Bifurcation and Deviation of Cleavage Paths at Through-Thickness Grain Boundaries

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The behavior of cleavage crack fronts at grain boundaries in free-standing silicon thin films is investigated through a microtensile experiment. In addition to the crystallographic orientation, the orientation of grain boundary plane also plays a critical role. With respect to the initial crack surface, if the inclination angle is relatively small, the crack tends to penetrate across the boundary; if the angle is large, the crack may either bifurcate along the boundary or turn back on another crystallographic plane. The former is triggered by crack front transmission, and the latter may result in a higher critical crack growth driving force.

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I. INTRODUCTION

OVER the years, fracture in silicon thin films has drawn considerable attention.[1] The study in this area has shed much light on understanding unexpected failures or malfunctions of microelectromechanical systems and integrated circuits. Because amorphous silicon films are of relatively poor electrical and mechanical properties, very often microfabrication is based on polycrystalline silicon.[2] The grain size ranges from a fraction of micrometers to a few millimeters. Most of silicon thins films are produced through deposition processes.[3] Initially, at the surface of a substrate, a large number of crystal nuclei would be formed. Because they are of different orientations, their growth rates can be quite different. The nuclei with favorable axes oriented along the film growth direction can eventually dominate, and the others would be buried.[4] Consequently, most of the grain boundaries are through thickness. If the substrate temperature is relatively low, the grains tend to be small.[5] If the substrate temperature is relatively high, the grain size can be quite large.[6] In either case, grain boundaries are major obstacles to cleavage cracking.

In a brittle material, when a crack propagates along a crystallographic plane, the local fracture resistance is nearly constant, if the dynamic effects and the influence of dislocation activities are ignored. Because cleavage cracking in silicon can take place on both {111} and {110} planes and the surface free energies of them are only slightly different,[7,8] within the same crystal, the crack surface may shift among cleavage facets that are of similar orientations, resulting in relatively wavy flanks. Once the crack front reaches a grain boundary, if the orientations of the two grains are considerably different, especially when the twist misorientation angle is relatively large, the crack must overcome a significant barrier effect to enter into the next grain.[9] As the orientation of cleavage plane changes, the total area of fracture surface can become larger, and thus the work of separation increases.[10] Moreover, as the cleavage planes are misoriented across the boundary, a certain amount of grain boundary must be geometrically necessarily separated apart so as to complete the fracture surface separation, which demands additional fracture work.[11–14] Most importantly, the involvement of grain boundary separation makes the motion of cleavage front highly nonuniform. In a previous fracture experiment on large iron-silicon bicrystals,[15,16] it was observed that the crack front first penetrated across the boundary at break-through points (BTPs), as depicted in Figure 1. The distance between adjacent BTPs distributed in a broad range from less than 1 μm to more than 50 μm. The cleavage front around a BTP first penetrated across the grain boundary, becoming the protruding part; the rest of the front was arrested by the persistent grain boundary islands (PGBIs) between BTPs, becoming the concave parts. Along the protruding parts, the local crack growth driving force was lower than that at the concave parts. However, to keep the crack advancing, it must equal to the local fracture resistance. Thus, the overall stress intensity at the crack tip was higher. The main factors that govern the effective grain boundary toughness include the twist and tilt misorientation angles, the grain boundary shear strength, the effective boundary surface free energy, and the characteristic length.[17]

The grain-boundary failure process discussed previously can be regarded as the regular mode. Researchers also observed a number of other failure mechanisms, such as the irregular break-through mode,[18] the boundary cracking,[19,20] *etc.* In the current study, through a microtensile experiment on free-standing silicon thin films, we investigate possible behaviors of a cleavage front when it encounters a through-thickness grain boundary. It is noticed that other than the crystallographic structure, the orientation of the boundary plane is also important.

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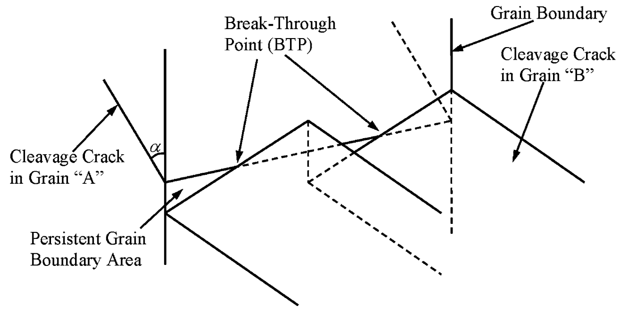

Fig. 1—Schematic diagram of the regular mode of crack front transmission across a grain boundary.

II. EXPERIMENTAL

In order to produce precracked testing samples with crack tips arrested by grain boundaries, a controlled temperature-gradient treatment was performed on a polycrystalline silicon wafer. The wafer thickness was 4.0 mm, and the grain size was around 5 to 15 mm. The material was heavily doped with boron so that it could be machined by electrical discharge cutting. The wafer was first heated uniformly in a tube furnace at 450 °C for 1 hour and then quenched nonuniformly by immersing one half of it in cold water. Among the large number of generated thermal cracks, the suitable ones that were arrested by through-thickness grain boundaries were chosen. The thermal cracks were usually generated in the section immersed in water, where the local temperature was relatively low and the dislocations should be inactive. Because the grain size is relatively large, the orientations of the grains could be determined through the Laue back-reflection technique.

Square pieces (15 × 15 mm) surrounding crack tips were harvested, sliced into 0.2-mm-thick films, and coated with silane groups. The coating was performed in a round bottom flask, with the opening of the condenser being protected by a drying tube. After drying and cleaning, the precracked silicon thick films were immersed in a dry toluene solution of chlorotrimethylsilane at 90 °C for 5 days. The chlorotrimethylsilane concentration was 20 pct. Its molecular size was quite small, so it could fully access the precrack surfaces,[21] forming protective surface layers.

The coated silicon films were mechanically ground to about 100-μm thickness by using an Unipol-300 polisher (MTI Co., Richmond, CA). During this process, the silane groups on outer surfaces were removed. The samples were then wet etched in a mixture of 7 pct of hydrofluoric acid, 75 pct of nitric acid, and 18 pct of acetic acid, until their thicknesses were thinned to 2 to 20 μm. The etching process took about 30 minutes. The film thickness was frequently examined using a laser interferometer. During etching, the etchant slowly flowed across the sample surfaces with the rate of 30 mL/min. The cross-sectional area of the etching chamber was 730 cm^2.

The silicon thin-film specimens were mounted on the self-aligning stages of a microtensile testing machine, which was developed previously for experiments on thin-film and soft materials.[22] Axial loading was

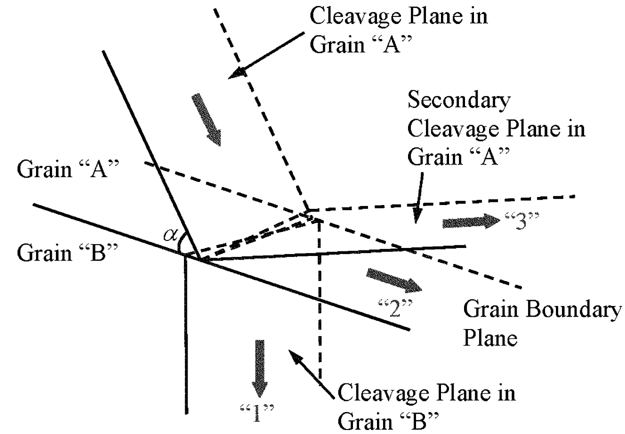

Fig. 2—Schematic diagram of cleavage cracking at a grain boundary in a free-standing thin film.

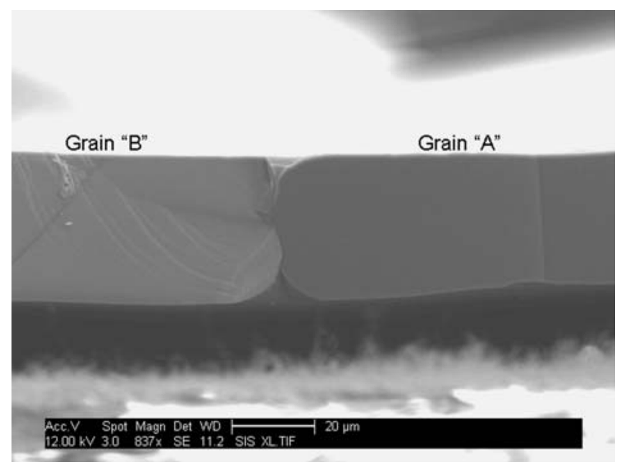

Fig. 3—SEM microscopy of cleavage cracking across a grain boundary. The crack propagates from the right to the left.

applied normal to the precrack path in displacement-control mode. The loading rate was about 170 nm/s. Once the critical condition was reached, the crack would overcome the grain boundary and propagate unstably. The fractured samples were thermal treated in a tube furnace in nitrogen environment at 400 °C for 2 hours, rinsed by acetone and ethanol, and dried in air. The fracture surfaces were observed in an environmental scanning electron microscope (SEM). Figure 2 depicts all the observed cracking modes. Figures 3 through 6 show typical fractography.

III. RESULTS AND DISCUSSION

Because no evidence of plastic deformation can be observed and the fracture resistance is relatively small,[23] the fracture mode is dominated by the cleavage process. A cleavage front may penetrate across the boundary from the grain behind the boundary (“A”) to the grain ahead of the boundary (“B”), as shown by

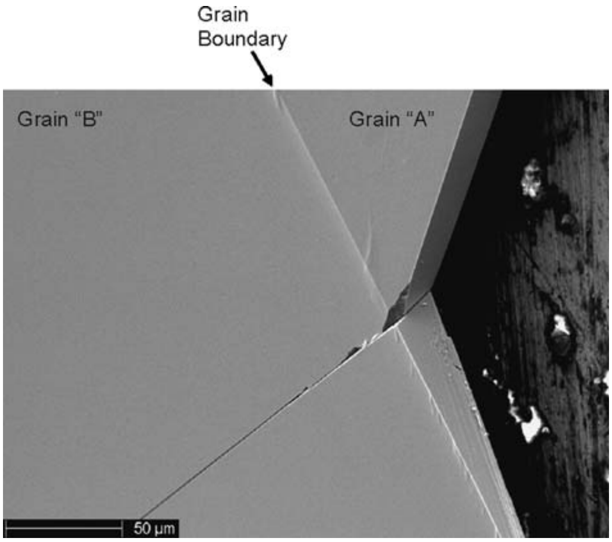

Fig. 4—SEM microscopy of cleavage bifurcation at a grain boundary. The crack propagates from the top to the bottom.

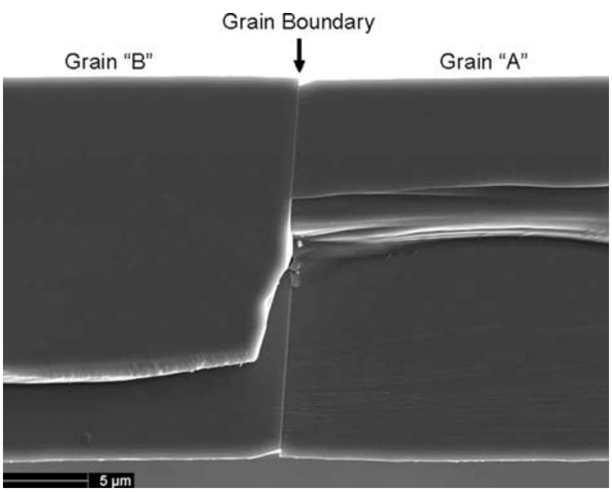

Fig. 5—SEM microscopy of the fracture surface of a bifurcated crack in the grain ahead of the boundary. The crack propagates from the right to the left.

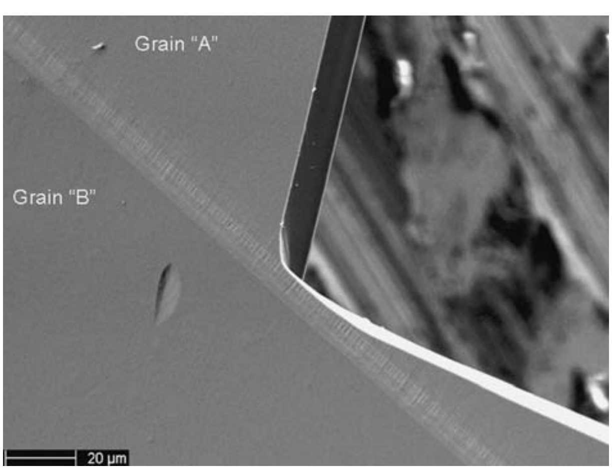

Fig. 6—SEM microscopy of a deviated cleavage path at a grain boundary. The crack propagates from the top to the bottom.

arrow "1" in Figure 2; advance simultaneously in grain B and along the boundary plane, as shown by arrows 1 and "2"; or propagate on another cleavage plane in grain A, as shown by arrow "3." The first mode (Figure 3) is compatible with the previous study on iron-silicon bicrystals.[15] It tends to be dominant when the incident angle, α, is large. About 80 pct of the grain boundaries break in this mode. The crack surface shifts from the cleavage plane of grain A to that in grain B at a BTP. Different from the breakthrough processes in large bicrystals or coarse-grained polycrystals,[15,16] where the cleavage front overcomes the boundary at a number of BTPs distributed quasi-periodically, in a thin film, the grain boundary width can be insufficient for multiple BTPs. As the film thickness is smaller than 20 μm, there can be only a single BTP along the entire crack front. Under this condition, if the film thickness is lowered, because the area of PGBI is reduced, the overall grain boundary toughness may decrease; that is, the front transmission process cannot be fully developed, primarily because of the confinement effect of the lateral film surfaces.[23] As the front penetration depth becomes larger, the shear stress on the PGBI connecting the fracture surfaces across the boundary keeps increasing. Eventually, the PGBI is separated apart, and the barrier effect of the grain boundary is overcome. The critical condition of the onset of unstable crack advance can be assessed in the framework of *R*-curve analysis.[24] With the increasing front penetration depth, the effective resistance offered by the boundary to cleavage cracking rises, because more grain boundary area is involved and more cleavage plane in grain B is exposed to the front. The crack growth driving force also increases. However, its increase rate must be smaller than that of fracture resistance; otherwise, the front penetration cannot be stable. Both of the increase rates of resistance and energy release rate are functions of the effective crack growth length. At the critical condition, they equal each other, and a small increment in front penetration depth would cause unstable crack behavior. When the boundary width, *i.e.*, the film thickness, decreases, it is easier to reach the critical condition, because the relative variation in grain boundary area involved in the front transmission is larger.

The BTP is always a certain distance away from the film surface. First, the PGBI area can be minimized if the BTP is at the center point of the boundary. If the BTP is at the intersection of the boundary and the lateral film surface, the total PGBI area would increase by a factor of 2. A larger PGBI area demands more work of separation, and thus is energetically unfavorable. Second, near a free surface, the local stress intensity at the crack tip tends to be lower.[25] For instance, for a cleavage front propagating along a crystallographic plane, the sections close to free surfaces are always behind the central part.[26]

If the incident angle is relatively small, as shown in Figure 4, the cleavage path may bifurcate, which is observed at about one-half of the grain boundaries that do not fail in the regular mode discussed previously. One branch is along the boundary plane, and the other is along the cleavage plane in grain B; that is, the failure mode of the grain boundary exhibits both intergranular and transgranular characteristics. As shown in Figure 5, the features of transgranular cracking are quite similar with those of the front transmission discussed previously. The front bypasses the boundary at a BTP in the central part of the boundary, and the PGBI is separated apart *via* shear deformation or shear fracture. The crack surface along the boundary is quite smooth (Figure 4), containing no defect sites that typically exist in grain boundaries of many alloys.[27] Clearly, because in silicon the grain boundaries are clean, they are not weakened by inclusions. Their surface free energies are about 80 pct of that of crystallographic planes,[28] which is related to the disordered atomic structure. Because the energy difference is relatively small, occasionally, river markings can be observed in isolated areas, indicating that the fracture surface can shift to cleavage planes in grain A or B nearly parallel to the boundary.

The two cleavage paths, one in grain B and the other along the boundary, must be formed simultaneously; that is, the crack branching is not caused by secondary cracking. If one of them is produced first, because the microtensile experiment is displacement controlled, the stress intensity at the verge of propagating front would increase rapidly. Most of the strain energy stored in the background would be released at the crack tip, and due to this shielding effect, crack branching behind the front is difficult. Because silicon is brittle, T stress (the nonsingular term of crack-tip stress field) should be of only a negligible influence.[29] The cracking behavior is dominated by the stress intensity factor as well as the angular distribution of stress. Because the degree of anisotropy of silicon crystals is quite weak,[30] as a first-order approximation, the crack-tip stress field can be assessed by using the solution for isotropic materials. Along a plane with a tilt angle of θ from the crack surface in grain "A," the crack opening stress is[31] $\sigma_\theta = \left(K_I/\sqrt{2\pi r}\right)\cdot f(\theta)$, where K_I is the stress intensity factor, r is the distance to the crack front, and f is a function with the peak value at 0. While σ_θ reaches the maximum value when θ is small, if there is no cleavage plane along this direction, the crack must advance along another direction. When α is small, the opening stress along the boundary is relatively large, and it may be close to that of the cleavage surface in grain B. Even when the crack opening stress of the boundary is slightly smaller, bifurcation can still occur because the cleavage path along the boundary is only tilted from the initial fracture surface, while there can be a considerable twist misorientation of cleavage planes across the boundary, which causes additional fracture resistance. Note that in the current study, boundary cracking without front penetration was never observed. That is, even though the two fracture paths are produced at the same time, the crack front transmission is a prerequisite for cleavage path bifurcation, which is likely associated with the stress concentration at the kink-like front segments around a BTP.

When the incident angle is small, another possible mode is cleavage path deviation, as shown in Figure 6. Instead of penetrating across the boundary, the crack front advances along another crystallographic plane, as if the boundary reflected the crack back into grain A. The crack opening stress along the cleavage plane should be relatively large. More importantly, the resistance of the boundary must be high, *e.g.*, when the twist misorientation is significant. Under this condition, the crack front penetration is difficult, and thus, the local stress concentration at the BTP that triggers crack bifurcation along the boundary is no longer available. The crack-tip stress intensity keeps rising until secondary cracking occurs, which first takes place along a plane close to the boundary and rapidly deviates away and is stabilized. The critical stress intensity factor, although higher than that for crack bifurcation (*i.e.*, cleavage cracking along the boundary), should be smaller than that of crack penetration.

There are a number of other factors that are not analyzed in the current study, *e.g.*, the variation in chemical composition at grain boundaries due to prolonged etching, the grain boundary grooves, *etc*. Because all the samples are treated through the same procedure, they should not be the major reasons causing the difference in cracking mode.

IV. CONCLUSIONS

To summarize, by observing cleavage cracking at through-thickness grain boundaries in a number of free-standing polycrystalline silicon thin films, it is discovered that in addition to the crystallographic orientation, the crack front behavior is also determined by the orientation of the grain boundary plane. Although the tilt misorientation angle is less important to the resistance of the grain boundary to transgranular crack advance, the incident angle of the cleavage crack is critical to crack bifurcation or deviation. When the incident angle is relatively small, when the boundary toughness is low, the crack front can penetrate through the boundary and simultaneously bifurcate along the boundary. If the boundary toughness is relatively high, the crack must shift to another cleavage plane in the grain behind the boundary. For crack bifurcation, the triggering event is still the front transmission, and thus, the overall fracture resistance should be similar to that of the regular crack penetration mode. As the front penetration does not take place, a higher crack growth driving force may be required for crack deviation.

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