Dark-field electron holography for strain and composition measurement with a sub-nanometre spatial resolution

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1. INTRODUCTION

Transmission electron microscopy (TEM) is a well-established tool for strain measurement that combines an excellent precision with a high spatial resolution. Strain maps can be obtained by different TEM techniques such as high-resolution transmission electron microscopy (HRTEM), high-resolution scanning transmission electron microscopy (HR-STEM), convergent beam electron diffraction, nanobeam electron diffraction, or dark-field electron holography (DFEH). All of the above-mentioned techniques provide high measurement precision in the range of 10^{-3} - 10^{-4}, but only HRTEM and HR-STEM are able to measure strain with a sub-nanometre resolution. Processing of HR(S)TEM images is done either in real or in reciprocal spaces, the latter approach called "Geometric Phase Analysis" (GPA) is widely used thanks to an efficient noise reduction. The highest possible spatial resolution available with GPA equals to a double interatomic distance. The highest spatial resolution of a strain map obtained with DFEH is limited by a double spacing between the hologram fringes, which can be tuned within certain limits by changing the biprism voltage. However a decrease of the fringe spacing is accompanied by a reduction of the hologram contrast, which leads to a drastic drop of the measurement precision. Consequently a spatial resolution of strain measurement with DFEH is typically in the range of 4 - 6 nm, with the best to our knowledge reported value of 1 nm.[1]

A recent development of the TEM equipment and appearance of microscopes specially designed to perform electron holography, namely I2TEM, opens exciting possibilities for the electron holography. We report on a new mode of DFEH, which allows to measure strain distribution with a sub-nanometer resolution. The capacities and the limits of a new technique will be compared with those of HRTEM by studying the InGaN/GaN heterostructures grown by molecular beam epitaxy (MBE) and metalorganic vapour phase epitaxy (MOVPE).

2. RESULTS

2.1 Experimental conditions

Off-axis electron holography experiments were carried out on the Hitachi I2TEM (in-situ interferometry TEM), an HF3300 equipped with a cold field emission gun, Aplanator image aberration corrector, 4 rotatable electrostatic biprisms and 4k x 4k Gatan CCD camera. Cold FEG emits electrons with a high degree of coherence, while multiple biprisms provide more flexibility in choosing the hologram width and fringe spacing and allow to eliminate the Fresnel fringes.[2] The Aplanator B-corr Cs-corrector not only compensate for the objective lens aberrations near the optical axis, but also corrects for the radial and azimuthal off-axis coma[3]. Consequently large field of view holograms do not suffer from the position-dependent axial coma, which would appear in case of elliptic illumination. Finally the overall stability of the microscope is so high, that the acquisition time of tens of seconds can be used. Owing to the above-mentioned factors it is possible to obtain holograms with a fringe spacing below 0.1 nm having both a high contrast and a wide field of view.

In this study we acquire holograms with a field of view of 70 nm x 70 nm and 0.15 nm fringe spacing using objective lens. The reference holograms recorded in vacuum typically have a contrast of about 20%, while the holograms acquired in dark-field mode have a contrast between 10% and 15%. Objective aperture having a 3.0 nm^{-1} size in the Fourier plane is used to cut off all the beams except the diffracted beam with g=0002, which is used to form the hologram. The geometric phase is calculated with a spatial resolution of 0.4 nm.

2.2 Experimental results

As a test structure we study a set of MBE grown $In_{0.18}Ga_{0.82}N$/GaN quantum wells (QWs). A preliminary study of the samples with STEM-HAADF (not-shown) indicated that all the QWs have an atomically abrupt interfaces and uniform composition. Figure 1 shows strain maps in the growth direction obtained using DFEH and HRTEM. Strain maps calculated with GPA (Fig. 1(b)) has the widest field of view, but contains artefacts due to the local attenuation of the HRTEM contrast. As opposed DFEH contrast is very uniform, which provides a reliable strain measurement throughout the whole field of view (Fig. 1(d)). In turn the real space treatment

(Fig. 1(f)) has the smallest field of view and it can be applied only to the HRTEM images with no contrast variations (the image processing algorithm can be found elsewhere).[4]

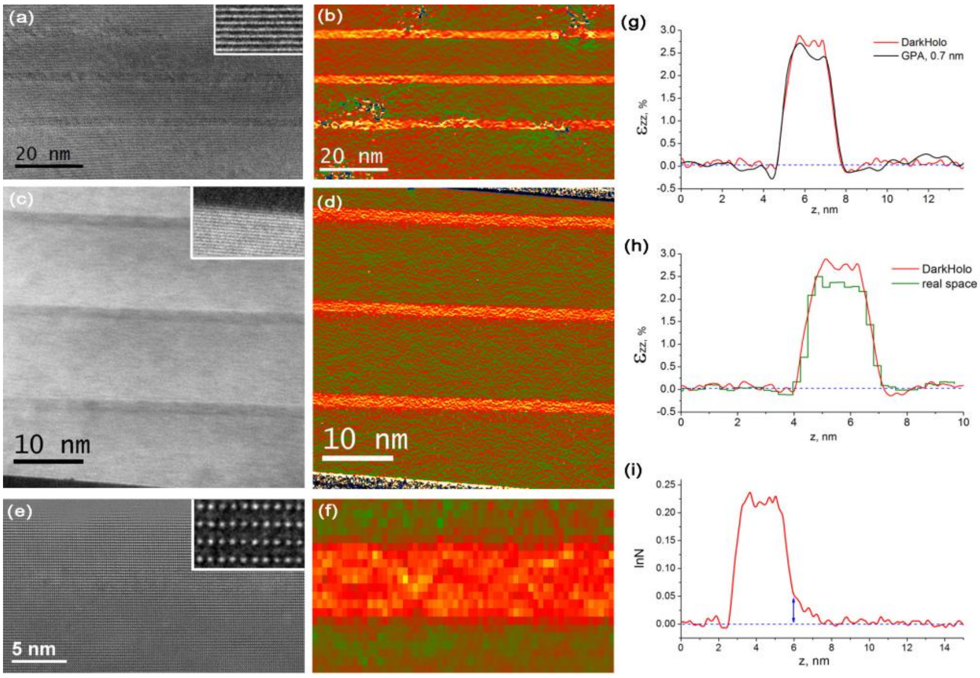

Figure 1. (a) HRTEM image of InGaN/GaN QWs taken along the [14-50] zone axis and (b) corresponding GPA strain map with a spatial resolution of 0.7 nm. (c) Dark-field electron hologram and (d) corresponding strain map calculated with a spatial resolution of 0.4 nm. (e) HRTEM image taken along the [1-100] zone axis and (f) magnification of the strain obtained using a real space treatment of 30 similar HRTEM images. All the strain maps are normalized to the same range from -10 to +10%. All the inserts are magnifications of the corresponding TEM images. (g) Comparison of strain profiles obtained by DFEH and GPA. (h) Comparison of strain profiles obtained by DFEH and a real space treatment algorithm. (i) Example of a composition profile from MOVPE grown InGaN QW obtained with DFEH.

Figures 1(g) and (h) show a comparison of DFEH and HRTEM strain profiles averaged over a distance of 25 nm. In a high-resolution mode DFEH has precision about 0.1%, which is better than that of GPA and comparable with precision achieved by the real space algorithm. The actual resolution of DFEH can be evaluated from the low interface broadening: the strain increases from 0% to 2.7% within 0.7 nm. The difference of the strain amount inside the QW measured by different techniques is attributed to the thin foil relaxation effect, which is higher in a thinner sample used for HRTEM. A combination of DFEH with the finite elements modelling constitute a powerful tool for the local composition measurement and it was used to study the surface segregation effect in MOVPE grown InGaN/GaN structures (see Fig. 1(i) for a composition profile example).

3. CONCLUSION

In conclusion, we show that electron holography is able to measure strain and composition distribution with a 0.7 nm spatial resolution while keeping a high measurement precision. It was found that the real DFEH resolution is lower than the spatial resolution of geometric phase (0.4 nm in our case, which can in principle be as high as 0.2 nm). The resolution reduction is attributed to several factors, such as finite size of the objective aperture, sample tilt or sample drift during the hologram acquisition.

REFERENCES

[1] A. Beche, J. L. Rouviere, J. P. Barnes, D. Cooper, Ultramicroscopy **111**, 227–238 (2011).
[2] K. Harada, A. Tonomura, Y. Togawa, T. Akashi, and T. Matsuda, Appl. Phys. Lett. **84**, 3229 (2004).
[3] E. Snoeck, F. Houdellier, Y. Taniguch, A. Masseboeuf, C. Gatel, J. Nicolai, and M. Hytch, Microscopy and Microanalysis **20**, 932-933 (2014).
[4] T. Schulz, T. Remmele, T. Markurt, M. Korytov, and M. Albrecht, J. Appl. Phys. **112**, 033106 (2012).