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To cite this article: Ashwin J. Shahani, Xianghui Xiao, Erik M. Lauridsen & Peter W. Voorhees (2020) Characterization of metals in four dimensions, Materials Research Letters, 8:12, 462-476, DOI: [10.1080/21663831.2020.1809544](https://doi.org/10.1080/21663831.2020.1809544)

To link to this article: <https://doi.org/10.1080/21663831.2020.1809544>



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Published online: 21 Aug 2020.



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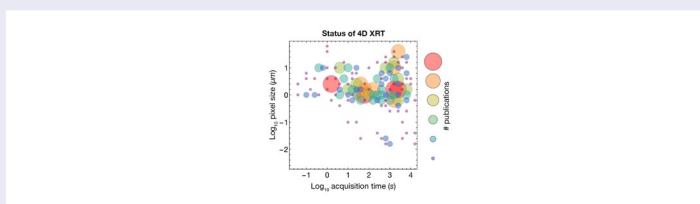
Characterization of metals in four dimensions

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ABSTRACT

The ability to watch the three-dimensional (3D) evolution of structural materials is a breakthrough in non-destructive characterization. In particular, X-ray tomographic imaging techniques have found success in revealing the underlying mechanisms of microstructural transformations in partially and fully solidified metals. Here we review the most important developments in four-dimensional X-ray microscopy, focusing on absorption- and diffraction-based techniques in the laboratory and the synchrotron. In light of recent progress in this area, we identify critical issues that point to directions for future research in imaging the evolution of heterogeneous microstructures at extreme space and time scales.



IMPACT STATEMENT

Four-dimensional X-ray tomography has opened a new paradigm in physical metallurgy, allowing us to characterize the various epochs of microstructural evolution in 3D and as a function of time.

ARTICLE HISTORY

Received 17 June 2020

KEYWORDS

Metals; solidification; grain growth; X-ray tomography; 4D imaging

1. Introduction

Our understanding of the ‘materials paradigm,’ that links structure, processing, properties, and performance of a material, has enabled the development of transformative technologies central to modern civilization. However, in order to assess the extraordinary morphological and topological complexity of technological materials, a 4D (i.e. time and 3D space resolved) analysis is needed. For this purpose, one can perform tomography with neutrons, electrons, or X-rays. In general, high-spatial resolution requires intense sources in order to supply sufficient signal per unit area. The flux of the most powerful neutron sources is several orders-of-magnitude lower than that of synchrotron radiation sources [1,2] and therefore neutron tomography does not compete with X-ray tomography (XRT) in terms of its spatial resolution (5 μm at best [3]). On the other hand, electron tomography offers a considerably higher resolution (down to

single Å [4,5]) yet it cannot accommodate large (mm-sized) samples. It is for these reasons that we restrict our review to XRT, which offers the best of both worlds: a reasonably large field-of-view as well as a high-spatial resolution (discussed in Section 2), both of which are amenable to the study of microstructural evolution.

While there are a number of excellent review articles [6–11] on 4D XRT (and its close companion X-ray radiography), there is no dedicated review on XRT for *metals* in particular. Even so, XRT is at the forefront of advanced analytical methods for studying metals and their evolution because it is the only technique that allows for 3D imaging across multiple length-scales and imaging modalities (*vide infra*), providing new information on local morphology, chemical composition, and crystal (grain) structure. Further developments in XRT will directly impact the field of metals processing: For instance, the National Institute of Science

and Technology's 'Measurement Science Roadmap for Metal-Based Additive Manufacturing' (AM) has identified nondestructive materials diagnostics as one of the key barriers to AM implementation [12].

At the heart of metals manufacturing processes (such as AM and casting) is solidification from a parent liquid phase. The process begins with the nucleation of nm-sized solid grains. Growth of the stable nuclei then proceeds through mass diffusion from the liquid to the solid. If the solid-liquid interfacial velocity is greater than a critical value, then an initially planar growth front will breakdown to form cells and dendrites [13]. At high volume fraction of solid phase, the grains interact with each other through 'soft' collisions (i.e. overlapping diffusion fields) [14,15] and 'hard' collisions (i.e. impingements) [16]. In the latter case, when two neighboring grains touch, a grain boundary (GB) is formed. Since GBs are non-equilibrium defects, they are expected to migrate over time through a process known as grain growth [17]. In parallel, a second solid phase may precipitate along the GBs or else nucleate directly from the liquid (polyphase solidification). Our goal in this review is to showcase recent applications of XRT in shining new light on these various epochs of microstructure evolution.

In what follows, we highlight three XRT-based techniques that may be of special interest to the metals community. The first and simplest is absorption-contrast X-ray microtomography (μ XRT), where the micro- prefix describes imaging with $1\ \mu\text{m}$ voxels (volume elements). Spatial resolutions substantially below $1\ \mu\text{m}$ are the province of X-ray nanotomography, which requires different instrumentation from the former technique and is therefore discussed in a separate section. It is also possible to go beyond absorption-based imaging, thanks to methods such as diffraction-contrast X-ray tomography, which is the subject of the third section. For each of these XRT techniques, we describe the operating principle and a few salient examples. Lastly, we offer an outlook on current and future trends in 4D XRT for metals research.

2. Emerging avenues for XRT

In the most general sense of XRT, an object is imaged nondestructively in many directions by passing X-rays through it and by measuring the modification of these rays caused by their interaction with the matter of the sample. The traditional approach to 4D XRT is to acquire a sequence of projection images of the object, which is rotated at constant speed, at progressively increasing view angles. 3D structures can be reconstructed from the set of projection images taken from every

180° angle range ('fixed blocks'). The scan time Δt for collecting images in 180° defines the temporal resolution. To reduce the reconstruction artifacts due to microstructural changes, the scan time Δt has to be fast enough to catch a 'snapshot' of the structure. In the 'dynamic *in situ* experiment,' the object is heated or cooled in a furnace while X-ray projections are recorded in parallel. For slower processes (e.g. grain growth), one can instead conduct an 'interrupted *in situ* experiment,' wherein the object is annealed, quenched, and imaged in series, without loss of any dynamic information [18].

Following data acquisition, volumetric data can be reconstructed from the projection images. Reconstruction relies on comparing the incident and exit beams. Here, we focus on X-ray absorption contrast revealed by differences in linear attenuation coefficient (Sections 2.1–2.2) and X-ray diffraction contrast achieved through differences in crystallographic (grain) structure (Section 2.3). Changes in other signals can be used for reconstruction [19], including refractive index (phase contrast), elemental distribution (fluorescence contrast), particle size/shape (scattering contrast), and local atomic environments (e.g. X-ray absorption near-edge structure or XANES).

Dynamic *in situ* XRT can deliver the much-needed insights on the mechanisms of microstructure evolution so long as the imaging approach is able to keep pace with the rate of interfacial motion. Blurring of the reconstructed domain occurs whenever $V(r) \cdot t_{\text{exp}} > \Delta r$ where $V(r)$ is the motion of a contrast object located at position r in the sample, t_{exp} is the exposure time per projection, and Δr is the spatial resolution [20]. Thus, higher spatial resolutions can be obtained only by reducing the exposure time or by slowing down the kinetics of the physical process. A stricter criterion is $\omega \cdot D/2 \cdot t_{\text{exp}} > \Delta r$, where ω is the angular velocity and D is the width of the camera field-of-view (FOV). This condition guarantees that blurring is less than the resolution everywhere in the images. Note the scan time Δt is a product of the number of projections N_p and the exposure time t_{exp} , i.e. $\Delta t \geq t_{\text{exp}} \cdot N_p$. With these relationships in mind, the total number of projections N_p that would need to be acquired for a given spatial resolution Δr and FOV D is, ideally, $\pi \cdot D/(2 \cdot \Delta r)$ [21]. Collecting this many images within a limited scan time Δt would necessitate a high-frame rate or short exposure time. Yet there are practical limitations on the frame rate determined by the specifications of the camera, the brilliance of the source, and the contrast provided by the sample. Selecting the optimal parameters is often a difficult task as there are competing objectives of spatial versus temporal resolution between consecutive reconstructions.

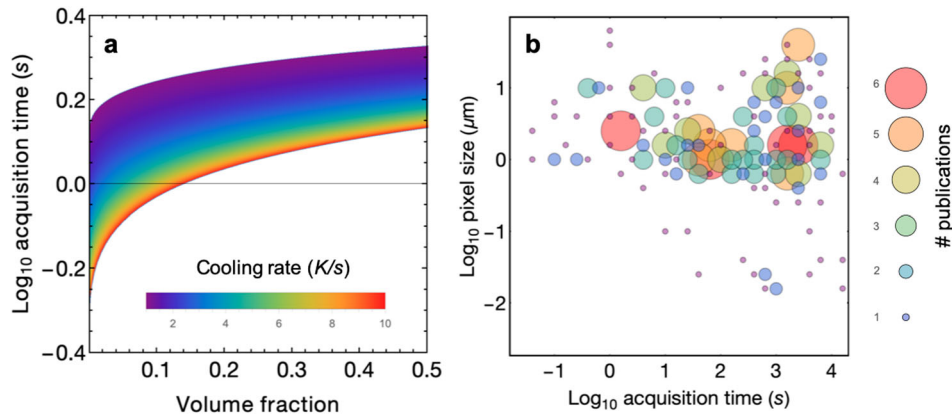


Figure 1. The state of 4D XRT: (a) the need and (b) our reality. Representative acquisition times in (a) computed as a function of volume fraction for an Al-20wt% Cu alloy undergoing solidification at cooling rates in the range 1–10 K/s. See text for details. In comparison, (b) shows the available acquisition times plotted against pixel size. Ultrafast tomography ($\text{Log}_{10} t(s) \leq 1$) is currently only attainable with μm resolution. Bubble size/color represent number of publications. This figure is based on data covering 1995–2016 retrieved from <http://tinyurl.com/Fast-Tomography> [25] with the addition of Refs. [26–44] from 2017–2019.

As an example, consider the imaging requirements for metals solidification, following Ref. [6]. Figure 1(a) presents the minimum acquisition time of a tomogram as a function of the volume fraction f of solid phase in an Al-20wt%Cu alloy, for various cooling rates. Here, we define the minimum acquisition time as the time it takes for a growth front to traverse $10 \mu\text{m}$. For the sake of simplicity, we assume that the solid phase is spherical with a density of $1000 \text{ grains per mm}^3$, and that the removal of heat is so rapid that it does not influence the growth process; we neglect also the influence of surface tension [6,22]. It can be seen that it is generally not worthwhile to perform 4D XRT with acquisition times longer than 10^1 s (for this particular example) or without reducing the exposure times t_{exp} , number of X-ray projections N_p , and/or cooling rates to unrealistically (low) values. This is because the solid–liquid interface would have propagated hundreds of μm within a span of 10 s (for cooling rates in the range of $1\text{--}10 \text{ }^\circ\text{C/s}$), and thus it would be challenging to follow the interfacial dynamics. Lowering t_{exp} or N_p will degrade the signal-to-noise ratio (SNR) of a reconstructed image according to $\text{SNR} \propto \sqrt{N_p \cdot t_{\text{exp}} \cdot I_0}$ [23], where I_0 is the mean photon count per second (assuming filtered backprojection [24] is used for reconstruction). If we want to study the early stages of solidification ($f \lesssim 0.05$) with cooling rate of $10 \text{ }^\circ\text{C/s}$, we should strive for *sub-second* acquisition times. Of course, requirements on temporal resolution are somewhat relaxed for solid-state transformations owing to the fact that diffusion in solids is typically orders-of-magnitude lower than diffusion in liquids.

A considerable amount of research in the last decade has been devoted to meeting these demands on imaging at extreme space and time-scales. Yet there is always an inherent trade-off between the image quality and

acquisition time, as alluded to above. The trends in spatial and temporal resolutions shown in Figure 1(b) highlight that, in general, higher spatial resolution necessitates longer measurement times. This is naturally an issue of limited signal-to-noise. To attain SNR from a smaller sub-volume that is similar to that from a larger sub-volume, a higher photon dose onto that sub-volume would be required [45–48]. Therefore, a higher flux is needed if the same temporal resolution is to be achieved. Nevertheless, improvements have been made owing to the higher photon flux afforded by synchrotron sources, faster CMOS detector technology, and new sampling and reconstruction strategies (see below). These developments have been charted in Figure 1(b) for more than 250 dynamic *in situ* XRT experiments [25]. Further improvements in scan time and spatial resolution are discussed in Section 3.

2.1. X-ray absorption-contrast microtomography

In absorption-contrast μXRT , photoabsorption is the dominant X-ray-matter interaction [49]. In the experiment, an incident beam of X-rays I_0 penetrates a sample with a spatial distribution of attenuation coefficients μ . Along the beam path z , the forward-transmitted X-ray intensity I will drop according to the Beer-Lambert law [50], $I(x, y) = I_0(x, y) \text{Exp}[-\int_l \mu(x, y, z) dz]$, where the coordinates x and y are in the plane of the detector and l is the thickness of the sample. With Z as the atomic number, ρ_a as the atomic density, and λ as the X-ray photon wavelength, μ can be shown [50] to vary as $\rho_a Z^4 \lambda^3$ (except at energies corresponding to photoelectric excitations, where it is discontinuous). Thus, attenuation increases for heavier elements and lower energies. The goal of μXRT is to reconstruct the 3D distribution

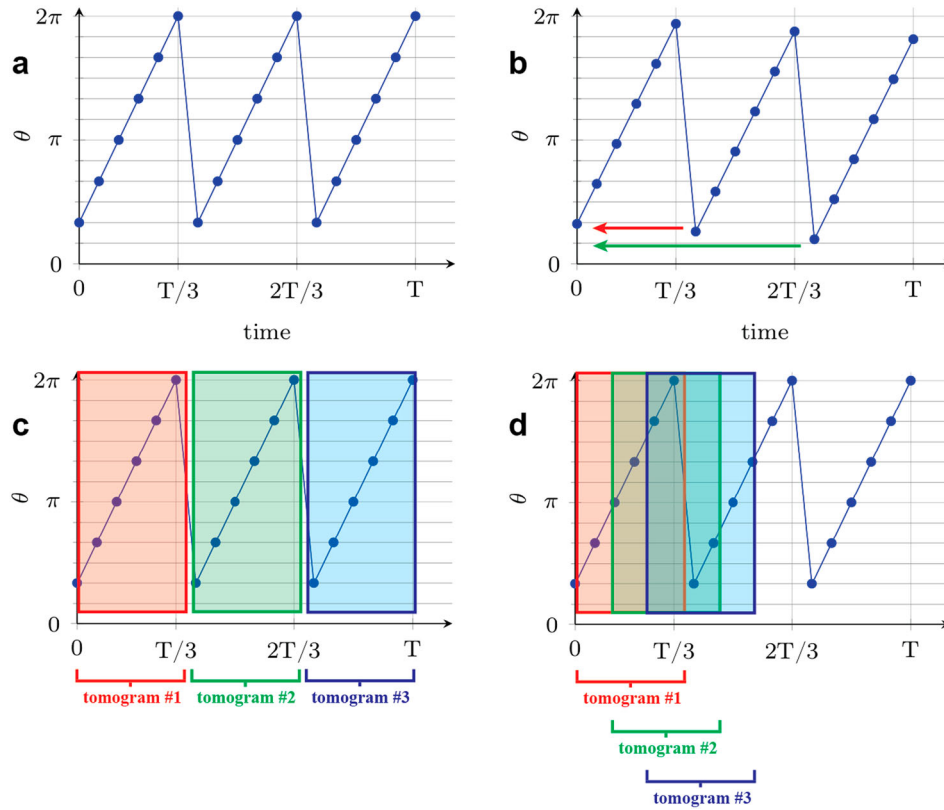


Figure 2. Various strategies for sampling and reconstruction. Projections (dark blue dots) are distributed over time (or sampled from the (θ, t) plane) as per the (a) progressive and (b) interlaced sampling scheme. Subsequently, they may be grouped together into (c) ‘fixed blocks’ (colored red, green, and blue) and reconstructed into tomograms, or else (d) reconstructed incrementally to improve the temporal resolution.

$\mu(x, y, z)$ from a set of X-ray projection images taken from different view angles θ .

Typically, the absorption-contrast μ XRT experiments are done with far fewer projections per tomogram than that required by the aforementioned criterion, owing to limitations on frame rate and/or disc space. However, this leads to severe aliasing artifacts in the corresponding reconstruction [51,52]. Instead, several new sampling schemes (beyond the traditional approach of *progressive sampling*, see Figure 2(a)) have been proposed to improve reconstruction quality [53–56]. One such strategy introduced in Ref. [56] is *interlaced sampling* (Figure 2(b)), wherein the projection angles for two consecutive tomograms are different but interleaved. In motion-compensated reconstructions (that regularize a dynamic reconstruction problem), the interlaced sampling scheme provides unique projection data of an evolving object, which altogether improves the quality of a time-sequence of reconstructed volumes. That said, interlaced view sampling when used with conventional reconstruction methods do not result in any gains since the number of projection images in each 180° rotation is insufficient to achieve Nyquist sampling for a single reconstruction of the object [56].

The μ XRT projections can then be grouped into fixed-blocks and reconstructed through filtered backprojection (FBP) [51] or direct Fourier methods [57,58]. One can subsequently concatenate these 3D tomograms to yield a 4D reconstruction, see Figure 2(c). Another related approach that has recently caught traction in the community is *incremental reconstruction* [59]. Here, the tomograms have a floating start angle, i.e. the ‘reconstruction window’ slides stepwise through the acquired projections (Figure 2(d)). The key benefit with this strategy is that sudden, critical events such as particle nucleation or film rupture [59] that might happen at the transition from one fixed-block tomogram to another will eventually move to the incrementally reconstructed tomograms in-between. The time-delay between subsequent incremental tomograms is (at best) the exposure time t_{exp} of the detector, instead of the total scan time $t_{exp} \cdot N_p$. Nevertheless, substantial blurring may influence the quality of the reconstruction and also our assessment of the critical event, if the objects-of-interest evolve too rapidly during a 180° rotation of the sample. Rather than reconstructing the tomograms independently, one can instead exploit time-space regularity in high-dimensional (e.g. 4D) datasets. Such is the focus of motion-compensated

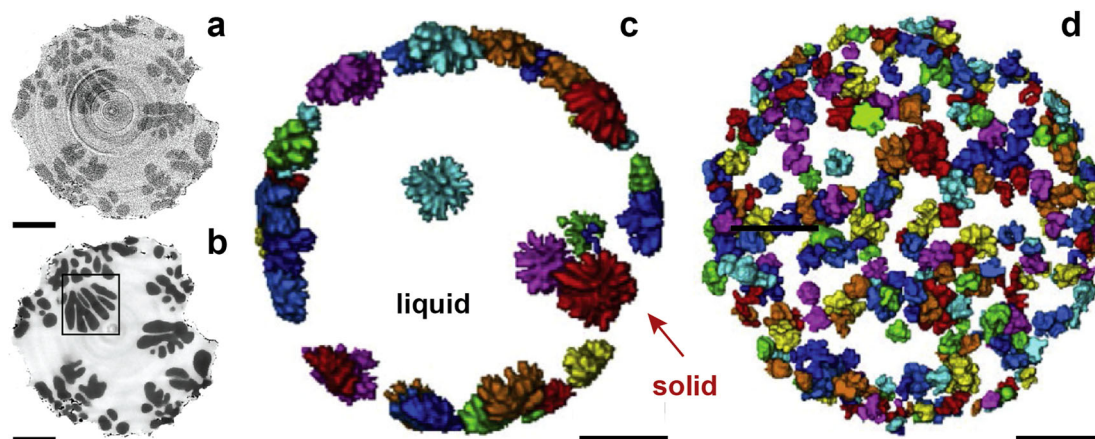


Figure 3. Visualizing dendritic solidification *via* μ XRT. Data quality depends on the reconstruction algorithm, e.g. (a) conventional FBP or (b) MBIR. Dendrites are dark grey while liquid is light grey. The latter approach suppresses the speckle noise and ring artifacts that are seen in the former. MBIR reconstructions were segmented and meshed to visualize the 3D grains in (c) NP-free and (d) NP-reinforced Mg-25wt%Zn-7wt%Al. Grains are randomly colored while liquid phase is white. Both experiments were conducted with a cooling rate of 3 °C/min. The tomograms show a nearly equivalent solid volume fraction of 2.5%. NPs cause substantial grain refinement. All scale bars measure 300 μ m. Retrieved from Ref. [76] with permission from Elsevier.

techniques such as *model-based iterative reconstruction* (MBIR), which enforces regularity using simple space-time prior models that penalize large movements of an object over time [56,60–62]. Other motion-compensated reconstruction techniques use *a priori* motion estimation [63,64] or estimate motion along with the unknown object [65].

Metallurgists have only recently taken advantage of the above innovations in sampling and reconstruction to achieve high temporal resolution in dynamic *in situ* μ XRT experiments. We highlight just a few examples from a larger body of work [66–74]. One of the earliest comes Gibbs and coworkers [75], who combined interlaced sampling and MBIR [56] to capture the 3D morphology of freely growing dendrites in an Al-24wt%Cu alloy. The Cu constituent provides a natural source of absorption contrast between solid and liquid phases (the partition coefficient is 0.15). Their observations revealed that the growth behavior of metal dendrites is remarkably different from that of transparent organic analogs, in terms of local curvatures, solid volumes, and interfacial areas [75]. Lee and coworkers [76] have extended our understanding of these phenomena by probing dendritic solidification in nanoparticle (NP)-reinforced alloys *via* μ XRT. Data were reconstructed through MBIR [62], which offered substantial improvement in signal-to-noise over FBP (compare Figures 3(a–b)). Analysis of the 4D data with and without NPs (Figures 3(c–d)) demonstrated that the NPs reduce the grain size by increasing the quantity of heterogeneous nuclei. The authors also speculate that the NPs reduce the effective solute diffusivity in the liquid, acting as a ‘growth restrictor’ [76]. This leads to compositional heterogeneities and variations in

driving force, leading to complex and hyper-branched dendrite morphologies (see also Ref. [31]). While these results are no doubt important in comprehending the processing of metal matrix nanocomposites, they do not show the NP-dendrite interactions *directly*. This is because of the limited spatial resolution associated with μ XRT.

2.2. X-ray nanotomography

While μ XRT has been used widely in studying the dynamics of solidification in metallic materials, the relative low spatial resolution precludes our observation of the early stages. X-ray nano-imaging provides resolving capabilities at tens of nm, which is roughly on the order of the size of the critical nucleus in metals. There are a few types of full-field microscope configurations suitable for nano-scale X-ray imaging [77,78]. Two of them are promising in studying fast dynamic behaviors: The first type is based on Fresnel zone plates (FZP) as X-ray objective lenses to magnify projection images [79,80]. The microscope based on this type of approach is called a *transmission X-ray microscope* (TXM). Similar to an optical microscope, a TXM is composed of an X-ray condenser and a FZP objective. A sample is placed between the condenser and the objective, and the magnified image of the sample is projected on an X-ray camera at the image plane. The achievable spatial resolution with TXM is limited to the numerical aperture of the zone plate. Typically, 30 nm resolution can be achieved easily at synchrotron sources. There are also demonstrations of sub-20 nm spatial resolution with TXM. Similar to an optical microscope, a TXM has limited FOV and depth of focus

that are determined by the numerical aperture of the objective lens.

The second type of full-field nano-imaging microscope, known as a *zoom microscope*, is based on X-ray focusing optics [77,78,81]. The incident beam is focused onto a small spot that is used as the secondary X-ray source to illuminate a sample downstream. A camera is located further downstream to record the sample's geometrically magnified images. By changing the sample location between the secondary source and the camera, the magnification of the projection images can be varied. The spatial resolution is ultimately limited by the size of the secondary source. With synchrotron radiation as X-ray sources, the incident beams are always partially coherent. Therefore, the images always have phase contrast due to X-ray beam interference [82]. The phase-contrast images have artificially enhanced edge features that can be used to identify the interfaces in materials. However, without further processing, the images are not suitable for quantitative imaging because the interference effects distort the gray-scale values of each phase, which are supposed to be linear with the attenuation coefficient of the phase. There are a number of so-called *phase retrieval* algorithms to extract electron density information of different phases in a sample. *Holotomography* is a technique that provides superior performance than most other phase retrieval algorithms [83]. However, to obtain a hologram, one must take scans at few different source-sample-camera configurations. Thus it is not suitable for dynamic *in situ* studies. Nonetheless, phase retrieval can still be done using single images [84].

As discussed above, the desired photon dose is inversely related to the achievable spatial resolution. Therefore, nano-imaging is slower than micro-imaging provided the X-ray source is the same. To harness nano-imaging in studying fast dynamical events, it is necessary to improve the photon delivery efficiency from an X-ray source to the X-ray camera. It is not until very recently that the application of nano-X-ray tomography (nXRT) in metallurgy has been demonstrated. One example with TXM was done at the Full-field X-ray Imaging beamline at National Synchrotron Light Source II at Brookhaven National Laboratory, USA [36]. Benefiting from the good match between X-ray source emittance and the FZP objective lens phase space, a one-minute nXRT scan of a metal substitution reaction was demonstrated. The scan was fast enough to catch the transient process wherein Ag whiskers grew rapidly on a Cu substrate in AgNO₃ solution. The experiment achieved 60 nm spatial resolution with 30 nm pixel size [36]. Recent work at this beamline further reduced the scan time down to a half minute per scan with even smaller pixel size, opening the door to further studies of kinetic phenomena. Another

two experiments were done with a zoom microscope by Villanova and coworkers [25] at Nano-Analysis beamline at European Synchrotron Radiation Facility, France. The microscope utilizes Kirkpatrick-Baez optics [83] to generate a quasi-monochromatic (1% bandwidth) and high-energy beam focused to a spot size of $50 \times 50 \mu\text{m}^2$. The experiments showed that it is possible to capture relatively fast processes in 3D, such as liquid droplet nucleation at about 100 nm pixel size in 20–30 s per tomogram [25]. This topic is particularly relevant to additive manufacturing (e.g. selective laser melting [85]), which involves the full or partial melting of powder particles; during the partial remelting of alloys, liquid droplets are formed [86,87]. Through *progressive sampling*, Villanova and coworkers were able to continuously scan an Al-Cu sample undergoing remelting, at a rate of 20 s per tomogram (Figure 4). The enhanced spatial and temporal resolutions afforded by 'fast' nTXM allowed the team to quantify the critical size and rate of nucleation, respectively, and compare their estimates to those made using quenched specimens [25].

Battery researchers, too, have benefited from developments in nTXM. *Operando* imaging is typically done in an 'interrupted' manner (*vide supra*), whereby a battery cell is imaged at discrete points in time during an electrochemical cycle. That is, the sample is probed only intermittently in order to reduce the total amount of X-radiation during the entire measurement and hence limit the beam damage [88]. One can probe a number of different anode and cathode architectures in this manner to identify and mitigate the dominant failure mechanisms, see, e.g. Refs. [89–91].

2.3. X-ray diffraction-contrast tomography

While the absorption-based techniques described above offer advantages for 4D metals research, they lack the ability to provide information on a very important feature of the material microstructure, namely the crystallographic orientations of the individual grains making up the specimen. If the strong transmission signal is blocked by a beamstop on the detector, the substantially weaker diffraction signals from the crystal lattices of the individual grains can be detected and used in a tomographic manner to reconstruct a so-called grain map of the grain shapes and crystallographic orientations. X-ray diffraction-contrast tomography (DCT) [92–94] and the related techniques three-dimensional X-ray diffraction (3DXRD) microscopy [7,95,96] and high-energy diffraction microscopy (HEDM) [97–99] were developed in parallel with the purpose to provide 3D grain maps of polycrystalline materials. Like their attenuation-contrast counterparts, these synchrotron-based techniques are all

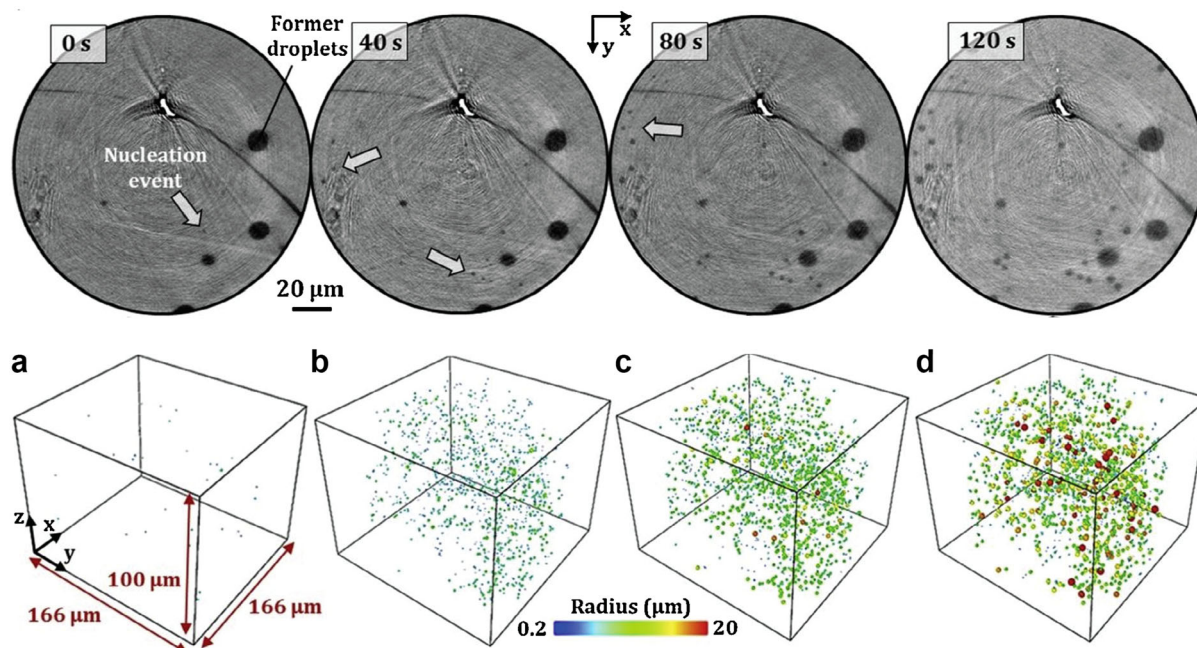


Figure 4. Visualizing liquid droplet nucleation *via* nTXM at the (a) 0 s, (b) 40 s, (c) 80 s, and (d) 120 s mark. *In situ* dynamic experiment was done by heating an Al-2.6wt%Cu alloy sample from 540 °C to 625 °C at a rate of 3 °C/min., while projections were recorded continuously. Top row shows 2D reconstruction slides while bottom row shows 3D renderings of nucleation events. Droplets colored according to their radius and solid phase is rendered transparent. Retrieved from Ref. [25] with permission from Elsevier.

non-destructive tools for bulk characterization, enabling 4D studies of grain structure evolution over time.

Grain mapping comes in a number of different varieties that can be grouped according to temporal resolution and spatial resolution. High temporal resolution at the expense of limited spatial resolution can be achieved with the so-called far-field variants of 3DXRD and HEDM. *Far-field* refers to the fact that a highly efficient detector with a pixel size in the range 50–200 μm is situated far away (20–100 cm) from the sample such that fast scans (of the order minutes) with a high angular resolution are possible. Information about grain shapes is lost due to the limited pixel size of the detector, while information on the center-of-mass positions, volumes, crystallographic orientations, elastic strains and stresses of the grains can be obtained. In the early days, this type of investigation was conducted to study grain nucleation and growth upon precipitation [100] and even solidification [101], but recently applications have moved toward deformation behavior of metals, e.g. tracking grain-resolved lattice rotations and stress evolutions during tensile deformation [102,103].

Near-field 3DXRD and HEDM, on the other hand, enables the user to measure full grain morphologies and intra-granular orientation distributions at the expense of limited time resolution (of the order hours). The name *near-field* refers to the fact that a high-resolution (1–5 μm pixels) detector is placed very close (< 10 mm) to the

sample. The line beam illuminating the sample cross-section is confined to a height of less than 10 μm to enable distinction between orientation and shape effects in the extent of the measured diffraction spots. Measuring a 3D volume thus requires stacking the reconstructions of a series of consecutively scanned layers. In this way, it is possible to monitor, for instance, the shape of growing grains during recrystallisation and recovery [104,105], grain rotations [106] and topological changes [107,108] during coarsening, and the nucleation of annealing twins during grain growth [109].

The synchrotron implementation of DCT uses a monochromatic box beam, as for the far-field 3DXRD/HEDM, and a high-resolution near-field detector. From a 360° scan comprising 3600–7200 DCT projections, the average crystallographic orientations and full morphologies of all illuminated grains can be reconstructed for samples with limited intra-granular misorientations. Recently, a comparative study using DCT and near-field HEDM to investigate the same sample, a slightly deformed (1%) aluminum alloy with an average grain size of ~100 μm, showed that DCT can detect sub-grain boundaries with disorientations as low as 1° and that HEDM and DCT grain boundaries are on average 4 μm apart from each other [110]. The unique set of key advantages of DCT—the ability to repeatedly cover large volumes (1000+ grains) in a reasonable time frame (hours) with a spatial accuracy of a few micrometers

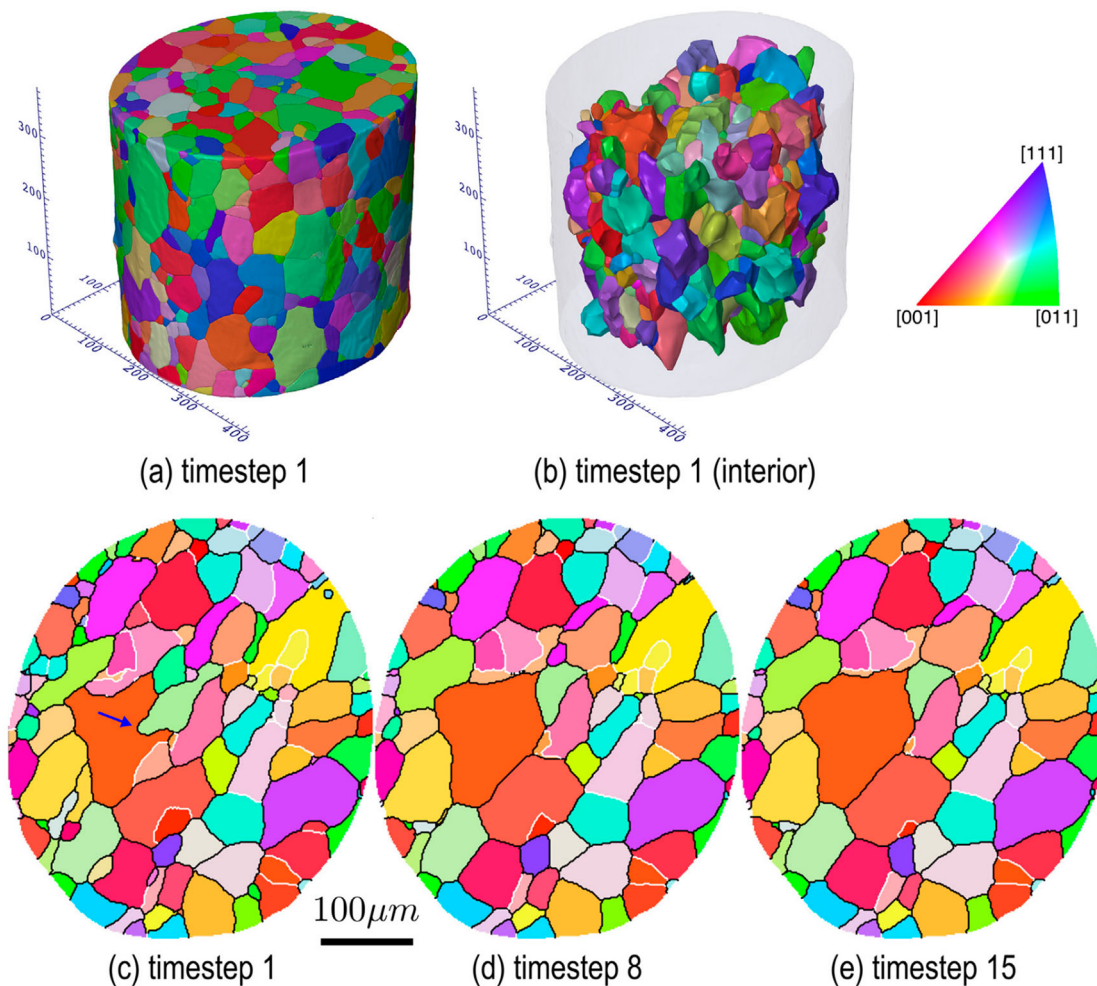


Figure 5. DCT results for grain growth in pure iron. Above: 3D grain maps for time-step 1, displaying (a) all grains and (b) only interior grains. Below: one section of the 3D grain map for time-step (c) 1 (0 min. of annealing at 800 °C), (d) 8 (40 min), and (e) 15 (75 min). Color represents the grain orientation along sample rolling direction (see the insert triangle), while black and white lines in (c)–(e) represent boundaries with misorientation above and below 15°, respectively. Retrieved from Ref. [112] with permission from Elsevier.

enabling extraction of experimental grain boundary characters—were exploited by Trenkle and coworkers to study 3D microstructural evolution in strontium titanate [111] and Zhang and coworkers in a study of grain growth in pure iron [112]. Zhang and coworkers measured DCT grain maps at 15 timesteps during interrupted annealing at 800 °C, with timestep 1 corresponding to the initial unannealed state and timestep 15 corresponding to 75 min. annealing. Example 3D grain maps and slices through these can be seen in Figure 5. Based on the grain maps, a comprehensive statistical analysis of the crystallographic, geometrical and topological evolution during grain growth was performed. It was concluded that the isotropic MacPherson-Srolovitz model [113] for grain growth described the experimental data well when averaging over the entire grain ensemble within short time intervals. At the level of individual grains, not surprisingly, the isotropic model failed to predict exact growth rates of the anisotropic iron grains.

Hence the anisotropic grain boundary mobilities and energies were extracted from the data by fitting to a phase-field model in a second paper [114], where it was found that reduced mobilities vary by three orders-of-magnitude and in general exhibit no correlation with the boundary's five macroscopic degrees of freedom, implying that grain growth is governed by other factors than the crystallography of the grain boundary.

3. Outlook and opportunities

4D XRT has come a long way over the past decade, in terms of our ability to probe microstructural changes at extreme space- and time-scales (Figure 1). We have little doubt that the use of XRT will continue to grow significantly, given the upgrade of synchrotron facilities, progress in data science, expansion of laboratory techniques, and development of correlative imaging platforms, all of which are discussed below.

3.1. Light source upgrades

Dynamic *in situ* 4D XRT is impossible without the bright synchrotron sources. The synchrotron-based techniques have been evolving rapidly owing to recent accelerator technologies, including the diffraction limited synchrotron ring (DLSR) [115]. Compared to the current third generation synchrotron sources, the DLSR sources have smaller source sizes but similar photon fluxes. This means DLSR sources provide a higher source brightness and higher spatial coherence. Higher coherence illumination would improve phase contrast imaging in term of the phase sensitivity. It would be particularly valuable to use phase contrast imaging to distinguish different phases in materials that have similar electron densities. Such is the case for white cast iron, which is comprised of two eutectic phases (γ -Fe and Fe₃C) that have similar real and imaginary components of the complex refractive index. That said, the DLSR sources may not help to improve temporal resolution in 4D μ XRT applications because the temporal resolution in μ XRT depends on the total photon flux incident upon a sample.

On the other hand, DLSR sources would have a major impact on nano-imaging. Since the emittances of DLSR sources will not match the FZP phase space, TXM will not be a suitable option with a DLSR source. However, with DLSR sources, it is possible to focus more photons into a small spot. Therefore, the *zoom microscopes* will largely benefit from DLSR sources. Consequently, the temporal resolution with zoom microscopes based on DLSR sources can be improved by a few orders-of-magnitude.

Another field of research that will benefit tremendously from the DLSR source is dark field X-ray microscopy [116,117], a non-destructive diffraction-based technique for the three-dimensional mapping of orientations and stresses on lengths scales from 100 nm to 1 mm within embedded sampling volumes. Dark field X-ray microscopy allows ‘zooming’ in and out in both direct and angular space, for instance to study the evolution of ultra-low-angle boundaries inside individual grains during recrystallisation [118]. It is estimated that with the advent of the DLSR source the data acquisition speed can be improved by several orders-of-magnitude, making this a promising technique for studying even more challenging dynamic processes *in situ* and *in operando*.

3.2. Data analysis techniques

Beyond improvements made to the source, advanced data analysis techniques are indispensable to dynamic *in situ* XRT experiments. New tomography reconstruction techniques such as the aforementioned time-interlaced MBIR have demonstrated success in decreasing the

number of projection images required for tomography reconstructions and hence improving the temporal resolution. With *incremental tomography*, one is able to catch rapid microstructural changes with a temporal resolution that is comparable the exposure time of a single projection image. Another recently developed algorithm [119] demonstrated that it is possible to extract dynamic structure evolution information quantitatively from continuous tomography scan data based on digital volumetric correlation (DVC) and differential data chunking scheme (Figure 2(d)). This algorithm can utilize motion-induced reconstruction artifacts in DVC analysis, facilitating the detection of fast dynamic processes and the rates at which they evolve. It relaxes the requirements on the sample rotation velocity ω to achieve higher temporal resolution Δt , and thus helps to reduce disturbances induced to the sample due to the fast rotation (e.g. convection currents that arise from centripetal forces).

Machine learning (ML) and artificial intelligence (AI) techniques also provide exciting opportunities to extract useful information from large tomographic datasets. There are quite a few studies [120–123] that deal with the missing angle issue, sparse angular sampling issue, and poor signal-to-noise issue, in tomographic reconstructions. These methods utilize prior information learned from a pool of existing data to either generate the missing information or remove the artifacts due to the imperfect data from dynamic tomography scans. Naturally, these techniques have also been applied to analyze and extract information from the reconstructed datasets. For instance, neural networks and random forests have both been employed to solve the segmentation problem, wherein each pixel is assigned to a particular phase [124,125]; evolutionary algorithms have also been used to solve the alignment problem associated with interrupted *in situ* scans, including DCT [126,127]; finally, deep learning has demonstrated success in detecting key features and tracking their evolution over time [128–130]. These examples represent only the tip of the iceberg. It is the authors’ belief that the impact of ML on metals characterization has not yet been fully realized.

3.3. Laboratory-based imaging

The recent advances of both hardware and software towards time optimization of synchrotron μ XRT also find counterparts in laboratory-based dynamic μ XRT [131,132]. Scan times less than 10 s can now be achieved, enabling studies of real time processes such as fluid flow through porous geological media [131], collapse of beer foam or muffin baking [133]. One particular challenge in such *in situ* or *in operando* investigations is the design of the cell to control the sample environment, for instance

constant fluid flow or elevated temperature. At the synchrotron, the cell must allow sample rotation, but in a laboratory setting one can choose to have the sample and cell stationary and let the source and detector rotate around the sample [133], which may be advantageous where the structure could be influenced by fast rotations (e.g. solidification fronts) or environments demanding substantial wiring (e.g. electrochemical cells). A final example on state-of-the-art metals research using laboratory μ XRT is the *in situ* observation of solidification in an Al-Zn alloy. The sample cell was set up with a temperature gradient enabling 3D investigations of the solid-liquid interface with a temporal resolution of 3 min. It was found that the growth velocity decreased continuously (at fixed thermal gradient) as the interface morphology transformed from dendritic to cellular to planar [134], consistent with theoretical predictions [13].

Also, grain mapping *via* DCT has moved into the laboratory under the name LabDCT, laboratory diffraction contrast tomography [135–138]. Where the synchrotron DCT uses a parallel, monochromatic beam of high flux, LabDCT is implemented on a μ XRT system with a divergent, polychromatic beam of limited flux. However, since LabDCT exploits the full energy spectrum of the laboratory source, only ~ 200 projections are needed for grain map reconstruction, overcoming the flux limitation and enabling LabDCT in the laboratory. LabDCT has been used to study microstructural evolution such as sintering of copper particles [139], annealing of industrial steels [140] and abnormal grain growth in Armco iron [141–143]. In the latter study, Sun and coworkers measured grain maps comprising 1200+ grains and 8000+ boundaries at three time-steps; the large datasets provided new vision on the balance between driving force (capillarity) and mobility during abnormal grain growth.

3.4. Correlative microscopy

As powerful as any one of the above XRT-based techniques may be, usually no single measurement is sufficient to fully characterize a material. Instead, *multi-modal* and *multi-scale* correlative tomography workflows [144] allow one to capture microstructural heterogeneities over multiple length-scales. Here are a few salient examples: μ XRT and DCT can be performed nondestructively and in series to probe a material that is simultaneously polyphase and polycrystalline [127,145–149]; likewise, nTXM and X-ray absorption near edge spectroscopy (XANES) can be combined to measure the 3D distribution of different chemical species at the nanoscale [150]; nTXM can also be combined with μ XRT to better understand how hierarchical structures come to be; and finally, 3DXRD and Bragg

coherent diffraction imaging [151] can be done in succession to ‘zoom in’ on defect motions and behaviors in larger-sized grains in polycrystals [152]. In the latter two cases, one can mitigate an inherent disadvantage of high-resolution techniques in that they focus on a region that may not be representative. By providing large-scale contextual information, one can ensure that the material characterized at the nanoscale is characteristic of the features-of-interest [153].

In order to precisely cross-correlate information obtained from multiple sources, we will need to determine very accurately the microstructural locations of the respective 2D/3D datasets. This can be done through automated registration procedures to correct for misalignment and distortion [154,155], yet the task is far from trivial. A related concern is the explosion of data from these multimodal sources, and the amount of time it takes to collect that data. For instance, a 3D XANES measurement can be done in a matter of minutes to hours [150], depending on the X-ray source flux and detector efficiency. At the Full-field Imaging (FXI) beamline at National Synchrotron Light Source II, 3D XANES scans can be routinely conducted within a single hour at 60 nm spatial resolution. Time reduction can be achieved by reducing the number of energy points to the minimum necessary to discriminate between phases (although this would require *a priori* knowledge of the sample chemistry) or by reducing the number of projection images (although this may influence the reconstruction quality). The former point can be addressed *via differential tomography*, which is especially useful in discriminating certain elements’ distributions in 3D [156]. As an example, suppose the investigated material is a Ni-based alloy. Ni has its K-edge energy at 8.333 keV. Two nXRT scans can be conducted at one X-ray energy just below (e.g. 8.3 keV) and another just above (8.4 keV) the Ni K-edge. Other elements have negligible changes in their X-ray attenuation coefficients at these two energies. Subtracting one tomographic dataset from the other would highlight Ni distributions in the sample. This approach holds promise in revealing microsegregation patterns in multicomponent alloys.

Acknowledgments

A.J.S. acknowledges support from the U.S. Department of Energy (DOE), Office of Science, Office of Basic Energy Sciences, under Award No. DE-SC0019118. P.W.V. acknowledges support from the U.S. Department of Commerce, National Institute of Standards and Technology as part of the Center for Hierarchical Materials Design (CHiMaD) under Award No. 70NANB19H005. This research used resources at 18-ID FXI beamline of the National Synchrotron Light Source II, a U.S. Department of Energy (DOE) Office of Science User Facility

operated for the DOE Office of Science by Brookhaven National Laboratory under Contract No. DE-SC0012704.

Disclosure statement

No potential conflict of interest was reported by the author(s).

Funding

This work was supported by the U.S. Department of Energy (DOE), Office of Science, Office of Basic Energy Sciences [DE-SC0019118]; Brookhaven National Laboratory [DE-SC0012704]; and the U.S. Department of Commerce, National Institute of Standards and Technology [70NANB19H005] as part of the Center for Hierarchical Materials Design (CHi-MaD).

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