Micron 113 (2018) 20–23

Contents lists available at ScienceDirect

Micron

journal homepage: www.elsevier.com/locate/micron

A geometric phase analysis method dedicated to nanomaterials orienting along high-index zone axis

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ARTICLE INFO

Keywords:
Interplanar spacing
Nanomaterials
Geometric phase analysis
Transmission electron microscopy

ABSTRACT

Strain within nanomaterials plays a crucial role in defining their physical and chemical properties. Geometrical phase analysis (GPA) was widely used to investigate deformation within nanomaterials. The traditional GPA method using geometric phases of two lattice fringes provides two-dimensional strain mapping, which is inapplicable to nanomaterials viewed along high-index zone axis. Herein using silicon nanoplate embedded in $Si_{0.5}Ge_{0.5}$ matrix as a test object, we illustrate a GPA method using single lattice fringes. The availability of this GPA method was testified by comparison with traditional GPA method. This work presents an opportunity to extend application of the GPA method.

1. Introduction

Strain within various nanomaterials plays a crucial role in defining the optical, mechanical, catalytic and electronic properties (Gilbert et al., 2004; Ouyang et al., 2010; Smith et al., 2009). Strain variation within nanomaterials is usually investigated using either coherent X-ray diffraction (XRD) (Gilbert et al., 2004; Pfeifer et al., 2006; Ulvestad et al., 2015) or Transmission electron microscopy (TEM). Compared to XRD, TEM can provide a better spatial resolution. In TEM, there are several methods that can be used to measure strain, including diffraction, holography and the analysis of high resolution images, as summarized by Béché et al. (2013). In recent years, a new method based on the combination of high-resolution transmission electron microscopy (HRTEM) and geometric phase analysis (GPA) has been developed to investigate strain distribution. The method of GPA, firstly proposed by Hÿtch et al. (1998), provides a powerful tool for measuring strains quantitatively as revealed in HRTEM. A precision of 3 pm was achieved during the study of strain field around dislocations in silicon by this technique (Hÿtch et al., 2003). The GPA method has been used successfully to quantify strain fields of quantum dots (Sarigiannidou et al., 2005), nanowires (Taraci et al., 2005), dislocations (Wang et al., 2015a, 2015b) and interfaces (Rösner et al., 2010). Strain fields around misfit dislocations at the α-Fe_2O_3/α-Al_2O_3 interface were systematically investigated using the GPA method by the authors (Wang et al., 2015a, 2015b). In the initial stage of growing epitaxial thin films, the interfaces are coherent with homogeneous elastic strain at the films. As thin films grow, the accompanying strain energy accumulation generates misfit dislocations, which can relax the mismatch between the film and substrate. And the interfaces become semicoherent, that is coherent regions separated by misfit dislocations. There is a convergence region of strain around the misfit dislocation core. On the α-Al_2O_3 side, the strains are compressive, and on the other side, the strains are tensile. The largest strain values occur in the immediate core region and the strain is smaller farther from the dislocation core.

As usual, the GPA method uses the geometric phases of two lattice fringes and provided two-dimensional strain mapping (Hÿtch et al., 2003, 1998; Rösner et al., 2010; Sarigiannidou et al., 2005; Taraci et al., 2005; Wang et al., 2015a, 2015b). Experimentally, however, various nanomaterials frequently show sole lattice fringe in HRTEM images, ascribed to their orienting along high-index zone axis. It is generally known that samples can be aligned along a low-index zone axis by double-tilt of the specimen holder. Nevertheless considering the small size of nanomaterials, it is indeed a highly challenging task to align them by tilting holder, even with the assistance of advanced nanobeam diffraction (NBD) technique. Furthermore during double-tilt of the specimen holder, nanomaterials may overlap with matrix at interfaces, leading to a failure in obtaining strain distribution at interfaces. This will restrict the application of GPA method in strain analysis of nanomaterials.

In this study, we illustrate a GPA method using single lattice fringes, by which strain mapping of naomaterials orienting along high-index zone axis can be obtained without requirement of tilting samples. Such a GPA mehod is dedicated to determining variation of interplanar spacings in nanomaterials. A silicon nanoplate embedded in $Si_{0.5}Ge_{0.5}$ matrix (in the context, it is abbreviated by SiGe) was used as a test object. Both the GPA with single lattice fringes and traditional GPA

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https://doi.org/10.1016/j.micron.2018.06.010
Received 1 June 2018; Received in revised form 14 June 2018; Accepted 14 June 2018
Available online 20 June 2018
0968-4328/

were applied to strain analysis and compared to each other. This paper attempts to present an opportunity to extend application of the GPA method.

2. Materials and methods

2.1. Specimen preparation and image acquisition

An array of 15 nm thick silicon nanoplates was fabricated by dry etching techniques, and SiGe was deposited around silicon nanoplates by reduced pressure-chemical vapor deposition. TEM samples were prepared using the conventional technique involving mechanical grinding and polishing until the electron transparency of a specimen is reached. HRTEM experiments were performed on a JEM-2100 F field emission transmission electron microscope at an accelerating voltage of 200 kV, with spherical aberration C_s 1.0 mm, chromatic aberration C_c 1.4 mm and point resolution 0.19 nm. Images were acquired using Digital Micrograph software (Gatan Inc.) on a 2k × 2k Gatan UltraScan 1000 CCD camera. Fast Fourier transformation (FFT) was carried out using a Digital Micrograph software package. TEM samples were left in the microscope for a sufficiently long time to reduce thermal drift, until no noticeable drifting was observed.

2.2. Geometric phase analysis (GPA)

Developed by Hÿtch et al. (1998), the GPA algorithm reconstructs the displacement field by two Fourier components in the power spectrum generated from a HRTEM image. In nature, the GPA measures the displacement of lattice fringes with respect to a reference lattice. The phase of these local Fourier components, or geometric phase $P_g(\boldsymbol{r})$, is directly related to the component of the displacement field, $\boldsymbol{u}(\boldsymbol{r})$, in the direction of the reciprocal lattice vector, $\boldsymbol{g}$ (Hÿtch et al., 1998):

$$P_g(\boldsymbol{r}) = -2\pi \boldsymbol{g} \cdot \boldsymbol{u}(\boldsymbol{r}) \quad (1)$$

Given the geometric phase of two Fourier components, the displacement field can be determined (Hÿtch et al., 1998):

$$\boldsymbol{u}(\boldsymbol{r}) = -\frac{1}{2\pi}[P_{g1}(\boldsymbol{r})\boldsymbol{a}_1 + P_{g2}(\boldsymbol{r})\boldsymbol{a}_2] \quad (2)$$

where $\boldsymbol{a_1}$ and $\boldsymbol{a_2}$ are the real-space basis vectors corresponding to the reciprocal lattice defined by $\boldsymbol{g_1}$ and $\boldsymbol{g_2}$ (i.e. $\boldsymbol{a_i} \cdot \boldsymbol{g_j} = \delta_{ij}$). Then, the derivation of the displacement field gives the strain field in two principal directions:

$$\varepsilon_{xx} = \partial u_x(r)/\partial x, \varepsilon_{yy} = \partial u_y(r)/\partial y \quad (3)$$

Thus the biaxial strains ε_{xx} (in-plane) and ε_{yy} (out-of-plane) are derived to illustrate the local lattice displacement relative to the reference lattice. For additional mathematical and theoretical discussion of GPA, see Refs (Hÿtch et al., 1998; Rouvière and Sarigiannidou, 2005). In this work, GPA was performed in Digital Micrograph using Koch's FRWR tools plugin (http://www.elim.physik.uni-ulm.de/).

3. Results and discussion

At the interface between the silicon nanoplate and SiGe matrix, there exist strain gradients. The accuracy of the GPA in areas where strain is varying abruptly was systematically investigated with experiment and image simulations by Chung and Rabenberg (2008). The interaction between strain gradients and gradient of objective lens transfer function at the fringe periodicity can introduce errors in the strain image, which appear as fluctuations across the entire strain images. The extent to which the strain gradient influences the accuracy of strain measurement depends on the defocus. Under the optimum defocus, which can eliminate gradient of transfer function, errors in the strain image will be smaller than the inherent inaccuracy in the GPA image processing. It should be noted that GPA has its own optimum defocus, which may differ significantly from the HRTEM optimum defocus, i.e. Scherzer defocus. If transfer function $\chi(\boldsymbol{q})$ has the simple form:

$$\chi(\boldsymbol{q}) = \pi\Delta f\lambda q^2 + (\pi/2)C_s\lambda^3 q^4 \quad (4)$$

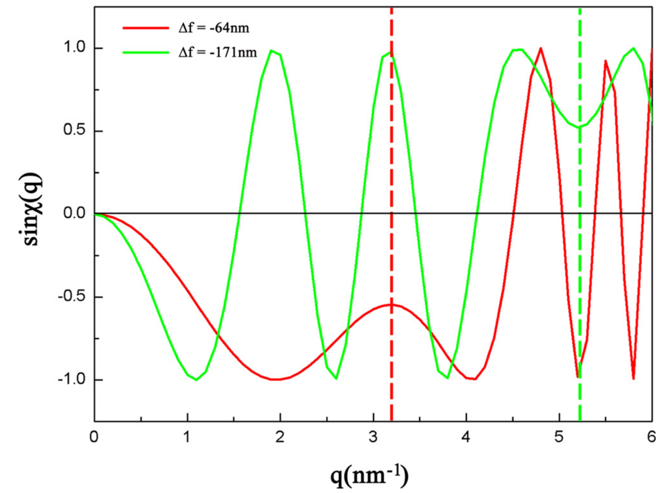

Fig. 1. The transfer function $\sin\chi(q)$ at the GPA optimum defocus $\Delta f_o = -171nm$ for imaging the (220) planes (green line), and at the GPA optimum defocus $\Delta f_o = -64nm$ for imaging the (111)/(11-1) planes (red line). The (220) and (111)/(11-1) reciprocal lattice vectors are indicated by green dash line and red dash line, respectively.

where λ, Δf and C_s are electron wavelength, defocus value and spherical aberration, respectively, Scherzer defocus $\Delta f_s = -1.2(C_s\lambda)^{1/2}$ and the GPA optimum defocus $\Delta f_o = -C_s\lambda^2 g^2$. It should be stressed that Scherzer defocus is more crucial, at which the contrast in HETEM image is interpretable. If the GPA optimum defocus is close to Scherzer defocus, then the GPA optimum defocus is preferred, which can give good, interpretable contrast with negligible errors in the strain image.

In this work, the (220) and (111)/(11-1) planes of Si/ SiGe were chosen for GPA. Imaging the (220) planes at 200 kV with $C_s = 1.0mm$ gives Scherzer defocus $\Delta f_s = -60nm$, and the GPA optimum defocus $\Delta f_o = -171nm$. It can be seen that the GPA optimum defocus deviates significantly from Scherzer defocus. Consequently at the GPA optimum defocus, transfer function $\sin\chi(q)$ starts to oscillate prior to the (220) reciprocal lattice vector, as indicated by green lines in Fig. 1, and contrast in HRTEM image becomes less interpretable. This can be attributed to the Si (220) interplanar spacing $d_{220} = 0.192nm$, which challenges the point resolution 0.19 nm. Therefore Scherzer defocus was preferred when imaging the Si/SiGe (220) planes. Imaging the (111)/(11-1) planes gives the GPA optimum defocus $\Delta f_o = -64nm$, close to Scherzer defocus. Therefore the GPA optimum defocus was preferred when imaging the Si/SiGe (111)/(11-1) planes, which can give good, interpretable contrast, as indicated by red lines in Fig. 1, with negligible errors in the strain image.

The HRTEM image of Fig. 2(a) shows a silicon nanoplate embedded in SiGe matrix at Scherzer defocus. The interface between silicon nanoplate and SiGe matrix is viewed edge on and free of second phases or intermediate layers. Both silicon nanoplate and SiGe matrix are imaged along a high-index zone axis as indicated by the sole (220) spots in the inserted Fourier transform of Fig. 2(a). The (220) spots look faint and blurred, which leads to very weak lattice fringes in HRTEM image. As can be seen in a magnified spot inserted in Fig. 2(a), two dots can be distinguished corresponding to Si and SiGe, respectively.

The sole (220) spot was chosen for GPA and Gaussian mask was used to reduce the GPA processing artifacts (Chung and Rabenberg, 2008; Hÿtch et al., 1998), as indicated in Fig. 2(a). The size of the mask determines the spatial resolution of results: in this case 0.5nm^{-1} in Fourier space, corresponding to 2 nm in image space. A larger mask