Simultaneous Dark- and Bright-Field X-ray Microscopy at X-ray Free Electron Lasers

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ABSTRACT

The structures, strain fields, and defect distributions in solid materials underlie the mechanical and physical properties across numerous applications. Many modern microstructural microscopy tools characterize crystal grains, domains and defects required to map lattice distortions or deformation, but are limited to studies of the (near) surface. Generally speaking, such tools cannot probe the structural dynamics in a way that is representative of bulk behavior. Synchrotron X-ray diffraction based imaging has long mapped the deeply embedded structural elements, and with enhanced resolution, Dark Field X-ray Microscopy (DFXM) can now map those features with the requisite nm-resolution. However, these techniques still suffer from the required integration times due to limitations from the source and optics. This work extends DFXM to X-ray free electron lasers, showing how the $10^{18}$ photons per pulse available at these sources offer structural characterization down to 100 fs resolution (orders of magnitude faster than current synchrotron images). We introduce the XFEL DFXM setup with simultaneous bright field microscopy to probe density changes within the same volume. This work presents a comprehensive guide to the multi-modal ultrafast high-resolution X-ray microscope that we constructed and tested at two XFELs, and shows initial data demonstrating two timing strategies to study associated reversible or irreversible lattice dynamics.
### 1 Introduction

Across materials science — from dislocation junctions strengthening materials to interstitial defects fracturing batteries over many charge cycles — defects change how materials respond to their surroundings\(^1,2\). Point defects are routinely used to finely tune material properties\(^3\), and defects extending across many unit cells (mesoscopic) can tune thermal or electron materials, among others\(^4,5\). For example, grain boundaries in bismuth selenide have been shown to create nanodomains that enhance their thermoelectric efficiency by orders of magnitude by decoupling the mean-free-paths of electrons and phonons\(^6\). Similarly, in metals grain boundaries, and dislocation networks govern bulk properties such as strength and ductility\(^7\). At this time, our understanding and control of mesoscopic defects and domains in bulk materials is primarily limited by our ability to probe their dynamics in a manner that is representative of bulk properties\(^8\). The multiscale structure often encountered implies that sample thicknesses of tens or hundreds of micrometers are required for representative sampling. Electron microscopy, field ion microscopy and atomic probe tomography can resolve defect cores with atomic resolution. However, they are intrinsically near surface probes and they rely on long raster scans to generate 3D maps, during which sample conditions are fixed\(^9,10\). Without \textit{in-situ} measurement tools that can resolve how mesoscopic defects with nanometer cores interact to form large 3D networks that evolve over hundreds of micrometers, our understanding of the dynamics has been limited to theory that is yet untested at the microscopic scale.

The primary challenge in detecting the mesoscopic structure lies in the wide range of length- and time-scales that must be probed to fully interpret the system. Lattice defects are comprised of local disruptions in the crystal packing — either a truncated plane (dislocation), missing or extra atom (vacancy, interstitial), or truncated domain of the crystal (grain boundary). While the cores of defects have Å-nm length scales, the long-range distortions from them that span micrometers to millimeters map key interactions that alter the macroscopic properties\(^11,12\). When these defects interact, the velocity of the property-transforming events can span from ballistic dynamics (ps-ns) through cumulative degradation (months to years), spanning \(> 13\) decades of timescales. A measurement tool to spatially and temporally resolve the evolution of plasticity \textit{in-situ} and, specifically, the interactions between adjacent strain or defects, requires sub-nanosecond imaging with nm-resolution\(^13,14\).

X-rays have been demonstrated to have the necessary penetration power to access this regime. X-ray microscopes can be divided into three main modalities: spectral imaging, absorption microscopy and diffraction-contrast microscopy\(^13\). Spectral imaging gives contrast that delineates the elemental composition and sometimes the oxidation state. Without a change in the photon energy, the absorption-contrast microscopes map attenuation based on the density of the material, while diffraction contrast ones map heterogeneity in the crystal structure. While the contrast mechanism defines the materials-specific information contained in X-ray images, the resolution vs field of view are set by the imaging optics. Scanning microscopy is performed by focusing the X-ray beam to a small spot, then rastering the spot across the sample to collect spatially-mapped signals; for this approach, the resolution is set by the beam’s spot size while the field of view is set by the number of points in the scan. Tomographic microscopy is also collected in a scanning modality, but with rotational scanning for a roughly cylindrical sample volume. The corresponding image stacks are compiled and Radon transformed to form the full 3D imaged volume.

By contrast, full-field imaging collects information about the entire sampling volume in a single acquisition, with projections along one of the three sample dimensions. Near-field imaging (e.g. radiography, topography) captures full-field images just behind the sample, with a field of view limited by the detector or beam size, while magnified imaging with focusing optics maps an image plane in the far-field. The information encoded in these X-ray images is dictated by the material attribute responsible for the light-matter interaction that produces the beam being imaged. Images collected along the X-ray transmitted beam (transmission X-ray microscopy, TXM) are collected along the \textit{bright-field} (BF), and map the attenuation of the beam from absorption (and XRD in special cases). Conversely, images collected along the diffracted beam are produced by the X-rays interacting with the undisrupted periodicity of a lattice plane, thereby mapping the crystallographic information along a specific symmetry of a crystal. These types of \textit{dark-field} (DF) images result in spatial maps of the disruptions to crystalline order in the lattice (i.e., the defects).

Dark-field X-ray microscopy (DFXM) is a full-field magnified microscope, collected by placing an X-ray objective lens along the X-ray diffracted beam. DFXM captures crystallographic distortions, i.e. the strain and mosaicity of the lattice, with a spatial resolution of 30-150 nm, with strain resolution of \(10^{-5}\) and mosaicity (orientation) resolution of \(10^{-3}\) rad. Recent work has used DFXM to characterize deformation texture\(^15\), dislocation structures\(^16\), domain boundary migration in ferroelectrics\(^17\), fatigue in polycrystalline metals\(^18\), among other phenomena. We recently established time-resolved DFXM at the European Synchrotron Radiation Facility (ESRF) and demonstrated its utility with a first-ever study of collective dislocation dynamics deep inside bulk aluminum from 97 – 99% of the melting temperature\(^19\). Today, DFXM has a temporal resolution that is limited by the integration time required to acquire the images with synchrotron radiation; access to material dynamics at sub-microsecond timescales requires more brilliant X-ray sources.

The time resolution of X-ray microscopy tools is dictated by the acquisition time and the type of system. For reversible and reproducible dynamics, a pump-probe modality may be used to excite a transient state into the material, then probe how that excitation drives the material then relaxes back to its ground state via millions of successive excitations and signal averaging.
By contrast, irreversible processes require single-shot acquisitions with sufficiently high frame rates and signal to noise that they may gather all the relevant information about the system in the time that follows a single excitation pulse. Full-field imaging approaches are required to study irreversible processes.

Since the development of hard X-ray free electron lasers (XFELs), ultrafast X-ray science has had breakthroughs across physics, engineering, biology and beyond as similar types of optical and synchrotron techniques have become achievable with single-shot acquisition capabilities\textsuperscript{20}. With $10^{12}$ photons per pulse, XFELs have shifted static techniques that are limited by the integration time to ultrafast pump-probe measurements where femtosecond single-shot measurements were able to overcome issues with radiation damage\textsuperscript{21}. For structural imaging, holography and X-ray coherent diffractive imaging (XCDI) have demonstrated this type of spatiotemporal resolution at XFEL sources\textsuperscript{22,23}, but the necessary apertures and foci at the sample prevent these methods from capturing sufficiently large field-of-view to capture statistical populations of nanoscale features\textsuperscript{24}. While Bragg-XCDI has been able to spatially resolve the strain fields in nanoparticles\textsuperscript{25}, the same lattice-resolution measurements have not been extended to real-space X-ray imaging. XFEL imaging in real-space (i.e. in $x, y, z$ coordinates as opposed to the Fourier transformed XCDI images) has only been performed in the transmitted beam with associated density contrast\textsuperscript{26}. This makes it insensitive to e.g. the sparse defects that initiate plastic transformations and to strain. To statistically probe the distortions that initiate large-scale material transformations, we need a technique with high spatiotemporal resolution and a large field-of-view that is sensitive to both density variation and localized strain fields inside the crystals.

To address this challenge, in this work, we introduce DFXM at XFELs and present a set-up that enable simultaneous density mapping using BFXM (in the TXM geometry). Using 32-fs x-ray pulses, local structural information is probed orders of magnitude faster than previously accessible\textsuperscript{19}. From experiments first at the the Pohang Accelerator Lab (PAL, 2019) and then at the Linac Coherent Light Source (LCLS, 2021), we present the instrumentation we have developed to build, align, acquire data, and analyze the results from the XFEL version of DFXM. To enhance the information-content of each frame and enable single-shot acquisitions of irreversible systems, we include simultaneous bright- and dark-field X-ray microscopy (BFXM & DFXM), as they afford complementary information about the strain states and defect populations in sample. In these experiments, we benchmark our measurements using diamond single crystals. We first present our instrument design, including important trade-offs that must be considered when designing XFEL-DFXM experiments. We then present the analysis approaches we used for our data analysis, and present the types of data that may be collected at each of our 4 detectors in the imaging setup. We conclude with a discussion about the future opportunities for this instrument at XFELs around the world, across many fields of science.

2 Dark-Field X-ray Microscopy Design

The DFXM geometry and direct and reciprocal space coordinate systems as used at synchrotrons are defined in Refs.\textsuperscript{27,28}. In this section, we introduce the design, coordinate systems, and axes for scanning, noting the key differences between the established synchrotron coordinate systems and the XFEL one for clarity. We use the same notation here for consistency, with one main exception related to the direct space: the XFEL is defined to follow the optical conventions that defines the $\vec{z}$ axis as the beam’s propagation direction. Given the complexity of the DFXM experiments, we detail a conversion from the synchrotron to XFEL systems here. For full understanding of how these coordinate systems convert to intensity and contrast mechanisms, see our previous work in\textsuperscript{27,29,30}. The full microscope setup in this orientation is shown in Figure 1, with diffraction in the horizontal scattering geometry, as we used at XFELs.

The analytical form to convert an arbitrary vector $\vec{v}$ from the laboratory coordinate system used in Ref.\textsuperscript{27}) into the XFEL one described in this work is

$$
\vec{v}_{\text{XEL}} = \begin{pmatrix} 0 & 0 & 1 \\ -1 & 0 & 0 \\ 0 & 1 & 0 \end{pmatrix} \vec{v}_{\text{synch}}.
$$

As shown in Figure 1, the laboratory coordinate system is defined by $\{x_l, y_l, z_l\}$ and rotations about those axes, $\{R_x, R_y, R_z\}$, defined by the Poynting vector of the incident beam, $\vec{k}_i$. When the beam reaches the sample, it interacts with the lattice planes in the relevant grain based on the orientation of the lattice. We have previously defined a crystal coordinate system to describe how the crystallographic vectors, $\vec{a}, \vec{b}, \vec{c}$ map onto the lab system using $\{x_c, y_c, z_c\}$, such that the transform matrix $M^{c\rightarrow l}$ converts any arbitrary vector from the lab to crystal system\textsuperscript{29}.

After traversing the sample, the diffracted beam propagates along a diffracted wavevector, $\vec{k}_d || z_l$, that defines the diffraction imaging coordinate system $\{x_l, y_l, z_l\}$.

With DFXM contrast is provided by minor angular offsets of the diffraction vector from the nominal value for the undeformed lattice $\vec{Q}_{hl}$ and by varying the scattering angle $2\theta$\textsuperscript{28}. The latter requires a coherent motion of the far-field (ff) detector and imaging lenses along the $2\theta$ arc. As can be seen in Fig. 1, a combined $2\theta - \phi$ scan will probe the local axial
Figure 1. (a) Schematic showing the DFXM geometry and scanning procedure, including the rotational axes, \( \chi \) (cyan), \( \phi \) (green), and \( \omega \) (magenta), the axial strain axis \( 2\Delta \theta \) (red), the orientation at the sample position \( \eta \) (yellow), and the alignment rotation axis, \( \mu \) (brown). The circular plot at the bottom illustrates how the \( \mu \) motor is required to rotate the scanning stages from their initial orientations, \( R_x \), \( R_y \), and \( R_z \) in the laboratory coordinate system, into the rotated sample coordinate system, \( \{x_s, y_s, z_s\} \) for which the diffraction vector \( Q_{hkl} \) are aligned to the \( z_s \) axis. We also illustrate (b) how the image stacks collected on points in a \( \phi \)-scan (a so-called rocking-curve scan) show the different spatial components of each displacement field, sampled by the scanning angles. The strong-beam condition for the central grain is enlarged and circled in red. Images in this stack were collected from single-crystal aluminum DFXM at the ESRF synchrotron at ID06-HXM.

| Lab System | Sample System | Crystal System | Imaging System |
|------------|---------------|----------------|---------------|
| \( \hat{k}_i = \hat{z}_i \) | \( \hat{Q}_{hkl} = \hat{z}_s \) | \( \hat{n}_{hkl} = \hat{z}_c \) | \( \hat{k}_d = \hat{z}_d \) |

Table 1. Coordinate systems to describe DFXM with relevant defining characteristics, as described in the text and in\(^{29}\).

strain. The local variation in orientation, as expressed by the pole figure of the reflection (and known as the mosaicity) is probed by varying the goniometer settings, corresponding to rotations \( R_x \) and \( R_y \). These are known as rocking and rolling scan, respectively.

The XFEL polarization and instrument constraints currently require diffraction in the horizontal scattering plane, shown in orange in Fig. 1. Since \( \hat{k}_i \) and \( \hat{k}_d \) are defined in that plane, the \( 2\theta \) value constrains a precise measurement of the \( Q \) vector defining the normal to the diffracting crystal plane. As shown by the blue plane in Fig. 2, the \{\( \omega, \chi, \phi \)\} rotation vectors that define the goniometer are explicitly defined in reference to the direction of \( \hat{Q} \) meaning that they are best defined in the sample coordinate system that is rotated about \( y_i \) by \( \theta \) with respect to the lab system. We note that the sample coordinate system \( \{x, y, z\} \) describes the motorized axes with respect to the orientation of the surface normal to the sample, while a separate crystallographic coordinate system \( \{x_c, y_c, z_c\} \) is required to account for the orientation of that crystal with respect to its \( hkl \) and \( uvw \) vectors. The components of \( M^{l\rightarrow s} \) detail the conversion from the rotations about the \{\( x, y, z \)\} \( \ell \) system into the goniometer axes, \{\( \omega, \chi, \phi \)\}, as is inspired by Busing and Levy\(^{31}\).

As in previous work, DFXM can collect image series’ along scans that sample reciprocal space while rotating the crystal about \( \omega, \chi \) or \( \phi \), or by translating the lens and detector along \( 2\theta \) and \( \eta \). It is worth noting that, while only 2 rotation stages are technically required to access the full sphere of rotation, the precision of this experiment really necessitates rotation stages about 4 axes for scanning. As described in Poulsen, et al.,\(^{27,29}\) a bottom rotation stage, \( \mu \), is required to orient the 3-axis Eulerian cradle (\( \omega, \chi, \phi \)) to align the \( \omega \) rotations to the \( Q \) vector. The vector defining the relationship between each coordinate system is given for the XFEL coordinate system in Table 1. We note that in contrast to the previous work, \( \chi \) does not rotate about the incident beam axis, \( z_i \), but instead rotates about an axis that is rotated by an angle of \( \theta \) with respect to \( \hat{k}_i \), to ensure the rotations occur about the principal axes of reciprocal space.
3 Experimental Methods

The experimental principle was implemented first at the PAL-XFEL and then at the LCLS; the setup configurations differed slightly between the two experiments, as the technique and instrumentation were refined. Both experiments studied diamond single crystals to illustrate the technique under different conditions. Initial experiments at PAL-XFEL used 9.7 keV photons with \( \sim 10^{11} \) photons per 32-fs pulse at a 30-Hz repetition rate; the XFEL beam was prefocused to a \( 30 \times 10^{-\mu \text{m}^2} \) spot at the sample. Subsequent experiments at LCLS used 10.1 keV photons with 1.6 mJ pulse energy, and 50-fs per pulse at 30 Hz. In both experiments, the photon flux was calibrated on each shot to correct for fluctuations and used to normalize the resulting images. The smaller spot size at PAL was able to demonstrate DFXM’s utility at resolving irreversible phenomena like radiation damage with the stroboscopic approaches used in synchrotron experiments\(^{19}\). Since our first experiment at PAL-XFEL informed our full design for the refined DFXM microscope designed for XFELs, we focus on detailing the setup in full for the latter experiment at LCLS.

3.1 Upstream Beam-Conditioning Optics

The experiment used a channel-cut Si monochromator to reduce the bandwidth to \( \Delta E/E \sim 10^{-4} \), and a divergence of \( 1.1 \times 1.1 \mu \text{rad}^2 \). A monochromatic beam with a stable spectrum is essential for interpretable results from DFXM because fluctuations in the incident beam’s photon energy change the \( d \)-spacing and orientation that are imaged by DFXM\(^{29,30}\). We discuss the tradeoffs for this in Section 3.5.

Before illuminating the sample, the X-ray beam is prefocused using a stack of condenser lenses placed 3.435-m upstream of the sample. Prefocusing lenses used a combination of 1D and 2D Be CRLs to horizontally focus the beam into a vertical line beam at the sample position. From front to back, the CRL stack included \( 2x100 \) 2D, \( 2x500 \) 2D, 1x200 1D, 3x300 1D, 1x500 1D lenslets. The resulting beam was then apertured further with power slits and then cleanup slits to reduce the size of the focused line beam to its minimum \( 3.4 \times 3805-\mu \text{m} \) dimensions at the sample (as measured via curve fitting of images). To observe the beam positioning and pump-probe overlap, we installed a viewing camera in reflection mode on the upstream side of the sample.

When selecting the prefocusing lenses, it is important to consider the narrowest size of the beam waist vs the depth of focus corresponding to the focal point. Placing the condenser lenses very near to the entry-surface of the crystal, one may achieve the narrowest waist and average over the smallest amount of material in the \( x \)-direction. However, introducing shorter focal lengths into the system also shortens the depth of focus, making the Rayleigh range over which the beam is considered “at the focus” significantly shorter. In general, we have found that at XFELs, the longer prefocusing distances tend to give the best results due to instrument design constraints and the large sizes of the crystals.

3.2 Multi-Modal Microscope Design

After the x-ray pulse traversed the Bragg-oriented sample, its transmitted and \{111\} diffracted beams were imaged onto far-field detectors along the transmitted and diffracted beams (Fig. 2). Along the transmitted (direct) beam, transmission X-ray microscopy (TXM) was installed. The imaging objective was a stack of \( N = 17 \) Be 2D CRLs (radius of curvature, \( R = 50-\mu \text{m} \), distance between lenslet centres, \( T = 2-\text{mm} \)), corresponding to an effective focal length of 42 cm and an effective aperture (FWHM) of 261 \( \mu \text{m} \). The working distance (sample to entry of CRL) was set to \( d_1 = 47-\text{cm} \) while the exit CRL-detector distance was \( d_2 = 8.437-\text{m} \) (and total sample-to-detector distance \( d_{\text{tot}} = 8.941-\text{m} \)). From this follows a theoretical magnification of the X-ray beam of \( \times 18.4 \). The relevant formalism for calculating the optical parameters are given in\(^{27}\). The unfocused light was blocked with 600-\( \mu \text{m} \) diameter pinholes at the front and exit surfaces of the CRL stack, cut from 1-mm thick copper.

For the DFXM imaging system, the CRL comprised \( N = 33 \) Be 2D CRLs (\( R = 50-\mu \text{m}, T = 2-\text{mm} \)), corresponding to an effective focal length of 20 cm and a Numerical Aperture (FWHM) of 370. This objective was aligned with the center of the \{111\} diffracted beam. With \( d_1 = 23-\text{cm} \) and \( d_2 = 6.532-\text{m} \) (and total sample-to-detector distance \( d_{\text{tot}} = 6.832-\text{m} \)) the calculated magnification is 27. As the larger stack produced a smaller effective aperture, we used smaller Cu pinholes of 300-\( \mu \text{m} \) diameter (1-mm thick) at the front and back faces of the stack to remove stray light.

The long \( d_2 \) distances along both imaging systems attenuated the beams significantly when propagating through air. Attenuation was mitigated using vacuum beam-tubes spanning the longest possible range of the \( d_2 \) beam path, though there was slight attenuation from the 125-\( \mu \text{m} \) thick kapton windows at each end. We placed a 50-\( \mu \text{m} \) thick sheet of aluminium foil at the front surface of our vacuum tube along only the significantly higher intensity transmitted beam to avoid saturating the detector with the unattenuated XFEL beam.

3.3 Imaging Detectors

Selecting the appropriate imaging detectors for this experiment requires careful consideration of the dynamic range, resolution, and frame rates. X-ray lenses currently cannot offer simultaneously high numerical aperture and magnification. To capture enough of reciprocal space to avoid artifacts, DFXM requires the largest possible numerical aperture. Indirect X-ray detection
(i.e. conversion of the X-ray to optical via scintillator crystals) can detect images with significantly higher resolution, as the magnification is amplified by a relay optical imaging system with higher magnification lenses and the visible-light cameras afford a smaller pixel size. To observe single dislocations and the onset of damage in diamond, this approach was important, though it mitigated the photon efficiency of the technique.

For these experiments, we used home-built indirect detectors (similar to those employed in\textsuperscript{32}) for the far-field cameras along both TXM and DFXM arms, using 50-µm thick Ce:LuAG scintillator crystals (thicker crystals mitigate resolution). Each scintillator crystal was placed into the X-ray image plane, then was relay imaged with \( \sim 1x \) magnification to form an optical image on the camera. We used an Optem 34-11-10 zoom-lens attached to an Andor Zyla 5.5 sCMOS camera for both detectors, with scintillator-lens distances of 285-mm along the TXM arm and 130-mm along the DFXM arm. This resulted in an optical magnification of 0.71 × for the TXM arm and 0.75 × optical magnification for the DFXM arm. A turning mirror was placed between the scintillator and zoom lens to avoid the unconverted X-ray beam burning the detectors. We used both cameras at full resolution, but over a limited region of interest (ROI) that spanned the spatial extent of the scintillator screen to achieve the full 30-Hz readout rate of the detectors. Since the light under-filled the detector’s active areas, this did not limit our field of view in the material.

Diamond single crystals cut with surface normals of [110] were placed into the interaction point with the XFEL beam; the 660-µm thick crystals were oriented to an angle of 37.5° about the \( R_y \) axis (i.e. rotated about \( y \) from being normal to the incident X-ray beam) in the direction opposite of the diffracting beam to meet the Bragg condition for the \{111\} plane. The 1D X-ray beam thus illuminated an angled but nearly planar sheet of 3.4 \( \times 3805 \times 1151 \)-µm\(^3\) volume in the sample, with the FWHM of the line focus of 3.4 µm. The ROI for each imaging system was centered on the beam, however, because the diffraction projects the wavefront along a new wavevector (i.e. deflects it by 2\( \theta \)), the integrated volumes differ between experiments. As such, the TXM images mapped the \((x,y)\) plane of the sample, integrating along the \( z \)-axis, while the DFXM images projected a map of the \((z,y)\) plane, integrating the images along \( x \) (the line-beam’s narrowest waist). The numerical apertures, pinholes, and magnification of the two imaging systems made each one’s field-of-view differ slightly: TXM mapped 4 \( \times 101 \)-µm\(^2\), while DFXM mapped 700 × 200-µm\(^2\) (or less, depending on the diffraction condition).

The DFXM had a total magnification of 54 × (corresponding to 0.11 µm per pixel), while the TXM along the bright-field had a total magnification of 14 × (corresponding to 0.44 µm per pixel). We note that the approximate magnification may be predicted based on the imaging equations and effective focal lengths described previously, however, since CRLs natively have aberrations, the actual magnification must be measured explicitly. At synchrotrons, this is often done either using resolution targets (e.g. Siemens stars, US Air Force Targets, TEM grids, etc.), or by moving translation motors to observe translation of the features on pixels. We note that at XFELs, the motorized approach is often highly inaccurate, as the precision, accuracy, and reproducibility of all motors are not reliable down to the sub-µm lengthscale; required by this experiment. As such, we strongly encourage the use of resolution targets, and used TEM grids (200- and 1500-mesh) for this work. Finally, we note the importance of understanding the orientation of the observation plane. As in XRT, the contrast mechanism giving rise to image signal in DFXM arises from scattering contrast along the \{Q\} Bragg diffraction vector of the lattice. That said, the \textit{spatial plane that is imaged by DFXM is always rotated by an angle of \( \theta \) with respect to the observation plane.}

### 3.4 Goniometer Design

For quick alignment of this sensitive 2-path (TXM, DFXM) imaging experiment, we designed a custom goniometer to streamline alignment and scanning precision. We defined our initial design criteria based on the points listed below:

- Fully self-contained goniometer assembly for accurate alignment before XFEL beam.
- Precise & reproducible positioning of sample translation \((x,y,z)\) & rotation \((\chi, \phi, \omega)\) with sub-µm and 0.005° precision, respectively.
- Motor controls for fast translation between alignment and scanning samples (without manual realignment).
- Rapid orientation of each crystal into its most optimized Bragg condition in the horizontal scattering plane.
- Scanning & data acquisition capabilities to collect series’ of images while scanning the sample & detector along \(x, z, \chi, \phi, \omega, 2\theta\).
- Motion control for each CRL stack along its local \(x_i, y_i, R_{xi}, R_{yi}\) with respect to the imaging beams.
- Viewing cameras near the Fourier plane of the imaging CRLs for alignment (possible future aperturing opportunities).
- Facile motion of the CRLs along an arc concentric with the samples for direct-beam alignment.

Based on the needs outlined above, we designed the goniometer assembly shown in Figure 2(a-b). We used a 12-axis goniometer, using (from top to bottom): 2 piezo motor translation stages \((x, y)\) to position each sample of interest into the center of rotation for the rotation stages, 3 rotation stages to mimic an Eulerian cradle with rotation about the \(z, x, \) and \(y\) axes (i.e. \(R_z, R_x, R_y\)), 3 linear translation stages to center the pre-defined center of rotation into the center of rotation for the CRL carousel, and finally, a 2-axis linear translation stage \((x, z)\) to position the goniometer’s center of rotation into the XFEL interaction point.

To simplify CRL alignment and strain scans, we used a motorized \(2\theta\) rail that can independently rotate 3 carriages about a
central position, as shown in Fig 2. The radius of this circle was selected based on the imaging distances for 10 keV photon energies and \( \sim 30 \) imaging CRLs, and the baseplate for each carriage was planned to integrate radial translation stages for further focusing of each lens stack. Each carriage was fitted with stage stacks to position the BF and DF stacks of CRLs along their respective \((x, y, R_x, R_y)\) axes, as shown in Fig. 2b. The third carousel was used for the near-field alignment camera, though in future this could be used for 2-phase DFXM (i.e. diffraction imaging for a second phase).

At the top of the goniometer, we required resolution targets, alignment guides, and single crystals for our measurements. For facile motion between these, we used a sample holder with interchangeable custom cartridges that could affix samples into the image plane of the microscope, either at an arbitrary geometry, or pre-aligned to the Bragg condition. The cartridges in our case were machined for high precision positioning, but in future we anticipate they could be 3D printed before or during an experiment for unexpected geometries. Pre-positioning the samples in this way simplified the goniometer design and alignment needs because it limited the range of angular sweeps to \( \lesssim \pm 5^\circ \) required to orient the crystals to only...
3.5 Alignment Strategy

As is described fully in previous work, the resolution function of DFXM enforces a strict Bragg condition, requiring very precise, accurate, and reproducible alignment. Due to the scarcity of XFEL facilities, no permanent and dedicated DFXM facility exists at the time of writing; this necessitates full alignment of the entire microscope and the samples within the beamtime accessible for one experiment. We detail alignments including 2 alignment cameras in the near-field (NF) and intermediate-field (IF), which will be discussed in full in Section 3.6. With experiments at PAL-XFEL and then at LCLS, our team assembled a basic alignment strategy that follows the steps below:

1. With the full 2D beam, align the direct beam to the far-field detector to align, position, and calibrate the beam size & optical magnification of the home-built BF detector.
2. Align the CRL assemblies along the x and y axes (dark-field, DF then bright-field, BF) using the “intermediate field” (IF) alignment camera, with each lens stack placed along the direct-beam to avoid spatial jitter in the beam positioning during alignment.
3. Watching on the far-field camera, align the focus (z-position) of the TXM camera, then calibrate the TXM imaging system’s resolution, field of view, and any aberration distortions with its full magnification.
4. Align the 1D prefocusing CRLs to achieve the narrowest possible beam waist for the 1D vertical line beam at the sample position (calibrated using a TEM grid at the sample position). Calibrate beam size with magnified and unmagnified images (i.e. with and without the objective) along the direct beam. (Comparison of the magnified and unmagnified images allows one to calibrate the divergence of the XFEL beam)
5. Orient the crystal of interest (or grain of interest in a polycrystal) into optimal Bragg condition along z, ϕ, ω directions (intrinsic motor rotations of $R_z, R_\phi, R_\omega$ from bottom to top) using a photodiode, then a near-field camera at the DF angle.
6. Orient the crystal’s diffraction peak into the horizontal scattering geometry by rotating in along $\omega$, confirming its position on the NF, then IF, then FF detectors along the DF path.
7. Rotate the laterally pre-aligned DF CRLs into the diffracted beam; adjust lateral alignment as needed for accurate positioning on the IF then FF cameras.
8. Focus the DFXM imaging system by translating that CRL stack along the z direction while monitoring on the DFXM far-field camera. Once aligned, calibrate the resolution, field of view, and distortions on the DFXM images using a crystal with a TEM grid affixed to the exit surface of the crystal.
9. For single crystals - translate the sample along z to identify the upstream and downstream faces of the crystal, then select your ROI to ensure the motor encoders can track the ROI’s precise position in the sample.
10. Rotate & translate the sample as needed for data acquisition scans

The specific points in this list have been identified with significant considerations, which we explain in more detail below:

CRL Alignment Precision. X-ray lenses are notoriously difficult to align due to their small effective apertures, long working distances, and the necessity of remote alignments. To align the 66-mm thick stack of imaging CRLs (280-μm aperture) along the principal axis of the X-ray beam requires precise alignment along 5-axes - four lateral (x, y, $R_x, R_y$) and one along the beam’s path for focusing (z) - and is highly sensitive. Lateral alignment of DFXM CRLs is particularly challenging at XFELs because of the microscope’s sensitivity; the high X-ray flux and strong interactions can thermally distort the material.

Imaging Axial Strains To position the far-field detector along the diffracted beam is also no small feat, as the sample-to-detector distance requires 4-8 m of travel that goes beyond most standard XFEL facility equipment. Since the spot size of the beam is quite small, we identified that a temporary optical table for the DFXM far-field detector was extremely difficult to position accurately. Our optimized design used the Large Angle Detector Mover (LADM) stage at XCS (LCLS) to position the far-field detector along the 2θ arc of the diffracted beam. Long-term, detector stages like this will be essential for strain scanning capabilities in highly deformed crystals, which requires coupled translations of the CRL and FF-detector along the DFXM imaging arm to capture the full 2θ content of the diffracting crystal.

Alignment Targets. The specifics of the alignment targets used in this work were carefully planned. Along the direct beam, standalone apertures and TEM grids were suitable to track the position, size, and distortions of the beam and imaging system. Optical magnifications of the detectors were measured with no sample, but with a TEM grid placed over the front face of the far-field scintillator crystal to allow us to compare the grid to the known camera pixel sizes. We calibrated the magnification similarly with the full imaging system, placing the TEM grids instead at the sample position for the full calibration of the total magnification. For the DFXM, we calibrated the magnification and FOV using TEM grids placed on the exit surface of a
diamond single crystal. While this only acts as a mask over the back face of the crystal, it does create a pattern on the imaged beam that is stretched along the same projection angle, enabling us to calibrate the distortions.

**Precision of Scanning Stages.** For DFXM to accurately map reciprocal space, it must collect accurate and precise (∼10⁻⁵ radian, ∼~500-nm) scans of the sample along \( x, \phi, \chi \), and \( 2\theta \), as shown in Fig. 1. This requires scanning stages with high accuracy and precision that are well defined along the required directions of travel. While many XFEL experiments take advantage of the continuous range of travel afforded by hexapods, the accuracy of travel through the entire scanning range is not as high as for classical stepper motors. As such, we have found that accurate selections of the scanning motors is essential to the success of DFXM experiments at XFELs. For the best data acquisition, precise rotations along \( \phi, \chi \) and \( \omega \) require an extrinsic \( \mu \) rotation stage at the bottom of the stack to align the \( \omega \) rotation to be about the diffracting \( Q \) vector. Since it takes only two angular axes to rotate about the sphere, the designation of the \( \chi \) and \( \phi \) axes may be defined for each experiment (i.e. they are only a formalism).

**Beam Stability.** The stability of the XFEL beam is important to this experiment because of the sensitive alignment of the microscope and the connection between goniometer angles and contrast mechanism. There are four aspects of the stability of XFEL beams: (1) the poynting, (2) the spatial mode, (3) the timing, and (4) the spectrum. We detail these points below.

1. Deviations in the pointing of the incident beam must be < 10% of the beam’s spatial profile; poynting stability is essential to define precise alignment of the < 500-\( \mu \)m apertures of the CRLs. For cases with poor spatial stability, we have used a large beam size with a smaller aperture, which converts the spatial instabilities to intensity instabilities.

2. Spatial mode stability is important to ensure that the interpretation of features in images arises from the material and not from the beam. This has been challenging for many XFEL imaging studies, especially for experiments whose features of interest span a wide range of intensity gradients. The setup described in this work uses the simultaneous images from TXM to self-calibrate for spatial mode and intensity jitter known to be prevalent at XFELs. This effect may in some cases by mitigated by demagnifying the source, but can also be sensitive to aberrations along the undulator and X-ray optics halls.

3. While timing jitter is known to be a challenge at XFELs for sub-100-fs measurements, the timing stability required for DFXM at XFELs usually would not require the use of the timing tools. The time resolution required for each DFXM experiment is dictated by the physics of the material studied and specifically by the velocity of the dynamics and associated spatial resolution. For example, to image a 10 km/s wave would traverse a 0.4-\( \mu \)m pixel in 40-ps.

4. The spectral stability is perhaps the single most important point for DFXM. Since \( \lambda \) dictates the strain states that give rise to contrast in DFXM, the random fluctuations in the photon energy inherent to SASE beams cause different \( d \)-spacings to diffract through the lenses - changing the materials information inherent to the measurement. At XFELs, the high brightness of the source arises from Stimulated Amplification of Spontaneous Emission (SASE), which amplifies spontaneous emissions and therefore fluctuates pulse to pulse in its spectral content across the entire range of the 0.3% bandwidth. This significant photon-energy jitter is greater than the \( \gtrsim \)18 eV required to entirely shift (in the present system) the \( d \)-spacings that accounts for the image out of the the 286-\( \mu \)m aperture of the imaging CRL, since \( 2\theta = f(\lambda,d) \).

In general, we find that DFXM experiments at XFELs require either a monochromator or a seeded monochromatic beam (i.e. filtering the SASE beam before amplification) are best to ensure high sensitivity. In highly deformed systems (e.g. shock waves, fracture), the very wide range of strain states present in the material make it advantageous to use the full SASE spectrum to ensure that a sufficient range of strain states are represented to understand the system. Reversible systems (i.e. those that are feasible to measure with pump-probe experiments) may use signal averaging over many XFEL pulses to mitigate the spectral jitter, though this imposes an unchangeable minimum \( \varepsilon \)-resolution based on the bandwidth of the SASE spectrum.

### 3.6 Alignment Cameras

As each imaging system had a long beam path, we installed alignment cameras at three additional viewing positions: (1) upstream of the sample, (2) immediately behind the sample, in the near-field, and (3) just downstream of the CRLs, in the “intermediate field.” The upstream camera was primarily for alignment of the sample positioning stages - to confirm pump-probe overlap, XFEL positions, and sample motion. The two alignment cameras downstream of the sample confirmed the orientation of the crystals for alignment to the far-field detectors. As the intermediate-field camera was located behind the imaging CRL for DFXM, it also served as an alignment guide for the imaging CRLs.

**Viewing Camera.** The first of these was a viewing camera, placed upstream of the sample holder, that was a simple optical camera with a Zoom lens that resolved the interaction point of the XFEL with our sample. This camera allowed us to effectively position the sample and identify the position of the XFEL on our holder mount.
Near-Field (NF) Camera. On the third carriage for the $2\theta$ stage, we placed a near-field camera 6.5-cm behind the sample, along the direct beam. We then positioned it using a $\pm 100$-mm linear translation stage, to identify the position of the diffracted beam, which appeared at the appropriate angle of 34.75° (with respect to the 0° reference along the transmitted beam). In this way, our NF detector could capture images along the BF and DF beams, allowing us to optimize the crystal’s orientation into the Bragg condition, which is quite strict with the coherent and monochromatic XFEL beam. This NF camera also enabled us to orient the CRL stacks to the appropriate angles around the carousel stage, and to optimize the $R_z$ alignment ($\sim \omega$) to orient the crystal’s $\vec{Q}$ such that the $\{111\}$ beam scattered into the horizontal plane. This low magnification detector used a 35-$\mu$m-thick Ce:LuAG scintillator crystal to convert from X-ray to optical light, then relay imaged the visible light with a Zoom lens onto an Allied Vision Mako g-319b camera. Its 3.45-$\mu$m pixels produced an ultimate magnification of 0.01× in that camera.

Intermediate Field (IF) Camera. Behind the CRLs, we placed an alignment detector at a position 1.32-m behind the samples, with rotation along the $2\theta$ axis on its own separate rail system that allowed it to shift between the BF and DF beams. This intermediate-field detector used a 35-$\mu$m thick Ce:LuAG scintillator crystal with a Navitar zoom lens and an Allied Vision Manta g419b camera. Our intermediate-field camera had a resolution of 32-$\mu$m per pixel, corresponding to an optical magnification of 0.17×.

4 Results

We present the results from our microscopy data in the following way. We first summarize the raw data we measure on each detector, and what the results of those datasets tell us about the material and its dynamics. Next, we describe the analysis necessary to extract the full information contained in different types of sample rotational scans and explain how this changes our view of the material by sampling its momentum components (i.e. reciprocal space) with significantly more detail. Finally, we present the results of rotational scans (rocking curve along $\phi$ and rolling scans along $\chi$) from our intermediate-field, and far-field detectors, and explain the scientific insights afforded by each detector.

4.1 Detector Calibration

For all detectors in this work, we calibrated the spatial resolution and magnification using TEM grids. For each detector, we placed a TEM grid (200-mesh) at the front surface of the scintillator crystal to calibrate the optical magnification based on comparisons to the pixel sizes of each camera. We then calibrated the magnification of each imaging system using an illuminated TEM grid placed at the sample position. As the sample holder (Fig. 2b) held all calibration and imaging samples at the same $z$-position along the image plane, we were able to calibrate the BF imaging systems with a bare TEM grid held at normal incidence to the sample, and the DF imaging systems using a TEM grid affixed to the exit surface of a single-crystal of diamond.

Figure 3. Schematic showing a view from the top of the microscope that illustrates how a single pixel is projected onto the detector when it faces at normal incidence to the direct beam ($\vec{k}_i \perp \hat{z}_{det1}$) as was done in, and with normal incidence to the diffracted beam ($\vec{k}_d \perp \hat{z}_{det2}$) for the projection of the gauge volume of a pixel in the sample.
Indeed beyond the image magnification, DFXM and X-ray topography images require additional image stretching to account for their image projection. Because diffraction-contrast images are measured along the X-ray diffracted beam, they map the \((z, y)\) components of the sample by projecting the illuminated observation plate at the angle \(\theta\). While the line-beam simplifies image interpretation, it also necessitates careful calibration of image stretching to correct for this scattering projection.

Note that because the LADM detector is oriented at normal incidence to the diffracted XFEL beam, the stretch in the images differs from what would be observed at ID06 at ESRF (shown in Figure 3 as \(z_{det}\)\(^2\)). If we consider that the detector orientation at the XFEL is perpendicular to the Bragg-scattered beam \((det_1)\), and that the standard detector orientation at ID06 is normal to the direct beam \((det_2)\), the appropriate expressions relating the pixel size on the detector \(p_{det}\) to the effective pixel size \(p_0\) in the observation plane along \(z_t\) is

\[
\begin{align*}
    p_{det_1} &= p_{0z} \cdot M \cdot \sin(2\theta), \\
    p_{det_2} &= p_{0z} \cdot M \cdot \tan(2\theta).
\end{align*}
\]

This is shown graphically in Figure 3. For the effective pixel size along \(y_t\), the equation is \(p_{det} = p_{0y} \cdot M\) in both cases. We note that the TEM grids affixed to the exit surface of the diamond crystals necessitate that the resolution calibrations be performed with an additional stretch of the image by a factor of \(\cos \gamma\) to account for the orientation of the crystal.

Finally, we note the importance of understanding the orientation of the observation plane. As in XRT, the contrast mechanism giving rise to image signal in DFXM arises from scattering contrast along the \(\{Q_hk_l\}\) diffraction vector of the lattice. That said, the spatial observation plane that gives rise to DFXM images is always rotated by an angle of \(\theta\) about the scattering plane normal with respect to the crystal’s diffracting plane normal vector. The images shown in this work are all offset by the \(\theta\) rotation for simplicity, but careful analyses of DFXM images requires calibration for the observation-plane’s orientational offset.

### 4.2 Single-Frame Images with Ultrasonic High-Resolution X-ray Microscopy (U-HXM) Instrument

For every pulse generated by the XFEL, the U-HXM instrument saves the: (1) pulse energy, (2) goniometer positions, (3) TXM image of the line beam, and (4) DFXM image related to the elastic distortion states. As DFXM is a full-field microscope, each pulse produces a full image on the camera detectors; with beam tubes in place, we mostly found that the \(\sim 1\text{-mJ}\) pulse energy gave DFXM and TXM images with appropriate signal-to-noise in a single-shot.

The pulse energy informs the incident intensity required for calibration of signal intensity, while the goniometer angles catalogue the information required to interpret the microstructure and displacement states being sampled by each DFXM image. It is most effective to monitor the pulse intensity monitor is downstream of the monochromator. In the case that the intensity monitor is downstream, it may be used to calibrate the DFXM intensity. However, if the intensity monitor is upstream of the monochromator, the intensity that reaches the sample will be related to both initial SASE spectrum of the pulse and the intensity at the wavelength selected by the monochromator. In the upstream pulse-energy measurement case, an in-line spectrometer is required, or conservation of intensity must be assumed across many averaged pulses between the BF and DF images in the far-field. We note that future DFXM experiments may benefit from the new seeded monochromatic capabilities at XFELs\(^\text{36}\).

As described above, images collected in the far-field along the BF (i.e. TXM images) may comprise information on density variations but are also important to calibrate the DFXM images. As shown in Fig. 4, the line beams collected from the TXM camera can provide information about the spatial mode of the XFEL beam, the amount of dynamical diffraction occurring in DFXM, and context about the non-diffracting regions of the crystal (e.g. beam damage, phase transitions, etc.). In samples with no significant density variation, the TXM data can calibrate the fluctuations in the spatial mode of the XFEL, allowing one to correct for artifacts from the illumination. The TXM can also be helpful as a guide to quantify the amount of dynamical diffraction that occurs in a given diffraction condition (see TXM in Strong-Beam in Fig. 4). For nearly perfect crystals of sufficient thickness, dynamical diffraction (multiple scattering) causes the line-beam to scatter multiple times; for even numbers of scattering, the beam remains along the \(k_l\) vector, while an odd number of scattering events returns the beam back to \(k_l\). In both cases, the beam is translated from its initial position due to these multiple scattering events. This effect, termed the Borrmann effect\(^\text{37}\), (like optical etaloning) makes the TXM line beam spread out laterally, as ghost-fringes appear next to the primary beam. Information about the extent of dynamical diffraction can be helpful in understanding how to interpret image features with DFXM, as they also complicate signal in DFXM images. Finally, the TXM images give important information about the structure and its evolution in the entire illuminate part of the sample. Notably, DFXM is set up to image a specific part of the crystal which gives rise to scattering within a narrow region in reciprocal space with extremely high resolution. In general it does not provide any information on other material phases and grains that are differently oriented as there either is no diffraction signal or the diffraction occurs at angle not compatible with the DFXM settings. By offering density contrast in these “missing parts,” TXM gives a complementary view of the material, giving important context for DFXM interpretation.

As described in Poulsen, et al.\(^\text{29}\), each raw DFXM image contains bright and dark features that map specific components of the crystallography, detailing components of the strain and orientation states along the lattice plane being measured, \(hkl\),
Figure 4. Sample images from each detector in the bright-field (above) and dark-field (below) configurations. These images illustrate how the near-field cameras gather imaging data over a wide field of view, but with much lower resolution. By contrast, the far-field images capture detailed information in the weak-beam and strong-beam images that show the full grain and defect-only information, respectively. Spreading of the line beam on the TXM detector displays the dynamical diffraction spreading the beam waist.

| Scan Parameters | Axial Strain | Orientation | 3D Structure | Time |
|-----------------|--------------|-------------|--------------|------|
| 2θ, λ           | φ, χ         | ω, x_ℓ     | l, τ_{pp}    |

Table 2. Scanning modalities accessible for use with DFXM

These maps are related to the spatial derivatives of the elastic displacement fields, which includes information about defects in materials and distortion waves like phonons, shock waves, and heat.8

A full comparison of the different types of images collected along the bright-field (BF) and dark-field (DF) imaging arms is shown in Figure 4, illustrating the change in resolution vs field of view afforded by each imaging modality.

4.3 Overview of DFXM Scanning Modalities

To collect the full information about dynamics of interest with DFXM, one must go beyond single frames, collecting scans along the scanning parameters outlined in Table 2. A full DFXM scan is collected by measuring images at a series of different motor positions, photon energies, and/or times as described in Table 2. Strain and orientation scans probe different populations of reciprocal space in a region around the diffraction vector; alternatively, this is presented in terms of axial strain and orientation (a local pole figure). The 3D and time scans capture the same structural information as a function of position and time, respectively (without changing the contrast mechanism). In most experiments, a combination of these scans is performed, where some combination of structure and dynamics are measured at a series of positions across different axes of a crystal’s diffraction extent (rocking or rolling curve).

For strain and orientation scans, each image spatially maps the populations of displacement fields that - within the reciprocal space resolution function of the microscope - meet the Bragg condition at that position in reciprocal space (as shown in Fig. 1b). As described fully in Jakobsen, et al,9 images collected at the brightest point along a rotational scan map the entire extent of the undeformed grain of the crystal, and are said to be in the strong-beam condition. Images collected on the edges of the scan only describe specific types of distortions in the lattice, showing defects or other sparse structural features in the sample; these are defined as meeting the weak-beam condition. The nomenclature in this terminology was selected to parallel the analogous...
dark-field TEM experiments in electron microscopy. At XFELs, the time-resolved scanning capabilities introduce another set of displacement states not described in previous DFXM work: temporally weak-beam states. We use this term to denote new weak-beam distortion states that populate during the transient dynamics probed by the experiment, but that are not populated at $t < 0$.

Finally, strain scans are typically performed at the synchrotron by the collective motion of detector and CRL stages in coherent motion along the $2\theta$ arc of the diffracting crystal. While the dedicated instruments at synchrotrons make this approach viable, the extra setup requirements at XFELs make it simpler to hold the entire experiment stationary, scanning the $d$-spacing with small changes to the photon energy for $\Delta \lambda$. For the case of probing diamond (111) planes by an X-ray pulse of $E = 10.1$ keV and $\Delta E/E = 10^{-4}$, a single X-ray pulse with a bandwidth of $\Delta E = 1$ eV covers a narrow range of $\delta d = 2 \times 10^{-5}$ nm and $2\delta \theta = 4 \times 10^{-3}$ $\circ$. For example, in the present experiment, a strain scan spanning a range of $\Delta \varepsilon = 1 \times 10^{-3}$ ($\Delta d = 2 \times 10^{-4}$ nm) would require 10 steps of $\Delta E = 1$ eV. This range of strain scanned by changing photon energy is equivalent to that scanned by changing scattering angle for $2\Delta \theta = 0.03^\circ$.

The reciprocal space resolution function for DFXM is highly anisotropic, with the beam divergence accounting for the highest sensitivity in $q$; this effect is amplified by the coherence of XFELs. As such, to map different strain components, it is important to consider the orientation of the lattice in relation to the axis of the resolution function, as is discussed fully in\textsuperscript{28,29}.

Different types of dynamics are more suitable to study in the strong-beam or weak-beam condition based on the grain sizes and the spatial population of states present. In past, we have found defect measurements in low-defect density materials are easiest to interpret in the weak-beam conditions. For deformed crystals the local orientation changes are typically much larger than the Darwin width and the diffracted signal can be considered to be in the strong-beam condition throughout. At the XFELs, it is easy to assume that the temporally-weak condition should be most helpful to study dynamics. While the temporally-weak beam condition is usually the easiest data to interpret during an experiment, they often lack sufficient information about how those dynamics were informed by the initial microstructural features in the sample. As such, we suggest a balance between strong, weak, and temporally-weak conditions when planning for XFEL-DFXM studies.

When studying dynamics at the XFEL, it is important to consider the tradeoffs between different approaches. For irreversible processes, real-time images must be collected to capture the dynamics of a material as it changes - this approach will always be limited by the camera acquisition speeds (frame rate and shutter speed), the inter-pulse separations for timing sequences, and the data transfer rates that are possible for the data acquisition system. Optical fibers for data transfer are essential for the data transfer rates required for the high-resolution and high dynamic range detectors necessary to acquire the full content of DFXM images (> 12 bit-depth per pixel). From our experience, the U-HXM instrument performs best when each camera is connected to its own data-transfer line to enable parallel data transfer capabilities. From our experience, the $10^{12}$ photons per pulse available at the XFEL are indeed able to acquire sufficient signal-to-noise for single-shot acquisitions, however, for weak-beam conditions, this can introduce significant challenges in interpretation (e.g. Fig. 2d). Our experiments at the PAL-XFEL observed damage bands appearing in diamond after 20,000 pulses of the XFEL, however, the low intensity at the DFXM detector presented challenges in distinguishing this signal from burns to the Kapton tape behind it.

By contrast, for crystals exhibiting reversible dynamics, a pump-probe modality may be employed. In that modality, an optical pump laser drives reversible dynamics in the same sample region illuminated by the XFEL probing laser, at a series of time delays, $\tau_s$, that can be stepped through the full dynamics of the system. This approach enables signal averaging over hundreds through thousands of pulses, as shown in Fig. 2c, enabling much higher signal to noise. That said, this approach often requires longer than anticipated acquisition times, as the XFEL pulse may have to be attenuated to avoid sample damage. We include an example of this in Fig. 2c showing a strain wave propagating in diamond, generated by laser irradiation of the gold surface transducer layer.

Finally, as presented above one DFXM image probes a single observation plane in the specimen. A spatial 3D map may therefore be acquired by collecting image stacks while stepping the sample along the $x_t$ axis. We note that the sensitivity of the DFXM microscope requires that the images in such $(x,y,z)$ scans are collected with rotational scans of at least one tilt axis for each plane - to correct for motor drift (> 0.0005$^\circ$) or changes to the microstructure of the sample over the course of the scan. As the reciprocal space resolution function is thinnest in direction $\phi$, it is natural to perform this integration over $\phi$.

As an alternative to layer-by-layer mapping, the condenser may be removed an the specimen therefore illuminated by a square shaped beam. The DFXM images then represent a projection. Similar to classical tomography, 3D mapping may then be facilitated in a tomographic fashion by rotating in $\phi$ around the diffraction vector. This so-called topo-tomographic DFXM modality can give faster 3D information but is more complicated in terms of sampling.

In large crystalline materials, combined analysis of the near-field, the intermediate-field and far-field images is quite helpful. The large difference in the field of view provides context as the near and intermediate field detectors can capture global dynamics occurring in volumes that are outside the field-of-view of DFXM. Changes to the real-space sample (e.g. thermal expansion, fatigue, etc.) would be evident in the nf and if cameras, showing images that clearly demonstrate a change in the strong-beam image. New components of reciprocal space that populate are most evident at the IF camera, where satellite
Figure 5. Raw data showing DFXM images captured in each experiment, using pump-probe and real-time imaging modalities. The image in (a) shows an integrated image of a strain wave traversing a diamond single-crystal, while (b) shows the incipient beam damage after 20,000 pulses from the XFEL at PAL-XFEL. We note that the crystallographic legends shown in all images are rotated by an angle of $\theta = 17.5^\circ$ about the vertical axis from the orientations shown. For description of the dynamics of the strain wave images and measurements, see Holstad et al.\textsuperscript{38}.

4.4 Analysis for U-HXM Data

In this section, we provide an overview of the data analysis approach to evaluate the crystalline domains sampled by different scan modalities. For a detailed overview of the analysis, we encourage the reader to explore more thorough guides of specific analysis tools developed in dарifix\textsuperscript{40} and elsewhere\textsuperscript{11}.

As in classical XRD, the diffraction condition in DFXM is determined by the conservation of momentum criteria that arise from the sum of the momentum vector for the incident, $\vec{k}_i$, and diffracted, $\vec{k}_d$, beams being equal to a lattice vector, $\vec{Q}$. While a perfect crystal would diffract only at a specific value of $\vec{Q}_0$, the defects and distortions within the lattice cause additional $\vec{Q}_i$ states to populate as well, mapping the vectors characteristic of the subtle distortions in the lattice spacing and orientation, i.e., the displacement gradient tensor field\textsuperscript{41}. The specific components of the displacement gradient tensor field that contribute to the diffracted signal arise from the crystal’s orientation and the position of the detector. For a full derivation of the contrast mechanism for DFXM, we refer the reader to recent work using ray optics\textsuperscript{29} and wavefront propagation\textsuperscript{42} with synchrotron radiation, and XFEL sources\textsuperscript{30}.

Experimentally, DFXM crystallographic scans are collected by capturing images produced by the crystal when it is rotated to each point along the rocking curve. The corresponding stack of images then has coordinates of $(z, y, \chi, \phi, 2\theta)$ (assuming $\omega = \mu = 0$), and may be reconstructed into a single mosaicity map by constructing an image for which each pixel $(z_i, y_i)$ has an intensity, $I(z_i, y_i)$, that corresponds to the maximum intensity observed at that pixel throughout the scan, $I(z_i, y_i) = \max_{\chi, \phi, 2\theta} I(z_i, y_i, \chi, \phi)$, in a color, $C(z_i, y_i)$, that encodes the $\chi, \phi, 2\theta$ orientation that diffracted with that intensity. A schematic is shown in Fig. 1 displays the scanning procedure and associated image sequences, with experimental data to show the characteristic images and reconstructed results.

We note that the above description of the Center-of-Mass (COM) images is relevant when measuring the average properties of the lattice locally, e.g. measuring the orientation of a local domain. When studying the distortion fields surrounding single defects, the average properties are not of interest, and therefore, single images of the weak-beam conditions are preferred\textsuperscript{11,16}.

For static or reversible systems, the detailed maps accessible from reconstructed DFXM scans along $\phi$, $\chi$, and $2\theta$ can give key information about unknown defect structures that are not well understood. For irreversible systems (e.g. fracture, radiation damage), a crystal grain with sufficiently well-understood defect structures may be measured in real-time, using simulations of all possible defect structures to evaluate the image features appearing in DFXM images collected at only a single crystal orientation. This may be done manually\textsuperscript{19}, or using Bayesian inference for physics-informed image interpretation\textsuperscript{43}. 

images may begin to appear to indicate new strain/orientation components that could also be of-interest to DFXM but would require realignment to the new $2\theta$ and $\eta$ positions.
4.5 Guide for Image Reconstruction from DFXM Scans

The full DFXM scans described above measure large datasets that may be nontrivial to interpret for materials science or physics applications. In this section, we introduce the general approach we used for the image processing treatments, and reconstructions to measure the observables for the basic rotational scans.

**Background Subtraction.** The XFEL beam intensity fluctuates from shot to shot due to the SASE fluctuations and the monochromator (which converts the spectral jitter into ∼100% intensity jitter). Therefore, if possible we average over multiple x-ray pulses at a fixed sample position to improve signal to noise. Dark frames, acquired without the x-ray beam, are periodically saved between the averaged bright frames and are subtracted from the averaged bright field images. For the XFEL beam, we account for the intensity variation on a shot-to-shot basis by integrating DFXM and TXM images over all like-types of images and subtracting the background intensity of the image closest to the image of interest. We then looked at the same sorting for the BF images and calibrate them as the inverse of the rocking curve for the DFXM. By fitting these to a roughly Gaussian function, we could account for frames with un-characteristically low intensity and multiply the intensity to be appropriate for a smoothly-varying rocking-curve function.

We detail the workflow for the background subtraction to plot images herein. One must begin by ensuring that they begin with the raw images (not the normalized ones often saved by the data acquisition systems at XFELs, e.g. psana at LCLS). We then estimate the detector noise for each pixel by taking the mean value of each pixel from all images saved with the X-rays off (darks saved every 13 images in our acquisitions). To remove the detector noise, we then subtract the mean detector noise array from all images collected with the X-rays on. To account for the 100% intensity jitter, we discard all images where the intensity is low compared to other images at that position or timestep (if the x-ray flux is low, we collect mostly noise). Finally, we sum all images from the same position or timestep to plot single-shot images (e.g. Fig 2c). We note that when plotting these images, it is important to ensure that the minimum and maximum color limits are set carefully, as hot pixels in the image make python/matlab choose poor colormap limits if not set manually. The 1st and 99th percentile are normally a good choice for the limits of the colormap.

**Scanning Reconstructions.** A powerful way to reconstruct the strain or mosaicity fields from the scans collected by DFXM is to compile a center-of-mass data reduction over one or more axes in question. Before starting the comparison between different motor positions of the scan, we first normalize the summed images to account for the intensity variation of the XFEL. For sufficiently sampled scans, this may be calibrated by comparison of the DF rocking curve integrated intensity to the BFXM integrated intensity across the rocking curve. If the BFXM shows a complementary Gaussian curve to the DFXM rocking curve, this confirms that intensity loss arose from only the change in intensity of the XFEL beam, and the multiplier may be deduced from curve-fitting of the integrated intensity rocking-curve functions. Because of the noise subtraction, there were pixels with a negative intensity, requiring that all negative values be thresholded to 0 before subsequent processing.

Noise in the images collected at angles far from the center of the rocking curve have an outsized impact on the center of mass (COM) calculation, and will bring the COM towards the center of the scan. We reduce the impact of this by setting the intensity of pixels below the nth percentile to zero, or alternately subtracting a fixed value from all pixels before normalization. Finally, we then calculate the center of mass by fitting the $I_n(\phi, \chi, 2\Delta \theta)$ function for each pixel to a Gaussian and taking the weighted average of each pixel for each scanning parameter. More detailed descriptions of how the strain and mosaicity are solved from this approach can be found elsewhere.

4.6 Example Image Reconstructions

To demonstrate the mapping of local orientation, we include set of representative reconstructed images in Figure 6, showing the relationships between the topography and DFXM images as well as the different types of scanning modalities. Fig. 6(a) shows the reconstruction from an unmagnified X-ray topography image, while (b-c) show a stretched TEM grid over a domain wall in diamond (111), measured along the $R_x$ and the $(R_y, R_z)$ rotation stages to map $\phi$ and $\chi$, respectively.

The XRT image shown in Fig. 6a illustrates the additional context provided by the IF scans. Fig. 6a shows the reconstructed image of a $\phi$ scan collected from an $R_y$ scanning acquisition. The ~1150-µm field of view along the horizontal axis shows the entire extent of the observation plane in the crystal that is viewed in our images, $\frac{D}{\text{um}}$ for $D = 660\mu\text{m}$ crystal thickness, as viewed by diffraction microscopy. The XRT image is highly pixelated, showing a significantly lower spatial resolution, but maps the domain contexts spanning the crystal extents and orientations throughout the entire extent of the crystal. In this way, XRT provides context at the IF detector for the full crystal information - guiding the imaging alignment and the interpretation of the microstructure in the DFXM images that follow.

Fig. 6(b-c) show the representative images that may be collected in the ff in the magnified DFXM geometry, showing rotational scan reconstructions from the $\chi$ ($R_x, R_y$) and the $\phi$ ($R_y$) directions, respectively. Both images map the exact same region of the diamond (though a different one than in XRT) - with a low-angle boundary bisecting the left and right regions of the crystal by an angle that maps between the two rotational directions. While Fig. 6(c) shows the $R_y$ rotations that map directly to the $\phi$ rotational axis described above, the reconstructed results in Fig. 6(b) included rotational offsets in $R_z$ to correct for the
Figure 6. Reconstructed diffraction-contrast imaging scans, collected (a) at the IF detector, producing X-ray topography (XRT) images that scan $\phi$, and (b-c) after magnification at the far-field detector, along (b) $\chi$ and (c) $\phi$. Scale bars for the DFXM images are identical, while the XRT image is $10 \times$ larger, but the crystallographic legends are consistent for all images.

fact that the $R_z$ rotation shown in Fig. 1 includes components of rotation in $\chi$ and $\omega$. The $R_z$ corrections were performed by ensuring that the same TEM-grid fiducial appeared over the same pixels in each frame of the rotational scan. As such, the image in Fig. 6(b) approximates a $\chi$ scan.

The results from Fig. 6b-c indicate that the misorientation of the diamond across the boundary in its center is $\sim 5 \times 10^{-1}$ degrees in the $\chi$ direction, and $-6 \times 10^{-3}$ degrees in the $\phi$ direction. More thorough investigation of this type of feature may inform the Burgers vector and spacing of the dislocations that comprise the boundary - serving as a direct measurement of the microstructure.

5 Discussion

Our results in this work demonstrate a new capability for X-ray science that holds opportunities to enhance DFXM from the 0.1 second timescale all the way to the 100 fs timescales. Using the instrument design we describe here, this setup allow simultaneous multi-beam imaging experiments that can probe multiscale dynamics in materials under a range of different conditions. Using the simultaneous dark- and bright-field imaging (DFXM and TXM, respectively), we have demonstrated a key new opportunity to explore the reciprocal space components that underlie a specific feature captured by a TXM image of the material. This is important in applications like fracture or amorphization from Fresnel and/or Bragg scattering as voids begin
forming that instigate the fracture process.\textsuperscript{44}

While the bright-field TXM spatially maps a material’s density by imaging along the transmitted beam (magnified version of radiography), DFXM resolves the local lattice distortions that underlie those density variations. Together, these techniques hold the opportunity to resolve snapshots of how defects and domains (and their associated strain fields) grow, propagate and interact as they cascade into large-scale material dynamics. This work sets the stage for a new host of experiments to study how mesoscale defects deep beneath a material’s surface initiate phase transitions, strengthen materials, modify their physical properties, and beyond, as they respond to external stimuli.

6 Conclusion

We have presented a full integrated design for DFXM and TXM at XFELs, providing opportunities to resolve dynamics that occur from slow to ultrafast timescales – spanning 13 orders of magnitude. The shift from stroboscopic integrated measurements to the single-shot ultrafast measurements that are now accessible at XFELs can enable a new class of experiments at the mesoscale. This new capability offers opportunities to understand ultrafast deformations (e.g. fracture), rapid phase transformation heterogeneity, thermal transport, and charge-density waves in superconductors, and important phenomena in many other fields.

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Author contributions statement

LDM wrote the manuscript with significant contributions from TH, HFP, MMN, KH and TR, then edits and correspondence from all co-authors. LDM designed the experimental configurations, refined experiments, led experiments and analysis. Spontaneous acquisition experiments were conceived of at LLNL, while the pump-probe experiments were conceived of at DTU. EF developed the 3D models and engineering designs for the U-HXM instrument and helped to construct them at LCLS and PAL-XFEL. BK and TMR performed data analysis and image acquisitions at the LCLS, supported by TSH, and EBK. Further post-processing data analysis were also performed by LEDM, KH, and ZS. XFEL experiments were carried out and supported by LEDM, BK, TSH, MS, DN, SB, PKC, JHE, EF, EG, LG, AG, KH, MH, KK, SK, SK, SK, HK, EBK, SK, H-JL, CL, RSM, BN, NO, RPP, HFP, MMN, TMR, AMS, TS, HS, TvD, BW, WY, CY, DN, SK, SN, MS, TvD, and MC supported the experiments as beamline scientists at the PAL-XFEL and LCLS, respectively. FS evaluated and selected the diamond samples for all experiments presented in this work. SB, AG, and MH supported the analysis of data collected at PAL-XFEL and developed optimization methodologies in support of the optics alignment in this work.

Additional information

The authors of this work have no competing interests in this work.