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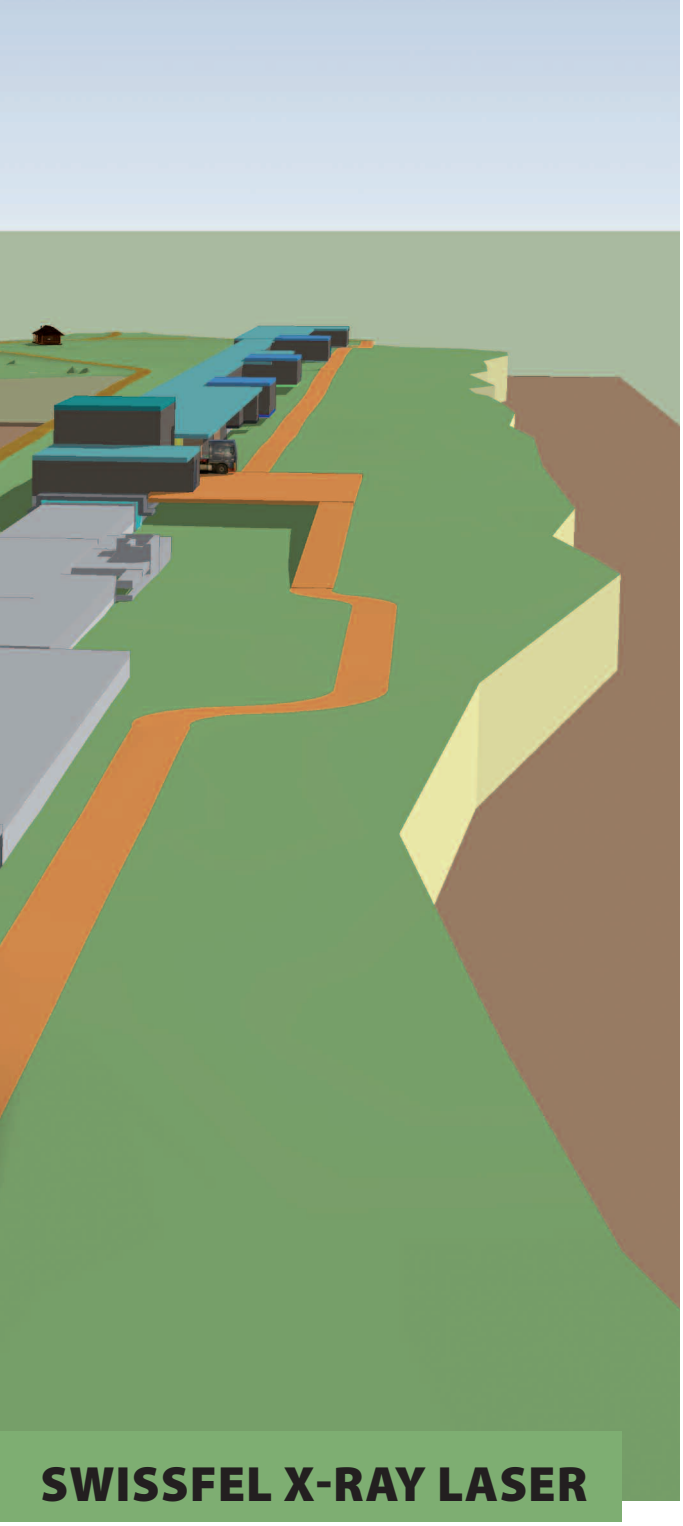
ULTRAFAST PHENOMENA AT THE NANOSCALE: NOVEL SCIENCE OPPORTUNITIES AT THE

Next generation X-ray sources, based on the X-ray Free Electron Laser (XFEL) concept, will provide highly coherent, ultrashort pulses of soft and hard X-rays with peak intensity many orders of magnitude above that of a synchrotron. These pulses will allow studies of femtosecond dynamics at nanometer resolution and with chemical selectivity, and will produce coherent-diffraction images of organic and inorganic nanostructures without the deleterious effects of radiation damage. The PSI SwissFEL is one of presently four XFEL projects worldwide.

Three enduring trends in investigations of structure and function in the natural sciences are "smaller", "faster" and "more complex". And by far the preferred tool of such investigations is electromagnetic radiation – light. Major advances in the investigation of matter with light have been the invention of IR and visible lasers, with their capability of bright, ultra-short (fs) pulses, and the advent of high-brightness synchrotron X-ray sources, which allow atomic-resolution imaging and chemically-specific spectroscopy. The new generation of light sources, the X-ray Free Electron Lasers (XFEL), will encompass all three above-mentioned trends in a single instrument. An overview of length and time scales (Figure 1) indicates where an XFEL will



◀ Preview of SwissFEL

SWISSFEL X-RAY LASER

make the strongest impact – in particular below the μm scale of optical microscopy and below the ns scale of pulsed synchrotron X-rays. We are speaking, for example, of biomolecular aggregates and small inorganic molecules, and of ultrafast magnetic recording and molecular vibrations.

An enduring wish is to image matter at X-ray resolution, a task which is complicated by the lack of low-aberration X-ray lenses. Also here, the XFEL offers a solution: coherent diffraction. By nature, XFEL radiation has a high degree of spatial coherence, implying that scattering by a thin phase object shows a rich "speckle" (Figure 2), which, using recursive phase-retrieval algorithms, permits a diffraction-limited, real-space reconstruction of the object [1].

Finally, a major difficulty in nm-scale studies of matter with both X-ray scattering and electron microscopy is sample degradation by radiation damage. Ironically, the ultra-short, ultra-bright pulses from an XFEL have the potential to circumvent this problem – by a technique called "diffract-and-destroy". Due to photoemission and Auger processes, a nanoscale object exposed to a single XFEL pulse (10^{11} – 10^{12} photons), will rapidly become positively charged and, on the scale of 50 fs, undergo destructive "Coulomb explosion" [2]. Hence a 10-20 fs XFEL pulse will record the scattering from, to a good approximation, as yet undamaged material. Note that approximately the same number of photons is supplied in a 20 fs XFEL pulse as in a full second of 3rd generation synchrotron radiation.

Operating principle

As in a synchrotron, XFEL light is created by the passage of a pulse of relativistic electrons along a periodic magnet array – an "undulator" (Figure 3a). For reasons of beam quality, a single-pass linear accelerator is required for the XFEL, in contrast to the circular storage ring of the synchrotron. And as for the synchrotron, the emitted wavelength λ is related to the undulator period λ_u, the maximum undulator field B_0 and the relativistic electron energy factor γ (total energy divided by rest mass energy) by the undulator equation:

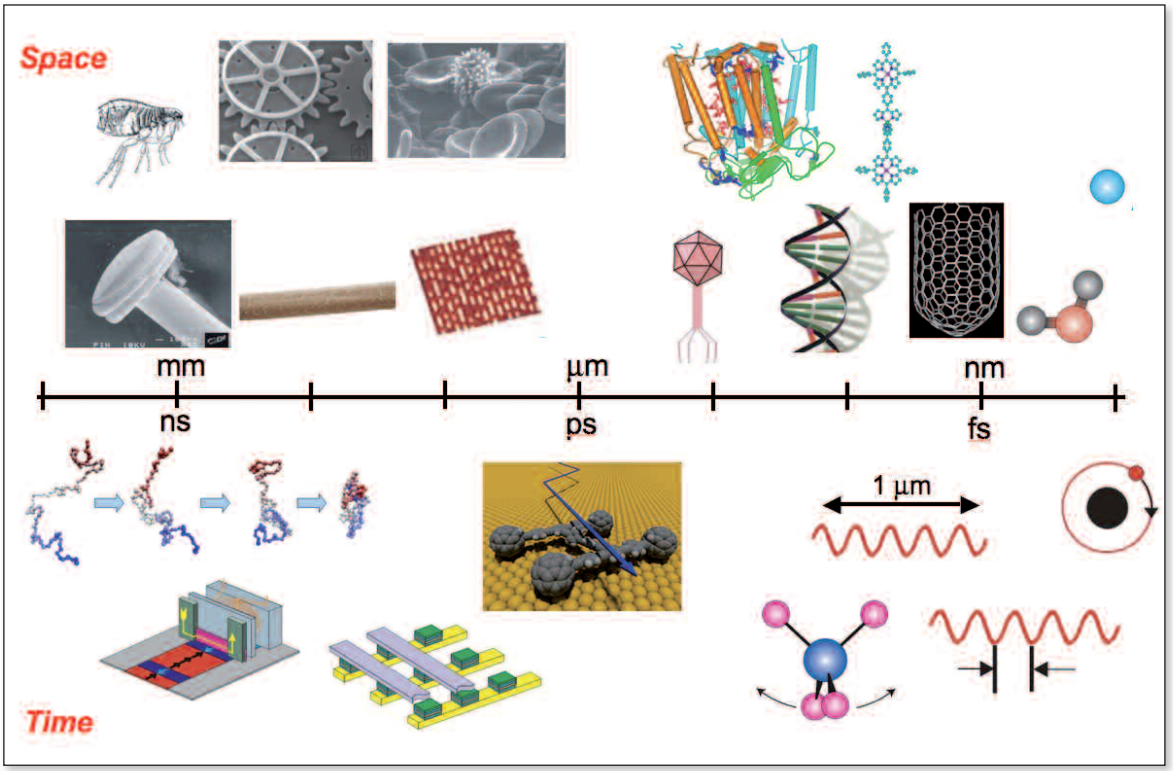
$$\lambda = \frac{\lambda_u}{2\lambda^2}\left(1+\frac{K^2}{2}\right) \qquad (1)$$

where the undulator "K-factor" is given by $K = eB_0\lambda_u/2\pi mc$, and m is the electron mass.

The long XFEL undulator length (60 - 100 m, vs. 2 - 4 m at a synchrotron) produces sufficiently strong undulator radiation to subtly alter the electron trajectory, causing a sub-division of the electron pulse into "micro-bunches", whose spacing is just the emission wavelength λ (Figure 3b). The resulting coherent radiation initiates a positive feedback effect – more radiation, better defined micro-bunches, more radiation, ... – which exponentially amplifies the light via "self-amplifying spontaneous emission" (SASE) (Figure 3c). It should be noted that SASE is a highly stochastic process, generally resulting in quite irregular X-ray profiles, in both time and frequency. Efforts are underway, also at PSI, to use laser-seeding techniques to produce Fourier-transform-limited XFEL pulses [4].

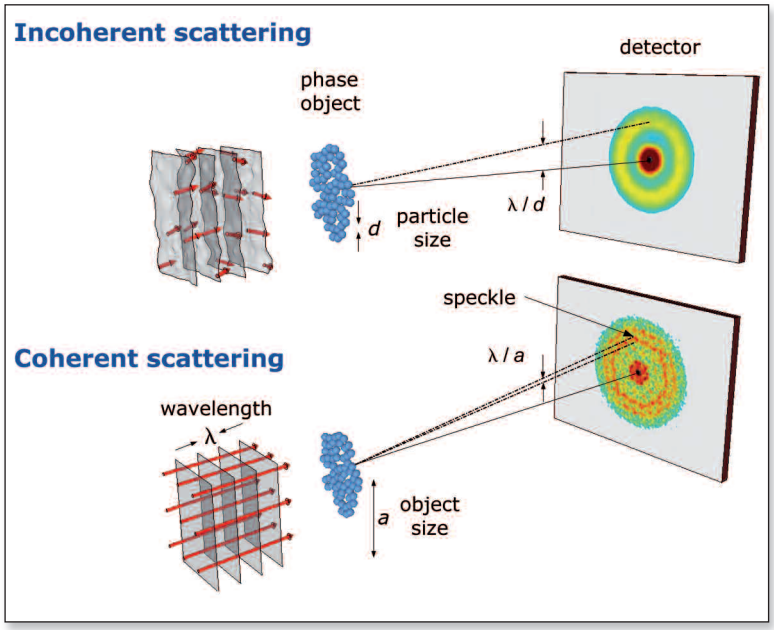
Other XFEL projects

Worldwide, there are presently three funded XFEL projects: the LCLS (US), SCSS (Japan) and European XFEL (Germany) [5-7] (see Table 1). Parliamentary approval of the SwissFEL project [8], at the Paul Scherrer Institute, Villigen-Würenlingen, Switzerland, is anticipated for mid-2011.



► **FIG. 1:** The XFEL will bring novel opportunities to study matter at µm to nm resolution and on the ps to fs time scale.

A major difference between the European XFEL and the SwissFEL is the use in the European project of superconducting accelerator technology. This dictates a pulse structure with many micro-pulses (2700 micro-pulses at 220 ns separation) per macro-pulse, resulting in a much higher average flux. Such a pulse structure is optimal for dilute samples, such as molecular gases, and coincidence experiments, but it complicates the recovery of a condensed matter sample, as well as the efficient readout of imaging array detectors.



▲ **FIG. 2:** Whereas the Debye-Scherrer rings of incoherent scattering only yield the particle size d, the rich speckle seen with coherent scattering allows an accurate reconstruction of the entire object [1].

SwissFEL

The 720 m long SwissFEL [8] (see Figure 4) at the PSI site in Villigen-Würenlingen, Switzerland, is the shortest of the current XFEL projects. Although it has the lowest electron energy, it will still be capable of producing hard (λ = 0.1 nm) X-rays. In the initial phase, it will provide simultaneous operation of two beam lines, the hard X-ray line *Aramis* (λ = 0.1 – 0.7 nm) and the soft X-ray line *Athos* (λ = 0.7 – 7 nm), with possible later extensions of both wavelength ranges. Each line will sequentially serve three experimental areas - *Aramis*: "coherent diffraction / scattering", "pump-probe" and "generic"; *Athos*: "pump-probe imaging / scattering", "pump-probe spectroscopy" and "inelastic scattering". *Aramis* will specialize on short pulse (10 fs) SASE operation, and *Athos* will offer variable polarization control and, with laser-seeding technology, Fourier-transform-limited pulses. Both beam lines will have as an option a "broad-band chirped" mode, with a bandwidth of 1%, for single-shot spectroscopic measurements involving an energy-dispersive detector. Among the synchronized sources of pump radiation pulses to be included in the SwissFEL facility will be an independent, accelerator-based terahertz source, capable of producing non-ionizing, half-cycle or wave-train radiation with high maximum peak field strengths (3 T, 10^9 V/m).

Science applications

The uniform 10 ms spacing of the X-ray pulses from the SwissFEL is particularly suitable for applications in condensed matter science. Whereas a higher repetition

Project	Length (m)	$E_{electron}$ (GeV)	λ_{min} (nm)	Peak brightness (ph/s/mm²/mrad²/0.1%bw)	Pulse structure	undulator beamlines
LCLS	1500	13.6	0.15	1×10^{33}	120 Hz	1
Japan XFEL	1000	8	0.1	0.5×10^{33}	60 Hz	1
Europ. XFEL	3400	17.5	0.1	5×10^{33}	2700 × 10 Hz	3
SwissFEL	720	5.8	0.1	0.5×10^{33}	100 Hz*	2

▲ **TABLE 1:** Comparison of XFEL projects. "0.1% bw" refers to a relative photon energy bandwidth of 10^{-3}. As mentioned in the text, the pulse structure of the European XFEL consists of trains of 2700 micro-pulses, repeated 10 times per second. * It is under consideration to operate the SwissFEL with two micro-pulses per macro-pulse, one for each undulator beam line.

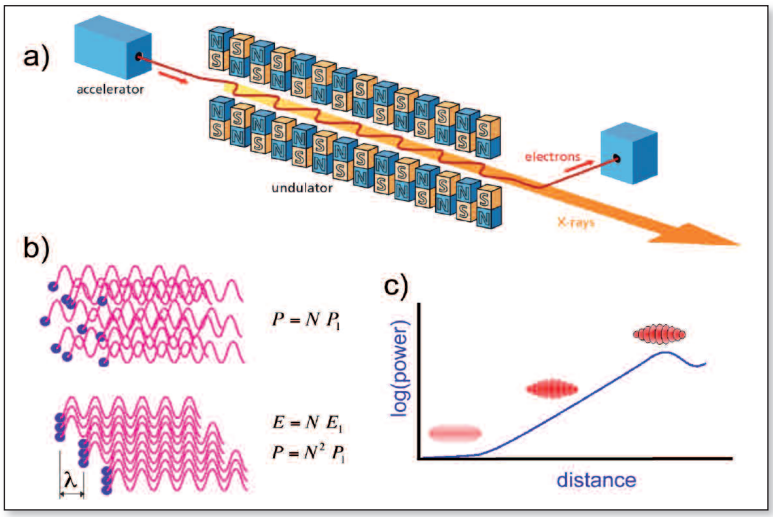
rate, such as will be produced by the European XFEL, is desirable for dilute samples such as gases and plasmas, the 100 Hz rate of the SwissFEL is well-suited to solid or liquid samples, which will require a certain recovery or repositioning time. Furthermore, the majority of experimental methods proposed for the SwissFEL incorporate imaging detectors with several million pixels, whose read-out rates are well-matched to the X-ray pulses. A survey of the prospective user community was carried out in the form of a series of Workshops, and the resulting SwissFEL Science Case [9] comprises five chapters: ultrafast magnetization processes, catalysis and solution chemistry, coherent diffraction from nanostructures, ultrafast biochemistry and dynamical effects in correlated electron materials. Here we present three selected proposals.

a) Switching dynamics in magnetic nanostructures

In the continuing effort to enhance the storage capacity and switching performance of magnetic media, the highly stable magnetic vortex states of thin disks (*e.g.*, Permalloy, 20 nm thick, 200 nm diameter) are being investigated both experimentally and theoretically [10] (see Figure 5). It is known that application of an in-plane field pulse can switch between the up (red) state and the down (green) state, but an experimental method with both the spatial and temporal resolution required to visualize this process is lacking. Using the magnetic contrast from X-ray Circular Magnetic Dichroism (XMCD), coherent, circularly-polarized synchrotron X-rays resonant with the L_2 and L_3 absorption edges of 3d-magnetic ions can produce static holographic images of ferromagnetic films [11]. A single pulse from the *Athos* beam line will suffice to produce such an image, and combined with rapid switching by a synchronized THz pump pulse, it will be possible to follow the switching process at the required resolution.

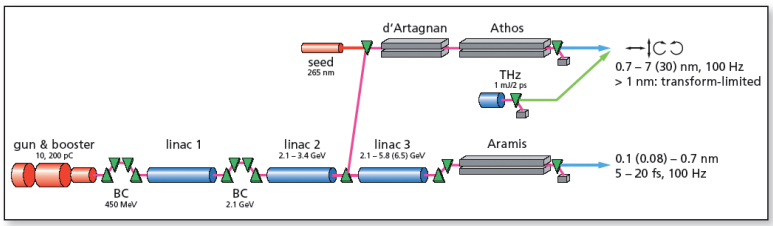
b) Intermediate states in heterogeneous catalysis

A further example of nanoscale dynamics with hitherto unobservable intermediate states is chemical reactions at catalytic surfaces (*e.g.*, see Figure 6). It has been proposed [12] that such a reaction can be initiated under

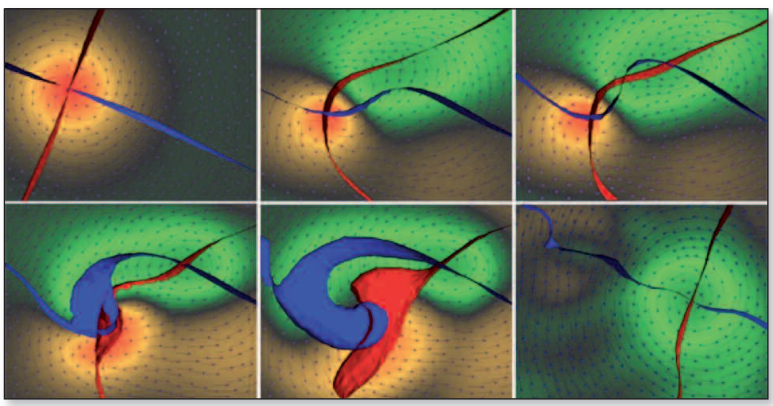


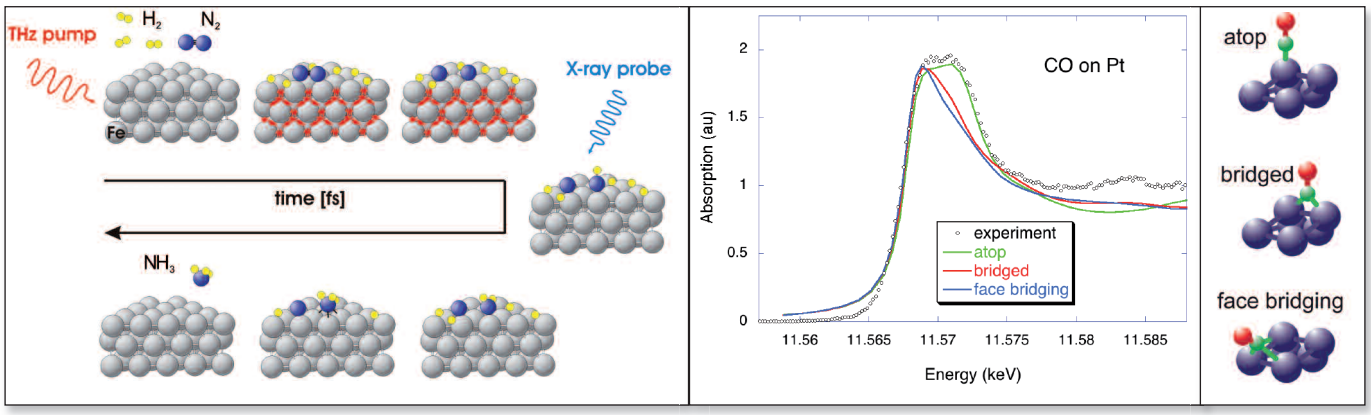
▲ **FIG. 3:** Schematic principle of XFEL operation [3]: A relativistic electron pulse from a linear accelerator (a) traverses a long magnetic undulator. The initially incoherent undulator radiation (b) becomes increasingly coherent as the "micro-bunching" process sub-divides the pulse into coherently-emitting slices. Saturation is reached at the end of the undulator (c), at which point the electrons are discarded. The relative power gain of this instrument over the incoherent synchrotron is equal to the electron number per pulse: N≈10^9 ! (E and P are the total field strength and total emitted power; E_1 and P_1 the field strength and emitted power from a single particle).

▼ **FIG. 4:** The schematic layout of the SwissFEL X-ray laser facility, including the electron injector (gun and booster), two electron bunch-compressors ("BC"), three linear accelerator ("linac") sections and hard (*Aramis*) and soft (*Athos*) X-ray beam lines. An external laser and the additional d'*Artagnan* undulator allow seeding of *Athos*, and a synchronized THz source is available for non-ionizing pumping.



▼ **FIG. 5:** Switching is simulated in a magnetic nanostructure [10]. Application of an in-plane field pulse (80 mT, 5 - 60 ps) to an up vortex (red) generates an intermediate vortex-antivortex-vortex state, which, after "annihilation", evolves to a down vortex (green).





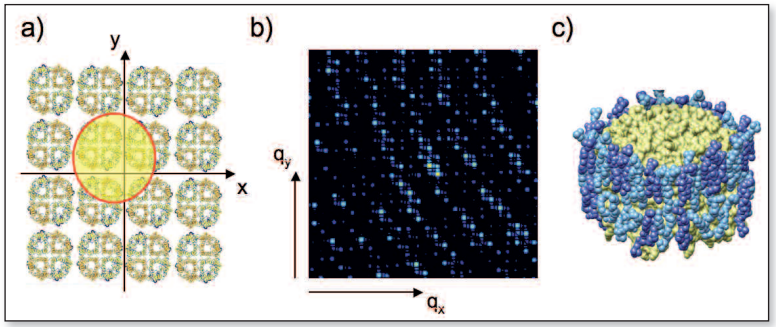
▲ FIG. 6: A catalytic process, in this case the Haber-Bosch production of ammonia, is initiated with a THz pulse and queried with a broadband X-ray pulse. The site-specific XANES spectrum, here for static CO on Pt [13], can be acquired with a single pulse from the XFEL.

nearly equilibrium conditions with non-ionizing THz radiation – either by resonantly exciting local vibrations or by physically displacing ionic species with a half-cycle pulse. The local environment of a selected chemical element can then be queried, with a single broadband XFEL pulse and an energy-dispersive detector, via its XANES (X-ray Absorption Near Edge Spectroscopy) signature.

c) 2D-crystallography of membrane proteins

With the completion of the Human Genome Project, the focus of microbiology has shifted from genomics to proteomics – what is the structure, and hence the function, of the proteins coded in our DNA? Of particular interest as targets for intelligent drug design are the membrane proteins, which control access to the cell and its nucleus. But since by nature these proteins prefer a 2D-environment, they are particularly difficult subjects for conventional 3D-protein crystallography. The low scattering power and high sensitivity to radiation damage further complicate X-ray and electron scattering approaches. With "diffract-and-destroy", it is believed that the XFEL will circumvent radiation damage, and advantage can be taken of its high spatial coherence to retrieve the missing crystallographic phase information. But even an ultra-bright XFEL pulse produces very little scattering from a 2D-protein crystal (see Figure 7) – of order 10 photons per Bragg reflection. One would like to combine the scattering from many separate XFEL shots, each performed on a fresh 2D-microcrystal, but account must be taken of the uncertain orientation of each sample. An elegant method [14] of combining coherent diffraction data from differently oriented 2D-crystals is "ptychography", from the Greek "to fold", and simulations show that repeated measurements at different tilt angles will yield molecular structures with atomic resolution.

▼ FIG. 7: 2D-membrane protein crystallography. A series of single-shot XFEL exposures (spot size: 100 nm) (a) are taken, each of which yields a coherent diffraction pattern (b). The data are combined, using the ptychographic method [14], to achieve a molecular structure at atomic resolution (c).



Outlook

Progress is continuing on the realization of several XFEL projects worldwide: the LCLS in California is producing prodigious amounts of spectacular data, and the interest in XFELs of the scientific community is rapidly growing. If parliamentary approval is granted for the SwissFEL during 2011, operation in Villigen-Würenlingen can begin in 2016. The Paul Scherrer Institute welcomes input from all interested parties. ■

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