Refinements for Bragg coherent X-ray diffraction imaging: electron backscatter diffraction alignment and strain field computation

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Bragg coherent X-ray diffraction imaging (BCDI) allows the 3D measurement of lattice strain along the scattering vector for specific microcrystals. If at least three linearly independent reflections are measured, the 3D variation of the full lattice strain tensor within the microcrystal can be recovered. However, this requires knowledge of the crystal orientation, which is typically attained via estimates based on crystal geometry or synchrotron microbeam Laue diffraction measurements. Presented here is an alternative method to determine the crystal orientation for BCDI measurements using electron backscatter diffraction (EBSD) to align Fe–Ni and Co–Fe alloy microcrystals on three different substrates. The orientation matrix is calculated from EBSD Euler angles and compared with the orientation determined using microbeam Laue diffraction. The average angular mismatch between the orientation matrices is less than ~6°, which is reasonable for the search for Bragg reflections. The use of an orientation matrix derived from EBSD is demonstrated to align and measure five reflections for a single Fe–Ni microcrystal via multi-reflection BCDI. Using this data set, a refined strain field computation based on the gradient of the complex exponential of the phase is developed. This approach is shown to increase accuracy, especially in the presence of dislocations. The results demonstrate the feasibility of using EBSD to pre-align BCDI samples and the application of more efficient approaches to determine the full lattice strain tensor with greater accuracy.

1. Introduction

Bragg coherent X-ray diffraction imaging (BCDI) allows 3D nanoscale strain measurements, with a typical spatial resolution of a few tens of nanometres and a strain resolution of the order of ~2 × 10⁻⁴ (Hofmann et al., 2017b). BCDI has been applied to study crystal defects and lattice strain in a variety of materials, including noble metals (Robinson et al., 2001), alloys (Kawaguchi et al., 2021), geological compounds (Yuan et al., 2019), semiconductors (Lazarev et al., 2018) and functional materials (Dzhigaev et al., 2021). An advantage of using BCDI is the ability to study 3D volumes up to 1 μm in size under ambient conditions. This has enabled BCDI to become an essential tool for probing how lattice strains evolve in \textit{in situ} and \textit{operando} studies, for example in battery charging (Singer et al., 2018), thermal diffusion (Estandarte et al., 2018), dissolution (Clark et al., 2015) and catalytic oxidation (Carnis et al., 2021).
BCDI involves fully illuminating a crystalline sample with a coherent X-ray beam and positioning the diffractometer such that the Bragg condition is met for a specific hkl reflection. The outgoing wavevector produces a diffraction pattern that is collected on a pixellated area detector positioned in the far field (Fraunhofer regime). By rotating the sample through the Bragg condition, a 3D coherent X-ray diffraction pattern (CXDP) is recorded as different parts of the 3D Bragg peak sequentially intersect the Ewald sphere in reciprocal space, which is projected onto the detector. If the CXDP is oversampled by at least twice the Nyquist frequency (Sayre, 1952), iterative phase retrieval algorithms can be used to recover the phase (Fienup, 1982). The amplitude and phase in reciprocal space are related to the real-space object via an inverse Fourier transform (Miao & Sayre, 2000) followed by a space transformation from detector conjugated space to orthogonal laboratory or sample space (Yang et al., 2019; Maddali et al., 2020; Li et al., 2020). The real-space amplitude \( \rho(\mathbf{r}) \), where \( \mathbf{r} \) is the position vector, is proportional to the effective electron density of the crystalline volume associated with the particular crystal reflection. The real-space phase \( \psi(\mathbf{r}) \) corresponds to the projection of the lattice displacement field \( \mathbf{u}(\mathbf{r}) \) onto the Bragg vector \( \mathbf{Q}_{hkl} \) of a specific hkl crystal reflection,

\[
\psi_{hkl}(\mathbf{r}) = \mathbf{Q}_{hkl} \cdot \mathbf{u}(\mathbf{r}).
\]

Since the development of BCDI in the early 2000s, most experiments have featured the measurement of a single reflection, providing only one component of the strain tensor. However, the analysis of a single strain component can be ambiguous as different information is obtained for different reflections (Yang et al., 2021). If at least three linearly independent reflections are measured, the full 3D strain tensor can be calculated. Before 2017, only three experiments (Beitra et al., 2010; Newton et al., 2010; Ulvestad et al., 2015) reported measuring more than one reflection on a single crystal. This is not surprising, as multi-reflection BCDI (MBCDI) experiments require prior knowledge of the crystal orientation (Newton et al., 2010) or the scanning of extensive volumes of reciprocal space until two reflections are found, upon which further reflections can then be located.

The development of a microbeam Laue X-ray diffraction pre-alignment procedure in 2017 (Hofmann et al., 2017a) enabled the direct determination of the crystal orientation matrix, such that crystals could be reliably pre-aligned for MBCDI. Recently, a double-bounce Si(111) monochromator that allows Laue X-ray diffraction to be performed has been commissioned on the BCDI beamline 34-ID-C at the Advanced Photon Source (APS), Argonne National Laboratory, USA (Pateras et al., 2020). Another method to determine the orientation of a sample is by indexing pole figures (Richard et al., 2018), but this method requires a Bragg peak with known Miller indices to be found. The indexing is performed using texture analysis and relies on the samples being well faceted to produce truncation rods in reciprocal space that are perpendicular to the facet surfaces. These pre-alignment protocols have not only led to the increased popularity of MBCDI for determination of the full strain tensor with respect to an arbitrary reference (Yang et al., 2022; Hofmann et al., 2017b, 2018, 2020; Phillips et al., 2020) but also enabled simultaneous multi-Bragg-peak phase retrieval procedures to increase reconstruction quality (Newton, 2020; Gao et al., 2021; Wilkin et al., 2021).

Here we present an alternative method of pre-determining crystal orientation for MBCDI alignment without relying on synchrotron X-rays. We use electron backscatter diffraction (EBSD) to determine the orientation (Adams et al., 1993) of randomly oriented Fe–Ni and Co–Fe microcrystals on three different sapphire substrates. EBSD instruments are much more widespread and accessible than synchrotron instruments and can be used as a valuable pre-screening tool for BCDI. EBSD measurements can produce 2D orientation maps with a high spatial resolution of \( \sim10 \) nm, thus enabling the selection of specific crystals with particular orientations or features such as twin domains. This allows the user to preserve synchrotron beamtime for BCDI measurements rather than performing...
pre-orientation measurements and analysis on the beamline. We compare orientation matrices found by EBSD with those measured by microbeam Laue diffraction and the ultimately measured reflection positions in MBCDI. Using the pre-determined EBSD orientation matrix, we measured five crystal reflections for an Fe–Ni microcrystal (Fig. 7, Section 3.2) and determined its full strain and rotation tensors with respect to the average structure of the crystal. We also implement an alternative approach using the complex component of the phase, rather than the phase alone, for the strain tensor determination and the more accurate interpolation of the recovered phase to sample coordinates.

2. Experimental methodology

2.1. Microcrystal fabrication

Samples were produced by sputter deposition of a thin film onto a single-crystal sapphire wafer (C-plane orientation). One substrate with a film thickness of 375 nm was produced for the Fe–Ni microcrystals. It was dewetted in a vacuum furnace purged with a gas mixture of 5% hydrogen, balance argon, at 1523 K for 24 h. The resulting crystals exhibit a face-centred cubic (f.c.c.) structure, range from 0.5 to 1.5 μm in size (Fig. 1(a)) and adhere to the substrate surface. The substrate was cleaved to make substrates 1 and 2, both containing Fe–Ni microcrystals. Substrate 3 contained Co–Fe microcrystals that were produced in a similar way. The procedure and details for substrate 3 can be found elsewhere (Yang et al., 2022).

Each substrate was coated with 10 nm of amorphous carbon via thermal evaporation using a Leica ACE600 coater to assist with scanning electron microscopy (SEM) imaging. To facilitate reliable measurement of multiple reflections from a specific microcrystal, a ZEISS NVision 40 Ga focused ion beam (FIB) instrument was used to remove the surrounding crystals within a 40 μm radius using currents from 6 nA to 150 pA and an acceleration voltage of 30 kV. Only SEM imaging was used to position the FIB milling scans to prevent large lattice strains caused by FIB imaging (Hofmann et al., 2017b). The isolated crystals on each substrate are shown in Fig. 2. Crystal 1B [Fig. 1(b)] was used for the computation of the strain and rotation tensors (Fig. 7).

Energy-dispersive X-ray spectroscopy (EDX) was used to determine the elemental composition of each crystal [Fig. 1(c)]. EDX showed a homogeneous distribution of all elements throughout the dewetted crystals (Fig. 3). EDX was performed on a ZEISS Merlin instrument using an Xmax 150 detector (Oxford Instruments) with an elliptical region encapsulating crystal 2B on the substrate for 16 s with an accelerating voltage of 10 kV.

2.2. Electron backscatter diffraction

Crystal orientation was determined by EBSD using a ZEISS Merlin instrument equipped with a Bruker Quantax EBSD system and a Bruker eFlash detector tilted at 4°. Electron backscatter patterns (EBSPs) were recorded with the sample tilted at 70° (Fig. 4) using an accelerating voltage of 30 kV and a current of 15 nA. The EBSPs were 800 x 600 pixels and a step size of 19.8 nm was used between consecutive points on the sample. The diffraction patterns were indexed and the Euler angles extracted for each pattern using the Bruker ESPRIT 2.1 EBSD software. The Euler angles were exported and analysed using MTEX, a MATLAB toolbox for texture analysis (Bachmann et al., 2011), to produce inverse pole figure (IPF) maps for all crystals (Fig. 2).

Here we use the Bunge convention (Bunge, 1982) to describe each crystal orientation (crystal frame) relative to the substrate (sample frame). The crystal orientation matrix \( UB \) is composed of \( U \), which describes the rotation of the crystal reference frame, and \( B \) [equation (2)], which characterizes the unit-cell parameters.

Using the same convention as Britton et al. (2016), the unit cell has vectors \( a, b \) and \( c \) with lengths \( a, b \) and \( c \), respectively. Angle \( \alpha \) describes the angle between \( b \) and \( c \), \( \beta \) the angle between \( c \) and \( a \), and \( \gamma \) the angle between \( a \) and \( b \). \( B \) is used to transform the base vectors to Cartesian base vectors:
Several different coordinate frames are used in EBSD that refer to different aspects of the measurement. In this paper, all coordinate systems and rotation matrices will be right handed and we will use the same notation as Britton et al. (2016). For EBSD, the following subscripts describe specific coordinate systems:

(i) ‘d’ is the detector frame that describes the EBSPs.

(ii) ‘s’ is the EBSD sample frame that is related to the detector frame by sample (θ_sample) and detector (θ_detector) tilts about the x axis. The x_s and y_s axes correspond to the directions of EBSD scan points.

(iii) ‘m’ is the SEM map frame. This corresponds to how the EBSPs overlay on the SEM maps and thus how the EBSD orientation is referenced.

The EBSD software returns the orientation matrix from EBSD measurements in the SEM map frame (Fig. 4). The corresponding orientation matrix is referred to as UB_{EBSD, m}. It can be constructed from a series of rotations using Euler angles, where each angle describes a rotation about a coordinate axis. Here we use right-handed rotation matrices to describe vector rotations about the x, y, and z axes,

\[ R_x(\theta) = \begin{bmatrix} 1 & 0 & 0 \\ 0 & \cos(\theta) & -\sin(\theta) \\ 0 & \sin(\theta) & \cos(\theta) \end{bmatrix}, \]

\[ R_y(\theta) = \begin{bmatrix} \cos(\theta) & 0 & \sin(\theta) \\ 0 & 1 & 0 \\ -\sin(\theta) & 0 & \cos(\theta) \end{bmatrix}, \]

and the z axis,

\[ R_z(\theta) = \begin{bmatrix} \cos(\theta) & -\sin(\theta) & 0 \\ \sin(\theta) & \cos(\theta) & 0 \\ 0 & 0 & 1 \end{bmatrix}. \]

and Bunge-convention Euler angles, \( \phi_1, \Phi \) and \( \phi_2 \) (Britton et al., 2016). Equivalently, equations (5) and (6) correspond to left-handed rotations through angle \( \theta \) for the coordinate system. To transform vectors from the crystal coordinate frame to the laboratory frame, we use (Britton et al., 2016)

\[ \mathbf{U}_{EBSD,m}^T = R_z(-\phi_2) R_y(-\Phi) R_x(-\phi_1). \]

We note that the Euler angles in equation (7) are negative because the original angles as defined are for left-handed rotation matrices. First a rotation of \( -\phi_1 \) is applied about the original z axis, followed by a rotation of \( -\Phi \) about the new x axis and finally a rotation of \( -\phi_2 \) about the new z axis. For consistency with the convention used here (Busing & Levy, 1967), we express equation (7) as

\[ \mathbf{U}_{EBSD,m} = R_x(\phi_1) R_y(\Phi) R_z(\phi_2). \]

Here, the EBSD software already accounts for the instrument tilts \( \theta_{sample} \) and \( \theta_{detector} \) and returns Euler angles in the SEM map frame (subscript m) (Fig. 4).

To input the orientation matrix into the spec orientation calculator on beamline 34-ID-C, we must define two \( hkl \) reflections, \( \mathbf{H}_{hkl} \), in laboratory coordinates (Busing & Levy, 1967),

\[ \mathbf{H}_{hkl} = \mathbf{U} \begin{bmatrix} h \\ k \\ l \end{bmatrix}. \]

Several different coordinate frames are used in EBSD that refer to different aspects of the measurement. In this paper, all coordinate systems and rotation matrices will be right handed and we will use the same notation as Britton et al. (2016). For EBSD, the following subscripts describe specific coordinate systems:

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(ii) ‘s’ is the EBSD sample frame that is related to the detector frame by sample (\( \theta_{sample} \)) and detector (\( \theta_{detector} \)) tilts about the x axis. The x_s and y_s axes correspond to the directions of EBSD scan points.

(iii) ‘m’ is the SEM map frame. This corresponds to how the EBSPs overlay on the SEM maps and thus how the EBSD orientation is referenced.

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and the z axis,

\[ R_z(\theta) = \begin{bmatrix} \cos(\theta) & -\sin(\theta) & 0 \\ \sin(\theta) & \cos(\theta) & 0 \\ 0 & 0 & 1 \end{bmatrix}. \]

and Bunge-convention Euler angles, \( \phi_1, \Phi \) and \( \phi_2 \) (Britton et al., 2016). Equivalently, equations (5) and (6) correspond to left-handed rotations through angle \( \theta \) for the coordinate system. To transform vectors from the crystal coordinate frame to the laboratory frame, we use (Britton et al., 2016)

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We note that the Euler angles in equation (7) are negative because the original angles as defined are for left-handed rotation matrices. First a rotation of \( -\phi_1 \) is applied about the original z axis, followed by a rotation of \( -\Phi \) about the new x axis and finally a rotation of \( -\phi_2 \) about the new z axis. For consistency with the convention used here (Busing & Levy, 1967), we express equation (7) as

\[ \mathbf{U}_{EBSD,m} = R_x(\phi_1) R_y(\Phi) R_z(\phi_2). \]

Here, the EBSD software already accounts for the instrument tilts \( \theta_{sample} \) and \( \theta_{detector} \) and returns Euler angles in the SEM map frame (subscript m) (Fig. 4).

To input the orientation matrix into the spec orientation calculator on beamline 34-ID-C, we must define two \( hkl \) reflections corresponding to out-of-plane [equation (13)] and in-plane [equation (14)] reflections (Hofmann et al., 2017a). One concern is the consistency in the indexing of crystals on the 34-ID-E Laue instrument and in EBSD measurements. Due to the cubic structure of the present crystals, there are equivalent orientation matrices that differ by 90° rotations about the crystal axes. These rotations are accounted for using an \( \mathbf{R}_{crystal} \) rotation matrix that captures rotations by \( 90(n_x, n_y, n_z) \), where \( n_x, n_y, n_z \in \{-1, 0, 1\} \), about the x, y and z axes,

\[ \mathbf{R}_{crystal} = \mathbf{R}_{x,90(n_y)} \mathbf{R}_{y,90(n_x)} \mathbf{R}_{z,90(n_z)}. \]

To align the EBSD map frame to the BCDI laboratory frame, a \( -90° \) rotation about the x axis is required [Fig. 5(a)]. Combining this with equations (8) and (9) leads to the formation of the UB_{34C,EBSD} matrix,

\[ \mathbf{UB}_{34C,EBSD} = \mathbf{R}_x(-90°) \mathbf{U}_{EBSD,m} \mathbf{R}_{crystal} \mathbf{B}. \]

These matrix-based orientation operators provide a generalized approach to the transformation of orientation, irrespective of the software implementation.
2.3. Microbeam Laue X-ray diffraction

Microbeam Laue diffraction was used to verify the lattice orientation of each crystal independently. This was performed on beamline 34-ID-E at the APS. Further details about the instrument can be found elsewhere (Liu et al., 2004; Hofmann et al., 2017a).

The sample was positioned with its surface inclined at a 45°/C14 angle to the incident beam [see Fig. 5(b)] and diffraction patterns were recorded using a Perkin–Elmer flat-panel detector above the sample. 2D fluorescence measurements of the Fe Kα1 peak (6.40 keV) were used to identify the spatial position of the crystals, using a monochromatic 17 keV (horizontal × vertical) X-ray beam focused to 0.25 × 0.25 μm using Kirkpatrick–Baez (KB) mirrors.

Next, a polychromatic X-ray beam was used to collect a Laue diffraction pattern of each crystal (Fig. 6). The pattern shows weak Bragg reflections from the microcrystals and strong Bragg peaks from the single-crystal sapphire substrate. The two sets of peaks were indexed and fitted using the LaueGo software (https://www.aps.anl.gov/Science/Scientific-Software/LaueGo). From the indexing, we could determine the UB matrix [equation (4)]. The UB matrix determined by Laue diffraction on the 34-ID-E instrument is referred to as UBLaue,

\[
UB_{\text{Laue}} = \begin{bmatrix}
a^* & b^* & c^*
\end{bmatrix},
\]

where \(a^*, b^*\) and \(c^*\) are the column (represented by vertical lines) reciprocal-space vectors returned by LaueGo in units of nm⁻¹ in the 34-ID-E laboratory frame.

To convert \(UB_{\text{Laue}}\) into a UB matrix for use on the BCDI instrument 34-ID-C, \(UB_{34\text{C},\text{Laue}}\), the 45°/C14 rotation of the sample in the Laue laboratory frame must be accounted for [Fig. 5(b)]. To align the microbeam Laue and BCDI laboratory frames, a rotation of the sample by 45° about the x axis is required (Hofmann et al., 2017a), leading to

\[
UB_{34\text{C},\text{Laue}} = R_x(45°) UB_{\text{Laue}},
\]

where \(R_x(45°)\) is the rotation matrix about the x axis.

2.4. Bragg coherent X-ray diffraction imaging

BCDI was performed on beamline 34-ID-C at the APS. An in situ confocal microscope was used to position the microcrystal within the X-ray beam (Beitra et al., 2010). The sample was illuminated using a 9 keV (\(\lambda = 0.138\) nm) coherent X-ray beam, with a bandwidth of \(\Delta \lambda/\lambda \simeq 10^{-4}\), from an Si(111) monochromator. The X-ray beam was focused to a size of 1.1 × 1.1 μm (horizontal × vertical, FWHM) using KB mirrors. Beam-defining slits were used to select the coherent portion of the beam at the entrance to the KB mirrors. For beamline 34-ID-C, the transverse coherence length is \(\xi_h \geq 10\) μm and the longitudinal coherence length is \(\xi_w \simeq 0.7\) μm at a photon energy of 9 keV (Leake et al., 2009).

The sample needs to be positioned such that a specific \(hkl\) Bragg diffraction condition is met to produce a diffraction pattern in the far-field Fraunhofer regime. The orientation
matrix determined by EBSD or Laue diffraction is communicated to the spec software used on 34-ID-C by defining two reflections that correspond to hkl values associated with laboratory x (in-plane) and y (out-of-plane) directions [note the angles referred to here are set elsewhere (Hofmann et al., 2017a)]:

(i) The primary reflection (out-of-plane, y direction), \( \mathbf{H}_y \), where the instrument angles are set to \( \delta_{\text{spec}} = 0^\circ \), \( \gamma_{\text{spec}} = 20^\circ \), \( \theta_{\text{spec}} = 0^\circ \), \( \chi_{\text{spec}} = 90^\circ \) and \( \phi_{\text{spec}} = -10^\circ \). The fractional \([h\ k\ l]^T\) is then
\[
\begin{bmatrix}
    h \\
    k \\
    l
\end{bmatrix}
= (\mathbf{UB})^{-1} \mathbf{H}_y = (\mathbf{UB})^{-1} \begin{bmatrix}
    0 \\
    1 \\
    0
\end{bmatrix}.
\]

(ii) The secondary reflection (in-plane, x direction), \( \mathbf{H}_x \), where the instrument angles were set to \( \delta_{\text{spec}} = 20^\circ \), \( \gamma_{\text{spec}} = 0^\circ \), \( \theta_{\text{spec}} = 10^\circ \), \( \chi_{\text{spec}} = 90^\circ \) and \( \phi_{\text{spec}} = 0^\circ \). The fractional \([h\ k\ l]^T\) is then
\[
\begin{bmatrix}
    h \\
    k \\
    l
\end{bmatrix}
= (\mathbf{UB})^{-1} \mathbf{H}_x = (\mathbf{UB})^{-1} \begin{bmatrix}
    1 \\
    0 \\
    0
\end{bmatrix}.
\]

Here \( \mathbf{UB} \) refers to \( \mathbf{UB}_{\text{spec, EBSD}} \) or \( \mathbf{UB}_{\text{spec, Laue}} \). These two fractional hkl vectors are then entered into spec as known reflections. On this basis, the expected angular positions of the \([111]\) and \([200]\) reflections from the sample were calculated. Not all \([111]\) and \([200]\) reflections could be measured as some may exceed the angular range of the sample and detector motors. Occasionally, manual motor adjustments of a few degrees were required to locate the Bragg peak. Once a Bragg peak was found, the sample tilt and positioning were refined such that the centre of mass was on the centre of the detector positioned 0.5 m away from the sample. At this aligned position, the angular and translational positions were saved for each Bragg peak and used to determine the true beamline orientation matrix \( \mathbf{UB}_{\text{spec}} \) by minimizing the least-squares error associated with the measured reflections,
\[
\min \sum_{hkl} \| \mathbf{UB}_{\text{spec}}[h\ k\ l]^T - \mathbf{H}_{hkl} \|^2,
\]
where \([h\ k\ l]^T\) are the Miller indices in crystal coordinates.

Differences between the true and predicted positions of the reflections using \( \mathbf{UB}_{\text{spec}} \) arise from a number of different sources. The largest error is the repeatability of the sample position in different coordinate systems. The use of Thorlabs 1X1 kinematic mounts, which have angular errors of less than a millidegree, helps with the precise re-mounting of samples. There is also uncertainty in the goniometer precision and alignment with the diffractometer, which can influence the angle readout. Furthermore, the centre of the detector may not be perfectly aligned to the calculated position for a given detector distance or angle. The position of the measured Bragg peak is limited by the energy resolution of the incident X-ray as it affects the Bragg angle.

CXDPs were collected on a 256 × 256 pixel module of a 512 × 512 pixel Timepix area detector (Amsterdam Scientific Instruments) with a GaAs sensor and a pixel size of 55 × 55 µm positioned 1.0 m from the sample to ensure oversampling. CXDPs were recorded by rotating the crystal through an angular range of 0.6° and recording an image every 0.005° with 0.1 s exposure time and 50 accumulations at each angle.

To optimize the signal-to-noise ratio and increase the dynamic range of the CXDPs, three repeated scans for each of the \([111], [200], [020]\) and \([002]\) reflections were performed and aligned to maximize their cross correlation. Once aligned, the minimum acceptable Pearson cross correlation for summation of CXDPs from a specific Bragg reflection was chosen to be 0.976, similarly to previous BCDI studies (Hofmann et al., 2018, 2020). CXDPs were corrected for dead time, dark field and white field prior to cross-correlation alignment. Details regarding the recovery of the real-space images using phase retrieval algorithms are given in Appendix A and the computation of the strain is given in Appendix B.

2.5. Sample mounting

For the SEM, EDX and EBSD analyses, samples were mounted on 12.5 mm diameter SEM specimen pin stubs using silver paint. For microbeam Laue X-ray diffraction, they were attached to a Thorlabs 1X1 kinematic mount. From here, a Thorlabs kinematic mount adapter between 34-ID-E and 34-ID-C was used to mount the samples for BCDI. This adapter consists of two 1X1 mounts sandwiched together to enable sample orientation to be well preserved between the beamlines. There is no kinematic mount adapter between the SEM and BCDI instruments. Moreover, the use of magnets in the kinematic mounts inhibits their use for electron microscopy. Across the different instruments, the sample orientation was maintained throughout as shown in Fig. 5, which has an arbitrary sample feature to illustrate the respective orientations.

3. Results and discussion

3.1. Orientation matrix comparison

The angular mismatch between two \( \mathbf{UB} \) matrices, \( \mathbf{UB}_1 \) and \( \mathbf{UB}_2 \), can be determined by converting \( \mathbf{UB}_i(\mathbf{UB}_j)^{-1} \) into a rotation vector. A rotation matrix \( \mathbf{R} \) can be converted using Rodrigues’ rotation formula in matrix exponential form,
\[
\mathbf{R} = \exp(\mathbf{w}_m),
\]
where \( \mathbf{w}_m \) is an antisymmetric matrix,
\[
\mathbf{w}_m = \begin{bmatrix}
    0 & -w_z & w_y \\
    w_z & 0 & -w_x \\
    -w_y & w_x & 0
\end{bmatrix},
\]
which contains the elements of the rotation vector \( \mathbf{w} = [w_x, w_y, w_z]^T \). The rotation vector is defined by a rotation axis \( \mathbf{w} \) multiplied by a rotation \( \theta \). If the two orientation matrices are different, \( \mathbf{UB}_i(\mathbf{UB}_j)^{-1} \) can be converted to a rotation vector where the angular mismatch is \( \theta \). If \( \mathbf{UB}_1 = \mathbf{UB}_2 \), then \( \mathbf{UB}_i(\mathbf{UB}_j)^{-1} = I_3 \) and therefore \( \theta = 0 \).
Table 1

Angular differences (°) between $UB_{34C}$, $UB_{34C, Laue}$ and $UB_{34C, EBSD}$ for substrate 1.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$UB_{34C}$</th>
<th>$UB_{34C, Laue}$</th>
<th>$UB_{34C, EBSD}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1B</td>
<td></td>
<td>9.72</td>
<td>11.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.37</td>
<td></td>
</tr>
<tr>
<td>1C</td>
<td></td>
<td>2.22</td>
<td>5.35</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3.28</td>
<td></td>
</tr>
<tr>
<td>1E</td>
<td></td>
<td>9.70</td>
<td>11.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.97</td>
<td></td>
</tr>
<tr>
<td>1F</td>
<td></td>
<td>7.14</td>
<td>9.56</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3.05</td>
<td></td>
</tr>
<tr>
<td>Average</td>
<td>$UB_{34C}$</td>
<td>7.19</td>
<td>9.23</td>
</tr>
<tr>
<td></td>
<td>$UB_{34C, Laue}$</td>
<td>2.67</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$UB_{34C, EBSD}$</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Here we set $UB_1$ and $UB_2$ as $UB_{34C, EBSD}$, $UB_{34C, Laue}$ or $UB_{34C}$. First we rearrange equation (16) to calculate $w_m$:

$$w_m = \log([UB_1(UB_2)^{-1}], \quad (18)$$

where log here refers to the matrix natural logarithm. Next we reconstruct the rotation vector using equation (17) and calculate its magnitude to obtain the mismatch,

$$\theta = ||w|| = \|([w_m(3, 2), w_m(1, 3), w_m(2, 1)]\|. \quad (19)$$

To calculate the angular mismatch between $UB_{34C, EBSD}$ and other orientation matrices, the permutation of $n_{x,y,z}$ that produced the smallest error was chosen. Tables 1–3 show the angular mismatch of $UB_{34C, Laue}$ and $UB_{34C, EBSD}$ compared with $UB_{54C}$ for substrates 1–3. The average angular mismatch for all crystals between orientation matrices $UB_{34C, Laue}$ and $UB_{34C}$ is 4.48°, that between $UB_{34C, EBSD}$ and $UB_{34C}$ is 6.09°, and that between $UB_{34C, Laue}$ and $UB_{34C, EBSD}$ is 1.95°.

Generally, $UB_{34C, Laue}$ is most similar to $UB_{34C}$. This is expected, as there is a Thorlabs kinematic mount adapter between 34-ID-E and 34-ID-C for the precise angular alignment of samples. A larger difference is observed when comparing $UB_{34C, EBSD}$ and $UB_{34C}$. This is due to the manual removal of the SEM pin stub from the electron microscope, which then needs to be re-secured to the kinematic mount. The cylindrical pin permits a greater degree of rotational freedom, thereby increasing the angular mismatch when $UB_{34C, EBSD}$ is considered. Despite this increased angular freedom in the pin, a 2° increase in the angular uncertainty when using $UB_{34C, EBSD}$ instead of $UB_{34C, Laue}$ is still a very accurate result. This means only a slightly larger angular range needs to be explored in alignment.

The alignment of crystals for BCDI using EBSD remains much more time efficient because microbeam Laue diffraction pre-alignment on 34-ID-E is no longer required. The use of EBSD for pre-alignment of BCDI samples also affords greater flexibility in experiment type. The ability to pre-characterize samples offsite should substantially increase throughput and make MBCDI a much more widely accessible technique, especially on beamlines without pink-beam capability or access to a nearby Laue instrument.

3.2. Determination of strain

MBCDI allows the strain and rotation tensors to be computed if at least three reflections are measured, thus providing more information about the crystal defects present (Hofmann et al., 2017b, 2018, 2020; Phillips et al., 2020). Fig. 7 shows the strain and rotation tensors reconstructed from five measured Bragg reflections of crystal 1B. The $\epsilon_{xx}$, $\epsilon_{yy}$ and $\epsilon_{zz}$ slices show defects close to the edge that may not be resolved if analysing a single reflection alone (Yang et al., 2021), as some crystal defects, such as dislocations, are visible only when
\[ Q_{hkl} \cdot b \neq 0, \] where \( b \) is the Burgers vector (Williams & Carter, 2009).

These results were produced using a refined method for the computation of the strain and rotation tensor. A general approach for the computation of both these tensors is described in Appendix B. This relies on the recovery of the phase of the CXDP. The intensity of the CXDP is the squared magnitude of the Fourier transform \( F \) of the complex crystal electron density \( f \). The solution for the recovered phase is non-unique, as global phase offsets \( C \) can produce the same CXDP, i.e., \[ |F[f(\mathbf{r})]|^2 = |F[f(\mathbf{r}) + C]|^2. \] After phase retrieval, the phase values are bound between \([-\pi, \pi]\), which describes the periodic nature of the crystal structure but not necessarily the true complex crystal electron density. For instance, if the projected displacement in the direction of \( Q_{hkl} \) is greater than \( |Q_{hkl}| \), then a phase jump, where the phase difference is \( 2\pi \) between two pixels, will occur. These phase jumps cause discontinuities in the derivatives of the phase \( \partial \psi_{hkl}(\mathbf{r})/\partial j \), where \( j \) corresponds to the spatial \( x, y \) or \( z \) coordinate, leading to spurious large strains. Typically, phase unwrapping algorithms can be used to remove phase jumps, but dislocations have characteristic phase vortices (Clark et al., 2015) that end at dislocation lines, meaning that phase jumps associated with dislocations cannot be unwrapped.

To account for this, Hofmann et al. (2020) demonstrated an approach that involves producing two additional copies of the phase with phase offsets of \(-\pi/2\) and \(\pi/2\), respectively. This shifts the phase jumps to different locations and, by choosing the phase gradient with the smallest magnitude for each voxel, the correct phase derivatives can be found. Here, we employ a more efficient method used in coherent X-ray diffraction tomography following Guizar-Sicairos et al. (2011), which has also been applied in the Bragg geometry using ptychography (Li et al., 2021). Rather than making multiple copies of the phase, we take the derivative of the complex exponential of the phase and determine the phase gradient using the chain rule:

\[
\frac{\partial \psi_{hkl}(\mathbf{r})}{\partial j} = \text{Re} \left\{ \frac{\partial \exp[i\psi_{hkl}(\mathbf{r})]/\partial j}{i \exp(i\psi_{hkl})} \right\}.
\] (20)

Here \( \exp[i\psi_{hkl}(\mathbf{r})] \) is a circle expressed using Euler’s formula, where the phase jumps disappear. Fig. 8 shows the difference between the two methods for computing the lattice rotation and strain tensors. We can see that the procedure based on phase offsets (Hofmann et al., 2020) fails to resolve the details.
From detector conjugated space to sample space, by interpolating the complex regions. Computing phase gradients successfully deals with these central regions of missing intensity. The new approach of avoiding the blurring of phase jumps that occurs during direct methods removes the need for BCDI and Laue instrument coordination, and enables MBCDI on BCDI instruments that do not have pink-beam capability or a Laue instrument nearby. Using the orientation matrix obtained from EBSD, five reflections have been located on an Fe–Ni microcrystal and full 3D strain and rotation tensors have been recovered. When computing the tensors, we have demonstrated a more efficient approach to resolving phase jumps, by implementing a complex phase quantity to calculate and interpolate the phase. This allows for the phase to be unwrapped and the correct strain to be resolved in the vicinity of dislocations. These refinements make BCDI a more accessible microscopy tool.

5. Data availability

The processed diffraction patterns, final reconstructions and data analysis scripts, including a script to compute the orientation matrix using EBSD, are publicly available at https://doi.org/10.5281/zenodo.6383408.

Also available are three supplementary videos, as follows:
(i) SV1 shows xy plane slices through the lattice strain and rotation tensors in Fig. 7.
(ii) SV2 shows yz plane slices through the lattice strain and rotation tensors in Fig. 7.
(iii) SV3 shows zx plane slices through the lattice strain and rotation tensors in Fig. 7.

APPENDIX A
Phase retrieval

The reconstruction process of CXDPs was done independently for each reflection, in two stages, using the output from the previous stage to seed the next phasing stage (listed below). The CXDPs have a size of 256 × 256 × 128 voxels.

(i) Each reconstruction was seeded with a random guess. A guided phasing approach (Chen et al., 2007) with 40 individuals and four generations was used with a geometric average breeding mode. For each generation and population, a block of 20 error reduction (ER) and 180 hybrid input–output (HIO) iterations, with β = 0.9, was repeated three times. This was followed by 20 ER iterations to return the final object. The shrinkwrap algorithm (Marchesini et al., 2003) with a threshold of 0.1 was used to update the real-space support after every iteration. The σ values for the Gaussian kernel shrinkwrap smoothing for each generation were σ = 2.0, 1.5, 1.0 and 1.0, respectively. The best reconstruction was determined using a sharpness criterion, as it is the most appropriate metric for crystals containing defects (Ulvestad et al., 2017). The two worst reconstructions were removed after each generation.

(ii) The reconstruction was seeded with the output from stage (i). This second stage was identical to stage (i), except now for each generation and population a block of 20 ER and 180 HIO iterations, with β = 0.9, was repeated 15 times, followed by 1000 ER iterations. Here the final 50 iterates were averaged to produce the final image.

The overall average 3D spatial resolution was 35 nm. This was determined by differentiating the electron-density

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<td>(b)</td>
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Figure 9
Phases for the 111, 111, 200, 020 and 002 reflections at $z = 2.5$ nm. We compare (a) the new method of interpolating the complex quantity of the phase $\psi_c$ with (b) interpolating the phase alone $\psi_0$. (c) The difference between $\psi_c$ and $\psi_0$, $\Delta\psi$, highlights the information that can only be resolved around phase jumps in $\psi_c$. The amplitude threshold for the reconstructions is 0.30 and the size of the coordinate axes is 100 nm. Here the average morphology was used. Hence the 111 reconstruction shows a region of missing intensity corresponding to a twin, which is not present in the original 111 reconstructed morphology (Fig. 10 in Appendix A).

4. Conclusions

We have demonstrated that the orientation of various microcrystals on different substrates can be found via EBSD and used to align BCDI experiments. The results indicate a ~2° increase in angular error when using EBSD alignment compared with Laue diffraction alignment, which is still within reasonable tolerance for the search for Bragg peaks. Importantly, using EBSD to pre-align crystals allows beamtime to be more effectively utilized for BCDI data set collection. It also removes the need for BCDI and Laue instrument coordina-
amplitude across the crystal/air interface for the five reflection directions and fitting a Gaussian to each of the profiles. The reported spatial resolution is the averaged FWHM of the Gaussian profiles. The average 3D spatial resolutions were 30, 38, 39, 31 and 39 nm for the 111, 111, 200, 020 and 002 reconstructions (Fig. 10), respectively.

APPENDIX B
Strain and rotation tensor calculations
The full 3D lattice strain tensor $\varepsilon(\mathbf{r})$ and rotation tensor $\omega(\mathbf{r})$ are given by (Constantinescu & Korsunsky, 2007)

$$\varepsilon(\mathbf{r}) = \frac{1}{2} \left\{ \nabla \mathbf{u}(\mathbf{r}) + [\nabla \mathbf{u}(\mathbf{r})]^T \right\}$$

and

$$\omega(\mathbf{r}) = \frac{1}{2} \left\{ \nabla \mathbf{u}(\mathbf{r}) - [\nabla \mathbf{u}(\mathbf{r})]^T \right\},$$

which rely on the reconstruction of $\mathbf{u}(\mathbf{r})$. With a single BCDI measurement, we can determine one component of the displacement field. For MBCDI, if at least three linearly independent reflections are measured, $\mathbf{u}(\mathbf{r})$ can be determined by minimizing the least-squares error (Hofmann et al., 2017a; Newton et al., 2010),

$$E(\mathbf{r}) = \sum_{hkl} \left[ \mathbf{Q}_{hkl} \cdot \mathbf{u}(\mathbf{r}) - \psi_{hkl}(\mathbf{r}) \right]^2,$$

for every voxel in the sample. Here, we use the modified approach of Hofmann et al. (2020), in which case the squared error between phase gradients is minimized,

$$E(\mathbf{r}) = \sum_{hkl,j} \left[ \mathbf{Q}_{hkl} \cdot \frac{\partial \mathbf{u}(\mathbf{r})}{\partial j} - \frac{\partial \psi_{hkl}(\mathbf{r})}{\partial j} \right]^2,$$

where $j$ corresponds to the spatial $x$, $y$ or $z$ coordinate, to find $\nabla \mathbf{u}(\mathbf{r})$ directly for the computation of $\varepsilon(\mathbf{r})$ and $\omega(\mathbf{r})$ in equations (21) and (22), respectively.

In Fig. 7, five linearly independent reflections were measured and reconstructed to assemble the strain and rotation tensor. To assess the reliability of the MBCDI measurements and phase retrieval procedure, we calculate the strain tensor using four out of the five reflections to predict the strain projected along the scattering vector of the fifth reflection.

Figure 11
A comparison between (a) the calculated and (b) the measured strain for the 111, 111, 200, 020 and 002 reflections at $y = 2.5$ nm. The calculated strain is computed from the strain tensor determined using the other four reflections following the methodology presented in the text. (c) The measured strain subtracted from the calculated strain. The amplitude threshold for the reconstructions is 0.30 and the size of the coordinate axes is 100 nm.

Figure 12
A comparison between (a) the calculated and (b) the measured strain for the 111, 111, 200, 020 and 002 reflections at $y = 2.5$ nm. The calculated strain is computed using the strain tensor determined using all five reflections following the methodology presented in the text. (c) The measured strain subtracted from the calculated strain. The amplitude threshold for the reconstructions is 0.30 and the size of the coordinate axes is 100 nm.
gradients using the approach presented in the main text [equation (20)].

The average strain error for each reflection is computed by summing the magnitude of the difference between the calculated and measured strain and dividing the sum by the number of voxels in the morphology of each reconstruction (Fig. 11). The average strain error for each reflection (listed in parentheses) is $3.5 \times 10^{-4}$ (111), $2.1 \times 10^{-4}$ (T1), $3.3 \times 10^{-4}$ (200), $4.0 \times 10^{-4}$ (020) and $2.9 \times 10^{-4}$ (002).

If all five reflections are used to predict the strain along the scattering vector for each reflection (Fig. 12), the average strain error for each reflection (listed in parentheses) is $2.0 \times 10^{-4}$ (111), $1.2 \times 10^{-4}$ (T1), $8.2 \times 10^{-5}$ (200), $1.0 \times 10^{-4}$ (020) and $9.6 \times 10^{-5}$ (002).

The average strain error is close to the strain resolution limit for BCDI of $\sim 2 \times 10^{-4}$ (Yang et al., 2021; Hofmann et al., 2020), demonstrating that the reconstructed tensors using all five reflections are accurate. The use of all five reflections reduces the average strain error by 62% compared with the use of four reflections.

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