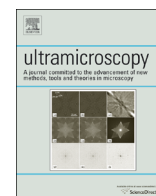




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Ultramicroscopy

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

Lattice and strain analysis of atomic resolution Z-contrast images based on template matching



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ARTICLE INFO

Article history:

Received 2 April 2013

Received in revised form

9 July 2013

Accepted 19 July 2013

Available online 7 August 2013

Keywords:

Strain analysis

Z-contrast

Template matching

STEM

ABSTRACT

A real space approach is developed based on template matching for quantitative lattice analysis using atomic resolution Z-contrast images. The method, called TeMA, uses the template of an atomic column, or a group of atomic columns, to transform the image into a lattice of correlation peaks. This is helped by using a local intensity adjusted correlation and by the design of templates. Lattice analysis is performed on the correlation peaks. A reference lattice is used to correct for scan noise and scan distortions in the recorded images. Using these methods, we demonstrate that a precision of few picometers is achievable in lattice measurement using aberration corrected Z-contrast images. For application, we apply the methods to strain analysis of a molecular beam epitaxy (MBE) grown LaMnO₃ and SrMnO₃ superlattice. The results show alternating epitaxial strain inside the superlattice and its variations across interfaces at the spatial resolution of a single perovskite unit cell. Our methods are general, model free and provide high spatial resolution for lattice analysis.

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1. Introduction

Atomic resolution images of crystals, recorded in a scanning transmission electron microscope (STEM) using a high angle annular dark field (HAADF) detector, provide a relatively uniform image contrast peaked at atomic columns and dependent on the atomic number (Z-contrast) [1–4]. The position of atomic columns can be determined using a peak finding technique. Once the projected atomic column positions are located, an analysis of atomic displacements and the related strain can be carried out directly in real space. There are several advantages in using Z-contrast images for measuring local displacements and strain. Firstly, the image contrast depends on the electron probe size and the best contrast is obtained using the smallest electron probe [5]. Secondly, the peak intensity at the atomic column position in Z-contrast images shows an almost monotonic dependence over a large range of sample thicknesses [6,7]. Atomically centered contrast is also available in high resolution electron microscopy (HREM), or bright-field STEM, but only when the sample is thin and at the right defocus [8]. However, most samples of practical

interest are not thin enough and in general in these samples HREM records lattice fringe images where lattice fringes are not registered with atomic positions. The analysis of lattice fringes has led to the development of the geometrical phase analysis (GPA) technique [9,10], which analyzes a few spatial frequencies and the associated phases recorded in a HREM image and uses these phases to measure lattice displacements and strain. GPA analysis has been applied to Z-contrast images [11–15]; however, this must be done with care especially near interfaces. A major drawback of Z-contrast STEM imaging is the noise present in images, including environmental noise [16]. The quality of STEM image is heavily influenced by scan distortions, beam and sample stability [17–19]. Because of this, frequency information recorded in Z-contrast images is not as reliable as those recorded in HREM images and their analysis requires remediation of the sources of noises. Furthermore, to take advantage of atomic resolution, an alternative method to GPA is required, since GPA by its design does not use all information recorded in an atomic resolution image.

Real space lattice analysis is based on locating peaks associated with the atomic columns that are recorded in a pixelated image. A column-by-column based analysis will take the full advantage of atomic resolution. The standard numerical techniques used for peak finding that have found applications in electron image processing include peak fitting using a model peak distribution, locating peak maximum using the parabolic curve fitting or fitting

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with other curve functions [20], the center of gravity method, and by fitting cross correlation function. These different approaches have been implemented in software packages that are available to the electron microscopy community. One is PPA (Peak Pair Analysis) developed initially by Galindo et al. [21], and the other is LADIA (Lattice Distortion Analysis) developed by Du et al. [22]. PPA measures the position of atomic columns directly on the image. To reduce the effect of noise, PPA relies on noise filtering to improve the measurement accuracy [21]. In LADIA, cross correlation function (XCF) between the mean subtracted template and image is used to measure the peak positions. The XCF is calculated by fast Fourier transform (FFT). As mentioned before, noise is a serious issue in Z-contrast images. Taking account of the noise in lattice analysis is critical to the measurement accuracy.

To take account of characteristics that are unique in Z-contrast images for quantitative lattice analysis, we have developed a Template Matching Analysis (TeMA) based approach. At the core of TeMA is the image processing technique of template matching. This technique is widely used in electron imaging of biological single particles, where images of particles recorded at different projections are located using a template and averaged to construct a 3-D structure of the particle [23]. The same pattern recognition capability in presence of noise has also been used in HREM and STEM Z-contrast imaging of grain boundaries where similar patterns can be located and averaged using template matching [24–26]. Here we show that template matching can be used effectively to filter a noisy experimental Z-contrast image into a lattice of smooth correlation peaks, which then is quantified for lattice analysis. The template contains a single, or a group of, atomic column(s). For the creation of the template, we use an iterative procedure based on template directed spatial averaging. For lattice analysis, TeMA uses a reference lattice to calculate local displacement and strain. TeMA is implemented in C as callable functions through DigitalMicrograph (DM, Gatan Inc) plug-in. The TeMA software will be made available free for academic research (for more information, email jianzuo@illinois.edu).

This paper describes the principle, design and application of TeMA. It follows from the application of template directed composition analysis that was reported previously [27]. Specifically, in developing atomic resolution image processing techniques for lattice analysis based on template matching, we have targeted following goals: (1) analysis at atomic resolution, (2) sub-pixel precision, (3) correction of scan noise and distortion, and (4) model free analysis. Realization of these goals will be demonstrated using experimental Z-contrast images recorded using aberration corrected STEM. Application in materials science is demonstrated through an analysis of epitaxial strain in a superlattice of LaMnO₃ and SrMnO₃.

2. Methods

2.1. Template matching of intensity images and peak finding

A typical atomic resolution Z-contrast image contains repeated patterns where the scattered intensity is peaked at the position of a column of heavy atoms. The repeated patterns can be detected using the image processing technique of template matching. This technique works by searching for location (r, s) within the targeted image I where the sub-image of $I(r, s)$ are most similar as the template image T . The similarity between T and the sub-image $I(r, s)$ is measured by the image distance $d(r, s)$ defined by:

$$\begin{aligned} d^2(r, s) &= \sum_{(i,j) \in R} [I(r+i, s+j) - T(i, j)]^2 \\ &= \sum_{(i,j) \in R} I(r+i, s+j)^2 + \sum_{(i,j) \in R} T(i, j)^2 - 2 \sum_{(i,j) \in R} I(r+i, s+j)T(i, j) \\ &= A(r, s) + B - 2C(r, s) \end{aligned} \quad (1)$$

Here $C(r, s)$ is the linear cross-correlation function between $I(r, s)$ and the template. $A(r, s)$ and B are the so-called signal energy of the sub-image and T , respectively. When $A(r, s)$ is constant, the minimum image distance is obtained when $C(r, s)$ is at maximum. In practice, the two images are often compared using the differences with respect to the average value of T and the local average of $I(r, s)$:

$$C_c(r, s) = \frac{\sum_{(i,j) \in R} [I(r+i, s+j) - \bar{I}(r, s)][T(i, j) - \bar{T}]}{\sqrt{\sum_{(i,j) \in R} [I(r+i, s+j) - \bar{I}(r, s)]^2} \sqrt{k\sigma_T^2}} \quad (2)$$

here k is the total number of pixels in T and

$$\bar{I}(r, s) = \frac{1}{k} \sum_{(i,j) \in T} I(r+i, s+j), \quad \bar{T} = \frac{1}{k} \sum_{(i,j) \in R} T(i, j) \quad (3)$$

and

$$\sigma_T^2 = \frac{1}{k} \sum_{(i,j) \in R} [T(i, j) - \bar{T}]^2 \quad (4)$$

The function C_c in Eq. (2) is known in statistics as the correlation coefficient (C_c) [28]. It has a value ranging between $[-1, 1]$, regardless of the intensities and sizes of T and $I(r, s)$. The maximum value of 1 is obtained when the intensity distribution of T and $I(r, s)$ are the same. An comparison of C_c as defined in Eq. (2) with the XCF [22] is given in appendix. We note that subtraction of local averaged intensity makes C_c less sensitive to variations in background in the recorded Z-contrast images due to either the change in thickness or the presence of surface amorphous layers.

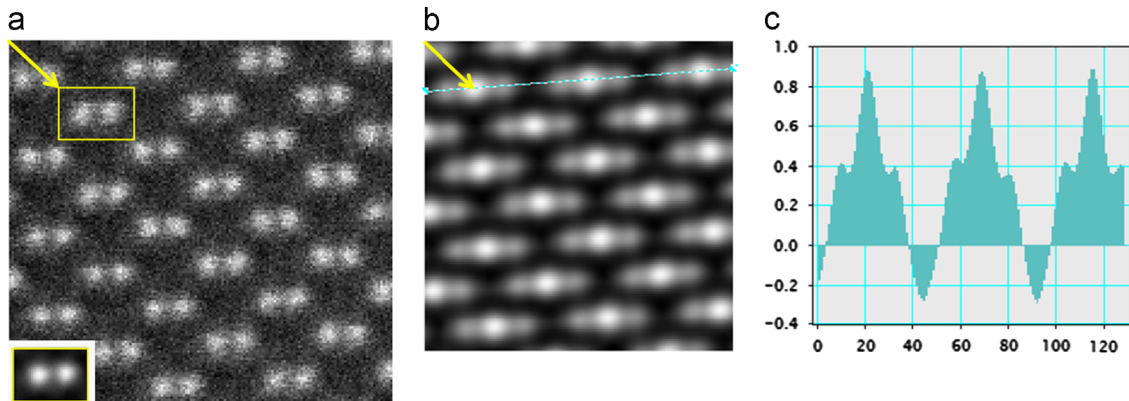


Fig. 1. An example of template matching for atomic resolution Z-contrast images: (a) the target image recorded from Si [110] using an aberration corrected STEM, (b) the calculated correlation function, (c) a profile of the correlation function taken along the line marked in (b). The template used for matching is shown at bottom-left of (a). The arrows mark the position of the correlation peak and the corresponding position of the sub-image.

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