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Quantitative strain mapping of InAs/InP quantum dots with 1 nm spatial resolution using dark field electron holography

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The optical properties of semiconductor quantum dots are greatly influenced by their strain state. Dark field electron holography has been used to measure the strain in InAs quantum dots grown in InP with a spatial resolution of 1 nm. A strain value of 5.4% ± 0.1% has been determined which is consistent with both measurements made by geometrical phase analysis of high angle annular dark field scanning transmission electron microscopy images and with simulations. © *2011 American Institute of Physics*. [doi:10.1063/1.3672194]

Epitaxially grown, self-assembled, III-V semiconductor quantum dots (QDs) are of interest for a number of applications, such as high-speed communication lasers, optical switching devices, and optical memories.[1] To be able to use these structures in devices, a detailed understanding and a precise control over parameters that affect their electronic and optical properties is required. Hence, a significant effort has been dedicated to understanding the characteristics of these QDs. An important parameter in determining their optical properties is the elastic strain distribution within and around the QD, as the strain affects the energy bands and the wave-functions of the confined carriers. A number of theoretical studies have dealt with calculating strain distribution in QDs, for example using atomistic or continuum methods, based on assumptions about their morphology and composition.[2] However, there is still need for quantitative experimental measurements of strain on these structures with nanometre spatial resolution, to compare with the theoretical models and help exploit the optical properties of these QDs.

Transmission electron microscopy (TEM) is well adapted for the measurement of strain with nm scale resolution. In the past, geometrical phase analysis (GPA) of high resolution images have been used to obtain the strain in nano-structured materials.[3] Although this approach can be used to acquire strain maps with a spatial resolution of around 1 nm, the signal to noise ratio can be poor, and an unstrained reference region is required to be present within a small field-of-view.[4,5] Recently there has been a lot of activity in the development of TEM-based techniques that can be used to measure strain, such as dark field electron holography and nanobeam electron diffraction (NBED). Although NBED has been shown to be a versatile technique that can be used to measure the strain in many different types of samples, at this time the spatial resolution of between 3 and 6 nm is not sufficient to measure the strain in these dots.[6,7] Dark field electron holography[8] is a variant of off-axis electron holography.[9] An electron biprism is used to interfere an electron wave that has passed through the region of interest with an electron wave that has passed through a reference region in order to provide an interference pattern known as the hologram. An objective aperture is used to select a given diffracted beam and the reconstructed phase image contains information about the variation of the diffracted beam with respect to its value in the reference region, therefore a map of the strain can be obtained.[10] A precision of 0.02% has been demonstrated previously in semiconductor specimens which is much better than can be obtained by either NBED or by the GPA analysis of high resolution images. A problem however, is that until now, the spatial resolution for dark field holography was typically in the range 4-6 nm which is not suitable for the measurement of these QDs.[8,11] For the analysis of these QD specimens, the objective lens has been used instead of the Lorentz lens in "free lens control" mode where the microscope projector and intermediate lenses are used to increase the specimen magnification and the diffraction lens is used to increase the electron hologram width in the intermediate image plane.[12] Electron holograms with a fringe spacing of 0.33 nm have been acquired using this approach which allows the reconstruction of strain maps with a spatial resolution of 1 nm.

The QDs studied in this work were grown in a vertical low-pressure metal organic vapour-phase epitaxy (MOVPE) TurboDisc reactor on InP (001) substrates. Arrays of QDs were formed by the deposition of 2.2 ML of InAs onto an InP buffer layer at 520 °C, with a V/III ratio of 4.7 and a growth rate of 1.32 Å/s and then overgrown with 100 nm of InP without any growth interruption. Specimens suitable for TEM examination were prepared using a FEI Strata FIB using *in situ* lift out. The specimens were prepared to a thickness of 200 nm using an operating voltage of 30 kV and then finished to a thickness of less than 50 nm at 8 kV which provides a compromise between reducing the surface damage

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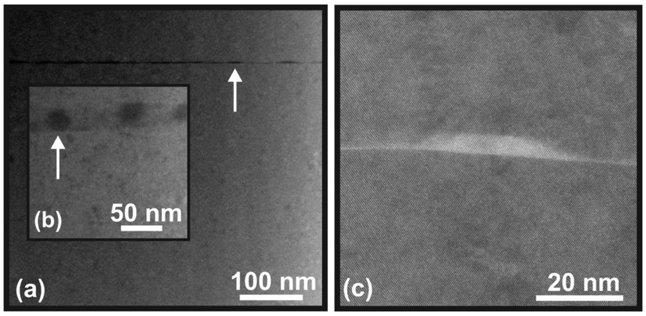

FIG. 1. (a) Low magnification dark field HAADF STEM image of the InAs QDs in the InP lattice. (b) Low magnification dark field HAADF STEM image with specimen tilted showing that the quantum dot examined is perfectly encapsulated in the InP. The arrow shows the individual QD that was selected for analysis. (c) A probe corrected HAADF STEM image of the quantum dot examined in these experiments.

and maintaining the parallel sides that are required for electron holography. An individual QD was selected where the whole dot was contained in the thin TEM specimen. Dark field electron holograms were acquired using a FEI Titan TEM operated at 200 kV with the (004) diffraction spot selected in order to assess the strain in the growth direction and the (220) diffraction spot to assess the strain in the in plane direction. After the electron holograms had been acquired, probe corrected high angle annular dark field (HAADF) scanning transmission electron microscopy (STEM) images were acquired at a TEM operating voltage of 200 kV. These additional experiments were performed both as an independent method of measuring the strain and also to provide accurate values of the dimensions of the QD in order to simulate the expected values of strain accounting for effects such as specimen relaxation.

Figure 1(a) shows a low-magnification HAADF STEM image of the region of interest and (b) a low magnification image at high tilt confirming that the QD is complete and located in the middle of the TEM lamella. The dot was found to be 32 nm in diameter in a TEM specimen of a total crystalline thickness of 35 nm. Figure 1(c) shows a HAADF STEM image of the QD.

Figure 2(a) shows a dark field electron hologram of the quantum dot with the (004) diffraction spot selected. To achieve a two beam condition, the dot was tilted by 0.5° from the [011] on-axis orientation. The electron hologram shown has a fringe spacing of 0.33 nm and a fringe contrast of 15%. Although the FEI Titan is stable enough to acquire electron holograms for one minute or more in order to provide phase images with excellent signal to noise ratio,[13] the stability of the specimen stage limited the acquisition time of the electron holograms to 16 s. As a consequence, the number of electron counts is lower than typically used for dark holography leading to a precision lower than has demonstrated elsewhere.[11]

Figure 2(b) shows a strain map for the (004) growth direction. The maximum value of strain measured in the centre of the dot, relative to the unstrained substrate is $5.4\% \pm 0.1\%$ where the error was obtained by taking the standard deviation of an unstrained region of the InP substrate. Simulations were performed using 3D-finite element modeling to verify that the measured values of strain were as expected for the dot in a 35-nm-thick specimen, the QD was assumed to be pure InAs with no intermixing of P atoms, except at the interfaces. The experimentally determined and simulated strain profiles are shown in Fig. 2(b) and the simulated value of 5.6% is consistent with the holography results. Due to the excellent signal to noise ratio in the strain map, information about the deformation around the dot was observed which was used to accurately adjust the simulations. In addition, the low values and distribution of strain in the wetting layer indicate a significant intermixing of As and P. From the measured strain of 2.0%, we can estimate an As concentration of only 29% in the wetting layer.[14] From the determined value of strain, the composition in the centre of the dot appears to be 100% InAs. Figure 2(d) shows a strain map of the quantum dot for the (220) in-plane direction acquired by dark field electron holography. It is important to emphasize that the measured values of strain shown here are relative to the unstrained substrate and therefore, as expected, no strain is seen as the dot is in perfect epitaxy with the reference substrate.

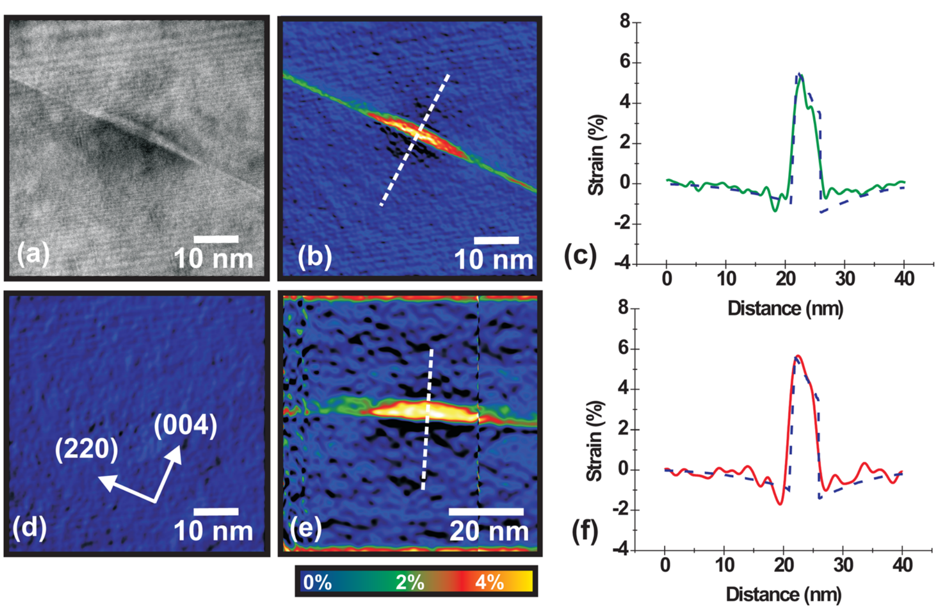

FIG. 2. (Color online) (a) A dark field electron hologram of a single InAs quantum dot encapsulated in InP. The (004) diffraction spot has been selected. (b) Strain map for the (004) growth direction. (c) Experimental strain profile extracted from the region indicated by the dashed line in the strain map (solid line) compared to simulations (dashed line). (d) Strain map for the (220) in plane direction acquired using dark field electron holography. (e) A strain map for the (004) growth direction calculated from the HAADF STEM image. (f) Experimental strain profile extracted from the region indicated by the dashed line in the strain map (solid line) compared to simulations (dashed line).

Strain maps, such as shown in Fig. 2(e), were also calculated from the probe corrected HAADF STEM images using a GPA algorithm. The algorithm measures locally the reciprocal lattice parameters and thus the strain measurement is not affected from the Z-contrast in the image, (although some artifacts can be introduced at interfaces). The advantage of using HAADF STEM images instead of HRTEM images is that the contrast is not reversed with thickness and defocus, and thicker samples and larger field of views can be obtained. However, problems from scan distortion and sample stability can lead to noisy strain maps as shown here. Despite these problems, the strain map obtained from STEM images is shown in Fig. 2(f) and it is consistent with the dark holography results and simulations. The spatial resolution of the HAADF STEM strain map is estimated to be 1 nm from the size of the mask that was used in Fourier space. The maximum value of strain recovered through the centre of the dot is $5.6\% \pm 0.4\%$ which is consistent with the values measured by dark field electron holography. The error was assessed by taking the standard deviation of an unstrained region of the InP substrate.

In conclusion, we have shown that by combining dark field electron holography with careful specimen preparation, the strain both in and around an individual InAs QD buried in a InP substrate can now be measured by dark field electron holography. Results that have been obtained both by HAADF STEM imaging and by simulations are consistent with the experimental results obtained here. The quality of this experimental data should help us to improve our simulations in order to provide better insights into the strain states and indirectly, the chemistry of these QDs. These results suggest that there is no great intermixing at the core of the dots, while there is some intermixing at the edges of the dot and significant intermixing in the wetting layer. Definitely these results are encouraging and suggest that dark field electron holography is a powerful tool to quantitatively measure the strain in objects with 1 nm spatial resolution and a high sensitivity. We expect to be able to improve the signal to noise ratio in these results by acquiring electron holograms for longer time periods using modern, more-stable specimen stages and by using the brighter electron sources that are now routinely supplied with the latest generation electron microscope. The ability to quantitatively measure the deformation of these QDs as a function of diameter and composition will allow their emission properties to be better understood which will directly lead to the development of more efficient optical devices.

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