

X-RAY

imaging detectors

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Advances in detector technology, in concert with new synchrotron sources, x-ray optics, and computational methods, are opening new ways to probe the structure and dynamics of matter.



A child in North Africa 160 000 years ago died and left a well-preserved jaw that was found in Jebel Irhoud, an archaeological cave in Morocco. Remarkably, we know the child died when it was about 2839 days old.¹ We know that because human teeth grow with microscopic, daily enamel accretions that are clearly visible in a micron-scale x-ray tomographic reconstruction of the child's tooth (see figure 1). Indeed, such microtomography provides one of the most accurate means to date of identifying incremental changes in the human fossil record, which are crucial to understanding the origins of anatomical modernity.

The tomographic reconstruction of a fossil is just one of many examples in which x-ray methods can nondestructively determine in exquisite detail the structural or chemical makeup of materials. Other examples include the revealing of paintings hidden under famous masterworks and of mathematical proofs, written in now-faded iron-laden ink, on the Archimedes Palimpsest (see the article by Reviel Netz in *PHYSICS TODAY*, June 2000, page 32); the tracking of migrating fish by measuring the accumulation of metals in their "earstones"; and the analysis of wood to determine the dates of famous volcanic eruptions. Modern x-ray methods also extend to transient phenomena as disparate as insects swallowing and breathing in real time, electromigration in nanowires, rapid phase transitions, turbulence in microjets, and shock waves in gases.

In all those cases, the x-ray detector limits what can be done. Ideally, detectors would offer high spatial, temporal, and energy resolution. But practically, they must be designed to optimize the resolution of some aspects, often by compromising resolution in others. Moreover, although x-ray detectors have improved generally in recent years, largely thanks to improvements in electronics, they have not kept pace with x-ray sources and optics. When Wilhelm Röntgen first investigated x rays in 1895, he used a

Crookes tube—an early version of today's electrical discharge tube—to generate them and photographic film emulsions to record them; the photograph of his wife's hand, shown above, is a famous example. Crookes tubes have been relegated to the museum, but film emulsions are still very much with us.

In this article I outline the development of x-ray detectors as guided by experimental opportunity—or, more specifically, the need for enhanced resolution in some aspects of the measurement. And to narrow the scope, I focus on imaging detectors at hard x-ray synchrotron radiation sources.

Classic medical imaging

Medical applications drove the historical development of x-ray imagers. Röntgen discovered x rays by noticing that some invisible rays emanating from his Crookes tube were exciting a glow on a nearby phosphor screen. Within days he was recording x-ray images on photographic film. Film and phosphors are still the most prevalent area detectors of x rays.

X-ray film dominates medical imaging. It is cheap, simple to use (if not to develop), and readily available in large sheets. Film emulsions are thin, so they provide excellent micron-scale spatial resolution, but they are also inefficient x-ray absorbers—that is, they have low stopping power. Most important, film stores the x-ray image in a compact, permanent form. It is only within the past two decades that mass digital storage has become sufficiently inexpensive to challenge film as an information storage medium.

Storage phosphors, also known as imaging plates and first commercially developed in Japan in

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the 1980s, are an alternative to film. In a typical luminescent material, electrons excited by ionizing radiation to singlet or triplet states promptly de-excite by emitting visible photons. But materials such as europium-doped barium fluoride bromide exhibit a photostimulated luminescence: Ionized electrons in the material are trapped at metastable, intraband states with lifetimes on the scale of days. The material thereby stores an image in the distribution of trapped states. Subsequent exposure to visible light excites the traps and leads to recombination of electrons and holes and the release of visible light.

In practice, the storage phosphor plate is exposed to x rays and is read out by raster scanning the plate with red laser light. The resulting blue photoemission at each location on the screen is filtered to exclude the scattered red light and then recorded with a photomultiplier. The number of blue photons is proportional to the local x-ray dose.

Storage phosphors are efficient because many optical photons are produced for each stopped x ray. The plates are reusable and easily reset by flooding them with visible light to empty the traps. Acceptable stopping power and spatial resolution are obtained by adjusting the thickness of the phosphor layer, typically to 50 μm . The output signal is proportional to the incident x-ray dose over many orders of magnitude, so the detectors have a very wide, linear dynamic range. The primary disadvantage is the need to read out the phosphor screen in a raster scanner, which takes minutes. That process is acceptable for medical imaging but limits scientific applications.

There's another important difference between medical and scientific imaging. Medical imaging requires that a radiologist be able to see sufficient detail and contrast to make a diagnosis. Human beings are good at recognizing subtle image features, even in detector materials whose response to x-ray dose is nonlinear and whose x-ray sensitivity varies slowly across the detector area. Scientific imaging places many more constraints on the detector. Ideally, it should measure as a function of position the absolute x-ray dose and wavelength in a given exposure period. In pursuit of that goal, researchers have adopted two different detection strategies.

The indirect and the direct

X-ray detectors may be thought of as a relay of components that serve to stop the x rays in a material and produce an initial signal that may then be amplified or converted to another form of energy before being detected as, for instance, an electrical signal. As outlined in figure 2, the energy converter may be a light-emitting phosphor layer, whose signal is amplified by an image intensifier before being captured on a CCD.² That process is an example of indirect detection: Signal quanta produced in the x-ray stopping medium (visible light in the phosphor) differ from the quanta ultimately recorded (charge in the CCD). In a direct detector, by contrast, the energy converter and the final imager are the same component—a strip of x-ray film, for instance.

Phosphors are still the basis of most scientific x-ray imagers, which typically operate indirectly.

The desired phosphor characteristics follow from the imaging requirements of the previous section: Linearity with dose implies a single, highly dominant pathway for excitation and de-excitation of energy levels in the material. Only a small number of the commonly used phosphors—for example, thallium-doped cesium iodide and terbium-doped gadolinium oxide sulfide—meet that constraint. For a high quantum efficiency one must maximize both the fraction of x rays stopped—which can be achieved using a high-density form of a material with a high average atomic number—and the number of optical photons produced per unit of stopped x-ray energy. A high spatial resolution dictates the use of a thin phosphor layer.

Indirect x-ray detection, though, is inherently complex and may give rise to nonlinearities and image distortions. To see how, consider the most common detector configuration used for scientific imaging at synchrotrons: a thin phosphor screen deposited onto a fiber-optic taper that guides the emitted light into a CCD. (Fiber-optic tapers are used because they transmit light more efficiently than lenses in demagnifying an image onto a CCD.) Let's examine each of those components in turn.

To start, the physics of phosphor luminescence is itself complex. Ideally, the light emission should be prompt and strictly linear with incident x-ray dose. But real phosphors contain solid-state energy traps and defects and thus multiple de-excitation pathways that reduce that strict linearity. Practical phosphor screens are considered good if the luminescent emission integrated over a few milliseconds

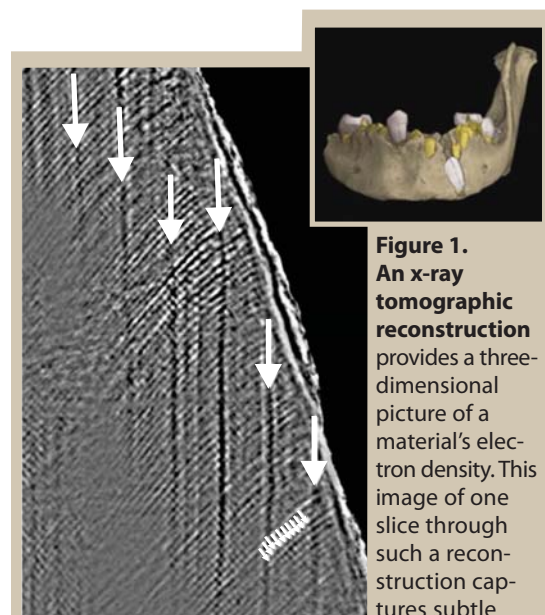


Figure 1. An x-ray tomographic reconstruction provides a three-dimensional picture of a material's electron density. This image of one slice through such a reconstruction captures subtle

density variations in the hard enamel of a living tooth—in this case, from the fossilized jaw of an ancient North African child (inset). The variations reveal periodic growth features known as Retzius lines (vertical arrows). The number of cross striations (small diagonal arrows)—daily accretions, each about 5 microns apart—between the Retzius lines provide an accurate record of the child's age. (Adapted from ref. 1.)

is linear with dose over four orders of magnitude.

Since the luminescence is emitted in all directions, only some of the light is captured by the fiber-optic bundle. Scattered light limits the lateral spatial resolution and may create halos around intense x-ray features. Worse, the fiber bundle's light-capture efficiency depends on the depth of the phosphor layer in which the x ray is stopped. The sum total of those effects is to increase the variance in the number of light photons per x ray that is conveyed to the CCD. The result is a noisy image.

Once captured in an optical fiber, light can leak into adjacent fibers, which may also produce halos, or it can be absorbed by defects in the fiber, which results in spatially varying efficiency in light transmission. Fiber-optic tapers are made by drawing down cylindrical bundles of hot, soft glass fibers into smaller-diameter fibers. That process can plastically distort a bundle, which then geometrically distorts the transmitted image.

Finally, not all photons transmitted out of the fiber produce charge in the CCD. Thus the numerous steps in converting the x-ray image energy to light produce several undesirable characteristics: increased noise, a nonlinear dependence of signal on intensity, spatial variation in sensitivity, and geometric image distortions. What's more, the entire detection process is inefficient: A 10-keV x ray might

produce a few tens of electron-hole (e-h) pairs in the CCD. Calibration of those various effects, though possible, is difficult.

The better approach

Fortunately, direct-detection imagers introduced in recent years have markedly reduced the undesirable characteristics. Consider, for example, the conversion of a 10-keV x ray in single-crystal silicon. The x-ray energy is converted in less than a nanosecond into e-h pairs in a region about a micron across, with one pair produced for each 3.64 eV of x-ray energy. Moreover, the statistics for the process are extraordinary: A 10-keV x ray yields 2740 ± 20 e-h pairs. Note that 20 is less than $\sqrt{2740}$, so the process is non-Poissonian, which implies that there are limited channels where the x-ray energy can go, except to the production of e-h pairs. If the x ray is stopped in the junction region of a reverse-biased diode, the pairs follow electric field lines and can be quickly collected at the diode terminals with limited lateral spread.

Therefore, not only are energy conversion and signal collection in a silicon diode simpler than that described for an indirect detector, they are faster (on the scale of nanoseconds) and offer high spatial resolution (on the scale of microns). Because thousands of e-h pairs are produced in silicon per x ray (compared with tens produced in phosphor-coupled CCDs), single x-ray detection is straightforward to achieve.

Of course, many things can still go awry. Electron-hole pairs can recombine, get trapped, or be produced thermally at surface or defect sites. The material must have a thickness L that is large enough to efficiently absorb x rays but smaller than the mean distance an electron can drift before being trapped or recombining with a hole: In general, if E is the magnitude of an electric field across the photoconductor, then L must be less than $\mu\tau E$, where μ is the drift mobility and τ is the mean carrier lifetime or trapping time.

Few available materials possess requisite characteristics, quality, and size needed for practical x-ray detectors. Commercially, large-area direct scientific imagers are based mostly on amorphous selenium (see the article by John Rowlands and Safa Kasap in *PHYSICS TODAY*, November 1997, page 24) and on single-crystal silicon. Photoconductive materials made of higher-atomic-number elements are especially desirable for their higher stopping power. Current R&D is focused on germanium, gallium arsenide, cadmium telluride, and cadmium zinc telluride.

Intelligent pixel arrays

Rapid advances in electronics have enabled engineers to design new x-ray detectors. One of the most versatile is the "bump-bonded," or hybridized, pixel-array detector (PAD), a technology that has been greatly advanced by the needs of high-energy physics. A PAD, shown in figure 3, consists of an upper sensor layer and a lower application-specific integrated-circuit layer of CMOS pixel electronics. The two layers are connected to one another via an array of pixel electrodes made

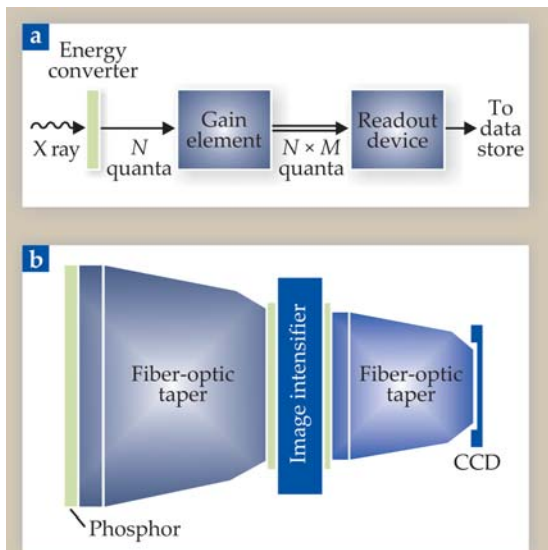


Figure 2. An imaging x-ray detector is essentially a relay of signal quanta. **(a)** The energy of an x-ray photon, for instance, may be converted in a phosphor to N visible photons, which are captured in a fiber-optic taper, amplified by an image intensifier with gain M , and then read out by a CCD and stored. **(b)** A specific configuration is shown. The intensifier works much like a photomultiplier: Visible photons strike a photocathode and generate photoelectrons that are accelerated across the intensifier's narrow gap and strike another phosphor layer, thereby producing many visible-light photons. The amplified image emitted from the intensifier is captured by another fiber-optic taper and conveyed to a CCD.

of indium or solder bumps between the layers.

X rays are incident on the sensor layer, which may consist of a 500- μm -thick, high-resistivity silicon. Absorbed x rays create electrons and holes that, provided a bias voltage is applied, are swept apart along field lines to top and bottom electrodes. Each bottom electrode is connected to its own pixel of processing circuitry in the lower integrated-circuit layer. A single hybrid module of that sort may be a few centimeters across and contain tens of thousands of pixels. Modules are arranged side by side to tile larger detectors with up to several million pixels.

Thanks to CMOS technology, an amazing amount of complex, radiation-hardened circuitry can be crammed into each of the pixels, thereby endowing them with considerable processing capability. Although x-ray pixels are currently relatively large, roughly 50–200 μm on a side, three-dimensional design techniques suggest that comparably powerful pixels only 20–30 μm on a side are feasible.³ For more detail on hybridized PADs in use or in advanced stages of development, see reference 4.

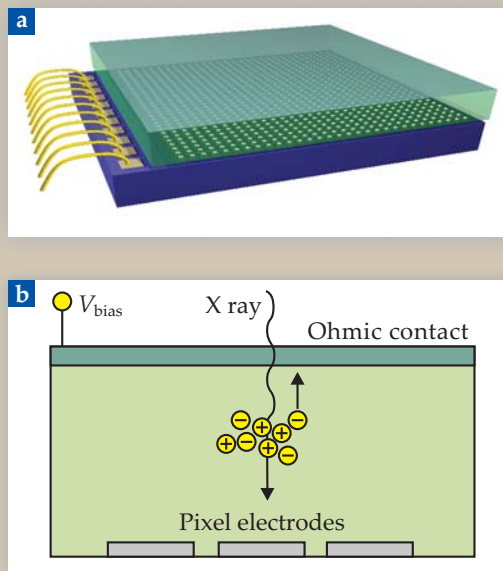
Imaging and electronics

X-ray imagers fall into two broad classes: Photon counters and integrators. Photon counters use pulse-height discrimination circuitry to detect pulses from individual photons. Such circuitry provides modest energy resolution, which is quite useful in monochromatic x-ray experiments for rejecting lower-energy Compton-scattered or fluorescent x rays from the sample. The need to process each x ray individually, however, typically limits the instantaneous count rate per pixel to a few megahertz. Another subtlety is that x rays captured near the boundaries of two neighboring pixels can contribute signal to both pixels. The challenge is to avoid overcounting, when both pixels record a count, or undercounting, when neither do because the signal falls below a given energy threshold.

Integrators avoid that problem by using an operational amplifier to sum the incident signal for an exposure period prior to digitization. Integrators are also advantageous in cases in which the instantaneous count rate is enormous; they are, in fact, the only practical approach with x-ray free-electron lasers (FELs), whose pulses are only femtoseconds in duration. A common misconception is that integrators are noisy. Because the signal from an x ray greatly exceeds typical amplifier noise levels, simple thresholding of the recorded image can easily provide single-photon noise performance with very high fidelity.⁵

On the other hand, integrators cannot reject Compton-scattered and fluorescent x rays in monochromatic experiments, because they sum the signal from all stopped x rays in a given pixel during an exposure. Integrators have a range of measurable dose defined by the voltage limits of the summing amplifier (typically the voltage of the power supply and ground) and to avoid saturating the amplifier must be read out and reset when the integrated signal approaches the maximum dose. Some designs, however, are “dead-timeless” because they remove

Figure 3. A pixel-array detector (a) consists of a layer of high-resistivity semiconductor (green) that is connected, pixel by pixel, by tiny metal bumps (silver) to a CMOS integrated circuit (blue). Each pixel in the circuit chip has its own electronics and contributes to signals in output wires (gold). **(b)** As shown in this cross section of the semiconductor layer, an absorbed x ray generates electron–hole pairs. If the conducting contact at the top of the semiconductor is positively voltage biased relative to the bottom pixel electrodes, electrons and holes drift vertically through the bulk along electric field lines, with the magnitude of the charge proportional to the energy of the x ray. Holes are collected on the bottom electrodes and passed through connecting bumps to a CMOS amplifier.



charge during the integration process and record the amount of charge removed. That workaround is effective as long as signal doesn’t accumulate faster than the circuitry can respond. The so-called mixed-mode PAD, for example, detects single 8-keV x rays with a signal-to-noise ratio of about eight, yet it has a maximum dose range per pixel that exceeds 10^7 x rays and can frame images at 1 kHz.⁶

Photon-counting detectors enable novel ways to acquire image data. For example, protein crystallography, one of the most prevalent and important x-ray imaging experiments, is typically done by rotating a protein crystal in a monochromatic x-ray beam. The normal operating mode with CCD detectors has been to rotate the crystal about one degree at a time. After each step the x-ray beam is shuttered off while the image is read from the detector. Sequential images make up the complete data set, which consists of hundreds of thousands of measurements of the relative intensities of Bragg-diffraction spots. PADs that stream data continuously from the detector now allow shutterless and continuous rotation of the crystal in the beam. Complete data sets have been acquired in just a few minutes, and acquisition times of tens of seconds are feasible.

That type of experiment still requires a protein crystal of sufficient diffraction quality and size—typically tens of microns across—to produce a complete data set of sufficient resolution before radiation damage destroys the crystal. For many proteins of interest, such as membrane proteins, growing that sort of crystal is difficult. There is evidence, however, that submicron protein crystals are easier to grow, and an alternative way to acquire complete data sets is to take diffraction images from many small crystals, each yielding a single diffraction image before the crystal is irreversibly damaged.

One implementation is the “diffract before destroy” method,⁷ recently performed at SLAC’s Linac Coherent Light Source (LCLS; see PHYSICS TODAY, April 2011, page 13). The basic idea is to hit a succession of submicron-sized particles, such as protein nanocrystals or even single protein molecules, with an ultra-intense x-ray pulse tens of femtoseconds in duration. That time is so short that the pulse is diffracted before photoelectron ejection causes the particle to “Coulomb explode” from the buildup of positive charge.

The need to capture an x-ray image in femtoseconds put extreme constraints on the detector design that was implemented at the LCLS. The rapid arrival of x rays required an integrating PAD able to tolerate a dose of several thousand x rays and yet operate with sufficiently low noise to be sensitive to single photons at the 120-Hz repetition rate of the LCLS.⁸

Spatial resolution

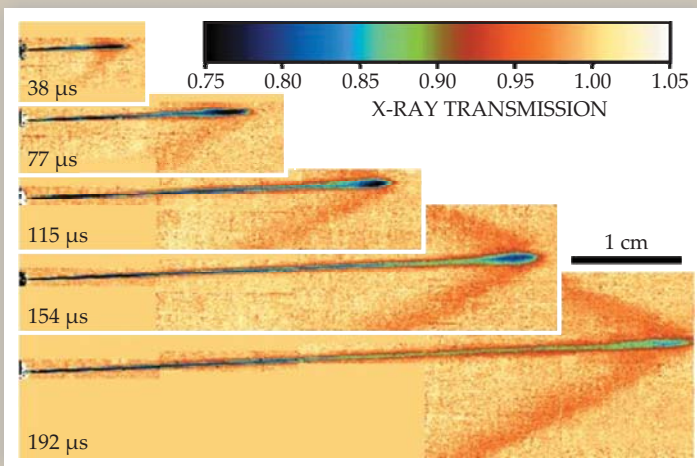
Much of the world is structured on submicron length scales; one need only look at the characteristic lengths of inhomogeneities in metals, composites, bones, and catalyst materials for examples. Modern synchrotron sources can produce intense x-ray beams that are superb structural probes at such dimensions. In diffraction applications, the x rays diverge from the specimen through a scattering angle, so increasing the distance between the specimen and the detector can spread the signal to accommodate a detector with a given pixel size.

In the microtomography example of the ancient tooth described earlier,¹ the x rays are barely deflected, so the resolution is set by the size of the detector’s pixels. In that case the structural information was encoded by x-ray absorption and phase shifts in scattered photons. So it is important to ask, What is the smallest practical size of a detector pixel?

In 2006, researchers at the European Synchrotron Radiation Facility (ESRF) introduced surface-doped luminescent crystals, lens-coupled to a CCD.⁹ In that project high-atomic-weight garnets were prepared as single-crystal plates about 0.5 mm thick. A thin layer doped with a few mole-percent Eu or Tb was grown onto the garnet surface, and only that doped layer, just 10 or 20 μm thick, was luminescent. The rest of the garnet acted as a substrate whose index of refraction matched the luminescent layer.

The researchers achieved submicron resolution from those crystals, but with low efficiency, using the hard x rays needed to penetrate the thick

Figure 4. Time-resolved radiographic images of a high-pressure diesel-fuel spray, as captured by a pixel-array detector. The fuel is doped with a cerium-containing compound to enhance x-ray absorption contrast, shown here in false color. The tip of the fuel jet is supersonic, and the corresponding shock wave, indicated by the Mach cone of compressed gas emanating from the jet tip, is clearly visible (red). (Adapted from ref. 11.)



specimens. Still, doped single-crystal films represent the state of the art in spatial resolution and were used for the tooth tomography. The inefficiency may be acceptable when radiation-hard materials are used, because they can tolerate the long exposure time required to do the experiment. But it severely limits what can be done with radiation-sensitive materials such as organic compounds. Achieving a spatial resolution substantially higher than that of the ESRF team will be difficult, given that the range of a primary photoelectron ejected when a hard x ray is absorbed is greater than a micron in most materials.

Response times

A central goal of countless experiments is to capture and resolve the rapid rearrangement of matter in response to stimuli. The ideal is to record an x-ray movie of some evolving specimen—the tip of a propagating crack, say, or a material passing through a phase transition in response to temperature, pressure, or light—with each frame acquired faster than the characteristic time of the process under investigation. Storage rings and FELs can generate intense x-ray pulses, each femtoseconds to tens of picoseconds in duration, at rates of up to tens of megahertz. And a new type of synchrotron x-ray source, known as an energy recovery linac, can push that rate into the gigahertz range.

Capturing movies of x-ray images at those rates is challenging. Limitations arise from the charge-collection, processing, and storage times of the electronics. Even among state-of-the-art imagers, a limited number of frames can be stored at a high rate (about 5–10 MHz) in analog form in the detector, typically as voltages on internal storage capacitors. The frames are then digitized and read into computer memory at slower rates after a sequence of

them has been acquired. That method, first demonstrated using an analog PAD to create x-ray images of fluid jets and shock waves,^{10,11} and exemplified in figure 4, is currently being developed for storage rings and x-ray FELs.⁴

An example of a rapid-framing imager is the adaptive-gain, integrating pixel detector being developed for the European XFEL. The light source, scheduled for completion in 2015, will deliver pulses shorter than 100 fs in trains of 5-MHz pulses. The goal of the detector project is a million-pixel instrument that can capture and store at least 200 diffraction patterns at that rate. The patterns will then be digitized and read out 10 times per second.

A dynamic range of 10^4 x rays per pixel per frame can be achieved by dynamically adjusting the gain of an amplifier connected to the x-ray diode pixel. The adjustment process relies on the fact that charge collection at the input of the amplifier is slowed by large signals. This provides sufficient time for fast electronics to dynamically switch in different feedback capacitors whenever the integrated output voltage approaches the amplifier's saturation limit.

New beginnings

Scientists know surprisingly little about the structure of many nonperiodic materials that make up our world, especially in the 10- to 1000-nm regime. Add in the variable of time and the list of unknowns becomes very long indeed, especially in the sub-picosecond regime: What is the structural basis for material failure under repeated stress? Why do catalysts die? Why do rechargeable batteries have a limited number of recharge cycles? What are the dynamics of processes in a biological cell? How do nutrients and pollutants migrate through soils? How are nanomaterials organized? The list goes on.

Our understanding of matter is always constrained by the availability of analytical tools through which we see the world. New x-ray sources in tandem with new x-ray optics and computational methods provide a window into the structural dynamics of matter at critical length and time scales. At the moment, the sources, optics, and methods are themselves constrained by the detectors used to capture the x rays. Fortunately, novel detectors are slowly catching up to the novel sources. Nonetheless, there is a long way to go.

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