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Novel electron microscopy method for accurate measurements of the lattice constant changes in layered structures

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In this work, we evaluate the viability of an improved stage-scan strain mapping method based on nanosized selected area electron diffraction. We demonstrate high accuracy and precision comparable with the other transmission electron microscopy methods for strain mapping. High characteristics of the stage-scan strain mapping method were achieved owning to idea of scanning the sample and acquisition of diffraction patterns under the fixed electron beam, and further post-processing of the raw data with Gaussian fitting for precise determination of the diffraction spot positions.

The rapid spread of nanotechnologies demands for the new methods of nanoscale strain mapping, because well-known macroscale methods do not provide the required spatial resolution. The use of electron diffraction and other transmission electron microscopy (TEM) techniques enables strain detection at nanoscale, however the conventional electron microscopy methods encounter difficulties narrowing their practical applications [1]. E.g. the use of convergent-beam electron diffraction and nanobeam electron diffraction methods is restricted to beam-insensitive samples due to electron beam damage, or in the case of high resolution imaging techniques either dark-field electron holography there are very strict requirements to the sample preparation. The use of selected-area (SA) electron diffraction eliminates these limitations, but arises problems of low spatial resolution and precision.

To address these issues, and extend the applicability of TEM based methods, we propose to use the stage scan mapping (SSM) technique (Fig. 1) based on the idea of two-dimensional (2D) scanning of the sample [2] and simultaneous acquisition of SA diffraction patterns at fixed beam conditions [3]. Unaltered beam enables direct comparison of the interplanar distances measured from diffraction patterns acquired at different positions of the sample. The relative interplanar distance change corresponds to the relative lattice constant changes within the scan area. Accurate determination of the relative lattice constant changes becomes possible owing to the 2D Gaussian fitting used to determine the exact position of diffraction spots [4]. High spatial resolution was reached by using a nanosized SA aperture in combination with aberration corrector [5]. Strain maps were plotted as the relative inter-planar distance variations at each scan position.

In this work we claim the viability of SSM method by evaluation of its precision or the reproducibility of the measurement, and accuracy or the degree to which the result of the strain measurement agrees with the actual value of the strain [1]. In the case of unstrained reference sample, the standard deviation of the lattice constant changes within the scan area will define a precision, and the spread of the data – accuracy of SSM technique [1].

Recently we have demonstrated the high precision of SSM technique by scanning an unstrained silicon sample [4]. At that work, we reached the best precision of 0.04%, but in many practical cases, precision was degraded especially when the spot shape elongated due to various reasons. In present work, we introduced anisotropic covariance matrix into the 2D Gaussian function used for the fitting. This change enables to fit those elongated spots more accurately and stabilizes the fitting even at low signal to noise ratio, resulting in the even data spread (accuracy) within ±0.1% and unvarying precision of 0.04% (Fig.2).

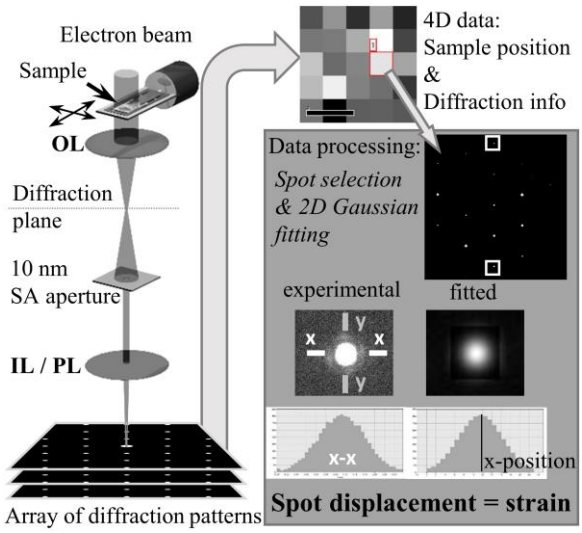

Fig. 1. Experimental setup and data processing. 2D stage scan enables the data acquisition at fixed beam conditions, thus, the distances between spots can be directly compared. Spot displacement indicates the lattice constant changes. Owning to Gaussian fitting spot positions can be determined with subpixel accuracy.

For sensitivity evaluation, we used epitaxial $SrTiO_3$ film deposited on a $SrTiO_3$ substrate by a pulsed laser deposition (PLD). The lattice constant of the film was locked to the substrate in the in-plane direction, but it was increased by 0.6% in the out-of-plane direction because of nonstoichiometry of PLD grown films [6]. The lattice constant changes were examined by SSM in both in-plane and out-of-plane directions, and plotted as 15×15 pixels maps (Fig. 3). In the in-plane direction, the average lattice constant was the same for film and substrate, but it was expanded by 0.45 ± 0.07% in out-of-plane direction, what is ~0.15% smaller than data derived from XRD spectrum. However, XRD result indicates average lattice constant through the whole thickness of the sample in a large area (ca. 5x5mm^2). Besides, lattice parameters of PLD films are not homogeneous in a large area. Thus, we can state that SSM technique can detect the local strain distribution with accuracy of ±0.1% and precision of 0.04%.

In summary, we improved the fitting stability of stage-scan strain mapping method by introducing anisotropic covariance matrix into the Gaussian function. This change makes the method a simple, accurate, and robust technique applicable to study beam sensitive battery materials.

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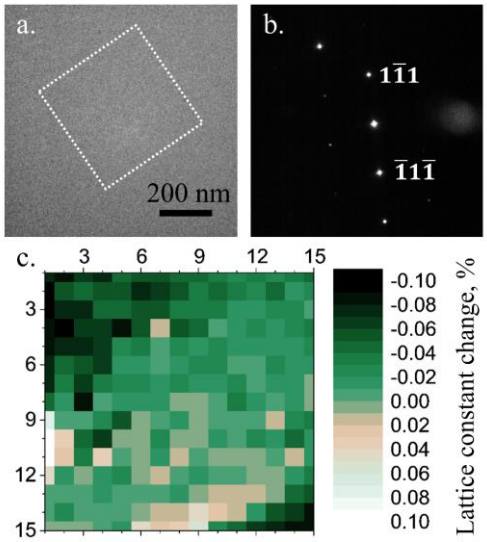

Fig. 2. Evaluation of precision on unstrained Si. a. – TEM image of the reference area with marked scan area; b. – Typical diffraction pattern. Marked spots used for the evaluation; c – Strain map. Standard deviation of the data defines the precision of SSM, spread of the data – accuracy.

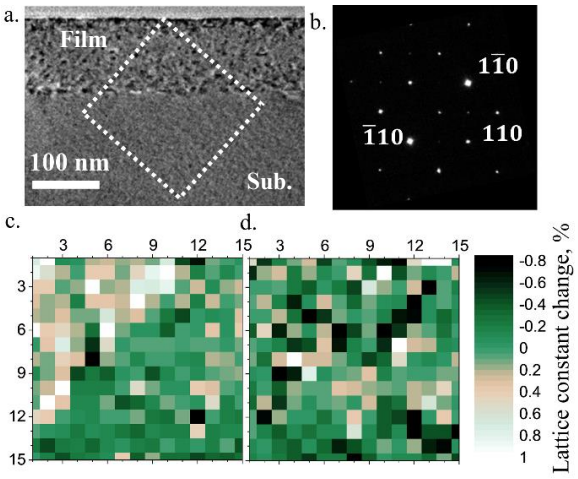

Fig.3. The sensitivity of SSM technique: a. – $SrTiO_3$ film grown onto $SrTiO_3$ substrate with marked scan area; b – Typical diffraction pattern and spots used for strain mapping; c – Out-of-plane strain map; d – In-plane strain map.

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