

High-Resolution HAADF STEM of Inversion Boundaries in Sb₂O₃-Doped Zinc Oxide

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In order to confirm the structure of inversion boundaries in Sb₂O₃-doped ZnO that has been investigated by HRTEM [1], we perform high-resolution high-angle annular dark field (HAADF) scanning transmission electron microscopy (STEM) with the aid of image simulation. The simulations are carried out using a program written by means of a new scheme we have developed [2]. The program code includes calculations for Bragg scattering and thermal diffuse scattering, based on Bethe method, providing fast ADF STEM image simulation due to its very efficient algorithm.

Fig. 1 indicates some examples of simulated images for an experimental [01 $\bar{1}$ 0] ZnO image shown in Fig. 2a, which was recorded using a 20-mrad-semiangle probe and a 100~200-mrad detector in a JEM 2010F-TEM/STEM ($C_s=0.5$ mm) operated at 200 kV. The simulation was performed by varying thickness t by an interval of 5 nm from 10 to 50 nm and defocus Df by an interval of 2nm from -25 to -45 nm. The computation time of an image is about 9.7 min on a personal computer with CPU of Intel Pentium 4 (2001) 2 GHz/400 MHz System Bus, 256 KB Cache, and additional calculation of an image at different thickness and focus takes only 11 s. Bright spots in these images exhibit the positions of Zn atomic columns, while no bright contrast is seen along O atomic columns. Fig. 2b is an enlarged image of the area surrounded by a rectangle in Fig. 2a. A processed image by the image smoothing method is shown in Fig. 2c. The processed image was quantitatively compared with the simulated images using the chi-square method, and the most probable thickness and defocus were evaluated to be $t=20$ nm and $Df=-27$ nm. The simulated image using these values is shown in Fig. 2d. A processed or smoothed image of the inversion boundary (IB) in Fig. 2a is shown in Fig. 3a. We simulated IB images using the parameters ($t=20$ nm and $Df=-27$ nm) that we determined for the matrix. A calculated image using an IB structure model proposed by Recnik *et al.*[1] is shown in Fig. 3b. The contrast in the images in Figs 3a and 3b was normalized so as to compare the two images. The intensity profiles along Zn-Zn-Zn lines and Zn-Sb-Zn lines in these images are shown in Fig. 4. Similar experiments for an IB in Zn(Sb)O in [2 $\bar{1}$ 10] orientation are shown in Figs 5 and 6.

From these HAADF STEM experiments, we have confirmed that the boundary plane itself consists of a highly ordered SbZn₂ monolayer [1]. Some disagreement between the experiment and simulation may be ascribed to instrumental and technical problem in field-emission HAADF microscopes of today.

References

- [1] A. Recnik *et al.*, J. Am. Ceram. Soc., **84** (2001) 2657.
- [2] K. Watanabe *et al.*, Phys. Rev. B, **64** (2001) 115432.

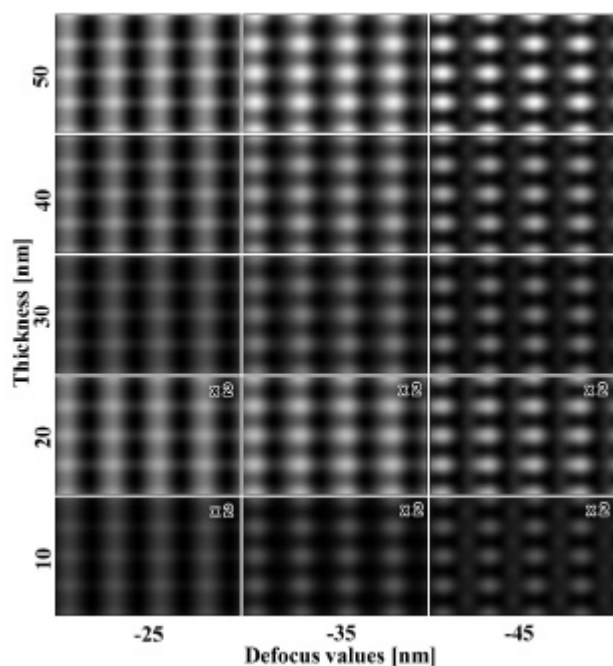


FIG. 1 Simulated HAADF STEM images of $[01\bar{1}0]$ ZnO. Total CPU time for these 15 images is 12 min. ($\times 2$: Intensity magnification)

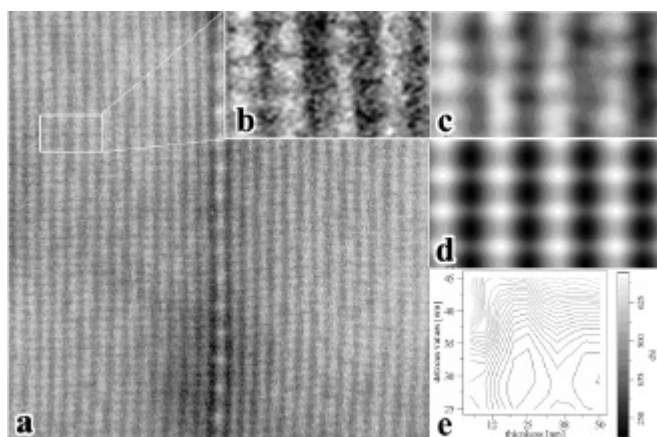


FIG. 2 **a** Experimental HAADF STEM image of $[01\bar{1}0]$ Zn(Sb)O. **b** Enlarged image of matrix area in (a). **c** Processed image of (b). **d** Simulated image for (b). **e** \mathcal{C}^2 of simulated images to (c). From (e), the defocus and thickness of (a) were evaluated to be $t=20$ nm and $D_f=-27$ nm.

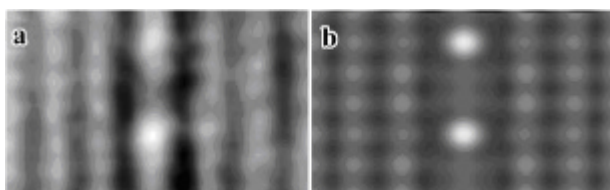


FIG. 3 **a** Processed HAADF STEM image. **b** Simulated image of IB in $[01\bar{1}0]$ Zn(Sb)O. In this projection, Sb columns exhibit strong bright spots on IB. The intensity of Zn columns on IB is weaker than that of Zn columns in matrix (see Fig. 4).

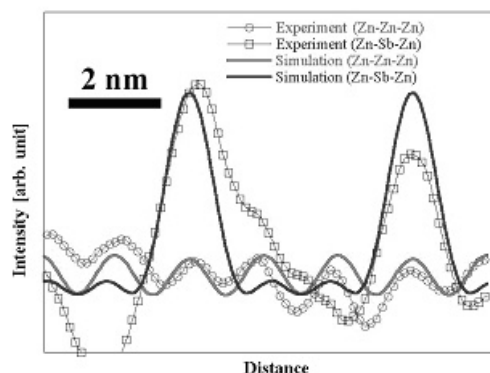


FIG. 4 Comparison between intensity line profiles from the images in Fig. 3.

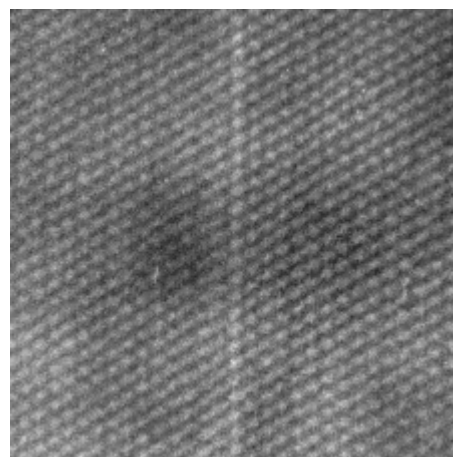


FIG. 5 Experimental HAADF STEM image of IB in $[2\bar{1}\bar{1}0]$ Zn(Sb)O. The boundary can be distinguished as a line of dots with higher intensity (presence of Sb atoms). In this projection, IB columns contain Sb:Zn = 1:2.

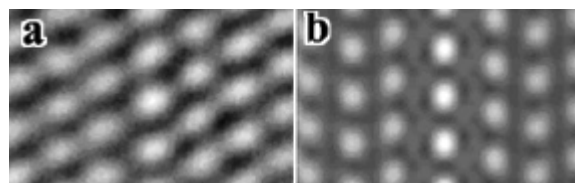


FIG. 6 **a** Processed HAADF STEM image. **b** Simulated image of IB in $[2\bar{1}\bar{1}0]$ orientation.