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The Effect of Amorphous Surface Layers on Images of Crystals in High Resolution Transmission Electron Microscopy

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1. ABSTRACT

The effect of a surface layer of amorphous silicon dioxide on images of crystalline silicon has been investigated for a range of crystal thicknesses varying from zero to $2\frac{1}{2}$ times that of the surface layer. It is shown that an amorphous surface produces fluctuations in image contrast which introduces difficulties in the interpretation of defects in very thin specimens. These difficulties are less pronounced but still present in thicker crystals. It is also shown that an edge smoothly approaching zero thickness produces an image that changes gradually from crystalline to amorphous character.

2. INTRODUCTION

It is well known that most materials under investigation in the electron microscope form a surface oxide layer or an amorphous layer which can affect the resulting image, Fig. 1 and Fig. 2. In fact, a noisy image is often attributed to surface effects without further explanation. Furthermore, very little work has been done to evaluate the extent to which an amorphous surface actually influences the nature of the image.

Krakow [1] calculated the image of crystalline gold showing reasonable agreement with an experimental image when the top gold layer

was substituted with a layer of randomly arranged gold atoms. However, when the same substitution was made for the bottom layer the computed image lost its crystalline appearance completely. In another study, Anstis et. al. [2] calculated the effect of an amorphous surface layer on the images of a 30 degree partial dislocation in silicon. They showed that the amorphous surface makes it impossible to distinguish between the shuffle and glide models of the dislocation core in a 28 Å thick crystal when the surface layer was 20 Å thick. Similar results have been quoted by Bourret et. al. [3].

The object of the present research program is to fully understand the contribution of such surface layers to high resolution image contrast, examining in particular the effect of amorphous layer thickness relative to the thickness of the crystal on which it resides. The problem of specifying suitable atomic coordinates in an amorphous material is addressed by choice of a model system, amorphous silicon dioxide on silicon, for which atomic coordinate data exists [4]. Calculations are carried out which simulate high resolution electron micrographs of a single crystal of silicon and then a $\Sigma=9$ bicrystal of silicon, the latter having a fully specified periodic defect structure [5]. Atom coordinates for the $\Sigma=9$ grain boundary were obtained by relaxation of a coincidence Site Lattice [6]. Images of crystals with different thicknesses are simulated for a constant thickness amorphous oxide film and compared to the case where no oxide is present.

3. IMAGE CALCULATION PROCEDURE

The images presented in this work were simulated by a multislice computation routine [7] written to run on a CDC 7600 such that any

number of different slices and slice thicknesses could be used. In addition, any number of sampling points up to 256^2 can be used and these can be arranged in any square or rectangular array. All adjustable microscope parameters were set to values characteristic of a JEOL JEM 200CX at Scherzer defocus; $C_s = 1.2\text{mm}$, $\Delta f = 660\text{\AA}$, $\delta = 50\text{\AA}$, $\alpha = 0.5\text{mrad}$.

Atomic coordinate data for the amorphous silicon dioxide layers was taken from the work of Bell and Dean [4] who used a random network theory model with a mean Si-O-Si bond angle of 153 degrees. They generated the positions of 614 atoms within a volume element of $31 \times 23 \times 22$ cubic $\text{\AA}$ Angstroms; this model closely matched the experimental radial distribution function to 8 $\text{\AA}$ detail. These atom positions were used in the calculations by dividing the above volume element into 7 slices of 3.14 $\text{\AA}$ thickness each, and using only the middle 5 slices for a total layer thickness of 15.7 $\text{\AA}$.

An oxide film was then hypothetically attached to a single crystal of silicon in [110] orientation by covering an area of 5×5 crystalline unit cells (Fig. 3). This required the selection of a rectangular slab of oxide with dimensions 27.1 $\text{\AA}$ by 19.2 $\text{\AA}$ which retained the same thickness of 15.7 $\text{\AA}$. Oxide films on the top and bottom surfaces were positioned such that they did not artificially superimpose; this was accomplished by simple translation of the bottom layer by $a/2[111]$ with respect to the top layer.

The potential from each layer of material in this sandwich was then calculated from its corresponding electron scattering factors out to a maximum reciprocal space dimension of $3.0 \text{\AA}^{-1}$. This corresponds

to an array of 182×114 sampling points such that every fifth sampling point along any direction was a Bragg reflection from the silicon matrix.

The oxide film attachment to the silicon bicrystal was accomplished in a slightly different way. The model of the silicon bicrystal shown in Fig. 4 covers an area larger than the size of the oxide such that a rectangular slab with oxide in the center had to be used. Two surface oxide layers were created, one $15.7 \text{ \AA}$ thick and another $9.4 \text{ \AA}$ thick. With the $9.4 \text{ \AA}$ thick surface oxide, unique top and bottom surface layers were constructed from the model of the oxide by simply using different slices for the two layers, whereas in the case of the $15.7 \text{ \AA}$ thick oxide the top and the bottom layers were rotationally displaced 180 degrees. Calculations of the resulting sandwiches were performed using 256×128 sampling points.

4. RESULTS

The results of the calculations are shown in Figs. 5 through 9. In Fig. 5 a set of computed images of the amorphous silicon dioxide is shown together with the corresponding projected potentials. Figure 6(a) shows a set of images calculated for perfect silicon with thickness varying from 0 to $38.4 \text{ \AA}$ and a constant total thickness of amorphous top and bottom surface layers of $31.4 \text{ \AA}$. The image of perfect crystal with no surface oxide varies negligibly with thickness in the range $8 - 38 \text{ \AA}$ and is shown in Fig. 6(b) for comparison.

Figure 7 shows the projected potential for the silicon bicrystal, while Fig. 8 and Fig. 9 show computed images. The $\Sigma=9$ grain boundary is a pure tilt-boundary with a rotation angle between the two grains of 38.9 degree. The boundary plane is (122) . Figure 4 shows a schematic

model of the unit cell used in the calculation. The thickness of the silicon varies from 8 - 38 Å and the total thickness of amorphous top and bottom surface layers is 31.4 Å and 18.8 Å respectively. These images should be compared to the set of images calculated for the $\Sigma=9$ silicon boundary without surface oxide, shown in Fig. 8. The rotational displacement used in the construction of the bottom surface oxide layer 15.7 Å thick produces an artificial mirror-symmetry when the top and bottom layer is viewed in projection, which can be noticed when the bicrystal is very thin (7.7 Å), but otherwise has no effect on the results. Because of the smaller size of the oxide, only the center portion of the images was considered.

5. DISCUSSION

i) Perfect Crystal

It is clear from Fig. 6(a) that the image changes from amorphous to crystalline in a gradual way. In fact, in the thin region of silicon, 8-16 Å, parts of the image appear amorphous, while another part appears more crystalline. Thus "islands" of crystalline material appearing in an apparently amorphous area near the edge of a foil could possibly be due to variations in the surface oxide alone. For "thicker" areas, the surface layers produce irregularities in the image, such as wavy lines of atoms and fluctuating contrast. As the thickness of the silicon increases beyond the total thickness of the surface oxide, the image is very closely that of a pure crystal with no oxide. In the case of silicon this corresponds experimentally [8] to a thickness of approx. 40 Å, see Fig. 2.

ii) Silicon Bicrystal

In this case, the most interesting result is the appearance of displaced atoms at the boundary plane which has significant implications for the general analyses of atomic positions near any such defect. Various models are usually possible for any given type of defect and the goal of HREM is to determine the correct model by matching computed images to experimental data. Sometimes the change in the image from model to model is subtle and "noise" in the image can make it impossible to distinguish between two models with any confidence. Unfortunately the presence of a surface layer will add to the difficulty of determining atom positions. As can be seen from Fig. 8, atoms may or may not show up due to surface contamination. The model of the $\Sigma=9$ boundary has a periodicity of $11.5 \text{ \AA}$ in the boundary direction, and there are two "unit cells" shown in Fig. 4. However, due to the presence of the surface oxide, the image does not reflect this periodicity and certain atoms could easily be thought missing, thus preventing a correct interpretation of the image. Naturally as the crystal gets thicker, the surface becomes less important, and for thicknesses slightly greater than the total surface-thickness, the surface effects have become negligible except at the dislocation cores where there still are small differences in contrast.

6. CONCLUSION

Because of dynamical interaction between Bragg scattered reflections HREM images is usually considered directly interpretable only for very thin specimens, typically less than $50 \text{ \AA}$. For thicker crystals computer matching of images is required to get detailed information

down to 2-4 Å resolution, but inelastic scattering may make even this approach unreliable.

In the presence of an amorphous surface layer, it is clear that the image of a very thin specimen is adversely affected by the surface. This may not represent a severe problem in imaging a perfectly crystalline structure, since one can always average over a number of identical unit cells to obtain a less noisy image. However the correct interpretation of a single isolated defect may be rendered impossible by the presence of an amorphous surface layer. With such surface layers of the order of 20 Å thickness, the crystalline material should be at least 40 Å thick to provide an interpretable image. It should also be noted that a uniform wedge-shaped edge produces an image that gradually changes from crystalline to amorphous character. A sharp change from crystalline to amorphous character indicates an edge that is approaching zero thickness in a discontinuous way.

ACKNOWLEDGEMENTS

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FIGURE CAPTIONS

1. High resolution electron micrograph of silicon in [001] orientation showing transition from crystalline to amorphous structure at a thin edge.
2. Native oxide on 3 degrees off (111) Si surface, thickness $20 \pm 3 \text{ \AA}$.
3. Projected unit cell of silicon in [110] orientation.
4. Model of $\Sigma=9$ grain boundary in silicon. The model shows the structural periodicity along the boundary plane.
5. Computed electron micrographs of amorphous silicon dioxide. The projected potentials are shown for comparison.
- 6a. Computed electron micrographs of perfect silicon with native oxide. The thickness of amorphous top and bottom surface oxide is $31.4 \text{ \AA}$. The thickness of the silicon matrix varies from 0. to $38.4 \text{ \AA}$.
- 6b. Computed electron micrograph of perfect silicon, thickness $23 \text{ \AA}$.
7. Computed projected potential for silicon bicrystal from model in Fig. 4.
8. Computed electron micrographs of silicon bicrystal with native surface oxide. The total thickness of the surface layers is held constant at $31.4 \text{ \AA}$, while the thickness of the bicrystal varies from 8 to $38 \text{ \AA}$.
9. Computed electron micrographs of silicon bicrystal with native surface oxide. The total thickness of the surface layers is held constant at $18.8 \text{ \AA}$, while the thickness of the bicrystal varies from 8 to $38 \text{ \AA}$.

VACUUM

AMORPHOUS

CRYSTAL

Fig. 1

XBB 840-8178

Fig. 2

XBB 834-3022

Si 110

XBL 8410-4105

Fig. 3

$\Sigma=9$

XBL 8410-4106

Fig. 4

AMORPHOUS SILICON DIOXIDE

Roar Kilaas

XBB 847-5399A

Fig. 5

SILICON WITH SURFACE OXIDE

XBB 840-8177

Fig. 6a

Fig. 6b

XBB 840-7433A

$\Sigma=9$

XBB 840-7431

Projected Potential

Fig. 7

**SILICON BOUNDARY
WITH SURFACE OXIDE**

W/OUT SURFACE OXIDE

7.7 Å

15.3 Å

23.0 Å

30.6 Å

38.4 Å

XBB 840-7432A

Fig. 8

**SILICON BOUNDARY WITH
SURFACE OXIDE (18.8Å)**

7.7 Å

15.3 Å

23.0 Å

30.6 Å

38.4 Å

XBB 840-7593A

Fig. 9

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