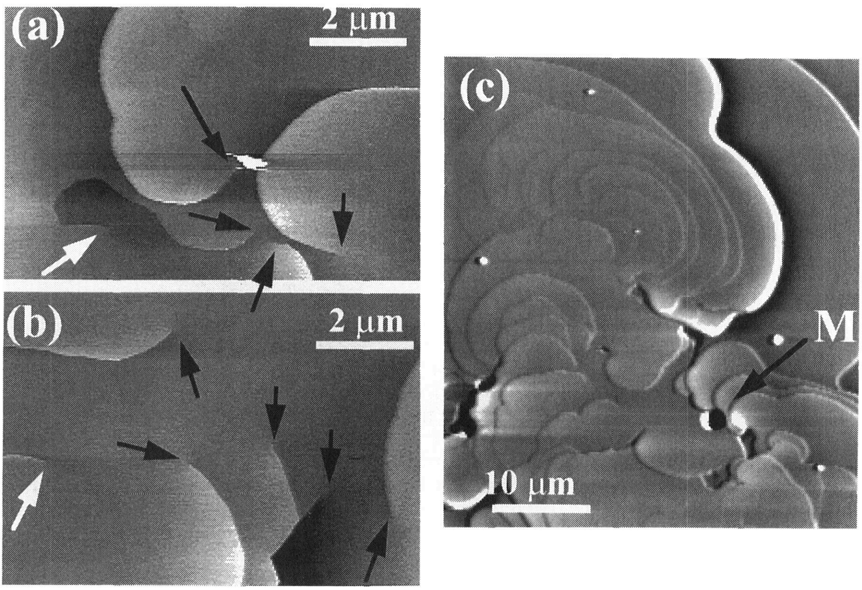

Figure 3. (a & b) AFM images showing the same surface/screw dislocation intersections (a) after the first 5 min of growth, (b) after the second 5 min. The dislocations with steps growing counter-clockwise are marked by a black arrow; white arrows mark those of opposite sign. (c) A micropipe (labeled M) on a mesa. The black-to-white vertical contrast is (a) 35 Å, (b) 30 Å, and (c) 140 Å.

DISCUSSION

Based on the experiments described here, the seeded sublimation growth of SiC starts while the system is being ramped to the growth temperature. The dislocation densities for typical Lely seeds are cited in the range of 10 to 1000/cm^2 [2]. However, after even very short treatments in the growth environment, the population of screw dislocations in the homoepitaxial layer is greater than 4 x 10^4/cm^2. Therefore, these dislocations are nucleated during the very earliest stages of the growth. Based on the observation that the concentration of these defects is highest in the vicinity of the voids that occur above the heterogeneous surface material, we assume that they are nucleated at this point by thermally induced shear stresses on the internal surfaces that bound the void (normal to the growth direction).

The presence of dislocations of both signs leads to step sources that produce circular steps during growth by the process originally described by Burton, Cabrera and Frank [7]. The continuous production of step loops creates regions around the defect initiation point with a relatively higher growth rate. As the steps expand outward, they apparently bunch to produce the steep walls that form the visible mesa boundaries.

We have also observed that the positions at which unit screw dislocations intersect the growth surface change as growth continues (see Fig. 3). Considering the fact that high temperature deformation tests have demonstrated that monocrystalline SiC deforms plastically by slip at 1300 °C, the motion of dislocations at 2300 °C is not too surprising [11]. There are several potential driving forces for dislocation motion. First are the elastic attractions and repulsions that exist between dislocations of opposite and like sign, respectively. However, these forces are very small at micron-scale separations. A second possible driving force is stress from thermal mismatch between the crucible lid and the seed, or from thermal gradients in the crucible. The magnitude of these forces has yet to be quantified. A third potential driving force for dislocation motion is the image force associated with steps moving across the surface. This phenomenon is outlined below [10].

Consider a step of height h (where $h \gg b_0$) that grows a very small distance (d) past the point where a screw dislocation intersects the surface. The new portion of the dislocation line feels an attractive image force, $-F_i = Gb^2/4\pi d$, which compels the surface/dislocation intersection to remain very near or even attached to the step as it continues to advance. For the dislocation line to advance with the step, it must eventually be bent to a minimum radius of h/2. Thus, there will also be a repulsive restoring force that tends to minimize the length of the line by pulling it away from the step so that it forms a straight line parallel to the c axis. This force will maximize at $F_r = \alpha 2Gb^2/h$, where α is a constant near unity, when the radius of curvature reaches its minimum value. Taking d to be a small number (i.e., we assume that very little new strained crystal is formed), equal to b_0, we see that as long as h is on the order of $8\pi b_0$ (approximately 400 Å), the step can "push" the dislocation line in a direction parallel to the step motion. When the minimum radius of curvature is reached, the step can grow only by breaking away from the dislocation line or by creating new edge segments parallel to the step growth direction. Because steps tend to bunch in SiC, steps of this height are not uncommon on the growth surface. The dislocation lines might also be moved significant distances by the consecutive action of many steps passing over the same point, advancing it by small amounts in the same direction.

We have also found that micropipes form during the very early stages of the growth. After the second 5 min treatment at 2300 °C, micropipes are observed near the centers of the mesas. The observation that a high population of unit screw dislocations is found in the presence of a void near the center of each mesa, and that the unit screw dislocations are able to move, adds support to the idea that micropipes form by dislocation coalescence. If the unit screw dislocations are driven to the voids either by large moving steps or by thermal stresses, then the strain associated with the dislocation core will prevent further growth in the empty core and a micropipe/super screw dislocation complex will be created.

As a final note, we should point out that the experiments described in this paper were carried out with the goal of studying defect initiation and not with the goal of growing the best possible crystal. In this case, we deliberately examined crystals with heterogeneous regions on the surface. In a state-of-the-art crystal growth, one would seed the process with a surface that has the lowest attainable defect concentration (actual details of the procedures used in commercial labs are closely held). However, surface defects are inevitably present on any seed and while our experiments certainly exaggerated the defect generation process, we expect that the mechanisms at work and the effects they have on the ultimate defect population are the same.

CONCLUSION

We have investigated the initial stages of 6H SiC PVT growth on Lely seeds. Extended defects such as dislocations and micropipes are generated during the first 10 min of growth. The concentration of these defects is especially high near heterogeneities on the seed surface. Screw dislocations of opposite sign continuously generate step loops which create mesa structures. The migration of unit screw dislocations during growth was observed. The migration of these defects to voids that are nucleated above surface hetreogeneities can lead to micropipe formation.

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