Comparison of the contrast in conventional and lattice resolved ADF STEM images of InGaAs/GaAs structures using different camera lengths

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Abstract. A procedure to quantify annular dark field (ADF) images in scanning transmission electron microscopy (STEM) has been applied to two 200kV transmission electron microscopes (TEMs), a JEOL 2010F and a double aberration-corrected JEOL 2200FSC. A series of ADF images is acquired as a function of the camera length (i.e. inner detection angle). Then the intensity ratio of InGaAs and GaAs is plotted vs. camera length and extrapolated to zero, at which point the contrast behaves exactly as predicted by Rutherford’s scattering. The linearity of ADF intensity ratio vs. camera length improves significantly by using the JEOL 2200FSC compared to the JEOL 2010F at medium resolution. A high-resolution ADF image at 2MX nominal magnification acquired in the JEOL 2200FSC shows the same linearity of intensity ratio vs. camera length, independent of whether the ratios of the average background intensities or the fringe amplitudes are used for the analysis. This is explained by both group III and group V atoms contributing to the {111} fringes observed, similar to low resolution data.

1. Introduction

Annular dark-field (ADF) scanning transmission electron microscopy (STEM) has been very successful because of the strong atomic number (Z) dependence. However, for crystalline specimens low angle Bragg beams complicate the contrast interpretation of the images. The image cannot be interpreted simply in terms of thickness and atomic number contrast. Therefore a high-angle annular detector (≥ 40 mrad) was proposed by Howie [1] with the aim to avoid the coherent Bragg scattering.

In ADF STEM the image intensity decreases while the contrast increases with increasing detection angle (decreasing camera length) [2]. For infinite scattering angle (camera length equal to zero), which cannot be achieved in any experiment directly, Rutherford scattering by the atomic nuclei is dominant and the intensity is proportional to the square of the atomic number (Z²) of the atoms at constant specimen thickness [3]. Therefore, a method was proposed to quantify the chemical composition in binary systems such as SiGe/Si, where only Si vs Ge atomic exchanges occur, by extrapolating the ADF image contrast as a function of camera length to zero [4]. The method was used in this work to evaluate the peak indium concentration in an annealed InGaAs/GaAs heterostructure, where only In vs Ga atomic exchanges occur.

2. Experimental

Eight layers of InAs/GaAs QDs covered by 5 nm InGaAs were deposited on a Si-doped GaAs (100) substrate using molecular beam epitaxy (MBE). These dot-in-a-well (DWELL) structures were
separated by 33 nm GaAs deposition. Then thermal treatment was applied to the sample in a N$_2$ ambient for 5 minutes at 750°C with the aim to minimize the compositional fluctuation in the DWELL structures. ADF STEM images were taken by using both the JEOL JEM 2010F TEM and the aberration-corrected JEOL JEM 2200FSC TEM while varying the camera length (i.e. inner detection angle) to investigate the peak In concentration in the DWELL structure.

For each acquisition, the detector gain was set at the initial stage of each experiment to ensure the highest intensity level (acquired at longest camera length) was just below detector saturation level and an associated dark current signal (intensity recorded with gun valve closed) was acquired as a reference (background intensity, which was subtracted before calculating the intensity ratios).

The JEM 2010F TEM is equipped with a Schottky-type field-emission gun (FEG) operating at 197 kV. In STEM mode, it normally provides an electron beam ~0.25 nm in size at ~9.5 mrad semi-angle of beam convergence with 0.5 mm spherical aberration constant.

The aberration-corrected JEM 2200FSC TEM is equipped with a FEG operating at 200 kV and two aberration correctors. The upper aberration corrector for similar STEM eliminates the opening error of the probe-forming condenser lens system. An electron probe with ~0.1 nm diameter can be formed and used to perform STEM. By applying a larger aperture, the electron beam can also be focused to a similar size provided by the JEM 2010F TEM, but with much more current in it. As a result the signal-to-noise ratio is improved.

3. Results

Figure 1 presents an ADF STEM image at 120 kX nominal magnification of the sample after annealing acquired in the JEOL JEM 2010F, with camera length decreasing from top to bottom during the acquisition. The bottom stripe was acquired while the gun valve was closed. The dark current signal, $I_0$, for each experiment was directly read from this kind of image stripe section using the Gatan Digital Micrograph software.

Figure 2 shows a high resolution ADF STEM image at 2 MX with the camera length decreasing from top to bottom during acquisition in the JEOL 2200FSC. Weak lattice fringes were resolved and the \{111\} lattice fringes are used to calculate the average intensity ratio ($R$) and the ratio of the fringe amplitudes ($Q$) in InGaAs and GaAs for each camera length.

![Figure 1. ADF STEM image at 120 kX magnification with camera length decreasing from top to bottom. The corresponding collection angle, $\beta$, from top to bottom are 18.0 mrad, 19.6 mrad, 21.3 mrad, 23.2 mrad, 25.2 mrad, 27.4 mrad, 29.8 mrad, 32.3 mrad, 35.1 mrad, 38.1 mrad, 41.4 mrad, and 45.2 mrad. The last stripe was acquired while the gun valve was closed.](image-url)
Figure 2. High resolution ADF STEM image at 2 MX magnification with the camera length decreasing from top to bottom during the acquisition in the JEOL JEM-2200FSC. The corresponding camera lengths for each stripe (collection angle, $\beta$) from top to bottom are 362 mm (20.7±0.8 mrad), 290 mm (26.2±0.9 mrad), 217 mm (39.3±1.2 mrad), 181 mm (47.1±1.6 mrad) and 145 mm (57.3±2.3 mrad). The last stripe was acquired while the gun valve was closed.

4. Discussion

The average intensity ratio ($R$) and the ratio of {111} fringe amplitudes of In$_x$Ga$_{1-x}$As to GaAs vs. camera length are plotted in figure 3. It shows that for the JEOL 2010F at the shortest camera lengths the intensity ratios calculated scattered dramatically, probably due to the poor signal-to-noise ratio. There is also a noted curvature around 250 mm camera length. This may relate to non-linearities in the electro-magnetic field of the inter-mediate lens used to alter the camera length [5]. Linearity has been improved significantly by using the JEOL 2200FSC at both medium and high resolution as the regressional coefficients now approach unity (cf. Table 1). That’s because the aberration corrected instrument is able to provide a smaller probe with higher current, giving a higher signal-to-noise ratio. For large camera lengths the ratio of {111} fringe intensity is generally low. It increases fast with decreasing camera length and finally yields a similar intercept for zero camera length. However, the relative compositional error evaluated by using fringe amplitude ratios is larger than that obtained by using an average intensity ratio, which again is due to the contribution of noise to the lattice fringes.

Assuming a Gaussian distribution of standard deviation $\sigma$ of the measurements, the 2$\sigma$-interval gives 95.4% confidence that the In peak concentration from the three separate measurements of $R$ is 0.291±0.062, 0.248±0.008 or 0.266±0.014 from the ADF images taken in the JEOL 2010F at 120 kX, the JEOL 2200FSC at 250 kX or the JEOL 2200FSC at 2MX, respectively. Consequently, the peak indium concentration in the In$_x$Ga$_{1-x}$As lies between 0.25 and 0.26.
Figure 3. Plots of the average intensity ratio ($R$) and the ratio of $\text{\{111\}}$ fringe amplitudes ($Q$) of InGaAs to GaAs vs. camera length.

Table 1. Results of linear regressional analysis of the plots in figure 3

| Condition | 2010F TEM | 2200 FSC TEM |
|-----------|-----------|--------------|
| nominal magnification | 120 kX | 250 kX | 2 MX |
| intensity ratio | average | average | average |
| fringe amplitude | $1.204\pm0.024$ | $1.174\pm0.003$ | $1.187\pm0.005$ | $1.202\pm0.093$ |
| intercept $b$ from lin. reg. fit | 0.752 | 0.998 | 0.995 | 0.935 |
| lin. regression coefficient | | | |
| $\text{In peak concentration, } x\pm\Delta x (\varepsilon=2)$ | $0.291\pm0.031$ | $0.248\pm0.004$ | $0.266\pm0.007$ | $0.288\pm0.132$ |
| relative error, $\Delta x/x$ | $10.7\%$ | $1.6\%$ | $2.6\%$ | $45.8\%$ |

5. Conclusion
Aberration corrected STEM improves linearity of the data, mainly due to improving the signal-to-noise ratio by forming a smaller probe with higher current compared to conventional STEM. This reduces the relative error of the evaluated chemical composition by using average intensity ratio significantly. This work also shows that the result can be confirmed by using the fringe amplitude but with a larger relative error due to the higher contribution of noise to the lattice fringes.

References
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