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Optimising multi-frame ADF-STEM for high-precision atomic-resolution strain mapping

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ABSTRACT

Annular dark-field scanning transmission electron microscopy is a powerful tool to study crystal defects at the atomic scale but historically single slow-scanned frames have been plagued by low-frequency scanning-distortions prohibiting accurate strain mapping at atomic resolution. Recently, multi-frame acquisition approaches combined with post-processing have demonstrated significant improvements in strain precision, but the optimum number of frames to record has not been explored. Here we use a non-rigid image registration procedure before applying established strain mapping methods. We determine how, for a fixed total electron-budget, the available dose should be fractionated for maximum strain mapping precision. We find that reductions in scanning-artefacts of more than 70% are achievable with image series of 20–30 frames in length. For our setup, series longer than 30 frames showed little further improvement. As an application, the strain field around an aluminium alloy precipitate was studied, from which our optimised approach yields data whos strain accuracy is verified using density functional theory.

1. Introduction

The measurement of lattice distortion and strain in materials at the nanoscale presents an important characterisation challenge. Strain is crucial in controlling the functional, electrical and mechanical properties of materials, and strain field design can unlock the engineering of new material properties, including in quantum-dots [1], semiconductor transistors [2], ferroelectrics [3], flexoelectrics [4], high-temperature superconductors [5] and alloy development [6]. High-resolution electron microscopy techniques can map strain information; however, the finite resolution and signal-to-noise ratio (SNR) of experimental images makes a quantitative measurement of these subtle displacements very challenging. Current experimental approaches include; conventional transmission electron microscopy (CTEM) [7–9], convergent-beam [10], nano-beam [11] or nano-precession [12] electron diffraction (CBED, NBED, NPED), dark-field electron holography [13] (DFEH), and annular dark-field scanning transmission electron microscopy (ADF-STEM) [14–16]. Concise reviews of these methods can be found elsewhere [17,18]; but notably, each of these methods come with their own unique complications. With CTEM, careful choice of imaging parameters must be used to avoid image artefacts or contrast inversions from (for example) sample-tilt, sample-thickness, defocus or aberration changes. CBED strain measurements can require significant sample tilt to achieve high-precision and can be vulnerable to crystal plane bending, while DFEH, NBED and NPED techniques each require additional hardware (a bi-prism, an additional condenser lens, and precession coils respectively) and more involved off-line data processing. Lastly, in STEM the serial nature of the image raster means that environmental effects can distort the image fidelity [19–21] and the necessary atomic resolution generally requires aberration correction.

Of the methods above, there has been increased interest recently in measuring strain from ADF-STEM images to make use of their Z-contrast and incoherent nature. The development of high-brightness electron sources and aberration correctors has improved image-resolution, while improvements in power-supply stability and acoustic isolation [19] have reduced problems from 50(60) Hz electrical, and few-kHz acoustic, *scan-noise* artefacts respectively [20,22]. As a result, and taking the nomenclature from [21], our attention now turns to the far harder to isolate low-frequency

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(few Hz) *scan-distortion* and unavoidable Poisson counting-noise. Strain mapping using ADF-STEM will only reach its full potential if the deleterious effects of low-frequency scanning-distortion can be minimised.

Previous scan-distortion correction efforts can be grouped into those using reference data or those using frame-averaging. Reference approaches have required instrument mode changes [23], or assumptions to be made that distortions remain constant between line-synced images recorded at different times [24,25], or within whole scan-lines of an image [16]. However, as practical environmental distortions cannot be assumed to be reproducible, such methods often retain scan-distortion artefacts even in their corrected data [16,24]. Alternative approaches using averaged multi-frame data may perform better and show increased signal-noise ratio but can have different limitations. Moire based methods surveying larger fields of view cannot visualise strain around localised defects [2,26], while simple rigid-alignment [15] or the more advanced rigid-plus-affine approaches [27], do not incorporate localised non-linear scanning-distortions [21,28]. These distortions, if not countered, then lead to a worsening of image resolution during averaging [29]. Lastly, unconstrained non-rigid registration [30] can lead to artefacts at sample edges [21] and appears to converge more slowly (Fig. 1).

In this work we do not introduce a new method of strain measurement, but rather we evaluate how to best utilise a fixed electron-dose-budget in the context of multi-frame acquisitions. We use existing methods of strain mapping from the literature to evaluate image precision, such as Fourier-space geometric phase analysis (GPA), which is fast to compute, but fundamentally not atomic-resolution [8], or real-space peak finding (atomic-resolution but more computationally intensive to extract) [14];. Non-rigid registration of multi-frame ADF data is performed using the Smart Align algorithm [21]; this approach does not make assumptions about crystal-periodicity or crystal-orientation, does not require that scanning-distortions remain constant across whole scan-frames or scan-lines, is able to incorporate non-linear distortion correction, and is robust to sample edges and local defects.

The dose-fractionation optimisation is evaluated using a model $SrTiO_3$ image series before it is deployed to obtain high-quality ADF images of an AlMgSi precipitate. These are then used to demonstrate the ability to extract strain data that is directly comparable with density functional theory (DFT) calculations. The experiment-design method presented here for spatial-precision optimisation is general across various STEM detector geometries; including, medium-angle dark-field (MAADF) [31], annular bright-field (ABF) [32], or even spectrum-imaging time-series [33].

2. Optimising dose fractionation

A collection of ADF-STEM image series were recorded from a [110] oriented crystal of $SrTiO_3$ (STO) with a total fixed dose, but differently fractionated across increasing numbers of frames. Imaging was performed using a double aberration-corrected JEOL ARM-200CF; for the STO sample the acceleration was set to 200 kV to obtain a high spatial resolution. Total electron-dose was maintained by varying the pixel dwell-time. The series recorded were for example, $1 \times 40\,\mu s$, $4 \times 10\,\mu s$, etc. through to $40 \times 1\,\mu s$. Other variables such as fly-back settling time or field of view [17] may affect the apparent strain, but these were all kept constant throughout this investigation. The data were realigned and non-rigid registered using the STEM robust mode of the Smart Align method [21] and then GPA was used to measure the apparent strain. Example ADF data recorded with differing dwell-times are shown in Fig. 1 (top row).

For a fixed total electron-dose budget we are able to record more frames when the pixel dwell-time is reduced, though the

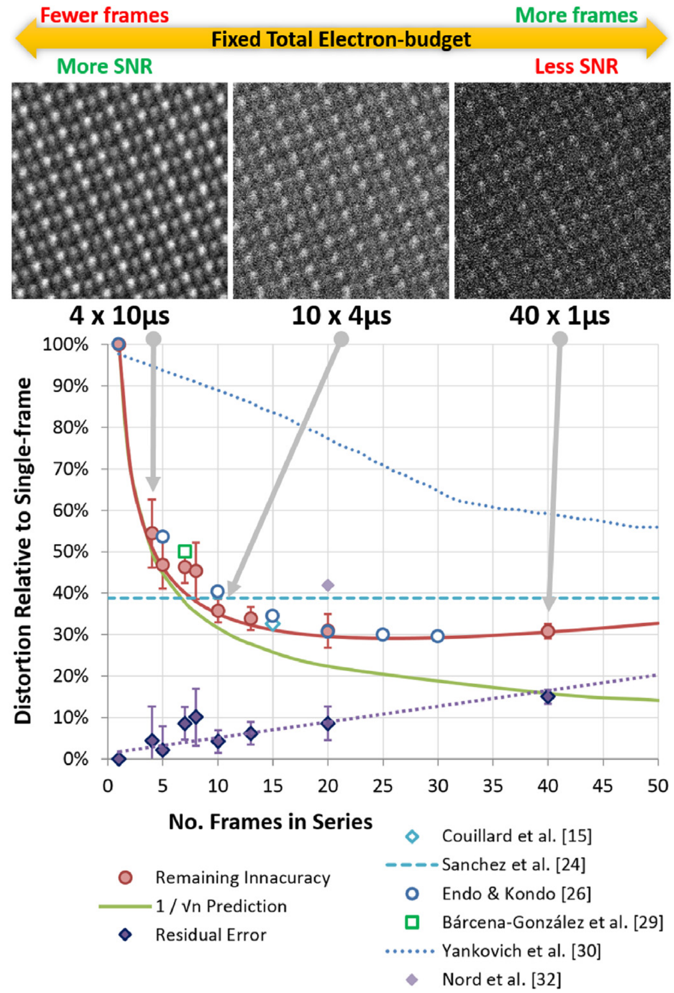

Fig. 1. Illustration of the experimental optimisation for a fixed total electron-dose; top row shows 200 × 200 px (1.84 nm^2) crops from example frames from ADF-series of [110] STO with dwell times of 10 µs, 4 µs, and 1 µs. Bottom, the distortion magnitude relative to a single 40 µs dwell-time slow-scanned frame. Filled circles show the reduction in relative scan-distortion with increasing number of STEM frames (fixed total dose). Five additional reference data are also shown normalised from references [15,24,26,29,30,32]. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

SNR of each of these individual frames is necessarily reduced. After each ADF-series has been non-rigidly aligned and averaged, ε_{yy} plots were calculated using GPA. Because of the inherent Fourier-filtering of GPA, GPA is highly robust to Poissonian noise, and is hence a robust and useful tool here for analysing the distortions across the varying dose-series. Fig. 2a) shows an example of such a plot for a conventional single scan with a 40 µs dwell-time. The strain in this large defect-free single crystal should necessarily be zero, as a result the ε_{yy} plot (which scrutinises the slow-scan direction) acts as a very sensitive metric of any imaging distortions present. As this ε_{yy} plot is purely a measure of scanning distortion, it does not depend on crystal orientation.

For each frame of the individual ADF scans in each series, the average standard deviation of ε_{yy} was measured to be 2.11% with no trend observed with respect to scan-speed (see supplemental information at [**URL**]). This is effectively the baseline measure of the combined stability of the microscope, sample, and EM-suite. The average ε_{yy} standard deviation of 2.11% reflects the environmental conditions present of the day of the experiments; from this we would expect genuine strains of less than 2% to be difficult to observe over these artefacts.