The role of rough surfaces in quantitative ADF imaging of gallium nitride-based materials

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Abstract. We have studied the surface of GaN after preparations commonly used to thin cross sectional samples to electron transparency with atomic force microscopy (AFM) and scanning transmission electron microscopy annular dark-field (STEM-ADF) imaging. Tripod polishing and ion milling leave the GaN surface in varying degrees of roughness and strain. By comparing the roughness seen in the AFM with the ‘roughness’ observed in STEM-ADF imaging, we show most nano-scale variations in ADF can be attributed to surface roughness alone. However, some systematic differences are seen for very thin specimens.

1. Introduction

Unlike its phase-contrast imaging counterpart, scanning TEM annular dark field imaging enjoys good agreement between simulations and experiments [1,2]. Nowhere is this quantitative assessment more important than for measuring the extent of atomic ordering where the brighter (or darker) contrast of the sample can be interpreted as precipitates, chemical ordering and point defects [2]. However, the preparation of a thin TEM specimen is a grievous process and the sample surface can be left with surface strains, rough surfaces, as well an accompanying amorphous layer, all of which introduce varying levels of point-to-point contrast that may obscure the features of interest.

We have sought to examine the influence of genuine surface roughness on the standard deviation of the ADF signal when measured in a fully quantitative way. We have measured the roughness of the GaN surface, in cross section, using the atomic force microscope (AFM). We proceed to use these preparations to produce a cross-sectioned sample for examination in the STEM. We compare the surface height and ADF standard deviations to assess the level of agreement.

2. Experimental details

All surface preparations studied were applied to a standard MOVPE-grown GaN film with a thickness of 8.5 μm on c-plane sapphire. Tripod polishing followed standard practice: Diamond impregnated mats with abrasive sizes 30, 15, 9, 6, 3, 1 and 0.1 μm were used with a Southbay Technology hand-held tripod polisher. Mechanical polishing was concluded with a Syton 50 nm silica suspension on a cloth mat. A Gatan Precision Ion Polishing System (PIPS) was used for ion milling at 5 keV with a beam inclination angle of 7° at room temperature.
A Veeco Dimension V atomic force microscope was used to measure the surface height profile of the prepared surfaces. It was operated in tapping mode using DESP SiN cantilevers; the fast scan direction was parallel to the c-axis of the GaN film. ADF imaging in STEM mode was performed on an FEI Tecnai F20 with an illumination angle of 12.9 mrad and collection angles of 45 to 230 mrad for high-angle ADF (HAADF) and 24 to 118 mrad for low-angle ADF (LAADF). The sampling rate and the defocus were adjusted to avoid lattice contrast that would artificially increase variance.

3. Surface roughness

Roughness measurements in AFM are based on the root mean square height difference from the mean: “RMS roughness”. Taken across the whole image, it measures the total variation, and therefore low-spatial frequency components are included. Fine-scale roughness can be measured by decomposing the one-dimensional power spectrum function:

$$\langle \rho(k) \rangle = \frac{1}{N} \sum_{i=1}^{N} \left| \int_{-L/2}^{L/2} h_i(x) \exp(ikx) dx \right|^2$$

into narrow and broad spatial frequency bands; $h_i(x)$ are the set one-dimensional height profiles that were sampled $N$ times across a square image of length $L$. Invoking Parseval’s theorem, we calculated the total variance associated with each band by fitting polynomials to the band shape and integrating over all spatial frequencies. This allowed us to extract the fine-scale roughness associated with the broad high frequency band. All roughness values quoted below were measured over 1 µm$^2$ scan area.

Tripod polishing alone gave extremely smooth, flat surfaces over the whole GaN surface (figure 1(a)). The height range was typically less than 20Å and the total RMS roughness was small (2.2 ± 0.4 Å). This roughness was dominated by polishing scratches in the surface, which left wide but shallow depressions on the surface. The fine-scale roughness was sub-Ångström (0.9 ± 0.02 Å).

Ion milling at a low dose of 4000 ions/nm$^2$ increased the total RMS roughness ten-fold to about 20±4 Å. However, most of this roughness was attributed to very large shallow pits (figure 1(b)). Spectral analysis showed that the fine-scale roughness was much more modest (1.1±0.03 Å), i.e. a small increase from the tripod-polished state.

A higher ion dose of 30000 ions/nm$^2$ lead to small increase in total RMS roughness (23±6 Å) as the shallow pits became channels (figure 1(c)). Spectral analysis showed that a much higher fraction of the total roughness was due to the fine scale roughness (17±8 Å). This was now seen as ‘islands’ that were typically 200 Å in width. Islands much smaller than this were difficult to detect because of the limited lateral resolution of the AFM, about 110 Å by our estimates.
Figure 2. Normalized power spectra showing the effect of progressive ion milling on the fine scale roughness (high frequency band) as measured in the AFM (a). A (11-20) dark-field image of a tripod-polished only GaN sample shows significant presence of dislocations in the near surface region (b).

4. Surface roughness in STEM-ADF imaging: Theoretical values
Variance in the surface height contributes twice over to the variance of the specimen thickness. Thus thickness fluctuations can be converted into ADF intensity fluctuations if we know the dependence of the ADF intensity and its gradient on the specimen thickness. For this, we used Kirkland’s STEM-SLIC code, based on the multislice method, to calculate the total ADF signal at high and low collection angles corresponding to those on our microscope. [11-20]-oriented GaN super-cells were constructed with random displacements included to account for thermal vibrations (Einstein model, 25 configurations). This approximation is not accurate for the azimuthal distribution of thermal diffuse scattering, but is accurate for the total ADF intensity [4]. The theoretical standard deviations in the ADF signals were obtained by multiplying the ADF intensity gradient by the roughness measured by AFM (with a factor of $\sqrt{2}$ to convert the surface height standard deviation into a thickness deviation).

5. Surface roughness in STEM-ADF imaging: Measured values
To compare values of standard deviations, the absolute value of the current detected on the ADF detector must be known, as well as the incident probe current. We calibrated the ADF detector by placing an empty CBED disc with a small condenser aperture onto the detector and measured the output signal in the ADF image for a variety of spot sizes (C1 lens settings). The probe currents were then measured using a pico-ammeter connected to the Gatan Imaging Filter, which was configured for use as a Faraday cage. With fixed contrast and brightness settings, all ADF images could be converted into absolute intensity levels (fraction of the incident probe current).

We attempted to use a tripod-polished only thin foil for the ADF measurements. However, routine examination showed that the GaN is neither very thin, nor strain free. It is plagued with very high densities of sub-surface dislocations (figure 2(b)), leading to a sample in a highly strained state. Only by ion milling the tripod-polished sample with a low dose, could we obtain smooth, dislocation-free regions suitable for imaging in the STEM. Reasonably straight but tapered edges were analyzed in a similar way to the AFM, i.e. one-dimensional power spectra functions, but the thickness gradient at the edge had to be factored out. Thus, we compared the power spectra expected for several different specimen thicknesses (figure 3).

Figure 3 shows the standard deviations in both the high and low angle ADF signals (HAADF: 45-230 mrad and LAADF: 24-118 mrad) as measured by our approach (solid lines) and compared to the values expected for the low-dose ion-milled sample. The two broken lines correspond to the ADF variation one would expect if only fine-scale roughness occurred (dashed), or was rougher to the same extent as the RMS roughness (dotted). It is easy to see that the ADF variations concur only with very rough surfaces for the thicker regions while, for the thinnest regions (< 50 Å), the agreement is somewhere between the fine-scale and coarse scale roughness values.
Figure 3. Measured standard deviations in the HAADF (a) and LAADF (b) at differing specimen thicknesses compared to that expected for the RMS surface roughness (20 Å, dotted line) and for fine-scale roughness (1.1 Å, dashed line). The ion-milling dose was 4000 ions/nm$^2$ (c.f. figure 1(b)).

6. Discussion and concluding remarks

It is surprising that the measured ADF variations cannot be explained by the fine-scale roughness alone. Several explanations are feasible: First, the presence of the amorphous layer and an ion-implanted surface region would lead to significant near-surface strain that relaxes when the sample is thin. Our comparison assumes that the amorphous layer is relatively thin and benign [5]. Given the sensitivity of the LAADF signal to strain, this might explain the excessive ADF variation seen here. Second, the roughness of the top surface may not be statistically equivalent to the bottom because of electron channeling in the deeper parts of the crystal. However, given the thinness of the samples, this is likely to be small. Third, our use of the AFM to measure fine-scale roughness may be too challenging. The tip radius imposes a minimum lateral resolution of about 10 nm that effectively low-pass filters the power spectrum and removing the higher frequency components that might contribute significantly more to the total variance. However, significant quantities of energy are contained in the very low frequency components and these are still present in all spectra shown in figure 2(a). Perhaps by modeling and measuring the variance in the low-frequency band and subtracting that from the RMS roughness would give us a better estimate of the fine-scale roughness.

However, we have seen that reasonable agreement does exist between the theoretical and measured variances. Thus the RMS surface roughness can account for contrast variations seen in STEM ADF images on a scale from a nanometre to hundreds of nanometers.

References

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