Strain quantification in epitaxial thin films

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Abstract. Strain arising in epitaxial thin films can be beneficial in some cases but devastating in others. By altering the lattice parameters, strain may give a thin film properties hitherto unseen in the bulk material. On the other hand, heavily strained systems are prone to develop lattice defects in order to relieve the strain, which can cause device failure or, at least, a decrease in functionality. Using convergent beam electron diffraction (CBED) and high-resolution transmission electron microscopy (HRTEM), it is possible to determine local strains within a material. By comparing the results from CBED and HRTEM experiments, it is possible to gain a complete view of a material, including the strain and any lattice defects present. As well as looking at how the two experimental techniques differ from each other, I will also look at how results from different image analysis algorithms compare. Strain in Si/SiGe samples and BST/SRO/MgO capacitor structures will be discussed.

1. Introduction
The amount of strain in a system affects the properties of the material. Crystal defects are sometimes created to relieve strain (e.g. if the lattice misfit is large). Edge and mixed dislocations, antiphase boundaries and other defects are commonly found in strained materials and can lead to device failure.

Recently, there has been research into the benefits of strain in a system. It has been found that, when compressively strained, SrTiO$_3$ is ferroelectric at room temperature [1] – surprising since this material is not usually ferroelectric at any temperature. It has also been found that ultrathin films of Pr$_{1-x}$Sr$_x$MnO$_3$ have strain-induced magnetic properties which are not found in the bulk material [2].

There are many techniques for determining strain in materials: X-ray diffraction and Raman spectroscopy are both non-destructive ways to look at strained systems, although the results are averaged over relatively large areas of the material, giving a poor spatial resolution. These methods are less spatially resolved and inadequate if the strain in a material is not homogenous.

For a quantitative approach (which requires a high spatial resolution), the most promising techniques are convergent beam electron diffraction (CBED) and high-resolution transmission electron microscopy (HRTEM), both of which have nanometre resolution. Using these techniques, it is possible to see the variation in strain across a film. In both cases, image analysis involves comparing experimental with simulated images.

2. Sample Information
So far, two types of sample have been investigated. The first were MgO/SrRuO$_3$/Ba$_{0.5}$Sr$_{0.5}$TiO$_3$ capacitor structures with Au contacts on the top surface [3]. These were prepared offsite using
molecular beam epitaxy. The BST layer thickness was varied from 6nm to 240nm. The SrRuO$_3$ layer is between 100 and 200nm thick.

Ba$_x$Sr$_{1-x}$TiO$_3$ (BST) is currently of interest for use in high-density dynamic random access memories (DRAMs), due in part to its relatively high dielectric constant. Strain in ferroelectric films (such as BST) is especially interesting as it can change the phase diagram, shift transition temperatures and can even alter the order of the phase transition [4].

The samples were prepared for viewing in the transmission electron microscope (TEM) by cross section encapsulation. The samples were encapsulated using a section of molybdenum (Mo) rod (with a 400 micron slot) and 3mm brass tubing. After baking to cure the epoxy resin, the cross sections were cut and thinned to approx. 120 microns, then dimpled and ion milled using a Precision Ion Polisher at 4kV and then gentle milling at 0.5kV to improve the surface of the sample.

The second type of sample was a Si/SiGe calibration sample (mag-i-cal™) which consisted of sets of five parallel bands of SiGe (thickness of 9.6-11.9nm) with a spacer layer of silicon (10.7-14.2nm) between them [5].

![Figure 1. Si (light) and SiGe (dark) layers in mag-i-cal™ sample.](image)

3. High Resolution Transmission Electron Microscopy (HRTEM)

In this section we look at the relevance of HRTEM for strain determination and discuss two methods of image analysis.

3.1. Introduction

High-resolution transmission electron microscopy (HRTEM) allows the direct imaging of the local crystal structure of a thin film, with atomic resolution. It can be used to determine lattice parameters across a film, allowing strain to be calculated, and can give information on any lattice defects that are present.

In a high-resolution image, there are two main aspects that require interpretation: the image contrast and the position of any structures present. Quantitative analysis of the image contrast is difficult in the context of strain determination as contrast features due to structure factor effects and those due to strain are difficult to separate. There are also difficulties in determining the imaging parameters to the accuracy required for image simulations.

Several techniques have been developed for analysing high-resolution images, taking these difficulties into consideration. Two techniques will be discussed here: the geometrical phase technique (Hýtch) and the “peak pairs” method (Galindo).

3.2. Geometrical Phase Method

The purpose of this program is to locally determine elastic strain in materials. It does this at the atomic scale by the analysis of high resolution images of the materials.

The first step is to filter the image with an asymmetric filter at one of the Bragg spots of the Fourier transform. An inverse Fourier transform is then performed and the phase component of the resulting complex image is analysed to give information on the local displacement in a given direction.
The local strain is found by looking at the derivative of the displacement obtained from two non-collinear components [6].

![Figure 2](image)

**Figure 2.** (a) BST/SRO interface and (b) e\textsubscript{xx} – visualisation of strain in the plane of the sample.

The interface between the SRO and BST is marked on figure 2(a) but can clearly be seen in the strain profile (generated using the geometric phase algorithm [7] in Digital Micrograph 2.5 for the Apple Mac). An in-plane compressive strain of 0.51% is expected in the BST layer but no strain is present in the strain map. It is most likely that the strain in the BST layer has relaxed during the sample preparation process.

3.3 **The Peak Pairs Method**

This algorithm is based on the peak finding technique, which involves the location of intensity maxima. The peak finding algorithm is particularly useful for the characterisation of interfaces, because of the global nature of the Fourier transform. The peak pairs algorithm is similar except it detects pairs of intensity maxima in a HRTEM image. It is more robust, as well as being faster and taking up less computer memory than its predecessor, the peak finding algorithm.

Galindo developed his algorithm into the “Strain Determination Software v.2” program [8], which has been used here to analyse HRTEM images of the SRO/BST interface in the capacitor structures and the SiGe/Si interfaces in the mag-i-cal sample.

After loading an image into the program and applying Wiener and Bragg filters (to eliminate background noise and mask the Bragg spots representing the periodic part of the image), the program calculates where the intensity peaks are, with sub pixel accuracy [9]. It is possible to set intensity thresholds, which eliminates the problem of extra, between layer peaks affecting the image.

The program then calculates the strain map for each direction (i.e. e\textsubscript{xx}, e\textsubscript{yy}, e\textsubscript{xy}), which gives a visual representation of the strain across a film.

![Figure 3](image)

**Figure 3.** The in plane strain map, out of plane map and strain profile. The maxima in the profile correspond to the SiGe and the minima correspond to the Si.
As before, no strain was seen in the BST layer of the capacitor structure. For SiGe on Si, we expect the SiGe to be compressively strained in the film plane to lattice match to the Si. This results in an out-of-plane strain in the SiGe as a Poisson’s ratio effect.

Figure 3(a) shows the in-plane strain map, which is featureless, except for around the edges – which can be ignored as they are calculation artefacts.

As expected, the out-of-plane strain map (Figure 3(b)) shows a clear positive strain in the SiGe layers in comparison to the Si layers, which show a corresponding negative strain. This result is even more clearly visible in Figure 3(c), which is a line profile of the strain variation across the Si-SiGe layers.

4. Convergent Beam Electron Diffraction (CBED)
Convergent beam electron diffraction (CBED) allows the local determination of strain by analysis of the higher order Laue zone (HOLZ) lines in the convergent beam disc. Information is taken from a relatively small area of the sample and nanometer resolution is possible. As well as having a potential strain sensitivity of two parts per 10,000, CBED patterns also give 3D information (for example, symmetry and crystal defects present).

As yet, we have been unable to prepare BST samples of a good enough quality to achieve any CBED results. On the analysis front, we are hoping to learn more about the multislice simulation of CBED patterns [10] and learn to generate experimental patterns in both normal and nanoprobe mode in the transmission electron microscope.

5. Future Work
As well as making headway with the CBED part of the project, we are also looking at other samples and sample preparation techniques. Also, as well as the high resolution image processing programs mentioned above, we will be using DALI [11] and CUSUM [12] algorithms to analyse images.

6. References
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