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Quantitative high resolution electron microscopy image matching applied to the strontium titanate $\Sigma 3(112)$ grain boundary

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Abstract. Quantitative image matching has been employed in order to compare experimentally observed grain boundary structure to predicted model structures for the strontium titanate (SrTiO$_3$) $\Sigma 3(112)$ grain boundary. Initially, quantitative image matching of image regions containing bulk crystal were used to determine the specimen and microscope parameters, including specimen thickness and tilt, and image defocus. Using the fitted values for thickness and defocus, images have been simulated from two proposed model structures of the SrTiO$_3$ $\Sigma 3(112)$ grain boundary and quantitatively matched to experimental images.

1. Introduction
High resolution electron microscopy is an established tool for characterization of defects in crystalline materials [1]. However, due to the complex electron–specimen interactions, image interpretation is not always intuitive. With the onset of aberration correction, image interpretation has become more straight–forward in many cases; however even with aberration correction, the image pattern observed near defect regions can be complicated and therefore image simulation may be necessary. Further, in order to provide reliable structural information for comparison with model structures, quantitative image comparison is often required. In this paper quantitative image matching has been employed in order to differentiate between two predicted model structures. Using parameters established from bulk crystal images, it has been possible to quantitatively assess the match between simulated images from two model structures of the SrTiO$_3$ $\Sigma 3(112)$ grain boundary in order to determine the structure which agrees best with experiment.

2. Experimental Methods
A $10 \times 10 \times 1$ mm piece of bulk SrTiO$_3$ $\Sigma 3(112)$ bicrystal material, which had been prepared by diffusion bonding, was obtained from a commercial source. The bulk material was heat treated as described in reference [2] and TEM specimens were prepared by using the methods described by Strecker et al [3]. Experimental data was collected using a JEOL 2200 MCO microscope operating at 200 kV. The microscope is fitted with CEOS aberration correctors [4] for both the image and probe forming optics. The third-order spherical aberration ($C_3$) was set to optimal conditions to balance fifth-order spherical aberration, $C_3 = -6 \mu m$ [5]. In certain circumstances, negative values of $C_3$ have the additional benefit of improving contrast of light elements such as...
oxygen in a multicomponent system such as SrTiO$_3$[6]. Twenty member focal series data sets were collected and three images from a focal series were selected for quantitative image matching first of the bulk crystal and then of the grain boundary.

3. Bulk Crystal Fitting

In order to determine specimen and imaging parameters, crystalline sub–regions of the experimental image were compared to a series of images of a bulk crystal model simulated using the multislice method [7] implemented in the QSTEM package [8]. Following the definitions of Hýtch and Stobbs [9], the mean squared difference between images $f$ and $g$ can be described as the sum of differences due to intensity, contrast and pattern mismatch between the two images. The pattern mismatch between $f$ and $g$ can be described by the cross–mean $C_{fg}$:

$$C_{fg} = \frac{\bar{f}g - \bar{f}\bar{g}}{\sigma_f\sigma_g}$$  \hspace{1cm} (1)

where $\bar{f}$ and $\sigma_f^2$ are the mean and standard deviation of pixel intensities in the image (for a complete derivation of equation 1 see reference [9]). In the case when the pattern of the two images are similar, $C_{fg}$ will be close to unity and in this case it is useful to define the angle of the cross–mean, $\theta_\rho$ [10]:

$$\theta_\rho = \cos^{-1}(C_{fg})$$  \hspace{1cm} (2)

In this case, the $\theta_\rho$ value will approach zero as image similarity increases. For this work, the $\theta_\rho$ value was used as a comparison metric because it is directly related to the pattern mismatch between the two images (for alternative metrics see references [11, 12]). The $\theta_\rho$ criterion has the additional benefit that it increases linearly in proportion to the difference between two images [10]. The difference could be between the defocus, thickness, crystal tilt, etc. of the two images.

Parallel MATLAB code was written to carry out the matching over a wide range of fitting values using sub–regions of the experimental images from bulk crystal in the grains on either side of the boundary. Following the procedure outlined in references [9, 13], the fitting process was broken down into four steps: defocus–thickness, crystal tilt, beam tilt and astigmatism fitting. The Parallel MATLAB code was run on a 64–node cluster within the Oxford e–Research Centre.

In the case of beam tilt, the best–fit beam tilt had magnitude of zero in all cases. This is likely to be due to the fact that beam tilt could be compensated slightly by the crystal tilt. Figure 1 presents the experimental sub–regions from three different images along with simulated images with the best–fit defocus and thickness values, while table 1 shows the best–fit simulation parameters for each image along with corresponding $\theta_\rho$ values. In the case of the defocus–thickness fitting, the code determined the best–fit thickness using all three images simultaneously, which effectively forces the best–fit thickness to be constant for all images. For all three defocus values, the change to the pattern of the simulated image due to incorporating crystal tilt and astigmatism was small and therefore these effects were not included in the simulations shown in figure 1 or for subsequent grain boundary simulations. In addition, because the model structures were produced from density functional theory relaxation [14], it was not possible to incorporate the best–fit crystal tilt values.

4. Grain Boundary Model Comparison

Using the best–fit simulation parameters determined from the bulk crystal fitting, images were simulated from two model structures proposed in the literature [14]. In one configuration the grain boundary was mirror symmetric with no translation, while the other had mirror–glide symmetry (see reference [14] for details). Figure 2 shows the experimental grain boundary images
Table 1. Best–fit thickness, defocus, crystal tilt and astigmatism values, with corresponding $\theta_\rho$ values, for three images from the focal series. In all cases the best–fit beam tilt had a magnitude of zero.

| Thickness (Å) | Defocus (Å) | Tilt About ⟨100⟩ (mrad) | Tilt About ⟨110⟩ (mrad) | Astigmatism Magnitude (Å) | Astigmatism Azimuth (°) | $\theta_\rho$ (rad) |
|--------------|-------------|--------------------------|--------------------------|---------------------------|-----------------------|-------------------|
| ±10 Å        | ±10 Å       | ±2 mrad                  | ±2 mrad                  | ±10 Å                     | ±10°                  |                  |
| 25           | 120         | 2                        | 5                        | 20                        | 130                   | 0.557             |
|              | 190         | 1                        | 5                        | 40                        | 130                   | 0.377             |
|              | 330         | 1                        | 5                        | 20                        | 80                    | 0.627             |

Figure 1. Experimental sub–regions taken from one side of the grain boundary with corresponding best–fit simulations. Simulated images shown in a–c have defocus values of 110, 120 and 330 Å, respectively, and thickness of 25 Å.

along with simulated images from the two model structures, with corresponding $\theta_\rho$ values shown below each of the simulated images. From both visual inspection and quantitative comparison of the images, the mirror symmetric model is found to be a relatively better fit to the experimental data. This result was confirmed by comparison with several different segments of the grain boundary (including data sets taken from several different thin regions of the specimen).

5. Conclusions
We have used quantitative image matching including eight fitting parameters to fit imaging parameters for images of bulk crystal near a defect region. Using the fitted parameters, we have simulated images from two proposed model structures of the SrTiO$_3$ $\Sigma3$ (112) grain boundary. Both quantitative and qualitative analysis have shown that the mirror symmetric configuration is the best–fit to the experimentally observed grain boundary structure. Quantitative defocus–thickness matching is also an effective method for confirming the specimen thickness in order to determine if advanced techniques such as focal series reconstruction can be applied [15, 16]. In the case of the thickness value fitted here, the images would be suitable for focal series reconstruction, which could be used for further analysis [2].

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Figure 2. Experimental GB images (centre column), and simulated images from the mirror symmetric model structure (left column) and mirror–glide symmetric model structure (right column). Rows a–c correspond to defocus values 120, 190 and 330 Å, respectively. \( \theta_p \) values corresponding to the model structures are shown below the image for each defocus value. All images are simulated with thickness of 25 Å.

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