Real-time SEM/FIB Studies of Whisker Growth and Surface Modification

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We report on real-time measurements that enable us to watch the morphology of whiskers and hillocks forming in real-time and provide insight into the mechanisms controlling their growth and initiation. These measurements show that whiskers appear to grow out of a single grain on the surface with little lateral growth. To understand why whiskers initiate at specific sites, we modified the surface using the focused ion beam to remove the oxide in selected areas. Whiskers did not grow out of these uncovered areas, indicating that the underlying grain structure is important to whisker growth and it is not sufficient to just remove the oxide barrier. In comparison with whiskers, we found that hillock formation is accompanied by a large amount of grain growth and often by grain rotation at the surface.

INTRODUCTION

Tin is used heavily in the electronics industry as a protective coating on copper conductors because of its excellent conductivity and resistance to oxidation and corrosion. Additionally, its low melting point and ready formation of intermetallic compounds makes it an ideal candidate as a solder material for joining interconnects. In the past, alloying with Pb has been used to lower the melting point further and impede the formation of Sn whiskers[1–3] (i.e., thin filaments of Sn that grow out of the surface and can cause system failures by creating short circuits).[4] However, the recent industry move to Pb-free processing has once again raised concerns about the reliability issues in electronic components due to Sn whisker formation.

Over the six decades since their initial discovery,[5] a large body of research has been dedicated to determining the mechanism of whisker formation.[2] Even so, the whole process is still not well understood and there is not an accepted whisker mitigation technique to replace the addition of Pb. To prevent whiskers, we need to understand the underlying driving forces and kinetic processes controlling their formation. This paper reports observations made using real time scanning electron microscopy (SEM) to monitor whisker and hillock nucleation and growth. These measurements provide a window into the detailed process of how the surface evolves and give insight into the controlling mechanisms.

See the sidebar for experimental background.

RESULTS

We observed two types of morphologies for features that grew on the surface that we classify as whiskers and hillocks. The term whiskers refers to long thin filaments that appear to grow out of a single grain on the surface and show little observable widening in the SEM images. Hillocks are more mound-like in shape. They also appear to initiate from a single grain but grow in both the lateral and vertical directions, consuming neighboring grains as they grow.

We continuously monitored the surface for periods of 2–6 days in the SEM/focused ion beam (FIB) to observe both whiskers and hillocks nucleating and growing on the Sn surface. On average, 11 features (approximately 4 of which were typically whiskers) were observed over each area of 215 × 185 μm. For comparison, samples with identical structures that were kept in air over the same length of time[3,8] had a density of 240 features in a 1 mm square region after 2–4 days (slightly more than half

How would you…

…describe the overall significance of this paper?

This study follows the evolution of the surface morphology of pure Sn coatings over Cu and shows how whisker and hillock features form. It shows that these features nucleate at specific "weak" grains that can plastically deform at lower stress than their neighbors. Just having a weak surface oxide layer is not sufficient for a whisker to nucleate.

…describe this work to a materials science and engineering professional with no experience in your technical specialty?

This study measured the real-time growth of whiskers and hillocks on Sn coatings over Cu in a FIB/SEM system. The surface features grow due to compressive stress in the layer induced by the formation of Cu-Sn intermetallic. Long whiskers form when the grain grows out of the film without lateral grain growth. Hillocks form when there is lateral grain growth accompanying the growth outwards. Features nucleate at specific grains that plastically deform at lower stress than their neighbors. There is no apparent weakness in the surface oxide or other defects before nucleation occurs.

…describe this work to a layperson?

Lead had been removed from tin coatings in electronics manufacturing because it is harmful to the environment. This includes the formation of tin whiskers that can cause system failures and a shorter product life. This study provides real-time observations of whisker growth on lead-free tin coatings in order to understand the cause of their growth and develop mitigation strategies.

of those features were hillocks as opposed to whiskers). In the following section we describe our observations regarding whisker and hillock growth with accompanying images of the evolving morphology.

Whiskers

Figure 1 shows the nucleation and growth of a whisker-type feature at different time intervals. The feature does not start to form until 14 h after the initial deposition. Before that, no changes can be seen to occur on the surface (Figure 1a) relative to the first measurements made at this position. After 14 h, we observe a rapid change in the surface morphology around the position where the whisker will grow. The image appears to correspond to the lifting of the oxide on the surface over the emerging whisker. We refer to this as cracking although the details of the change in the oxide cannot be clearly observed. The crack spreads rapidly around the base of the forming whisker, appearing to follow the boundary of the original grain on the surface. After 20 min. (Figure 1b) it has spread roughly around half of the grain out of which the whisker is forming. After 40 min. (Figure 1c), the crack encompasses the entire whisker grain and we can observe the surface of the growing whisker detach from the surrounding film. For this particular whisker it took roughly 40 min. for the crack to fully propagate around the grain and detach from the Sn surface; in other cases the cracking process took from 10 to 70 min. No surface contamination or other defect was observed on the grain before it started to form a whisker or on the surrounding grains. Also no obvious surface morphology changes were observed in the surrounding grains after the whisker started to grow.

One possible cause of whisker initiation is a weakness in the oxide above the grain so that it can crack more easily and release the whisker. To determine if this was the case, we deliberately removed the oxide by using the FIB to sputter away circular regions to a depth of 10 nm with various diameters (0.5 μm, 2 μm, and 5 μm). The sample remained in the FIB after oxide removal so that no fresh oxide would grow over the sputtered holes. An example of one of these sputtered regions is shown in Figure 2, where the circle drawn on the figure highlights the region that was sputtered. We found that the Sn did not extrude through the holes that were made in the oxide, indicating that the underlying grain structure is critical for whisker nucleation, not just a weak oxide. The implications of this measurement are discussed later in this paper.

After the nucleation (oxide-cracking) stage, we find that the whisker grows at a nearly uniform rate and in a nearly constant direction for all the whiskers observed in these experiments. In contrast, in other experiments[2,7,9–11] whiskers have been observed to grow intermittently with pauses and/or change direction (i.e., form kinks). In our experience, we observed this to occur in whiskers grown from samples kept in air or measured in an SEM instrument with a poorer quality base vacuum (4×10^{-4} Pa) than the current experiments. This suggests that the presence of oxygen, water vapor or other gas may play a role in the non-uniform growth of whiskers seen in these cases, perhaps by regrowing a surface oxide that retards or modifies the whiskers growth.

From the measurements of the whisker length vs. time (Figure 3a), we can quantify the whisker growth kinetics. The whisker length is estimated by measuring the SEM image which does not account for the angle of growth of the whisker. Therefore it only provides a lower bound for the actual length. As seen in the figure, there seems to be an incubation period of 14 h, which is consistent with our previous findings.[3,8] The whisker grew to 14 μm in next 34 h, in Figure 3b we plot the instantaneous growth rate estimated from the length measurements. As shown in the figure, the growth rate is initially high then drops to a steady state rate of 1.14×10^{-10} m/s, similar to the rate found in the literature.[9,12,13] With a diameter of about 1.1 μm, the volume of Sn ex-

EXPERIMENTAL BACKGROUND

Bilayer samples of Sn and Cu were prepared on Si substrates in the form of 25.4 mm × 12.5 mm rectangles, The Si was (100) oriented with a 100-nm-thick oxide and cleaned before deposition by 5 min. each in acetone, methanol, and isopropanol bath with ultrasonic agitation, followed by drying with compressed nitrogen.

The Cu layer of 1000 nm was deposited using electron-beam evaporation (pressure during deposition = 4×10^{-4} Pa). Sn layers of the desired thickness (either 2000 nm or 4000 nm) were electroplated over the Cu using a commercial plating solution controlled by a potentiostat. A 15 nm Ti layer was evaporated on the Si prior to Cu to enhance adhesion to the substrate. Before electroplating Sn, the Cu samples were dipped in concentrated sulfuric acid for 15 s followed by rinsing twice in de-ionized (DI) water for 30 s each to remove any copper oxide. The resulting Cu films had a fine grain microstructure with grain size on the order of 100 nm as seen by transmission electron microscopy[6] while the Sn layers were columnar with a grain size that was comparable to the film thickness.

The CuSn samples were kept in air for 4–6 h to allow SnO_2 to form on the surface. Previous work[6] indicates that a 5–8 nm native oxide forms under these conditions. The sample was then mounted in the focused ion beam/scanning electron microscope (FIB/SEM) and was monitored over several days at high vacuum conditioins (6×10^{-6} Pa).

Because whiskers are small but widely separated, it is necessary to monitor a large area with high resolution in order to capture their growth process. We modified the acquisition program of the SEM on the FIB system to capture and save images over a wide area by automatically moving the sample stage and allowing multiple areas on the same sample to be measured over the same time period. Using this program five areas of 215 × 185 μm which were 1 mm apart were monitored. The stage was moved every two minutes and thus each area was revisited every 10 minutes. The images were captured at the maximum resolution (4096 × 3536) allowing us to clearly distinguish the grain boundaries and other surface features.

From the series of images small sections which show whiskers and hillocks nucleation and growth were then selected and are presented in the Results section. Individual animated movies made from the sequence of captured images can be seen at the link provided[7] for all the features discussed in this paper.

In addition to monitoring the surface, we used the FIB to remove the oxide from selected circular regions with different diameters. These regions were then also monitored for surface feature formation. This allowed us to determine the role of the oxide in facilitating whisker or hillock growth, as discussed elsewhere in this article.

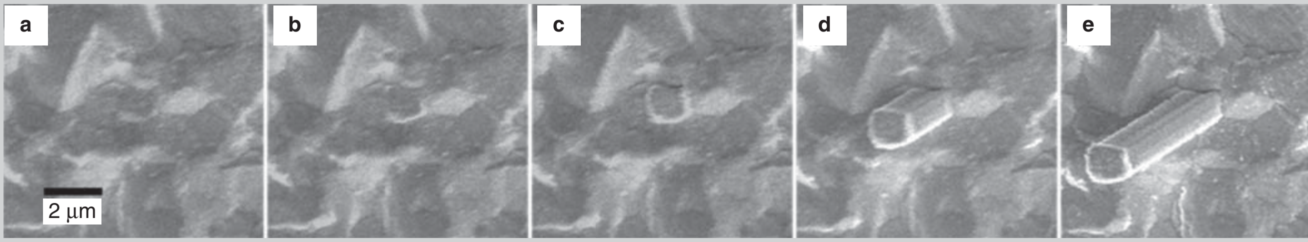

Figure 1. Time series SEM images showing whisker nucleation and growth. Time after deposition: (a) 14 h, (b) 14 h 20 min., (c) 14 h 40 min., (d) 17 h, (e) 21 h. View on-line to access movie or go to *http://www.engin.brown.edu/faculty/Chason/research/whisker1.html*.

truded from the surface occurs at a rate of 1.08×10^{-22} m^3/s. A similar rate was calculated for the other whiskers found on the same sample in different areas.

Hillocks

The key difference between whiskers and hillocks seems to be that the grains which form hillocks undergo lateral grain growth whereas the grains forming whiskers just grow in the upward direction. To illustrate this difference, images taken from regions where hillocks form are shown in Figure 4 for various time intervals after the initial deposition. These images are taken from a sample with Sn thickness of 4 μm which had a similar ratio of whiskers to hillocks as the 2 μm Sn samples. The hillocks start nucleating after an incubation period of 8–10 h after Sn deposition. We have included images from several hillocks to illustrate different features of growth.

In the first hillock example (Figure 4) the nucleation appears to start at a single grain, similar to the initiation of the whisker. However, unlike the whisker, the top surface of the hillock rotates as it grows until the initial oxide-covered top surface of the hillock is oriented approximately 90° relative to the surface it started from. To highlight this, a line drawing of the hillock is shown in the inset in Figure 4a–d. The rotation appears to occur due to one side of the hillock growing outward faster than the other. The extrusion of material is clearly occurring by addition of Sn at the base of the hillock—the Sn that is in the hillock above the surface does not change its morphology after it first forms.

After the initial rotation, the base of the hillock starts to widen at the same time that it is pushing up (Figure 4d–h), indicating an extensive amount of lateral grain growth by the hillock grain. As the hillock consumes adjacent grains, the horizontal growth is roughly constrained by the grain boundaries on the surface, appearing to consume an entire neighboring grain and then slowing down before consuming the next grain. Some of the neighboring grains are incorporated into the growing hillock while other grains remain unchanged and determine the hillock's horizontal boundary.

The sequence of growth often proceeds in a step-wise fashion, with an increment in horizontal grain-growth followed by an increment in vertical growth. This leads to the formation of horizontal steps (striation marks) on the side surface of the hillock as it grows. These striations correspond to the size of the hillock base at the time when it was pushed out of the surface so that, like growth rings on a tree, they can be used to recreate the history of the hillock's morphology. Similarly, the vertical ridge (as pointed to by the arrow in Figure 4h) forming on the hillock appear to be the remnants of grain boundaries between the adjacent grains (pointed to in Figure 4d) which, as the hillock grain grew laterally, got absorbed into the hillocking grain.

It is interesting to note that the surface oxide around the hillock's base does not seem to be preventing it from growing in the upward direction. As the hillock grows, it carries the oxidized surface with it. The features that were present on the Sn surface (for example the white particle circled in Figure 4e and h) stay there and get lifted with the hillocks, and thus the surface of the hillock carries with it the history of the Sn surface before the hillock appeared. After 76 h the lateral grain growth slows down and stops; at this point the grain boundaries might have become pinned. After this, the hillock is only observed to grow in the upward direction for the duration of the measurement.

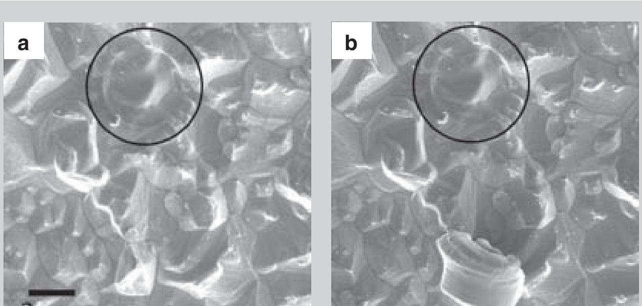

Figure 2. (a) SEM image of Sn surface with a hole in oxide layer made by FIB at 6 h after deposition, (b) image after 138 h; no growth is visible where oxide was removed but hillock is observed within approximately 10 μm.

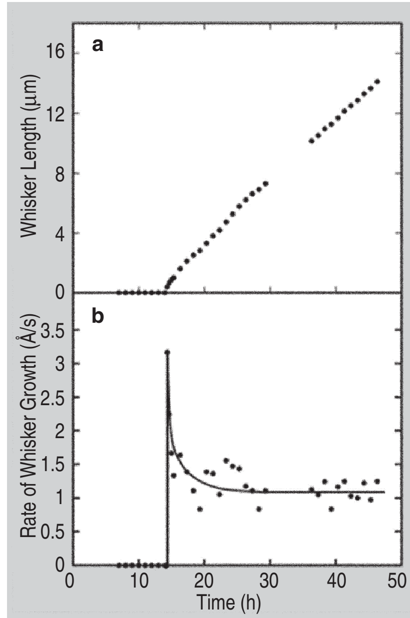

Figure 3. Measurement of: (a) whisker length vs. time; (b) instantaneous growth rate of whisker vs. time.

Figure 5 shows a sequence of images from another hillock on the same Sn sample, captured over the same time period but on a different area. In this case, the hillock appears to start growing from only a section of a single grain on the surface. The oxide breaks in the middle of the grain (highlighted by the circle) and the part on the left side starts growing upwards leaving the remainder of the grain behind (Figure 5b and c). Similar to the previous example, after the oxide cracks the original surface of the grain rotates by approximately 90° relative to its starting orientation (Figure 5d and e). After this rotation, the hillock grows primarily in the vertical direction with little lateral grain growth (Figure 5f–h). However, the tilt angle of the hillock relative to the surface changes several times during the growth as the growth rate at the base varies. The horizontal marks on the hillock (one of which is shown by the arrow in Figure

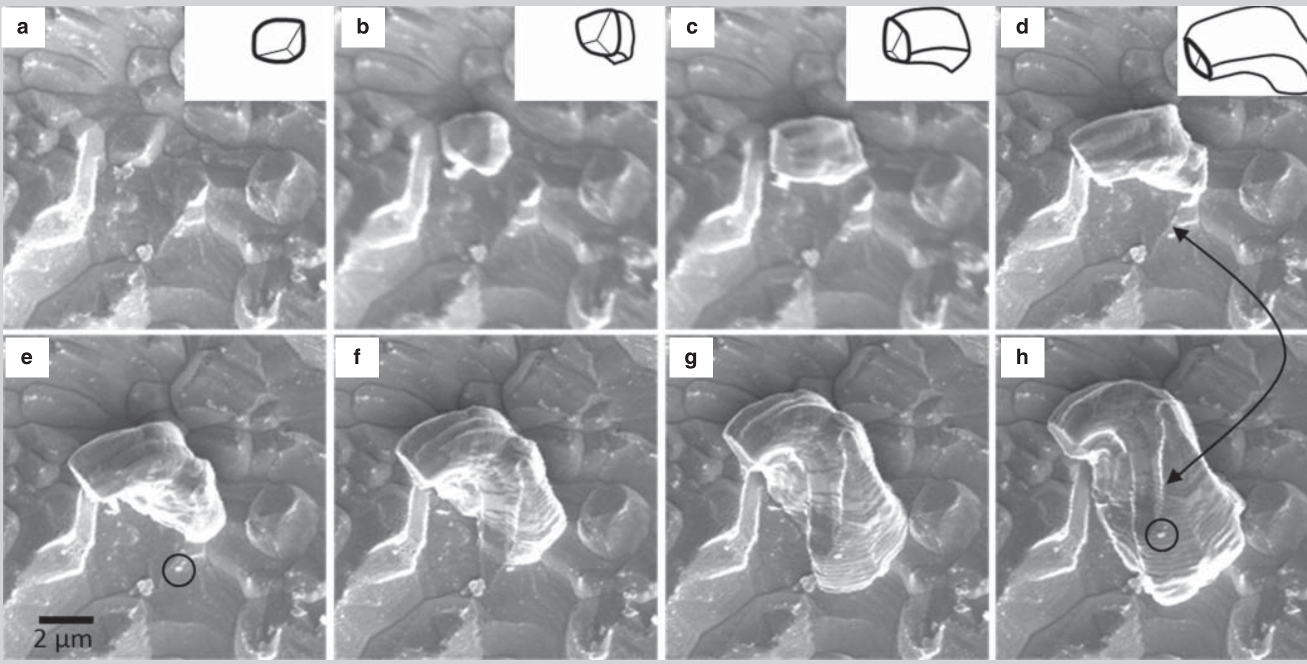

Figure 4. Time series SEM images showing hillock growth with surface rotation and extensive lateral grain growth. Time after deposition: (a) 6 h, (b) 12 h, (c) 18 h, (d) 32 h, (e) 44 h, (f) 56 h, (g) 76 h, (h) 138 h. Insets shows schematic of shape evolution highlighting rotation of the original surface. Arrows point to grain boundary features in (d) that are visible as ridges on side of the hillock in (h). View on-line to access movie or go to *http://www.engin.brown.edu/faculty/Chason/research/Hillock2.html*.

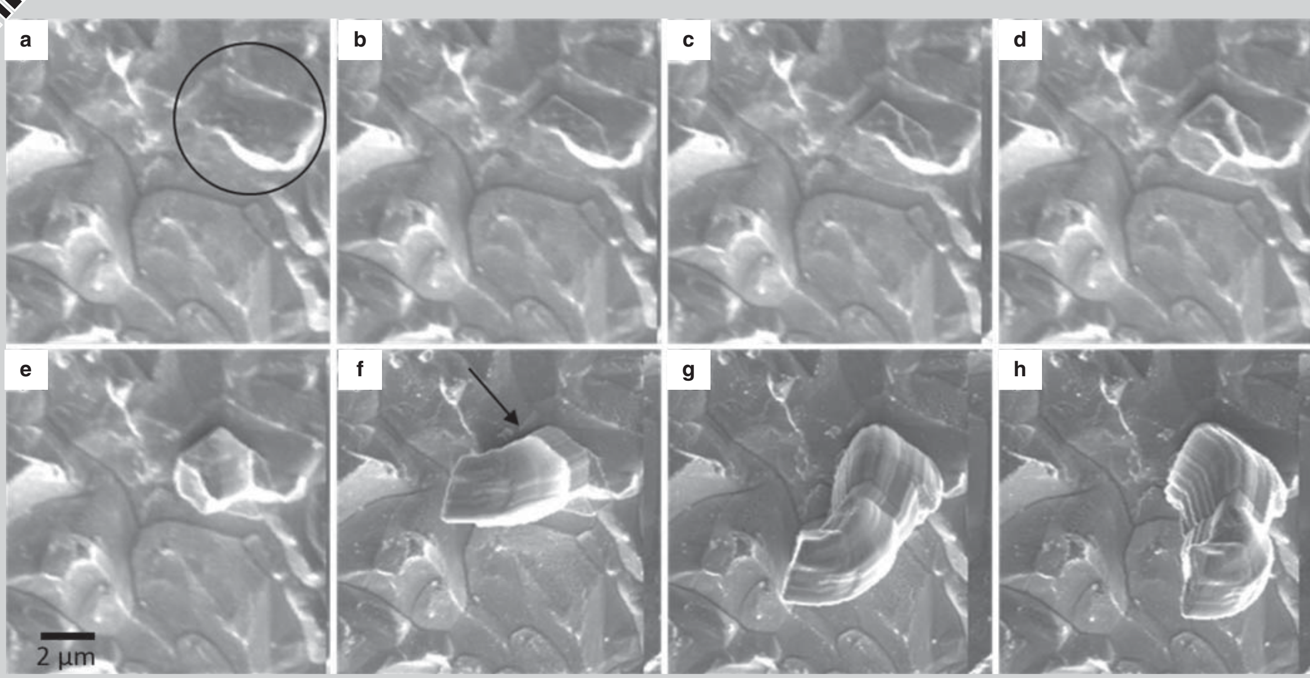

Figure 5. Time series SEM images of hillock that appears to emerge from only a portion of the original grain. Time after deposition: (a) 6 h, (b) 13 h 30 min., (c) 13 h 40 min., (d) 15 h, (e) 20 h, (f) 40 h, (g) 96 h, (h) 138 h. Circle in (a) highlights region of interest. Arrow in (f) points to horizontal band that forms when growth direction changes. View on-line to access movie, or go to *http://www.engin.brown.edu/faculty/Chason/research/Hillock3.html*.

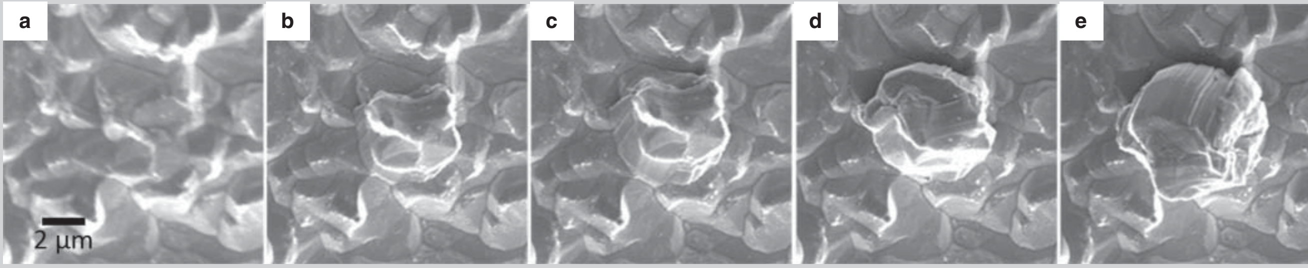

Figure 6. Time series SEM images of hillock growth comprising multiple grains on original surface. Time after deposition: (a) 6 h, (b) 31 h, (c) 56 h, (d) 81 h, (e) 106 h. View on-line to access movie, or go to *http://www.engin.brown.edu/faculty/Chason/research/Hillock4.html*.

5f) are indications of the point where the direction of the hillock growth changed. After roughly 40 h the remainder of the grain that did not grow initially also starts growing.

Figure 6 shows surface images from another hillock (same sample), in which the growing feature appears to be the result of 3 or 4 initially separate grains growing outward together in the form of a pillar with little lateral growth beyond what occurred before the hillock started to grow. After 81 h the hillock starts to consume an adjacent grain which changes the growth mode and leads to rotation of the hillock.

Figure 7 shows one more hillock taken from the same sample. In this case one side of the hillocks seems to remain attached to the surface, perhaps due to its incapability to completely break the surface oxide. As the hillock grows, the surface curves but there is no vertical growth. Finally it stops after roughly 50 h. The halt in the growth may be due to the fact that the curving surface curved by 180° and hit the starting surface. This appears likely as growth stopped immediately after hitting the surface. In comparison, other hillocks on the same sample continued to grow suggesting that the driving force for hillock formation had not been depleted. This again points out that the surface oxide is important in deciding the fate of the hillock.

Finally, we found regions in which grain growth could be observed underneath the surface with very little upward movement. This caused enough change in the surface structure so that the grain growth could be observed but no formation of a surface feature could be seen.

DISCUSSION

The growth morphologies can provide insights into the mechanism controlling whisker/hillock growth since their shape and orientation is intimately related to the way in which atoms are incorporated into them. In the first part of the discussion, we consider the significance of the fact that whiskers don't start to form in regions where we have removed the surface oxide. In the second part, we present a brief overview of the driving forces and mechanisms that we believe control whisker growth (based on our own work and that of others) and explain how we believe they relate to the morphologies that we observe.

Role of the Surface Oxide in Whisker/Hillock Nucleation

Tin surfaces exposed to air grow a tenacious native oxide which plays an important role in stress evolution by suppressing relaxation via diffusional creep of atoms to the surface.[10,14,15] Indeed, it has been shown that removal of the surface oxide by sputtering[3] or chemical etching[16] leads to relaxation of the stress in the layer. Therefore, it has been suggested[10,15,17] that whiskers form preferentially at weak spots in the oxide which can be more easily cracked to allow material to flow out of the coating.

To address the role of the oxide in nucleation we used the FIB to remove the oxide layer at selected regions on the surface as described above and shown in Figure 2. Importantly, we found that no whisker or hillock-type features grew out of these holes. Moreover, we found that a hillock-type feature did form at a distance of only 10 µm from the hole (Figure 2b) which shows that the surface modification did not remove the driving force for hillock formation. We believe this result clearly indicates that it is not sufficient to weaken the oxide to initiate the growth of surface features. Instead, whisker nucleation is determined by something in the underlying film.

We also looked at the effect of removing a larger area of the oxide by sputtering a region of size 50 × 50 µm. Even though we would have expected to see some features forming in a region of this size, we didn't find any. We interpret this to mean that modification of a large area of the oxide can relieve stress and hence remove the driving force for whisker/hillock growth.

Growth Modes of Whiskers

Although it is by now generally accepted that stress is the driving force for whisker growth,[2,3,8,10,11,17–20] this knowledge alone does not explain how whiskering occurs. To understand it, we must consider how the stress gets generated, how this leads to the transport of material to the whiskering grain and how this material gets incorporated into the whisker. At the end of the section, we discuss how these mechanisms are related to the results of our FIB/SEM measurements.

The fundamental source of stress in the Sn layer is the chemical reaction between the Sn and Cu to form the Cu_6Sn_5 intermetallic compound (IMC). In Sn coatings on Cu, IMC formation occurs primarily on the Sn side of the Cu-Sn interface due to rapid diffusion of Cu into Sn.[21] Because of this diffusional asymmetry, the IMC growth is accompanied by a large volume expansion that generates stress in the Sn layer. In previous work,[8,22] we have used finite element analysis (FEA) to simulate the evolution of the resulting stress field throughout layers of Sn with columnar grain structures assuming that stress relaxation can occur by elastic and plastic deformation and by grain boundary diffusion. Two important results of this work are the average stress saturates in the Sn (at approximately –12 MPa) due to the onset of plastic deformation; and the stress is distributed throughout the Sn layer due to the stress-driven diffusion of Sn along the grain boundaries. Without rapid grain boundary diffusion, the stress would remain much more localized near the growing IMC particles.

We extended our FEA model of stress evolution to include whisker growth by assuming that a whisker forms at a grain that is "weak." By weak we mean that this grain has a stress relaxation mechanism that becomes active at a lower stress than its neighbors (not a lower elastic modulus). As the IMC continues to expand and create stress in the Sn, the stress in the weak grain remains lower than the surrounding material which leads to a persistent stress gradient. This gradient drives diffusion toward the whisker base so that the whiskering grain is continually fed material which can then be incorporated into it and moved out of the film. The FEA work showed that measured values of the grain boundary diffusivity[15,23] and the IMC growth rate[3] are sufficient to explain observed whisker growth rates and therefore mechanisms of anomalously fast diffusion are not required.

There are multiple reasons why a grain may plastically deform at lower stress than its neighbors (i.e., be "weak"). Smetana[19] has proposed that whiskers grow where there are horizontal grain boundaries (HGB), (i.e., grain boundaries with a component parallel to the surface of the film). Addition of material at the grain boundary results in an upward force that can cause the whiskering grain to slide out of the region in which it is forming. The addition of extra planes at the interface can also be thought of in terms of the nucleation of dislocation loops in the grain boundary which grow by diffusion-controlled climb, a non-conservative process that adds material to the growing whisker. In support of this picture, many cross-sections of whiskers show the presence of horizontally-inclined grain boundaries near the base of the whiskers. The grain boundaries may be created during the plating process or could be the result of recrystallization. Similarly, Vianco and Rejent[20] have proposed the importance of dynamic recrystallization (DRX) in the formation of whiskers. They suggest that a new recrystallized grain nucleates to lower the strain energy created by dislocations in the existing film. The recrystallization process creates additional grain boundaries that have a component parallel to the surface of the film. Incorporation of material into the strain-free growing grain at these boundaries transports material out of the underlying coating and into the whisker.

We refer to the HGB and DRX mechanisms as grain-growth based mechanisms for whisker formation. They have in common that additional planes of atoms are added to the growing whisker at the interface between the growing grain and the surrounding material (a schematic of which is shown in Figure 8a). This generates an upward force on the grain to push it out of the film which may occur by grain boundary sliding.[19]

Whisker growth can also be explained by an extrusion-based mechanism if the whisker grain undergoes plastic shearing at a lower stress than the surrounding grains. The anisotropic plastic flow stress of Sn could give rise to such a strength contrast for a grain with anomalous orientation relative to the preferred crystallographic orientation of the film. In an extrusion-based mechanism, plastic flow within the whisker grain due to dislocation glide can cause extension of the whisker and transport material out of the film. This mechanism is analogous to the process of forming material with a die,[24] or squeezing material from a toothpaste tube.[20] This mechanism does not require the presence of horizontal grain boundaries which may explain how whiskers can grow without HGBs, as has been observed experimentally.[25] As the grain deforms, the adjacent grain boundaries remain at the yield stress. This induces a stress gradient which drives long-range diffusion to the grain along the grain boundary network. As material arrives it is incorporated into the deforming grain along the vertical grain boundaries, thus providing a continual source of new volume to replenish the volume removed by the growth of the whisker from the surface of the film. The flow of material within the deforming grain is represented schematically by the block arrows in Figure 8b. In this mechanism, the whisker is the same size as the deforming grain.

A key feature common to all of the mechanisms of whisker/hillock growth proposed above is the presence of a weak grain, which can relax stress more effectively than the surrounding grains. As a result, normal stress across the grain boundaries adjacent to the weak grain remains lower than the normal stress across more distant vertical grain boundaries within the Sn film. This sustains a steady-state, non-diminishing stress gradient that causes material to be continually transported from surrounding grains to the whiskering grain via long-range stress-driven diffusion.[26] Growth of a surface feature (whisker or hillock) from this grain occurs because the grain deforms so that it can accom-

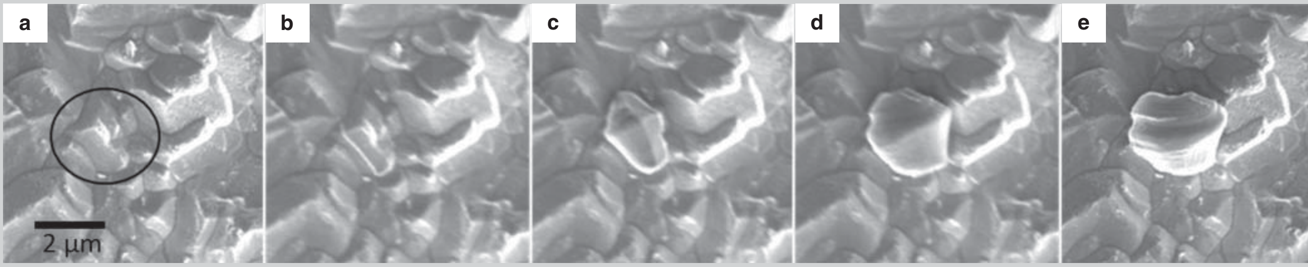

Figure 7: Time series SEM images of hillock growth showing extensive rotation and little vertical growth. Time after deposition: (a) 6 h, (b) 8 h, (c) 12 h, (d) 18 h, (e) 50 h. View on-line to access movie, or go to *http://www.engin.brown.edu/faculty/Chason/research/Hillock5.html*.

modate the material added to it at the grain boundaries. The stress-induced deformation may occur by dislocation-mediated glide or by grain-growth processes with grain boundary sliding. Most likely both of these mechanisms are active and work together to produce the complicated evolution seen on the surface. As pointed out previously,[9,20,27] dislocation-mediated plastic deformation is probably not the only mechanism because many whiskers grow in directions that are not aligned along the slip systems and they can also change directions (form kinks) after a period of growth. However, the complex morphologies and rotation that we observe in hillocks would be difficult to describe by pure grain growth without plastic deformation occurring as well.

To understand our current work, it is not necessary to distinguish among these different deformation mechanisms since each can produce a flow of material into whiskers and hillocks that can be spatially inhomogeneous.

Pure Whisker Growth

In this case, exemplified by the growth in Figure 1, material that goes into the whisker appears to come from deformation of a single grain. There is no lateral grain growth and the velocity field is uniform across the whisker so that it grows in a constant direction (see Figure 8a and b for schematic illustration). The lack of lateral grain growth may coincide with our observation that the whiskering grain is often smaller than those surrounding it which would suppress its tendency to consume its neighbors. Note that we do not observe any change in the region of the whisker before it nucleates, suggesting that the whiskering grain did not form by recrystallization though this is not certain; nucleation below the surface may not have been visible.

The tilt of the whisker relative to the surface may come about from the orientation of the grain boundaries feeding material into it (grain-growth mechanism) or the active slip systems in the grain (extrusion mechanism). In vacuum, where our measurements were made, the whiskers grow at a constant rate with little kinking, suggesting that the flow of material to the whisker stays uniform and there is little driving force to change the orientation. In other cases where kinks do form, this may be due to a re-orientation of the underlying grain boundaries or it may indicate a retarding effect due to formation of oxide at the surface. It is also possible that an alternate slip system has been activated or that the underlying grain has been rotated by formation of subgrain boundaries as seen in TEM.[6]

Surface Rotation

During hillock formation, we often see the surface of the growing feature rotate significantly (180° rotation in Figure 7), indicating that the rate of volume accumulation on one side of the hillock must be faster than on the other (schematic in Figure 9). Rotation of the crystal planes in the hillock suggests that significant numbers of dislocations are being injected into the material as it grows to change the growth direction. If the feature is being produced by the extrusion process, then the rotation may indicate non-uniformity in the stress surrounding the grain. Alternatively, if the feature is growing by a grain-growth process, the rotation may occur due to a reorientation of the underlying grain boundaries feeding material at the base, much as changing the direction of the nozzle from a hose can change the direction of the spray of water. Unfortunately, we cannot directly observe subsurface grain boundary changes with the SEM. However, in several cases (Figures 4 and 5), we observe that the hillock surface rotates by 90° in the early stages of growth and then grows out in the vertical direction. This suggests that the rotation may occur due to subsurface motion of the grain boundary which eventually becomes fixed and therefore leads to constant vertical growth.

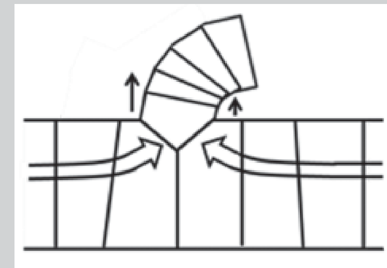

Figure 9. Schematic diagram showing how the hillock can curve due to unequal rate of growth across whisker cross-section. A similar mechanism may occur in extrusion-based deformation.

Hillock Formation

The morphology of hillock features is much more irregular than whiskers because the shape of the extruding region can change during their growth. This occurs because the vertical growth of the hillock is generally accompanied by lateral grain growth. The decrease of the strain energy density in the whiskering grain (either due to recrystallization or other forms of stress relaxation) lowers its chemical potential so that the whiskering grain may expand by consuming its neighbors. There is therefore a dynamic competition between vertical growth and lateral growth of the growing grain (shown schematically in Figure 10) that leads to an alter-

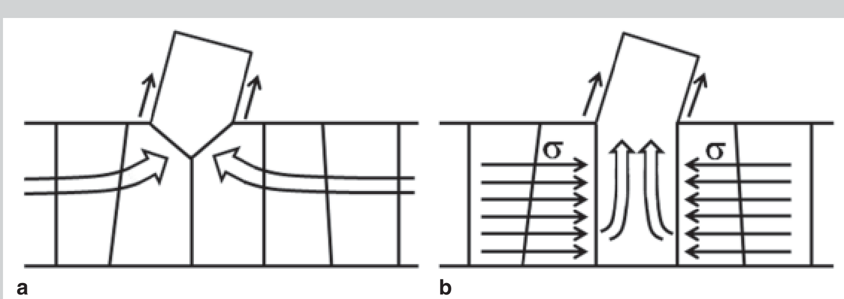

Figure 8. Two proposed mechanisms for whisker growth. (a) In grain-growth-based mechanism atoms are added to non-vertical grain boundaries at whisker base. The broad arrows indicate long-range diffusion along the grain boundary network that transports material to whisker grain. (b) In extrusion-based mechanism the whisker grain has a lower yield stress than the surrounding grains. Plastic shearing within the whisker grain carries material out of the plane of the film while also maintaining low biaxial stress within the whisker. The resulting stress gradient surrounding the whisker drives the transport of new material at the whisker via grain boundary diffusion has plastic deformation induced by stress field surrounding the whisker, shown by horizontal arrows; long range diffusion is essential to maintain local stress. The vertical broad arrows show the extrusion of Sn atoms.

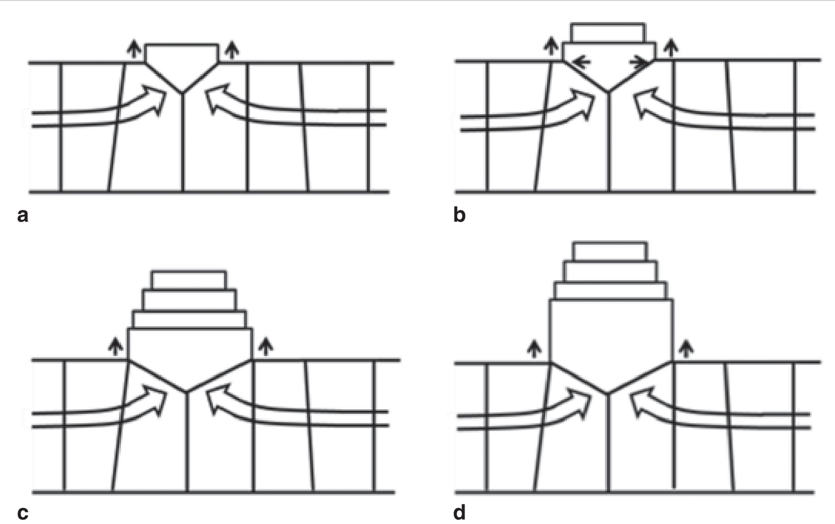

Figure 10. The formation of hillock with lateral grain growth: (a) vertical growth followed by (b) lateral growth resulting in the "wedding cake" morphology. (c) Lateral growth comes to an end when the grain boundary gets pinned. (d) After pinning the hillock may continue to grow in an upward direction. A similar mechanism may occur in extrusion-based deformation.

nation between horizontal and vertical growth, resulting in a "wedding cake" morphology (also described by Pedigo et al.[28]). Rapid lateral grain growth along the boundaries between columnar Sn grains leads to the ridges on the side of the hillock as has also been pointed out previously.[28] The sequence of alternation between vertical and lateral growth can vary at different sites. In some cases we observe extensive lateral growth followed by primarily vertical growth (Figure 4) and in other cases the opposite sequence (Figure 6). Therefore, we do not think that there is a prescribed sequence of grain growth and lateral growth; the actual morphology depends upon a balance between the different processes determined by the local microstructure and stress fields.

Unlike whiskers which always seem to start from a single grain, hillock growth can start from a variety of configurations. In some cases, the hillock originates from a single grain (as in Figure 4) with no apparent change in the surface or grain structure before it starts to grow. In other cases (Figure 5) we have seen the hillock form out of only part of a grain, suggesting that there was likely recrystallization of a new grain below the surface before the growth started. In other cases (Figure 6), several grains appear to have grown together before the hillock starts to grow.

CONCLUSION

We have measured the evolution of whiskers and hillocks on Sn coatings over Cu. Whiskers grow outward from a single grain while the more complicated morphologies of hillocks can be attributed to a balance between outward expansion and lateral growth into the surrounding grains. Our results are consistent with a picture in which whiskers and hillocks initiate at certain "weak" grains that can activate stress relieving mechanisms at lower values of stress than their neighbors; such mechanisms occur by adding atoms to the base of the grain (grain-growth based) or initiating glide processes (extrusion-based). The hillock shape is difficult to predict because of its reliance on the details of the underlying flow field of material into the whisker. Additional modeling work is needed to understand how factors such as the spatial distribution of the stress and the accommodation rate on different surfaces of the underlying grain can alter the morphology of the growing feature. In terms of mitigation, these results suggest that the best strategy would be to develop microstructures or alloys that better relax stress without the formation of surface features. If the stress can't be removed, then better understanding of the nucleation process may allow the development of microstructures that promote hillock formation over the long whiskers.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge the support of the NSF-supported Brown MRSEC (DMR0079964), NSF (DMR0856229) and help from L.B. Freund, S. Kumar, and G. Barr.

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