Fresnel Coherent Diffractive Imaging from Periodic Samples

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In recent years the increased proliferation of high coherence X-ray sources such as third generation synchrotrons has helped in the advancement of new forms of microscopy. Among the techniques which are currently being developed, coherent diffractive imaging (CDI) is emerging as one of the most promising, offering the potential of very high resolution lensless imaging of non-crystallographic samples. Conventional experiments using CDI assume a perfectly coherent beam and utilize plane wave illumination of the sample. Recently it has been demonstrated that introducing curvature into the illumination can greatly assist in the recovery of a unique solution from the diffraction pattern.[1] A further interesting modification to the standard method for carrying out CDI is possible in the case of samples which are periodic.

Previous experiments using Fresnel CDI have relied on an iterative reconstruction of the exit surface wave (ESW) in which the entire illumination is reconstructed[2] and a support constraint applied to the amplitude. However, in experiments where the sample is known to have periodicity it is no longer necessary to reconstruct the entire object. Instead, an analysis of the sample can be carried out based upon only a single isolated unit cell. This scheme involves making an initial guess for the unit cell and replicating it to give a transfer function, which is multiplied by the incident illumination to give the ESW for the sample. The ESW is then propagated to the detector plane where consistency with the measured intensity is enforced. Finally, the ESW is propagated back to the sample plane where a single unit cell is isolated and the illumination is removed. The whole process is then repeated until convergence of the algorithm.

Studies utilizing simulated data have shown that this iterative phase retrieval scheme could, in principle, provide a significant enhancement in the reconstruction of periodic or even quasi-periodic objects compared to conventional CDI. Here we present experimental data from optical CDI measurements using curved beam illumination of periodic samples which test the effectiveness of this method of iterative image reconstruction. Among the issues we wish to address are the extent to which the method relies on the strict periodicity of the object, the quality of the reconstruction as a function of the number of unit cells which are 'well illuminated' in the sample plane and the effect of varying the curvature across a single unit cell compared to varying it across the entire object. We also outline a plan for extending these ideas to the X-ray regime and discuss possible applications of the technique, including a potential method for aiding in the recovery of information from biological samples.

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Femtosecond pump-probe coherent X-ray diffraction imaging of ultra-fast processes

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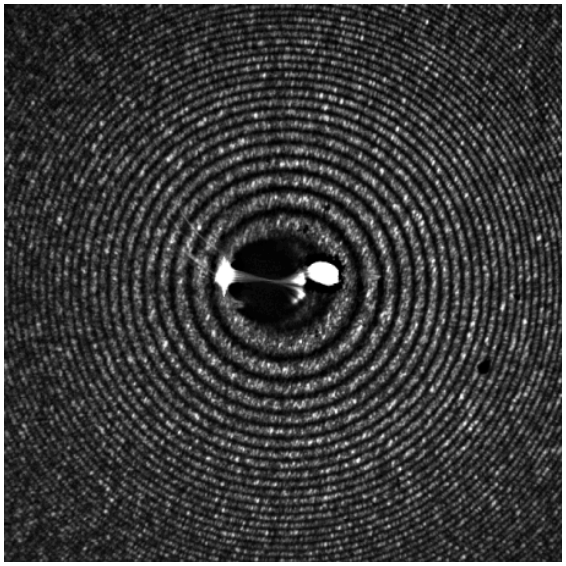
The intense, ultra-fast X-ray pulses produced by free electron lasers offer unique opportunities to study the dynamics of complex transient phenomena in materials. In particular, the high spatial coherence of these sources permit the use of coherent diffraction imaging to produce snapshots of the evolution of fast dynamic processes through time. The short wavelength of X-ray radiation allows probing the evolution of processes throughout the bulk of a sample, at spatial resolutions from the nanometer scale using current sources through to inter-atomic length scales using future hard X-ray FEL sources.

In the X-ray regime, time-resolved pump-probe experimental methods are challenging, requiring complex X-ray optical systems and diagnostics to synchronise the X-ray pulses that initiate a process and then probe it at a precisely defined time delay. We have developed and demonstrated a simple holographic technique in which the same X-ray pulse both pumps and probes the sample. The sample is placed near an X-ray mirror, which reflects the pulse back on to the sample after it initiates (or pumps) the reaction. The time delay is encoded by mirror separation to an accuracy of 1 fs, the sample structure is holographically recorded to sub-wavelength accuracy, and analysis of holograms recorded at different time delays yields information on the sample evolution through time. We have applied the technique to measure the explosion of hydrocarbon particles and other samples in intense FEL pulses. We observe sample explosion occurring well after initial sample heating, supporting the notion that X-ray flash imaging can be used to achieve high resolution beyond radiation damage limits for biological and other samples.



Shotgun Femtosecond Diffractive Imaging of Free Nanoscale Biomaterials

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Femtosecond diffractive imaging is a new x-ray microscopy technique for imaging non-crystalline objects beyond the damage threshold. Part of a new era of X-ray science delivered by X-ray free electron lasers (FEL), this technique has the potential to deliver three-dimensional atomic resolution structures of non-crystalline nanoscale biomaterials such as single biomolecules using images collected from reproducible copies exposed to the beam one by one. Experimentally, container-less delivery of single biomolecules to the X-ray pulses is imperative because any atom present in the X-ray path will contribute to the diffraction pattern. Here we use the soft-X-ray FEL in Hamburg (FLASH) to perform the first demonstration femtosecond diffractive imaging of free nanoscale biomaterials via a shotgun approach-a continuously refreshed stream of single nanoparticles synthesized *in situ* at atmospheric pressure by charge-reduced electrospray of a sucrose solution containing megadalton DNA origami complexes. In our method, the aerosol is transformed into a tightly focused particle stream in-vacuum using a set of aerodynamic lenses and single events of the interception of individual nanoparticles with an intense 10 femtosecond pulse, containing $\sim 10^{12}$ photons at 13.5 nm, results in a coherent diffraction pattern captured with a single photon sensitive X-ray camera. Reconstructed images of intercepted particles are obtained by phase retrieval through oversampling. Ions generated from the explosion of the nanoparticle are detected by a miniature mass spectrometer, providing insights to chemical composition. In its current configuration shotgun femtosecond diffractive imaging operates at 0.1 Hz and is directly transferable to future hard-X-ray FEL sources.

Iterative phase retrieval in one dimension for studying confinement induced ordering in microfluidic arrays

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Arrays of microfluidic channels lend themselves, e.g., to studying confinement induced ordering in model liquids or as orienting template for macromolecular small angle x-ray scattering (SAXS) studies in transmission geometry. We report on the microfluidic array phase profiling (MAPP) technique, focusing on iterative retrieval of one-dimensional complex-valued exit fields, and its application to studies of confinement induced ordering in model fluids.
The technique has several advantages. In contrast to traditional lensless imaging fully coherent illumination is not required. The ensemble averaged structure rather than individual realizations are investigated and thereby the dose on the specimen is reduced by orders of magnitude. By statistical averaging of solutions we show that the resolution is currently in the 10 nm range.

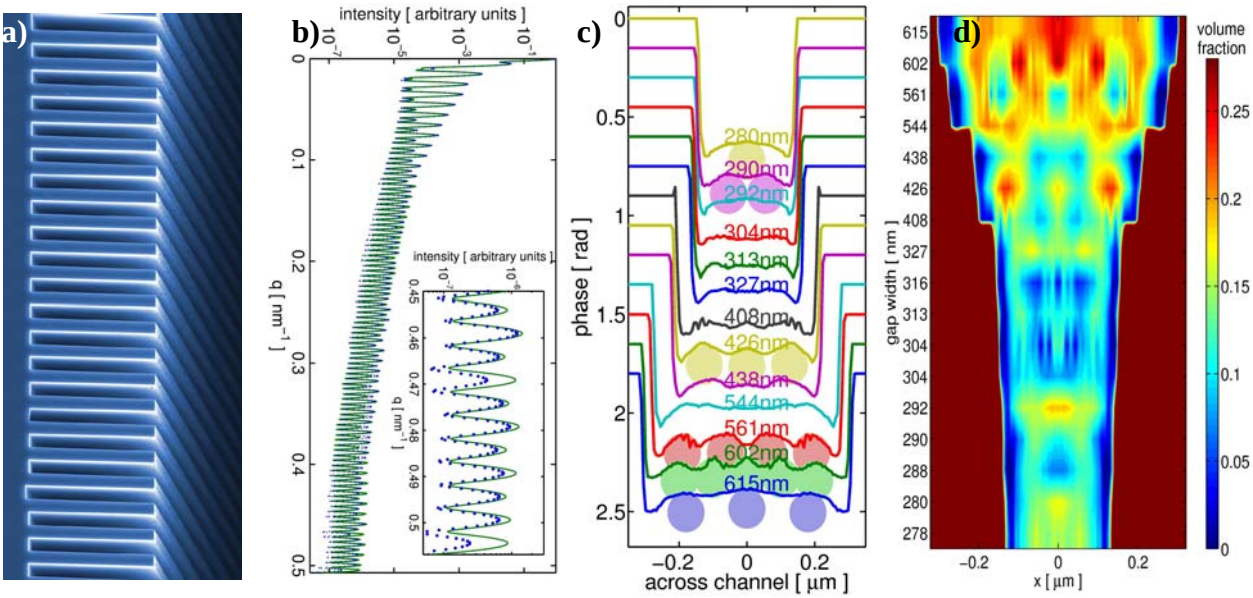

Figure 1: a) An array of microfluidic channels with a period of 1 µm. b) Measured and from the retrieved complex-valued field calculated grating diffraction intensities for a single array of 1.25 µm period and 615 nm channel width. c) Retrieved phase profiles of silica colloids (109 nm diameter) in microfluidic arrays. The profiles are offset vertically for clarity. The determined channel widths are shown for each profile, and layering, i.e., pronounced increased concentration is indicated by spheres. d) The silica concentration inside the microfluidic channels is shown as a contour plot for several channel widths. The excluded volume effect of 0% concentration at the confining walls as well as pronounced concentration increases due to the confinement are clearly visible. The dark red outer region is the area of the confining silicon walls.

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Ptychographic iterative diffraction microscopy with laser light

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The ptychographic iterative engine (PIE) technique [1] is a promising combination of the shifting illumination approach proposed by Hoppe [2] and standard iterative phase retrieval techniques [3,4]. It converges rapidly and eliminates the ambiguity between the 'true' solution and its complex conjugate. Meanwhile proof-of-principle studies have been published for visible laser light [5] and hard x-rays of about 1.5 Å wavelength [6]. Here we report on systematic studies of the error of the recovered amplitude and phase images as a function of the amount of overlap between individual probe positions. The simulations presented here are evaluated using a phase offset invariant error metric [7]. Experimental data are shown for a HeNe laser light PIE setup. The results of this study are of potential interest for PIE at all wavelengths.

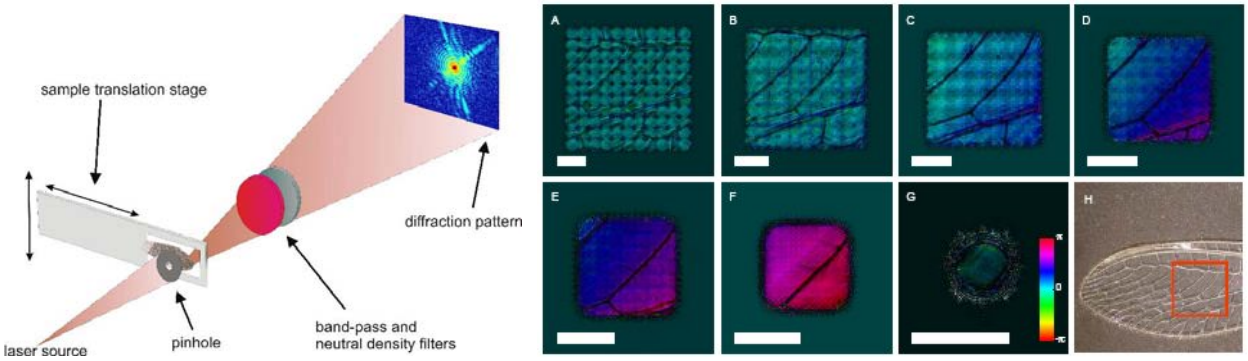

Figure 1, Left: The scanning diffraction microscopy setup. Right: Reconstructed phase profiles for different overlaps of the individual probe positions (A-G) and the sample under an optical microscope (H).

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Femtosecond diffraction imaging and coded aperture holography of a single cell

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The fourth generation light sources or free electron lasers possess many unique characteristics which promise to lead to interesting developments in the field of diffractive imaging of biological samples. Above all else, the greatly increased brilliance of these new sources compared to third generation sources appears most attractive. However, in itself, the increased intensity of the beam produced by FEL sources does not help eliminate the main limiting factor for imaging of biological material, namely the damage caused to the sample by the beam. The problem is indeed magnified for FEL sources as the damage occurs even faster. Only when combined with extreme short pulse duration (on the order of 100 femtoseconds or less) does the increased brilliance become an asset for biological imaging. The radiation damage to the sample can be circumvented by simply having all the x-rays in the beam interact with the object of interest in a period of time shorter than the time required for the damage to occur. We will present the first experimental demonstration of this flash imaging technique on a whole biological cell. While a single femtosecond x-ray pulse scattering from a single cell deposited on a membrane completely destroyed the sample, our results show that a two dimensional diffraction pattern could be measured and phase retrieval algorithms used to reconstruct an undamaged density of the cell to within the measured resolution. This technique could prove, at least in two dimensions, to be a powerful tool to study the structure of cells at near atomic resolution when shorter wavelength FELs become available.

Some of the inherent difficulties with iterative phase retrieval techniques such as the occasional lack of convergence and the difficulty to determine when the solution has been found can be overcome by using known reference objects, i.e. using holographic references. Using a single small pinhole as a reference object allows for high resolution structure determination but leads to a small signal level. A clever choice of a complex reference object can preserve the high resolution while boosting the holographic signal dramatically. We will present results from single FEL shot coded aperture holography of a single biological cell and discuss how such a technique at fourth generation sources can beat radiation damage while avoiding computationally intensive phase retrieval object reconstruction.

Focused X-ray vortices with high topological charge

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Vortices are characterized by a true zero in the intensity distribution that is stable under propagation; the phase at these points is therefore undefined. These structures have a "doughnut" like transversal intensity distribution and carry an orbital angular momentum, proportional to *l* [1]. Vortices are obtained for visible wavelengths using proper designed diffractive optical elements (DOEs) which convert the Gaussian beam generated by normal laser sources [2] and are used for optical particle manipulation. Same techniques used to design DOEs for visible light can be generalized for shaping X-ray beams [3].

Theoretical fundamentation and experimental observation of x-ray vortices at 9 keV were reported recently using phase plate fabricated in polymide as a spiral staircase structure approximating a spiral ramp [4]. Nevertheless, this phase plate is a non focusing element which generates a vortex beam observable only far away from it. Due to the limited thickness of the material, only vortices with low TC can be generated with phase plates. Moreover, phase plates can not be fabricated for soft x-ray since the material thickness is too small to be controlled.

We designed and fabricated amplitude DOEs to generate x-ray vortices with high TC and tested them at TWINMIC station at Elettra Syncrotron, in scanning [5] and full field microscope setup configurations. DOE function is calculated summing a helical wave with a desired TC to a spherical wave. In the scanning setup we used a the DOE as microscope objective and demonstrated the doughnut intensity pattern characteristic to the vortex but we could not characterize the phase distribution. In the full field X-ray experiment we used larger DOEs that generate a high focused TC vortex (*l=32*). The vortex DOE, fabricated at TASC-LILIT beamline, has the following characteristics: 220 nm gold on 100 nm silicon nitrade membrane, 300 nm resolution, 614 um diameter, 111 mm focal length at 720 eV. The DOE, placed in the condenser position converts the X-ray input beam into a helical beam focused in the sample plane where the intensity pattern is a doughnut with radius proportional to the square root of the topological charge. In order to characterize also the phase distribution characteristic to the vortex field we used the interference between the zero order beam and the vortex. The obtained interference pattern demonstrates the phase distribution characteristic to the optical vortex with the topological charge l=32. Possible applications of the interaction of these beams with the matter [6] are also discussed.

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Multiple pinholes to produce a clean hard-x-ray beam for coherent diffractive imaging

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Signal intensities for a coherent diffractive imaging (CDI) experiment decrease in proportion to the fourth power of the scattering angle. High-resolution information is encoded in this high-angle data. Accordingly, it is necessary to use a clean x-ray beam with minimal high-angle intensity.

We have investigated the production of a clean beam using multiple pinholes to select a highly coherent portion of the x-ray beam and to remove divergent beam components. These divergent components arise from the regular shape of the aperture - resulting in an Airy diffraction pattern, for instance - and from aperture imperfections, resulting in parasitic scattering.

Figure 1 shows the arrangement that we have employed in a recent experiment at 32-ID-B of the APS. A 10-μm pinhole was used to select the coherent beam fraction. However, use of such a pinhole produces higher-order background. This background would overwhelm the high-angle signal if allowed to reach the detector.

In order to place the pinholes at the optimum location we have calculated near-field intensities along the beam axis. The first propagation (Fig. 1a) shows that a 25-μm pinhole can be located so that its perimeter coincides with the second minimum of the Airy pattern. By choosing this location we are able to reduce the amount of scatter originating from this second pinhole. Figure 1(b) shows the intensity after propagation to the plane of a 45-μm pinhole that was used to guard in the first minimum of the Airy pattern. Further propagation to the specimen plane shows the intensity profile of the illuminating beam. We have found that such simulations are very useful for visualising the experimental illumination conditions.

Use of Advanced Photon Source is supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-06CH11357

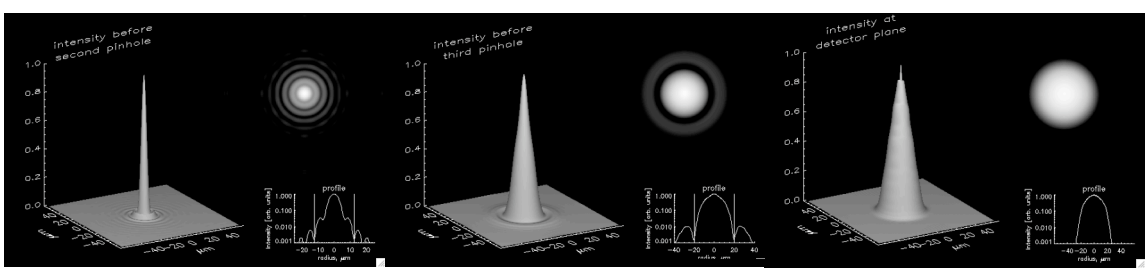

Fig. 1. Three pinholes are used to produce the illuminating beam. Pinhole 1 is selects the coherent beam fraction. Pinholes 2 and 3 remove the higher-order Airy fringes and unwanted pinhole scattering from the incident beam. The diameters and position of pinholes 2 and 3 are chosen so that their perimeters coincide with the second and first minima of the Airy pattern, respectively. The three figures show the intensity distribution at (a) the plane of the second pinhole, (b) the plane of the third pinhole, and; (c) the sample plane.

Electron Diffractive Imaging of a Single Nanoparticle

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The coherent electron diffractive imaging offers a promising path toward characterization of the individual nanostructures at the near atomic resolution [1]. In this microscopy, the complex exit surface wavefunction can be recovered from the far-field diffraction pattern using iterative algorithms.

In this present work, the diffraction intensities of the individual MgO nanoparticle with a size of about 24nm were recorded along [001] on the imaging plate (see Fig. 1a). The FEG-transmission electron microscope (JEOL 2010F) was operated at 200-keV accelerating voltage in the nanoarea electron diffraction regime. Due to short exposure time, the scattering from the carbon substrate [2] is localized within the central beam and can be removed directly from the measured pattern. Enlarged image of the one Bragg peak (see Fig. 1b) shows the intensity modulations along the four fringes oriented perpendicularly to the faces of cubic-shaped nanoparticle. The detailed experimental procedure will be discussed in the workshop.

For the phase recovery we utilize the charge-flipping algorithm which does not require a priory knowledge of the support [3]. Figure 2 shows the phase (2b) and modulus (2c) of the reconstructed wavefunction. This result is in agreement with the high-resolution image of the particle. In the workshop, we will also present preliminary study for the 3D particle shape recovery from the oversampled intensity in the region of the one reciprocal lattice point.

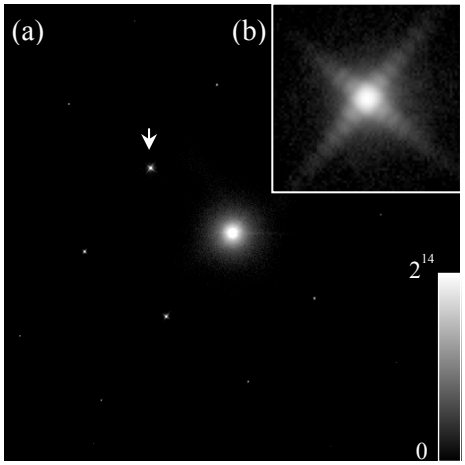

Figure 1: (a) Measured diffraction pattern from MgO nanoparticle near the zone axis of [001]. (b) Enlarged image of the Bragg peak labeled by the arrow.

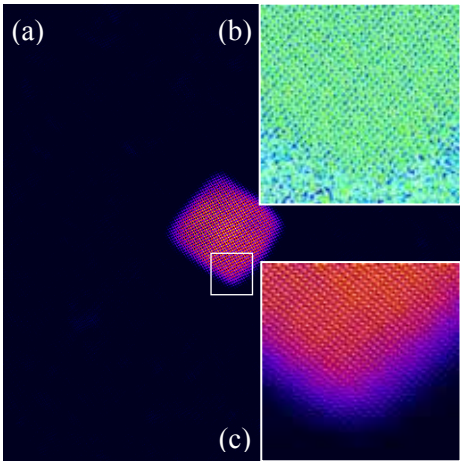

Figure 2: (a) Reconstructed modulus of the exit wavefunction. (b) Enlarged image of the phase and (c) modulus of the wavefunction from the selected region.

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A study of Laser-Generated Strain Fields with Coherent X-ray Diffraction Imaging and Microdiffraction.

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Abstract.

In its 324-bunch mode of operation, the Advanced Photon Source (APS) allows new femtosecond (fs) laser pump/x-ray probe experiments to be developed. In this mode of operation, if one uses the tightly focused low pulse energy (nJ), high-repetition-rate fs-laser Ti:Sapphire oscillator on beamline 7ID, every laser and X-ray pulse can be temporally delayed with respect to each other, as the frequency of the laser oscillator and the x-ray bunches are both 88 MHz. This can result in a high repetition rate pump-probe experiment which uses X rays from every bunch. This presentation describes an example of how coherent X-ray Imaging and Microdiffraction experiments may be used to study laser-generated strain fields in semiconductors. With an oscillator beam focused to 7 micron onto GaAs, we have observed coherent X-ray diffraction patterns with a high-resolution camera. We have also studied the strain fields with a focused x-ray beam generated by a long-working-distance Fresnel zone plate. Results from the two x-ray techniques will be compared. These experiments may help to develop techniques that will be used at the future Free Electron Laser sources where coherent and pump-probe experiments can be done simultaneously.

Iterative Extrapolation for Image Reconstruction by Phase Retrieval from Truncated Far Field Intensity Measurements

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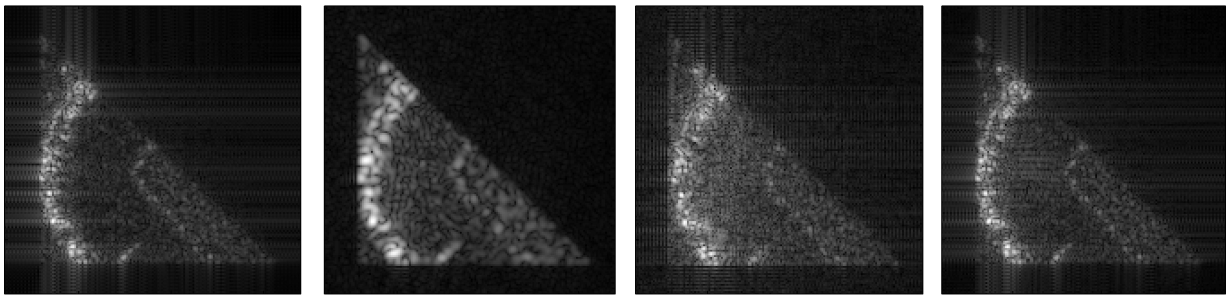
Phase retrieval algorithms have been shown to be able to reconstruct an image from a measurement of the far field radiation intensity pattern, if the object is illuminated with coherent source, by using *a priori* knowledge of the object support [1-3], *i.e.*, a set of points outside of which the object is known to be zero. When a diffuse, reflective object is coherently illuminated, it naturally back-scatters energy that falls out of the intensity detector array and any image formed from this truncated measurement will present side lobes that extend beyond the object support even if the object is spatially finite. In this situation image reconstruction algorithms are presented with an ill-posed problem since a spatially finite function is not consistent with a truncated, or finite, far field intensity pattern. As the hard-edged object constraint is continually reinforced, iterative algorithms tend to wraparound the phases at the computational window edge and are found to stagnate on a solution that has significant artifacts, as shown in Fig. 1(c).

Satisfactory reconstructions have been previously obtained by applying low-pass windows on the measured data. This reduces the image side lobes below the noise level and allows the algorithm to reach a faithful solution at the expense of resolution in the final image [Fig. 1(b)]. We propose a modified iterative transform algorithm that allows an analytic continuation of the far field measurement by applying weighted projections in Fourier space, thus allowing the far field data to be consistent with a finite support and achieving fully resolved reconstructions [Fig. 1(d)]. This approach is robust in the presence of noise and allows for a loose support constraint.



(a) (b) (c) (d)

Figure 1. (a) Speckled image. Image reconstructions (b) with and (c) without side lobe reduction filter. (d) Reconstruction with iterative extrapolation.

Iterative extrapolation methods for image super-resolution have been proposed previously [4]; in contrast our approach is not to achieve super-resolution but to use the slightly extrapolated data to improve image reconstruction algorithm convergence.

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Imaging Switching Behavior of Magnetic Nanostructures by resonant X-Ray Holography

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We report on magnetic imaging by x-ray spectro-holography. On the basis of a coherent scattering experiment, it is possible to image a specimen with high spatial resolution using a holographic approach.[1] A nanostructured mask makes it is possible to couple a reference beam to the object wave, thus allowing to solve the phase problem (and at the same time reducing the beamstop problem). We take advantage of multiple reference waves [2] and multiple objects [3] in a multiplexed experiment. An image is obtained directly by a Fourier transformation of the recorded scattering pattern, which constitutes a Fourier transform hologram. We exploit circular magnetic dichroism when scattering resonantly at the Co L edge in order to image the switching behavior of magnetic nanostructures with a spatial resolution of 40 nm.

We investigate the switching behavior and switching field distribution of magnetic multilayers on polystyrene spheres of 110 nm and 58 nm diameter. The magnetic caps on the spheres form exchange isolated magnetic islands with perpendicular anisotropy and are of interest for applications in magnetic data storage.[4] Magnetic contrast is generated by Co with an integrated thickness of 2.4 nm in the sample. The magnetic state of each nanosphere is imaged holographically as a function of applied field strength as well as of the direction of the applied field with respect to the anisotropy axis. On this basis, we draw conclusions on the microscopic switching mechanism and dipolar interactions between individual nanospheres, which are of importance in nanomagnetism and for data storage applications.

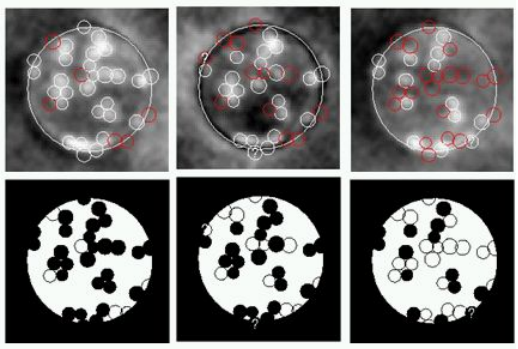

Figure 1. Example for the switching of 58 nm diameter spheres when the magnetic field is increased from 1.51 kOe to 2.16 kOe. From the magnetic image (top) we derive a model (bottom, inverted greyscale). The circular field of view is 500 nm in diameter.

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Investigations of complex objects through coherent x-ray diffraction in the Bragg geometry

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Our group, operating the coherent X-ray Diffraction (CXD) Beamline 34-ID-C at APS, has systematically studied the structure of nanocrystals by inversion of three dimensional coherent diffraction measured in the Bragg geometry. For various interesting reasons, including lattice distortions within the crystal, the measured diffraction is often not symmetric about the Bragg spot. This in turn leads to the recovery of complex density functions upon successful Fourier inversion.

We have frequently found that this phasing/inversion step fails to converge to acceptable error metrics, while at the same time producing acceptable looking images of smoothly connected electron density. Careful inspection of the resulting images found them to contain phase vortices, previously known to cause 'stagnation' in the convergence of iterative algorithms. In this case however the vortices occur in direct space outside of the support region defining the complex object. We have therefore introduced an adaptation of the Error Reduction(ER) algorithm. This form of ER removes the phase vortices in direct space while not destroying the recovered object, as traditional Error Reduction seemed to do. The details of these technique developments, illustrated with inversions of diffraction from a variety of nanocrystal and nanowire samples, will be presented.

Imaging Magnetic Nanostructures using Soft X-ray Spectro Holography

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I will present how to exploit the coherence and tunable polarization of soft X-ray synchrotron radiation for imaging magnetic nanostructures via Fourier Transform Holography [1-3]. This new lensless imaging technique is based on the direct Fourier inversion of a holographically formed soft x-ray interference pattern. Our implementation is particularly simple and is based on placing the sample behind a lithographically manufactured mask with a micron-sized sample aperture and a nano-sized reference hole. By exploiting the magnetic dichroism in resonance at the L_3 edges of the magnetic transition metals (wavelength ~ 1-2 nm (700-900 eV), images of magnetic nanostructures have been obtained with a spatial resolution of 50 nm. Different examples will be presented, ranging from stripe domain structures, over self assembled nanocap arrays, to lithographically defined patterned media. The technique is transferable to a wide variety of specimen, allows high magnetic fields and variable temperature, appears scalable to diffraction-limited resolution (about 2 nm), and is well suited for ultra-fast single-shot imaging with future X-ray free electron laser sources.

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3. *Multiple Reference Fourier Transform Holography with Soft X-rays*
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Sub-angstrom Resolution Electron Diffraction Imaging of Nanoclusters

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Diffraction imaging with a coherent electron beam is a promising approach to provide diffraction-limited atomic resolution. However, realizing this requires solving a number of issues, including experimental recording of high quality electron diffraction patterns, the robustness of reconstruction algorithms in the presence of experimental noises, the support and its effect on reconstruction, last but not least important, the dynamic scattering of electrons. In this presentation, we demonstrate the experimental approach for coherent diffraction of individual nanoparticles and implementation of phase retrieval algorithms to achieve atomic resolution by diffractive imaging. The results of a series of simulations will also be presented to show the effects of dynamical scattering, noise and interference with substrates on phase retrieval, and to establish effective inversion procedures. A new iterative algorithm, which uses a low-resolution image in the object domain, is proposed. With the help of the new algorithm, experimental diffraction patterns from individual nanoclusters can routinely be inverted.

Signal to noise considerations in diffraction and conventional microscopy

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In x-ray diffraction microscopy, the x-ray beam diffracted by the specimen is collected directly on an area detector such as a backside-thinned CCD with high sensitivity. However, the diffracted magnitudes must be phased to obtain a real-space image. In conventional microscopy, the beam diffracted by the specimen is instead collected and phased by a lens, to yield a real-space image at the detector plane. Unfortunately, present-day x-ray lenses have finite numerical aperture cutoffs and poor efficiencies, limiting the efficiency with which the diffracted signal is exploited.

In this work we address the following question through simulations: which approach yields more information for a given exposure on an example biological specimen? This is an important consideration for radiation-sensitive specimens such as frozen hydrated cells, since in electron microscopy it is the case that radiation damage is *the* resolution-limiting factor. Factors entering into the comparison include:

- How does one expect the signal to noise ratio α of an image to scale with photon number? A simple model suggests that α scales as $N^{1/2}$.
- How do you measure the signal to noise ratio of an actual, noisy image, or pair of images of identical objects with different noise? We discuss the use of both real space and Fourier space correlation methods.

We then carry out simulations of x-ray imaging of a simulated cell consisting of a lipid membrane, low concentration protein solute, and protein bars of various diameters and orientations inside the "cell." For diffraction microscopy, we simulate the illumination of the cell by a coherent x-ray beam, calculate the resulting diffraction intensity distribution, add simulated noise according to Poisson statistics, and reconstruct the image using finite support constraint iterative phase retrieval. For conventional microscopy, we calculate the absorption profile of the cell, convolve it with the point spread function of a representative soft x-ray zone plate, reduce the image intensity according to the efficiency of the zone plate, and add simulated photon noise to the resulting intensity image. We will discuss results of these comparative simulations and the implications for x-ray microscopy of frozen hydrated organic specimens.

Development of three-dimensional architecture of bone at the nanometer scale using X-ray diffraction microscopy

Huaidong Jiang[1], Damien Ramunno-Johnson[1], Changyong Song[1], Bagrat Amirbekian[1], Yoshiki Kohmura[2], Yoshinori Nishino[2], Yukio Takahashi[2], Tetsuya Ishikawa[2], Jianwei Miao[1]

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We have studied the 3D spatial relationships of mineral crystals to the collagen matrix of Alewife herring bone by using X-ray diffraction microscopy for the first time. We performed nanoscale and stereo imaging of the mineral crystals inside collage fibrils at different stages of mineralization. The origin and distribution of mineral crystals inside the collagen matrix has been identified. Based on the mechanism of biomineralization and our experimental results, we have developed a dynamic 3D structural model of bone to account for the nucleation and growth of mineral crystals in the collagen matrix with maturation of bone. The results obtained from this study will not only contribute to understand the complex 3D microstructures of bone, but also provide a basic principle with which to design and fabricate highly ordered organic-inorganic hybrid biomaterials.

Coherent Diffractive Imaging with Multiple Phase Fronts

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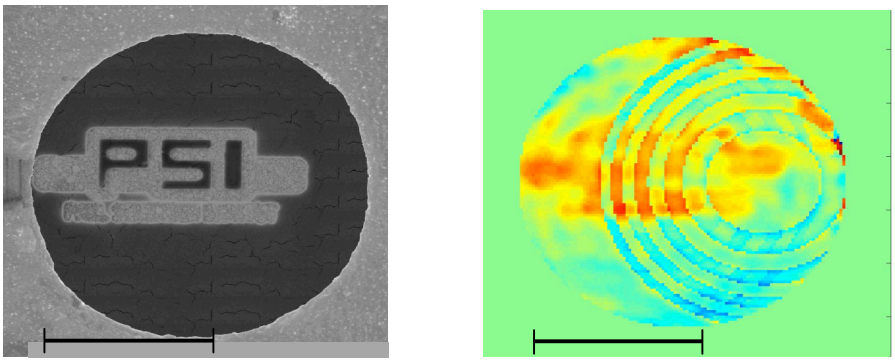
X-ray diffraction imaging is a promising technique for biological samples on the nanometer scale. The key task is to determine the absent phase information in the recorded diffraction pattern. Phase retrieval methods employ iterative algorithms [1-4] to computationally solve for the phase of the diffracted wave, however some techniques require a substantial number of iterations and may not lead to a unique solution. We are developing a technique that utilizes multiple exposures with different impinging wavefronts on the sample to compensate for the lack of the phase information. The complementary, though independent, exposures direct the convergence and solve ambiguities in the reconstruction. This technique is optimal for few micrometer size samples, with a potential niche for biological samples like small cells and viruses. Both the data acquisition time (a few exposures) and reconstruction time (tens of iterations) are short enough such that many projections of a sample may be efficiently recorded, reconstructed and rendered into a three dimensional image. The technique, simulations and the first results from a demonstration experiment with 6 keV X-rays will be discussed in this presentation.



Left: An SEM image of a 700 nm thick "PSI logo" and the illumination area defining pinhole. Right: The reconstructed phase profile of this test sample.

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Focusing hard X-rays with profile-coated mirrors

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Raymond Conley, Chian Liu, Wenjun Liu

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We report our progress in simulating the focusing of X-rays by elliptical mirrors, using wave-optical calculations. The simulations take into account the phase change of coherent hard X-rays on propagation and reflection from a mirror surface, which is described using microstitched interferometry data.

Profile-coated mirrors can be routinely fabricated with RMS residual height compared to the best-fit ellipse of the order of Angstroms. Simulated intensity profiles, and contours (isophotes) around the focal plane of such mirrors will be presented for coherent illumination by a 15 keV point source, which predict diffraction-limited focusing. The correspondence between the simulated results and the experimental focusing performance of the mirrors will be discussed.

This work supported under contract number DE-AC-02-06CH11357 between UChicago Argonne, LLC and the Department of Energy.

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Coherent X-ray Diffraction Microscopy with High Precision Aperture

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Small apertures are often necessary in coherent x-ray diffraction microscopy for illuminating small areas around the small samples. However, it is extremely difficult to fabricate hard x-ray pinholes with the diameter of smaller than 10 μm with sufficient precision. High precision cross slits, composed of four tantalum polished blades, were fabricated by TDC Corp., Sendai, Japan. The cross slits had the width of 10~20 μm (width down to 1 μm is possible). We believe that precise aperture will enable us to minimize the noise in the coherent X-ray diffraction microscopy.

Coherent X-ray diffraction microscopy was performed using 9 keV x-rays by taking the Fraunhofer diffraction pattern of the cross slit by the downstream image detector. Experiment was done at about 80 m from the light source of BL29XU/SPring-8 with well reduced upstream slit sizes. Fraunhofer patterns from the aperture included negligible scattering from the edges of the aperture. By placing various samples very close to the aperture, the complex transmissivity of the samples was also measured. The distance from the aperture to the sample and to the imaging detector was chosen to be approximately 3 mm and 4.3 m. Phosphor coupled CCD camera with the effective pixel size of 4.5 μm (Hamamatsu Photonics Corp., Japan) was used as the image detector. We have succeeded in retrieving the complex transmissivity of thick objects (e.g. distributed 8 μmϕ latex particles corresponding to 1/5 λ phase shift). Difference map algorithm was applied to a single diffraction pattern. For objects with complex transmissivity, we have found that the difference map algorithm is more powerful than the HIO algorithm.

Quantitative Comparison of Phase Retrieval Methods in In-Line Phase Tomography

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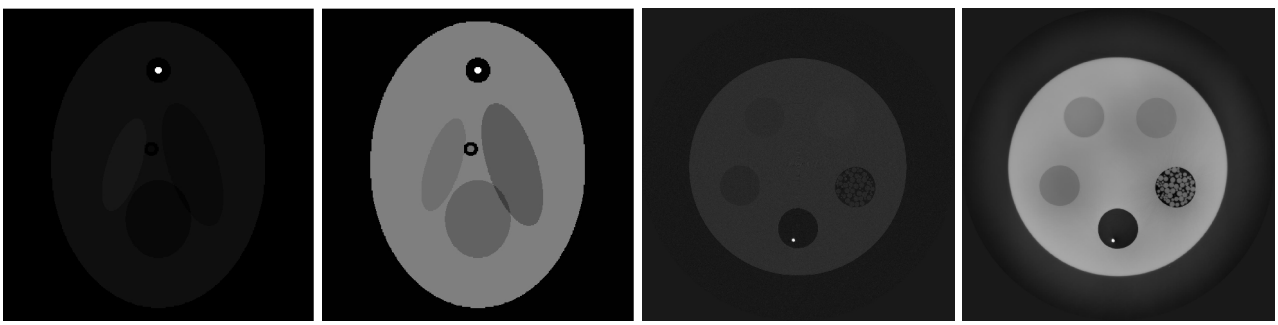
X-ray phase contrast tomography potentially offers several advantages over tomography based on absorption contrast, such as greatly improved sensitivity and lower dose. If the coherence of the X-ray beam is sufficient, a simple mode of phase contrast imaging, based on free space propagation, becomes possible [1]. Phase contrast is achieved by moving the detector downstream of the object and a tomographic scan is then recorded at several sample-to-detector distances. The reconstruction is usually divided into two steps. The phase shift induced by the object is retrieved for each projection angle from the radiographs taken at different distances. This is then used as input to a standard tomographic reconstruction algorithm such as filtered backprojection. This yields a reconstruction of the 3D refractive index distribution in the sample.



(a) (b) (c) (d)

Fig. 1 Phantoms used for evaluation. (a) Simulated absorption and (b) refractive index. Tomographic reconstruction of constructed phantom (c) absorption and (d) refractive index using the Mixed approach.

In this work the quantitativeness of the phase retrieval step for mixed absorption/phase objects in propagation based phase contrast imaging is addressed. Two tests were developed (Fig. 1). First, propagation based imaging was simulated using a mathematically defined phantom. Second, a phantom was constructed from materials of known specification and subsequently imaged using the propagation based phase contrast setup. The simulated and experimental data are then used to evaluate the phase retrieval by comparing the reconstructed to the theoretical values. We consider three phase retrieval methods, the Transport of Intensity Equation [2], Contrast Transfer Function [3] and a Mixed approach [4] between the two. We also present recent improvements of the Mixed approach.

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Three-Dimensional Imaging of Single Large Macromolecules Using Equally Sloped Tomography

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We report the development of equally sloped tomography for the reconstruction of the 3D structure of single large macromolecules. In a combination of pseudo-polar fast Fourier transform and the oversampling method with an iterative algorithm, equally sloped tomography makes superior 3D reconstruction to conventional tomography which has an intrinsic drawback due to the use of equally angled 2D projections. By employing equally sloped tomography and cryo electron microscopy, we have obtained the 3D structure of single hemocyanin protein molecules and HIV viruses at ~ 5 nanometer resolution. Preliminary analysis based on cross-correlation has indicated that the 3D images using equally sloped tomography are superior to those of the conventional method. We believe this general approach will find broad applications in high-resolution 3D imaging of large macromolecules.

Effects of X-ray Optics on the Visibility of Coherent X-ray Diffraction

Mengning Liang[1]**,** Ross Harder,[2] Ian Robinson[2] ***Mengning Liang***

[1]University of Illinois Urbana-Champaign, [2]University College London

Coherent X-ray diffraction (CXD) provides a non-invasive method of imaging the internal structure of nanoscale crystalline materials. A coherent x-ray diffraction image is obtained when the coherence of an x-ray beam is greater than the spatial dimensions of the object being imaged. In this scenario, the intensity distribution surrounding each Bragg peak of a crystal is the magnitude squared of the three-dimensional Fourier Transform of the shape. The phase information needed to invert the diffraction pattern to obtain an image of the original object is lost but may be recovered using iterative phasing algorithms. The success and accuracy of the inversion depends strongly on the quality of the initial diffraction image. The coherence properties of the incoming x-ray beam play a crucial role in determining whether a diffraction pattern can be inverted.

In coherent x-ray diffraction, the standard constraint is to have the coherence lengths of the x-ray beam be larger than the spatial dimensions of the crystal. These coherence lengths depend on machine parameters but also on the optics in the beam path. The propagation of coherence along this multitude of optics is difficult to quantify and it is hard to ensure that the x-ray beam incident on the sample has the necessary coherence condition to obtain an invertible pattern.

An alternative way of judging the coherence qualities of the beam is to look at the diffraction image itself. The traditional figure of merit for the coherence of a light wave is the complex degree of coherence which measures the degree to which two light waves interfere. This complex degree of coherence is coupled to the visibility of a diffraction image and is a quantity that can be experimentally obtained. By changing the beamline optics, we can alter the coherence properties of the x-ray beam. These changes can be clearly seen in the changing complex degree of coherence but would be difficult to quantify by using the coherence lengths of the source. We present a study of the effect of optical elements in altering the coherence of an x-ray beam by using the coherent diffraction images from gold nanoparticles.

Coherent scattering by isolated dislocations in a Si single crystal

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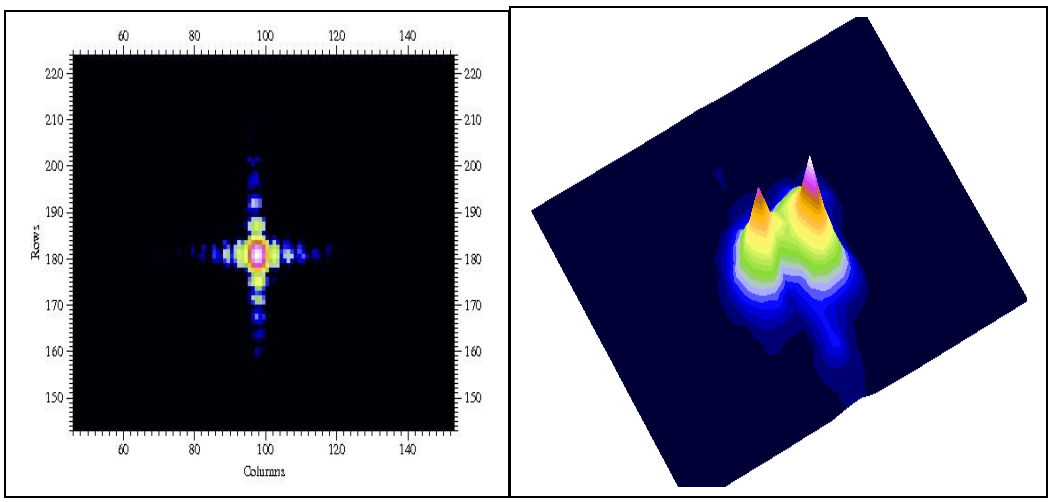
Coherent X-ray scattering opens the possibility of studying crystal defects of a "mesoscopic" extension, like dislocations, subgrain boundaries... As the beam extension is of the order of a few microns, first studies have to be carried out in systems with individual isolated defects. Silicon single crystals have a highly perfect lattice, with a very low amount of dislocations.

A float-zone single crystal has been studied, where oxygen inclusions have precipitated in Frank loops with a diameter of the order of 0.1 millimeter. A topographic study of this sample showed that Frank loops were of suitable size and concentration. X-ray coherent scattering experiment were performed at the BM2 beamline of the ESRF. A high degree of transverse coherence was obtained in a beam of 2 microns diameter by using carefully polished precision slits. As silicon is a light element, the penetration depth of x-rays is of the order of 10 microns, and the Si333 bragg diffraction of the monochromator was used for improving the longitudinal coherence length to the micron scale, in order to observe interferences in the diffracted beam.

In the vicinity of the distorsion induced by a dislocation, the change in the shape of the Bragg peak has been observed, and a typical double peak was observed close to the dislocation core. The general shape of the observed peak profile is nevertheless difficult to interpret, and needs some further modelisation.



a) b)

Figure 1: *a) Diffraction from 2x2µm slits; b) double peak observed at the dislocation core.*

Reconstruction of thin-film magnetic domains using an illumination mask

Duane Loh[1*], Veit Elser[2], Larry Sorensen[3]

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X-ray diffraction reconstruction usually requires the specimen to have a bounded support. Together with the measured diffraction amplitudes, the absence of scatterers outside the support provides sufficient information for the reconstruction. We present a method for speckle reconstruction which implements a pinhole's Fresnel diffraction pattern as an illumination mask, avoiding the use of the bounded support. This method is used to image thin-film nanoscale magnetic domain patterns[1], an area of research which commands significant theoretical and applied interests. Successful reconstructions from noisy diffraction data simulated from magnetic circular dichroism scattering [2] are presented.

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Practical Image Reconstruction

Filipe Maia[1] , Janos Hajdu[1,2]

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Phase retrieval is an extremely difficult optimization problem due the fact that its search space is at the same time non-smooth, non-convex and very large. Throughout the years much attention was paid to the problem of reconstructing an image from its oversampled diffraction pattern alone. This has led to well known algorithms like the Hybrid Input Output. But most of these efforts did not try to incorporate in the algorithm knowledge about the noise that is associated with the acquisition of diffraction images. Here we present current phase retrieval algorithms so that they minimize the effect of noise associated with short exposures CCD data acquisition. This leads to more accurate and faster reconstructions for real data.

Experimental Phasing with FLASH diffraction

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Single particle diffraction has the potential to revolutionize macromolecular structure determination, due to its ease of use, as compared to classical crystallography, and resolution, as compared to electron microscopy, as well as introducing the possibility of studying extremely fast phenomena. But to be able to realize all this potential there needs to be a reliable method for phasing the diffraction patterns obtained. Ab initio phasing that takes advantage of the pattern oversampling has made significant progress[1, 2] but it requires significant human intervention and often it fails to phase certain images. Here we present experimental phasing methods that could be used in conjugation with ab initio phasing to improve its speed and convergence capabilities.

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Towards Three-Dimensional Imaging of Yeast Spores Using Coherent X-ray Diffraction Microscopy

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The three-dimensional imaging of whole cells at around 10 nm resolution is of great biological interest. Using coherent x-ray diffraction microscopy, we demonstrate high-resolution imaging of an unlabelled yeast spore at different angles of tilt. The experiments were carried out on an undulator beamline at SPring-8, Japan. The collected diffraction data were analyzed using an explicit account for detector pixel size. Reconstructions of projections of the yeast spore, at differing angles, have been performed based on a guided hybrid input-output algorithm (GHIO). These reconstructions are shown, along with an account of the reconstruction method. This work represents a practical step toward routine three-dimensional imaging of unlabelled biological cells using X-ray diffraction microscopy.

This work was supported by the National Science Foundation (NSF), the Department of Energy (DOE), and the California NanoSystems Institute (CNSI). Use of the RIKEN beamline, BL29XUL at SPring-8 was supported by RIKEN.

Phase contrast x-ray microscopy with singular optics

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Optical schemes that enable imaging of the phase shift produced by an object have become popular in the x-ray region, where phase can be the dominant contrast mechanism. Singularities in the phase have been found to be a common feature of phase retrieval and imaging with coherent fields [1]. Optics that introduce phase singularities can also play a unique role in microscopy [2]. Production of x-ray vortices containing phase singularities was recently demonstrated with refractive optics [3]. Here, we report on use of diffractive optics to produce intense x-ray vortices and their application to hard x-ray phase contrast imaging in a full-field transmission microscope. This method offers some advantages for phase contrast x-ray microscopy and does not require a Zernike phase ring. This work is supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract DE-AC02-06CH11357.

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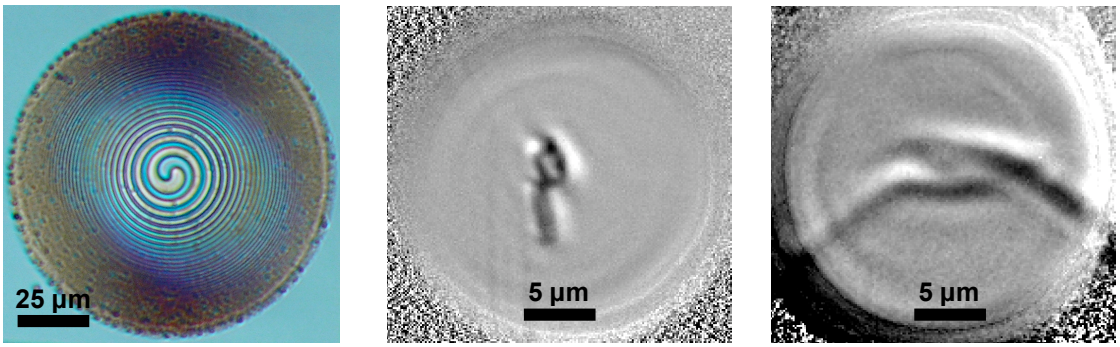

(a) Focusing diffractive x-ray vortex optic. Full-field phase contrast micrographs of (b) a silicaceous diatom and (c) two strands of spider silk recorded with 9 keV x-rays.

Applicability of an Iterative Inversion Algorithm to the Diffraction Patterns from Inhomogeneously Strained Crystals

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A lot of progress has been made lately on the development of iterative inversion algorithms for retrieving the shape of crystals directly from their diffraction pattern [1, 2]. In the case of strained crystals the convergence of the algorithms is more difficult. It is, however, a very important issue to evaluate local strains in nano-materials since these strains can have a great influence on various physical properties. We present here new numerical procedures, which allow for direct inversion of displacement fields in highly inhomogeneously strained objects.
In the case of a strained object, the direct space density can be expressed as a complex-valued number, whose phase provides information, within some small approximations, about the projection $u_{||}$ of the displacement field along the direction of momentum transfer vector $\vec{q}$. The iterative algorithm mentioned above for inverting reciprocal space maps from such types of objects has been investigated. It is observed that in the case of complex valued numbers expressing the highly inhomogeneously strained crystal density the convergence of the algorithms is problematic since ambiguity in solutions appears yielding to enormous numbers of local minima with very small error metric [3]. It means that different combinations of amplitudes and phases in direct space yield very similar Fourier transform amplitudes images. Therefore without additional *a priori* knowledge the "phase problem" proves to be difficult to solve by this approach in the case of inhomogeneously strained crystals.
Here a new method is proposed to break this stagnation. It is based on the use of additional but reasonable constraints in direct space, such as the maximal value of the displacement field derivatives [3]. This information can be approximately estimated from the diffracted intensity map. The method is not sensitive to some variations of these parameters.
The proposed algorithm was successfully tested on various numerical samples. We will also show inversion from highly resolved diffraction measurements from Si lines on SiO_2/Si substrate and arrays of oxide-filled trenches in silicon (BM32 beamline, ESRF). The retrieved displacement fields with a resolution of about 8 nm are in excellent agreement with those calculated by finite element modelling based on continuum elasticity [4]. These results offer important perspective for local strain determination at the nanoscale. The work is currently concentrated on the investigation of the general applicability of the method to different types of structures in different geometries.

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Localized phase retrieval algorithm for oversampled intensity measurements

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We present an algorithm for phase retrieval from oversampled intensity measurements in 2D. In particular we consider the case of oversampling by factor of 2 in each dimension. We use the relations between Fourier modes that result from the Nyquist-Shannon sampling theorem to establish a system of equations that can be solved approximately by taking only a few terms in an expansion of the support window. Within this approximation we introduce a method for phase retrieval that consists of two stages: In the first one, a few of the phases in a localized region in reciprocal space are obtained. These phases are then used to obtain the rest of the unknown phases by means of a simple propagation method on the second stage. We present an example in which we use a downhill minimization method to solve the system of non-linear equations that arises on the first stage.

X-ray Diffraction Microscopy at SPring-8

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We report various recent activities on x-ray diffraction microscopy at SPring-8. In materials science applications, we are especially interested in mesoscopic size structure in metallic materials, which often controls important functions in practical aspects. As an example, we present a 3D visualization of a sub-micron size precipitate in an Aluminum alloy particle. Such structure is difficult to observe non-destructively in other methods. In biological applications, we are pursuing visualization of organelles, and we have started measurements of chromosomes. In parallel with application works, hardware improvements have been made. Recently, a new function was added to our microscope instrument to largely change the sample-to-detector distance for measurement of various size samples at various x-ray energies.

At SPring-8, the construction of VUV free electron laser (FEL) has completed, and they succeeded in laser amplification at a wavelength of 49 nm. In addition, the construction of Japanese x-ray FEL has started last year. We have begun studies toward the use of x-ray FEL for diffraction microscopy. In order to make maximum use of coherent x-rays of FEL, x-ray focusing optics is essential. We consider that a total reflection mirror is a good candidate for it because of its radiation hardness. Preliminary studies of diffraction microscopy with mirror focused x-rays have started. We also show simulation results for a biological molecule in a very simple case of neglecting radiation damages, and some discussion regarding imaging detectors will be given.

Determining the Relative Orientations of Single-shot Diffraction Patterns from Individual Macromolecules and Nanoparticles

Ravi Shankar Vaidyanathan, Valentin Shneerson, Dilano Saldin, and ***Abbas Ourmazd***

Department of Physics, University of Wisconsin-Milwaukee

In about an hour, 10^5 "shots" from the LCLS XFEL should scatter ~10^3 photons into each pixel of the high-q regions of a diffraction pattern from a 500 kDa biological molecule - if only we could integrate the scattered photons from successive shots. In the absence of orientational alignment, each single-shot diffraction pattern will emanate from an unknown random orientation of the molecule. A key problem therefore concerns determining the relative orientations of the individual diffraction patterns in the presence of noise; a "high-q pixel" of a single-shot diffraction pattern is expected to contain an average of ~10^{-2} photons.

This presentation addresses the critical issue of finding the relative orientations of individual diffraction patterns at low signal-to-noise ratios in the absence of molecular orientational order.

We have developed and tested an algorithm that can reliably reconstruct the 3D electron density of a molecule from a set of ideal (noise-free) diffraction patterns, each emanating from an unknown random orientation of the molecule, and investigated its performance in the presence of shot noise, modeled by a Poisson distribution. A modified implementation of the algorithm to orient diffraction patterns by the so-called common-line method used in 3D electron microscopy functions satisfactorily for average signal levels as low as ~10 photons/pixel, excluding a (high-intensity) central region subtending a semi-angle of 6˚ around the undiffracted beam. This performance is comparable with that achieved in 3D-EM, where starting images (which contain phase information) have typical S/N ratios of ~$\sqrt{10}$ [1]. However, this is still ~ 3 orders of magnitude away from that required to orient single-shot XFEL diffraction patterns from single biological molecules.

The noise robustness can, in principle, be further improved by schemes, such as Principal Components or Correspondence Analysis, which consider the totality of the diffracted data without any knowledge of underlying correlations. Experience in other fields has shown that information theoretic limits can be approached by algorithms which explicitly consider inherent correlations within the entire data set, using physically based models.

We will describe the capabilities and limitations of each approach for practical orientation of individual diffraction patterns and structure recovery in the presence of severe signal-to-noise limitations. This is an essential pre-requisite to any experimental determination of single-molecule structures in the absence of molecular orientational order.

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Fluctuation x-ray microscopy: a novel approach for the structural study of disordered materials

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Materials research has increasingly focused on developing a better understanding of the disordered state of matter. Many x-ray techniques exist to probe long- and short-range order in matter, in real space by imaging and in reciprocal space by diffraction and scattering. However, the characterization of medium-range order (MRO) is a long-standing problem that current x-ray techniques cannot effectively probe.

We have developed fluctuation x-ray microscopy (FXM) based on fluctuation electron microscopy [1]. This novel approach offers quantitative insight into medium-range correlations in materials at nanometer and larger length scales. FXM examines spatially resolved fluctuations in the intensity of x-ray speckle patterns. Measuring the speckle variance as a function of scattering vector and illumination size produces a fluctuation map that reveals MRO and correlation lengths. FXM can explore MRO and subtle spatial structural changes in a wide range of disordered materials from soft condensed matter to nanowire arrays, semiconductor quantum dot arrays and magnetic materials.

FXM has been demonstrated at micron correlation length scales in studies of a model system comprising polystyrene latex spheres [2]. The theory underlying FXM, the data analysis and the quantitative determination of MRO correlation lengths are discussed. Efforts to develop FXM to study MRO with correlation lengths down to 50 nm are presented.

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Refractive Contrast X-ray Imaging with Laboratory Sources

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Phase contrast imaging with laboratory sources provides a promising means of visualizing low atomic number materials that are difficult to image with conventional absorption-contrast imaging. The phase contrast in most non-periodic materials results almost solely from refraction in the object, and thus such imaging can be termed “refractive contrast imaging.” The authors will present models of several refraction contrast imaging techniques using both sources adapted to have long coherence lengths and imaging optics. Long coherence lengths can be achieved by using collimating optics to provide a parallel x-ray beam, or through the use of small sized sources and long source-object distances. The relative merits of these systems are discussed. Imaging optics allow refraction contrast imaging with lower coherence sources. A benefit of using imaging optics, such as compound refractive lenses or Fresnel zone plates, is that the required resolution on the x-ray camera is greatly reduced. Neither Talbot-Lau gratings nor analyzer crystals are used. Experimental data are presented for each technique, both to confirm the predicted performance, and to demonstrate their relative advantages and disadvantages.

Neutron Phase Imaging and Tomography using a Grating Interferometer

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We report how a setup consisting of three gratings yields quantitative two- and three-dimensional images depicting the quantum-mechanical phase shifts of neutron de Broglie wave packets induced by the influence of macroscopic objects. Since our approach requires only a little spatial and chromatic coherence it provides a more than two orders of magnitude higher efficiency than existing techniques.

This dramatically reduces the required measurement time for computed phase tomography and opens up the way for three-dimensional investigations of previously inaccessible quantum-mechanical phase interactions of neutrons with matter.

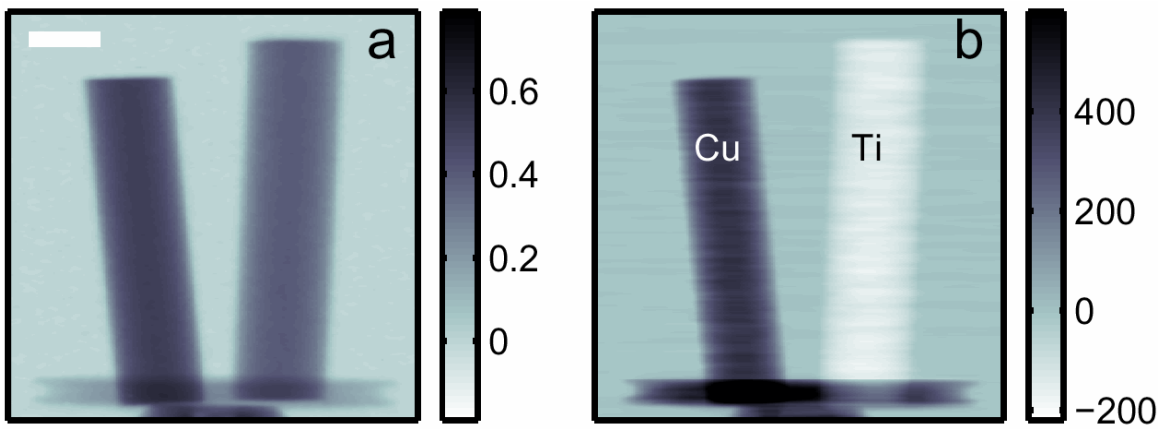

Figure: Conventional neutron attenuation image (a) and retrieved phase image (b) of two metal rods. It is interesting to note that Ti has a brighter color compared to the background, whereas Cu appears darker. This is due to the negative neutron scattering length density of Ti. Consequently, a negative phase shift is measured in the material. The scale bar corresponds to 5 mm.

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Advances in X-ray Phase Tomography using a Grating Interferometer

F. Pfeiffer[1], O. Bunk[1], C. Kottler[1], C. Grünzweig[1], C. David[1], M. Bech[2], G. Le Duc[3], A. Bravin[3], and P. Cloetens[3],

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We report on significant advances and new results concerning the extension of a recently developed *radiographic* phase contrast imaging method to three-dimensional *tomographic* phase imaging. In the case of x-ray synchrotron applications, we demonstrate how the soft tissue sensitivity of the technique is increased and show in-vitro tomographic images with unprecedented quality and resolution of a tumor bearing rat brain sample, without use of contrast agents. In particular, we observe that the brain tumor and the white and gray brain matter structure in a rat's cerebellum are clearly resolved.

To show that the results are potentially interesting from a clinical point of view, we present experimental results, which demonstrate that a similar approach can be implemented with more readily available x-ray sources, such as standard x-ray tubes [1].

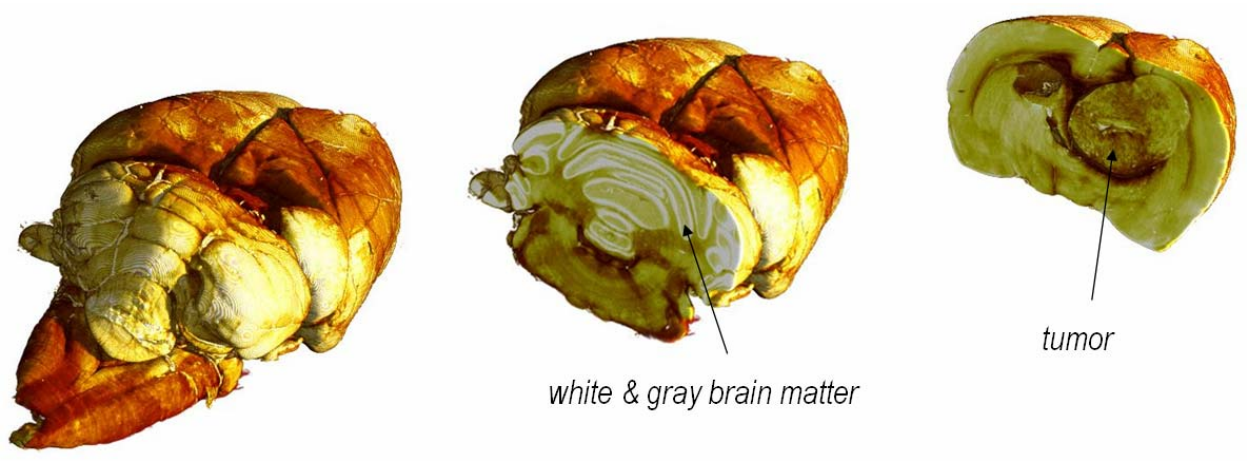

Figure: In-vitro X-ray phase-contrast tomography synchrotron results obtained on a rat brain fixed in formalin. Clearly visible is the white and gray brain matter in the region of the cerebellum and a tumor.

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Using the Loss of Coherence as Signal for Hard X-ray Dark-field Imaging

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Imaging with visible light today employs numerous contrast mechanisms, including bright- and dark-field contrast, phase contrast schemes, and confocal and fluorescence based methods. X-ray imaging, on the other hand, has only recently seen the development of complementary contrast modalities. While x-ray phase contrast imaging has successfully been implemented using several techniques, dark-field imaging, or more generally, scattering based imaging, with hard x-rays in practice still remains a challenging task even at highly brilliant synchrotron sources.
In this contribution, we report a new approach based on a grating interferometer that can efficiently yield dark-field scatter images of high quality, even with conventional x-ray tube sources. The image contrast is obtained by a spatially dependent measurement of the loss of coherence, which is reflected in the decrease of the visibility of an interference pattern [1]. The method provides complementary and otherwise inaccessible structural information about the specimen at the micron and sub-micron length scale. Our approach is fully compatible with conventional transmission radiography and a recently developed hard x-ray phase contrast imaging scheme [2,3]. Applications to x-ray medical imaging, industrial non-destructive testing, and security screening are discussed.

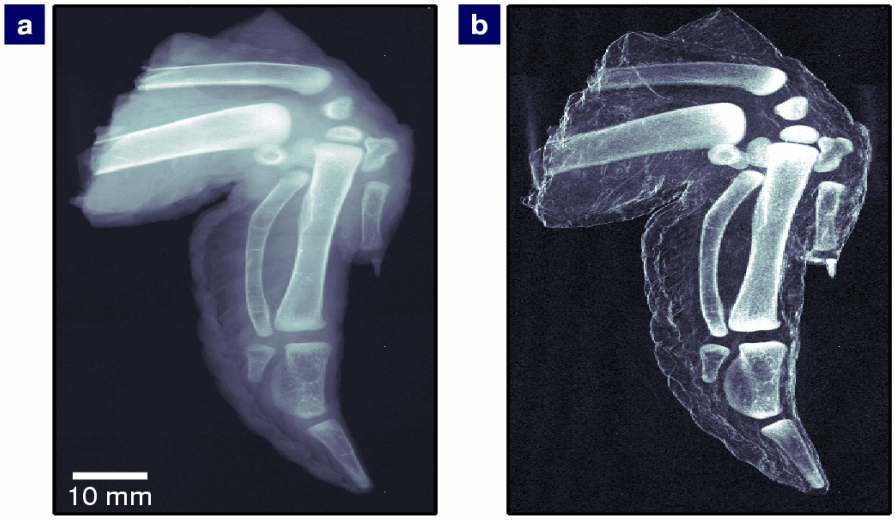

Figure: (a) Conventional x-ray transmission image. (b) X-ray dark-field scatter image. The x-ray scattering due to the porous microstructure of the bones and the reflection at internal or external interfaces produce a strong signal in the dark-field image.

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Theoretical and Computational Methods for Fresnel Diffractive Imaging.

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Conventional approaches to coherent diffractive X-ray imaging are based on the assumptions of plane-wave illumination of the sample and weak X-ray scattering described by the first Born approximation. The retrieval of complex electromagnetic wavefields from oversampled far-field diffraction data and *a priori* information about the finite support of the diffracting object using iterative projective algorithms is now a well-established computational procedure. The convergence of these schemes is often poor because the operators that define such schemes do not represent projections onto convex sets and, viewed as a problem in optimization theory, minimization of the error involved in fitting the diffracted intensities leads to multiple minima and iterative stagnation. A range of relaxation schemes has been proposed that provide practical, and frequently successful, solutions to the stagnation problem, but the formal non-convex structure of the underlying inverse problem remains. In addition, the use of plane-wave illumination necessitates the use of beam stops to protect the detector, resulting in a loss of some small-angle scattering information that plays an important role in determining the overall normalization of the wave.

The Centre for Coherent X-ray Science has developed techniques that employ illumination structured in both phase and amplitude. Experiments based on this approach generate both a low-resolution holographic signal as well as large-angle scattering data carrying high-resolution structural information. No small-angle scattering information is lost, because beam stops at the detector plane are not required. The phase structure of the illumination removes the transverse translational degrees of freedom inherent in far-field Fraunhofer diffraction, and profoundly changes the mathematical and computational characteristics of existing phase retrieval algorithms.

This presentation examines the theoretical and computational advantages inherent in the use of illumination possessing phase structure, including the effect on conventional projective iterative methods such as the Error Reduction, Hybrid-Input-Output and Difference Map algorithms. We explore the use of approximate solutions of the Transport of Intensity equation to provide initial trial functions for these iterative algorithms and demonstrate how reconstruction ambiguities may be resolved by employing independent solution algorithms in parallel. The hypersurface in the active parameter space characterizing the mean-square fitting error in fitting the experimental data is finally examined in the context of the modification caused by the application of phase curvature to the sample, demonstrating the dramatic reduction in the density of local minima and the increased likelihood of finding a solution corresponding to the global error minimum.

Full-Field Imaging with Coherent Hard X-Rays

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We report about full-field imaging with in-line phase contrast. The technique is applied to study the structure of a mammalian cochlea. The objective is to measure the motion patterns in a closed cochlea. This is in particular challenging, because soft (and low absorbing) tissue and strong absorbing structures, such as bone, are covering each other. In addition we report about recent imaging in cone-beam geometry using Kirkpatrick-Baez mirrors. The method has a strong potential for high-resolution and dose efficient imaging.

Soft x-ray phase contrast microscopy using a spiral zone plate

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Spiral zone plates[1], forked gratings, and spiral phase plates have been used in the visible light regime as optical elements because their angular phase pattern is useful for many applications: the generation of optical phase singularities, particle manipulation, interferometry, and microscopy. It was demonstrated in the visible light regime that using these elements as spatial filters is a way to optically implement radial Hilbert transform image processing[2,3].

Phase sensitive imaging is also useful for contrast enhancement in x-ray microscopy[4,5]. We use the spiral zone plate as a single element objective which is equivalent to optically combining the radial Hilbert filtering operation and the imaging operation into a single step. Spiral zone plates were fabricated using electron beam lithography as seen in figure 1. Initial experiments imaging $Gd_{25}Fe_{75}$ thin films with perpendicular magnetic anisotropy were performed using a full-field soft x-ray transmission microscope XM-1 at the Advanced Light Source (ALS) in Berkeley. A resulting image showing the edge enhancement is shown in figure 2. This is similar to that of Normanski phase contrast imaging except with isotropic sensitivity. Other x-ray phase contrast microscopy experiments will also be discussed.

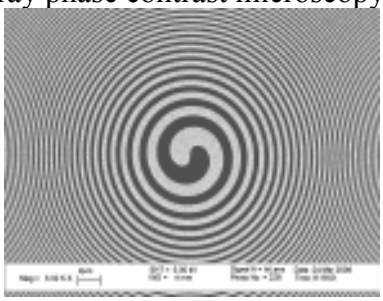

Figure 1

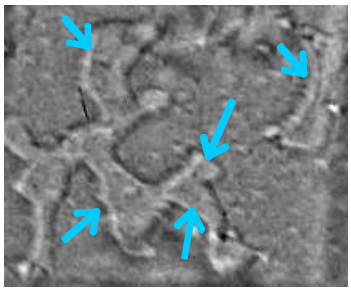

Figure 2

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Crystallography without Crystals: Structure from Diffraction Patterns of Randomly Oriented Molecules

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Amongst the promised capabilities of fourth generation x-ray sources currently under construction is the ability to record diffraction patterns from individual biological molecules. One version of such an experiment would involve directing a stream of such molecules into the X-ray beam and sequentially recording the scattering from each molecule of a short, but intense, pulse of radiation. Due to the shortness of the pulse, the diffraction patterns may be thought of as arising from molecules of different, frozen, random orientations. It has been proposed that the data from such diffraction patterns may be pooled to reveal the 3D structure of the molecular species. However, to this date, there has been no demonstration of such an algorithm.

We propose an algorithm based on the method of “common lines”, first developed for 3D electron microscopy. In that field, the raw data are projections of electron microscope images of "identical" objects in multiple orientations. In our problem the available data are a set of diffraction patterns from unknown orientations of an object. Due to the absence of direct phase information, there is less information in the diffraction patterns, resulting in a “Friedel Law ambiguity” in the determination of common-line directions by comparison of sinograms from different diffraction patterns. We show that this ambiguity may be removed by imposition of consistency relations amongst different diffraction patterns, a process that ensures that the common-line directions are determined by more than just the data from two diffraction patterns. The common-line directions determine two of the three Euler angles specifying the relative orientations of any two diffraction patterns in reciprocal space. Given three or more diffraction patterns, the remaining “hinge” Euler angle may be determined by a formula derivable from spherical trigonometry. By systematically cycling through all available diffraction patterns, we will show that it is possible to correctly orient the majority of these in 3D reciprocal space.

A gridding algorithm then interpolates the diffracted intensity data onto a regular Cartesian grid in reciprocal space at a sampling frequency greater than the Nyquist frequency for the expected size of the object. An iterative algorithm, which cycles between reciprocal and real space, applying appropriate constraints in each, is then able to determine the phases associated with the reciprocal space amplitudes, and simultaneously the electron density of the object under investigation.

We will demonstrate this algorithm for simulated diffraction patterns from a small protein. The molecular electron density is recovered with excellent fidelity. We have also investigated other methods of structure solution, e.g. one based on a “projection matching” approach, progress on which will also be reported.

Iterative 1D phase retrieval for studying the structure of confined fluids

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The study of the structure of a fluid under confinement is interesting from a fundamental and from a technological point of view. Theoretical studies have shown that the molecules of a confined liquid order in layers parallel to the confining surfaces, resulting in an oscillating density profile along the confinement direction. Experiments confirming this effect are scarce due to the difficult access to a small amount of fluid confined between two solid walls.

Microcavity Array Phase Profiling (MAPP) is a promising new technique for the direct determination of the 1D density profile of a confined fluid in the confinement direction [1,2]. The fluid is confined in an array of identical cavities and x-ray diffraction from the array is recorded in the far field with a strip detector. By means of a phase retrieval algorithm, the average density profile of the fluid in the confinement direction can be uniquely retrieved in a model independent way.

Different colloidal systems with a particle size of approximately 110 nm have been investigated with this method, showing an ordering of the colloidal particles in two dimensional sheets parallel to the confining walls, as predicted. Depending on the ionic strength and the volume fraction of the colloidal particles in the solution, different phenomena from liquid ordering to crystallization have been detected, even when the volume concentration of the bulk solutions remained lower than the freezing point. This poster comprises complementary information to the contribution by O. Bunk et al., which is submitted as an abstract for an oral presentation.

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[2] O. Bunk, submitted.

Phase imaging with resonant soft x-ray Fourier transform holography

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We demonstrate quantitative phase imaging using resonant coherent soft x-ray scattering. This lensless imaging technique has the benefits of phase contrast imaging while resonantly tuned to an absorption edge which inherently enhances the absorption and limits the probing depth. The method is illustrated by use of a Co/Pd multilayer sample which exhibits non-periodic magnetic domains. Magnetic contrast is obtained from x-ray magnetic circular dichroism at the Co $L_{2,3}$ absorption edges. The strong energy dependence of the real part (phase) and imaginary part (absorption) of the refractive index is utilized to selectively enhance image contrast at different energies in vicinity to an absorption resonance. An absorption-dominated diffraction pattern is directly obtained at the absorption peak, whereas phase-dominated diffraction is obtained slightly before and behind the resonant peak. Therefore, this method facilitates high contrast imaging while decreasing the radiation dose of the exposed object. The phase and absorption images of the magnetic domains are reconstructed by means of Fourier transform holography[1], where the diffracted wave is encoded by a reference wave in the Fraunhofer regime of the object. The real space image is readily obtained by Fourier transform inversion. The absorption and phase information about the object is separated and retrieved in the respective real and imaginary part of the complex object-reference convolution and cross-correlation in a quantitative manner. In addition, combining the phase and absorption information available in the diffraction patterns will improve current phase retrieval algorithms and the symmetry properties of phase holograms lessen the impact of beam stops.

This work has been supported by DOE (SSRL/PULSE).

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Iterative Phase Retrieval from Incomplete Coherent Diffraction Patterns

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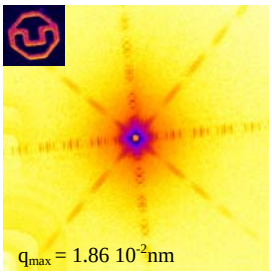
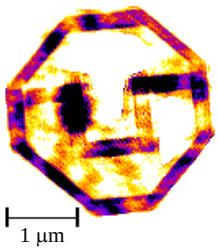
Coherent x-ray diffraction imaging (CXDI) is a suitable method to determine the structure of small, non-periodic objects up to a resolution on the nanometer length scale [1,2]. It can be considered as a microscopic technique providing information on structural properties of micro- and nano-objects in a variety of scientific fields, e.g. whole-cell imaging [3] and mesoporous structures.

In order to develop the method a new setup was designed and built. It allows for the in-vacuum sample positioning with nanometer accuracy in all three dimensions. Furthermore, a high resolution x-ray detector with an effective pixel size of 0.65µm is available to monitor the incoming beam intensity distribution as well as for slit and sample alignment. The diffraction patterns were recorded by a directly illuminated CCD with a pixel size of 20µm. Additionally, a rotation stage is foreseen for tomographic 3D-methods. Using this movable setup a first experiment was carried out at beamline ID10C, ESRF.

The reconstruction results of experimental data are strongly influenced by the quality of the measured coherent diffraction pattern. First of all, partial coherence effects reduce the visibility of interference fringes which can lead to artifacts in the reconstructed image. Furthermore, a certain amount of low frequency data is lost due to the beam-stop, additional parasitic scattering from the slit blades or a contribution of third harmonic photons in the central part of the diffraction pattern. The missing data can be provided either directly by using simulated diffraction data from a low resolution image, e.g. a SEM-image, or by using a tight support in order to confine the number of possible solutions at low frequencies.
In fig. a) the measured diffraction pattern of a lithographically prepared gold structure (size of 3µm and thickness of 150nm) is shown.
A first reconstruction result from this data set is presented in fig. b).



a) Diffraction pattern of a gold pattern (TU-Dresden logo)



b) Reconstruction of a)

In this presentation we will show the reconstruction results from experimental data obtained at the ESRF and discuss the reconstruction artifacts that can emerge if the data are incomplete.

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On The Validity Domain of the First Born Approximation in X-ray Diffraction Microscopy

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We are investigating the validity domain of the first Born approximation as applied to coherent x-ray diffraction microscopy. We simulate the x-ray diffraction patterns of objects of varying size and composition by multi-slice propagation of a plane x-ray wavefield through the object followed by a Fourier transform. We then reconstruct the object using the RASR algorithm both with and without applying a reality constraint. We find that the convergence error rises steadily with object thickness when a reality constraint is applied while it remains reasonably flat without the constraint. Furthermore, we have identified a thickness threshold, above which, the reconstruction algorithm is no longer capable of solving asymmetric structures when a reality constraint is applied. This threshold is plotted as a function of material density and x-ray wavelength. It is interpreted to be the point at which the far-field diffraction pattern becomes so significantly asymmetric that it can be assumed to not satisfy the first Born approximation and may serve as an upper bound on its validity. In fact, the complex values of the exit wavefield, which are used to generate the diffraction patterns, fit nicely within the first Rytov approximation. We discuss the implications of these results for experimenters wishing to do three-dimensional reconstructions from a tilt series of two-dimensional diffraction patterns.

X-ray Diffraction Microscopy Across the Spectrum: Soft (1~2.6 keV) and Hard X-rays for Materials Science and Biology

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The X-ray diffraction microscope is a versatile probe to characterize nano-scale materials and biological specimens. With significant effort over the last several years it has now reached a point that this novel imaging technique can be applied to address certain scientific questions. Our recent progress includes the exact oversampling of diffraction patterns for improved phase retrieval [1] and the implementation of a guided hybrid-input-output algorithm (GHIO) for consistent image reconstruction. By using equally sloped tomography, we carried out a quantitative 3D imaging of a heat-treated GaN particle with each voxel corresponding to 17x17x17 nm^3. We observed the platelet structure of GaN and the formation of small islands on the surface of the platelets, and successfully captured the internal GaN-Ga_2O_3 core shell structure in three dimensions [2]. Furthermore, we have developed a labeling technique to tag specific target proteins in yeasts and chromosomes using quantum dots. These developments allowed us to tackle various intriguing scientific problems in nano-structured materials, bio-minerals and single biological cells at an appropriate length scale.

This work was supported by the U.S. Department of Energy, Office of Basic Energy Sciences (DE-FG02-06ER46276) and the U.S. National Science Foundation, Division of Materials Research (DMR-0520894). Use of the RIKEN beamline, BL29XUL at SPring-8 was supported by RIKEN. Use of APS was supported by the U.S.DOE under the contract No. W-31-109-ENG-38.

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Coherence, Multiple Scattering and Scaling in Diffractive Imaging

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Diffractive imaging allows the formation of new kinds of images, not possible with a conventional microscope. Since one has access to the complex wavefield, a dark-field image may be formed which is linear in the charge-density (for X-rays) or electrostatic potential (electrons). The physical introduction of a beamstop in conventional microsopy produces non-linear dark-field images containing artifacts such as maxima where either minima or maxima in density occur in the sample.

An arrangement in which the X-ray source is conjugate to the sample may have advantages, and will produce partially coherent illumination if the source is extended. We analyse the degree of coherence in this case, where the effect is equivalent to lateral translation of the sample. We also show that for the case where the source is conjugate to the detector, the coherence patch for diffractive imaging must be larger than the object, corresponding to the requirement that the beam-divergence must be less than the oversampling angle (for critical Shannon sampling of the diffracted intensity). This has implications for exposure time calculations.

In the early days of LEED theory it was thought that absorption effects might be more severe than multiple elastic scattering. In that case, the ratio of the strength of the two effects is controlled by a complex optical potential, and this ratio does not depend on beam energy. For X-rays, where it does, the question arises as to how the relative importance of the two effects varies across an absorption edge, and whether beam energies may therefore be found where the attentuation length (due to photoelecton production) is much less than the multiple scattering extinction distance. Soft-Xray multislice calculations will be used to investigate this question.

If time allows, a new application of the iterative Ozlayni filipping algorithm (a HiO relative) to powder X-ray diffraction data will be shown to have advantages. (Wu et al, Nature Materials, 5, 647 (2006)). The dependance of exposure time on beam energy and on resolution for the Serial Crystallography scheme may also be reviewed. This scheme sprays hydrated, laser-aligned proteins in single-file across the ALS synchrotron in order to determine their structure (Starodub et al , J. Chem Phys. 123, 244304 (2005)) . Supported by ARO and NSF.

Coherent Diffraction Imaging of Nanostructures with Hard X-Rays

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Modern high-brilliance synchrotron sources provide intense X-ray beams possessing a high degree of coherence which allows for realising lensless X-ray microscopy using the oversampling technique. Thereby the coherent diffraction pattern is sampled on a scale finer than $2\pi/D$, with D denoting the size of the object. A model-free reconstruction of the object is obtained by using iterative phase-retrieval algorithms that transform the image back and forth between direct and reciprocal space while applying appropriate constraints in each domain[1]. The spatial resolution of this approach is limited by the wavelength of the used radiation only, and the imaging process does not suffer from lens errors. Moreover, one can take advantage of the high penetration depth and element specificity of X-rays.

Model-free structure determination of isolated non-periodic objects on the nanoscale has been demonstrated in the past by using soft [2, 3] and hard X-rays [4, 5]. Experiments with hard X-rays in SAXS geometry are, however, difficult due to the missing forward data caused by the beamstop protecting the CCD camera. Thus, essential information on the overall dimensions of the sample may be lost and there are endeavours to overcome that problem by means of clever algorithms [6, 7]. One of those is the so-called “shrink wrap” (SW) algorithm [6], which iteratively constructs a new object support based on the actual image estimate [6, 3]. However, information on the limit up to which the SW algorithm reliably finds the true object support is missing in the literature.

Here, we report on coherent diffraction imaging experiments of model nanostructures using hard X-rays at beamline ID10C at the ESRF. Beside concrete reconstruction results that are obtained using SW, we present an analysis of the obtained contrast based on coherent diffraction patterns from nanometre-sized colloidal particles deposited on a Si_3N_4 membrane.

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Dose, Resolution and Exposure in Serial Crystallography

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Resolution of biological diffractive imaging is severely limited by the sample damaging prior to the accumulation of statistically accurate data. In serial crystallography hydrated macromolecules pass successively across an X-ray or electron beam giving a diffraction pattern. The macromolecules are brought into alignment by the electric field of a high power laser. Since the molecule is exposed to the beam for a period of only a few hundred nanoseconds, the probability of an ionization event leading to damage is small, while the scattering from many molecules can be summed to achieve the desired signal. In order to provide a critical dose during molecule transit through an x-ray beam, its intensity should be much above the capabilities of any existing or projected x-ray sources. The effect is the same as having an aligned molecule in the beam for any given time without damage, and the resolution limit is determined only by the exposure time. The purpose of this work is to establish the relationship between exposure time, the required resolution, and the number N of molecules in the beam at any instant.

The real space charge distribution is found using the Hybrid Input Output (HIO) algorithm, where the object is embedded in a support that provides enough constraints to solve the phase problem. To optimize the HIO algorithm the pixel size on the detector should be matched to $2^{1/3}\times D$, where D is the object size, by adjusting the detector position. Then the resolution d in the reconstruction is related to the maximum scattering wave vector, which is governed by the detector size. This means that for a given detector the resolution is fixed both by the geometry and the number of pixels. The counting time can be estimated by setting the minimum acceptable number of counts at the maximum scattering angle, corresponding to the highest spatial frequency or best resolution. An analytic solution for the required exposure can be derived for a spherical macromolecule, and this gives a d^{-4} dependence on desired resolution and an inverse square dependence on wavelength. We estimate that the counting time for 0.5 nm resolution with the 8-kV X-ray flux of 3×10^{8} photons $\mathrm{nm}^{-2}\,\mathrm{s}^{-1}$ (for the projected Energy Recovery Linac source) is 470 s for $N = 1$, which is sufficient to delineate secondary structure.

This approach gives a lower limit for the required X-ray fluence, because it does not account for convergence stability of a phase retrieval algorithm with respect to a shot noise of the scattered photons. For more realistic estimations, we perform simulations of the diffraction pattern for GroEL-GroES, a chaperone protein complex, using coordinates taken from Protein Data Bank (1SVT). After adding a shot noise, corresponding to different data collection times, the object is reconstructed by the HIO algorithm with additional reality and positivity constraints. The resolution in each reconstructed object is evaluated from the spatial frequency cut off for the corresponding transfer function. Then the counting time scales with resolution as $d^{-3.99}$ in exact agreement with analytic solution. However, this time is found to be higher than predicted, likely because variations in scattered intensities rather than absolute count rate must be reliably detected. That requires collecting the higher number of counts at the maximum scattering angle. The data acquisition time can be reduced by using lower X-ray energy, increasing the number N of simultaneously exposed molecules or addition of convex constraints (such as histogram) to reduce the degree of oversampling required. Supported by ARO and NSF.

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Theory of a unified field: standard file formats and subroutine libraries for diffraction imaging

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Within a generation, protein crystallography has gone from tens of structures solved per year to thousands. In the same period, single particle methods in cryo electron microscopy have moved from being the subject of development in a few labs to something that is widely applied by many researchers. Advances in both of these fields have been due to a number of factors, but one of them is the adoption of a suite of highly developed programs and subroutine libraries which are able to work with common file formats.

We have begun to develop some interoperable capabilities between diffraction microscopy data handling and reconstruction software developed at Stony Brook, Lawrence Livermore National Laboratory, Lawrence Berkeley National Laboratory, and Cornell University. With this software (made available through a CVS or concurrent versioning system archive), we are able to automate the collection of data, combine raw diffraction patterns, run iterative reconstructions in 2D and 3D, and display the results. We propose here to increase the degree of interoperability, and reduce the work of building diffraction reconstruction software, by encouraging the community to contribute to the development of certain common standards for software. By using the widely-available, platform-independent HDF5 hierarchical data file format with a set of specific groups for different data functions, it will be possible to pass data files among different reconstruction programs to compare their success in reconstruction. By using a subroutine library written to carry out basic array operations such as FFTs, addition/subtraction, and array masking, it should be possible to develop C language programs that will compile and run both on single-processor laptops as well as MPI-enabled computational clusters. We describe our present efforts to develop file and array operation subroutines, and encourage community participation in developments towards common software structures.

We thank the Division of Materials Research, Office of Science, US Department of Energy for support of basic technique development in diffraction microscopy.

Progress towards x-ray diffraction microscopy of frozen hydrated cells at ALS 9.0.1

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The method of x-ray diffraction microscopy[1] has been used to image an entire freeze dried cell, unstained, using 750eV X-rays, with an image resolution of 30 nm[2]. The method is currently being extended to hydrated and 3D samples. The extension of the work has several challenging areas including sample preparation and phase reconstruction.

For application to hydrated organic specimens such as in some soft condensed matter studies and especially for biological applications, it is essential to work with specimens in the frozen hydrated state at liquid nitrogen or lower temperature. For this reason we have built a specialized apparatus with a cryo specimen mount and airlock sample exchange mechanism[3], and we are now working with an automated plunge-freezing system. With these improvements, we are beginning to collect tilt series x-ray diffraction data. We will discuss steps taken to improve ice thickness uniformity, frost reduction, and specimen pre-alignment, and present our current progress on x-ray diffraction imaging the yeast *Saccharomyces cerevisiase* carrying the wi5 mutation, and the bacterium *Deinococcus radiodurans*. We will also discuss the effects of ice thickness variations on the compact support requirement of finite support iterative phase retrieval algorithms.

We thank the Division of Materials Research, Office of Science, US Department of Energy for support of basic technique development, and the National Institute for General Medical Services, National Institutes of Health, for support of biological specimen preparation and imaging.

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X-ray Holographic Microscopy with Total-reflection-mirror Interferometer

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Holographic microscopy with a wavefront-dividing-interferometer has been developed. The concept of optical system is the same as that of electron holographic microscopy. A magnified image of object is generated at an image plane of objective lens, and a refractive prism (or a total-reflection plane mirror) is placed behind the back-focal-plane of objective lens to configure the wave-front-dividing interferometer, as shown in the figure. A half of divergent wave from the focal point is deflected by the total-reflection mirror, and superimposed on the magnified image of object. The interferometer with total reflection mirror is an analogue of Lloyd's mirror interferometer that is well known in the visible light optics.

We have already reported holographic microscopy with Fresnel zone plate objective and refractive prism interferometer [1]. Here, we will present hard X-ray holographic microscope with total-reflection-mirror interferometer, and quantitative phase measurement will also be shown.

A coherent illumination over the acceptance of objective lens is needed for holographic microscopy. The coherent illumination is achieved at BL20XU of SPring-8 by using the undulator light source and the long beamline (experimental station located at 248 m from source points). X-ray energy of 12.4 keV is chosen with a Si 111 double crystal monochromator. A Fresnel zone plate made of 1 μm-hick tantalum is used as an objective lens. The focal length is 155 mm at 12.4 keV, and the outermost zone width is 100 nm. The total reflection mirror is a conventional plane mirror made of borosilicate crown glass (figure error of about 30 nm).

The required spatial coherence for the imaging holography is only one-dimensional coherence, while the two-dimensional coherence is needed for projection-type holography. Asymmetric coherent property of synchrotron radiation light source, i.e. high spatial coherence in the vertical direction and low coherence in the horizontal direction, is used to suppress the speckle noises [2]. Phase retrieval has been performed using two methods: fringe scanning algorism and Fourier transform method. Typical result of phase retrieval by 4-step fringe scan is shown in the figure. The sample is latex sphere with a diameter of 8 μm.

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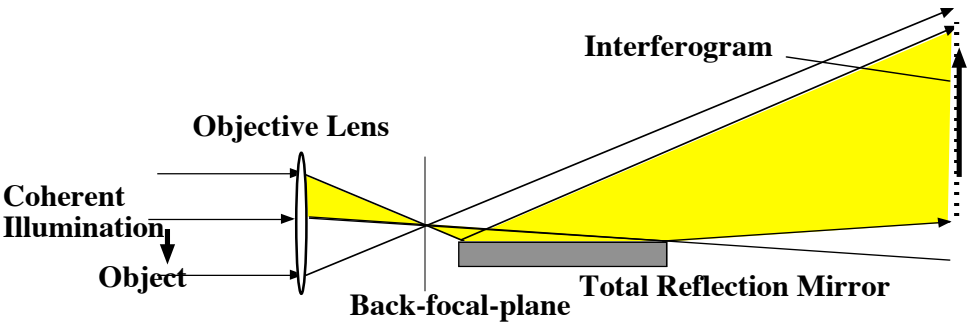

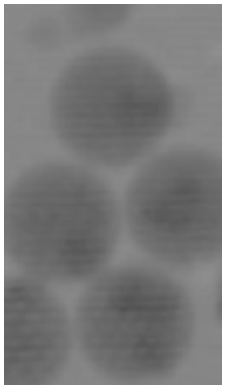

TITLE: Coherent Diffractive Imaging and Fluctuation Microscopy

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Fluctuation microscopy is a statistical analysis technique which examines mid-range order present in a partially ordered sample. Information about the presence of mid-range order may be determined by examining the variations in diffraction patterns from distinct neighbouring regions of a sample. By altering the size of the probing beam it is possible to extract length scales of mid-range order. At present it is not possible to invert fluctuation microscopy results to determine an exact model of the sample. By using the iterative imaging techniques of coherent diffractive imaging it is hoped to determine the exact structure of a sample at the mid-range.

Coherent diffractive imaging techniques require some a priori information about a sample. It is common to use the support constraint or physical extent of the sample to enable reconstruction. Algorithms will be investigated using the length scales of the medium as an additional constraint to aid reconstruction of the object function.

Samples have been investigated at optical wavelengths using fluctuation microscopy experiments. Simulations will be performed to investigate the feasibility of recovering the structure of a sample using a speckle pattern obtained during a fluctuation microscopy experiment. The structure of samples consisting of disordered arrangements of microspheres will be determined using a combination of fluctuation microscopy and coherent diffractive imaging. The techniques explored are fully applicable to shorter wavelengths.

Phase Retrieval from the Coherent GISAXS Measurements

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Coherent X-ray Diffractive Imaging (CXDI) is a new and fast developing field of lenseless x-ray microscopy. It has a potential to reach diffraction limited nanometer resolution at least for non-biological samples. Applications of this method to the study of nano-systems will be given in this talk. One of the well established methods to provide information on the shape and the size of the quantum structures is a Grazing Incidence Small Angle X-ray Scattering (GISAXS) technique. However results of this method are based on a delicate fitting procedure when a complicated models of x-ray scattering and different type of correlation functions are used to obtain reasonable fit to experimentally measured data. Here we propose to use coherent x-rays in GISAXS geometry in order to get all structural information about the sample in a model independent way by phase retrieval of scattered intensity.

Recently we studied theoretically [1] the image formation and reconstruction of coherently illuminated islands of nano-size dimensions in GISAXS geometry. In this scattering geometry the 1-st Born approximation (kinematic scattering) fails to describe correctly the scattering process and instead Distorted Wave Born Approximation (DWBA) has to be used. As a consequence there is no simple relationship between the scattered amplitude and the electron density of the sample in the form of Fourier transform. Total amplitude in DWBA is a coherent sum of several scattered amplitudes. It means that iterative phase retrieval technique that is based on application of Fourier transform many times to reconstruct the phase will fail to give correct image of the sample. However it was demonstrated that there are some favorable conditions (with incident angle close to the critical angle) when GISAXS diffraction pattern originating from individual island of nano-size dimensions can be inverted using iterative phase retrieval technique and can give a reliable image of the island shape. It was also shown that due to interference effects reconstructed image contains superposition of two images: island itself and a standing wave formed by interference of two scattered waves.

These theoretical ideas were tested in our experiments performed at BW4 station at HASYLAB [2] and ID01 beamline at ESRF. GISAXS diffraction patterns were measured from SiGe quantum dots grown on a (001) surface of Si by the Liquide Phase Epitaxy technique. All islands have narrow size distribution, same orientation and are randomly distributed on the surface. Different size of the islands was probed with the Coherent GISAXS technique: 50 nm, 140 nm, 200 nm and 1000 nm. First results of reconstruction from the measured data show all predictions revealed by theoretical analysis.

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Coherent X-ray Scattering and Lenseless Imaging at XFEL Facilities

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Coherent X-ray Diffraction Imaging (CXDI) is a rapidly advancing form of lenseless microscopy. The phase information of the diffraction pattern is embedded in a sufficiently oversampled coherent diffraction pattern. Using advanced computational methods this diffraction pattern can be inverted to produce an image of a sample with diffraction limited resolution.

In that respect future X-ray Free Electron Lasers (XFEL) based on Self-Amplified Spontaneous Emission (SASE) provide the best opportunity for the future development of CXDI methods and its applications to material science and biology. Several similar XFEL sources are at the moment at its construction and planning stage. These are LCLS (Linac Coherent Light Source) in the USA [1], SCSS (Spring-8 Compact SASE Source) in Japan [2] and European XFEL Facility in Germany [3]. Comparing to other projects the European XFEL Facility is unique due to its time structure. It will produce 30 000 pulses per second, to be compared to 120 of the LCLS and 60 of the SCSS.

Lenseless imaging is particularly well suited to the unique capabilities of the XFEL Facilities. As it is expected from theoretical calculations [4] the incoming beam will have nearly full transverse coherence (80% transverse coherence, with a beam of a few mm^2 section). Other unique properties of the XFEL, such as the high peak brilliance (we expect 10^{12} coherent photons in a single pulse) and ultrashort pulse time structure (100 fs pulses separated by 200 ns at European XFEL), allow us to consider applications of CXDI to structural analysis of nanometer-scale particles, inaccessible using third-generation undulator sources. The possibility to study dynamics at such resolutions and on the sub-picosecond time scale with the XFEL opens up entirely new research horizons.

In this talk we will present the scientific case, requirements and the possible realisation of the coherent x-ray diffraction experiments at XFEL Facilities.

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Serial Crystallography of Proteins

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Electron or X-ray diffraction from a beam of laser aligned molecules has recently been proposed as an approach to structure determination of proteins, particularly those that are difficult to crystallize and are too large for structure determination by NMR. In our method, a beam of hydrated proteins (or possibly microscopic protein crystals) is injected into an interaction region at the intersection of two crossed beams: a diffracting probe beam (continuous electron or X-ray beam) and an alignment beam (continuous polarized IR laser). The interaction region will usually be in a high vacuum environment to allow for evaporative cooling of the droplet beam and to minimize scattering of the diffracting beam by residual gas molecules. In the interaction region the proteins are aligned via torque exerted on the molecule by the electric field **E** of the laser beam, acting on an **E**-field induced dipole moment of the molecule. If N identical aligned molecules are in the interaction region at any instant and if the coherence patch of the probe beam is not appreciably larger than the target molecule, then the probe beam produces a diffraction pattern having N times the intensity of that from a single molecule. Since the molecules are constantly replenished (transit time across the interaction region ~200ns), a serial diffraction scheme of this sort allows much longer exposure times than with a static N-molecule crystal. Moreover each molecule receives a dose far below the critical dose that would cause damage at atomic resolution. A transmission diffraction pattern for one orientation builds up at the detector as scattered intensity from successive identical single proteins is recorded. After a useable diffraction pattern has been accumulated, the detector is read out, the alignment laser polarization rotated, and a new diffraction pattern is recorded for a different molecular orientation. In this way a 3-D diffraction dataset can be recorded. From this dataset the charge density of the molecule can be reconstructed with an iterative Gerchberg-Saxton-Fienup algorithm. Serial diffraction requires an injection method to deliver protein molecules or microcrystals - preferably uncharged, fully hydrated (in detergent micelles if membrane proteins), spatially oriented, and with high flux - into a focused probe beam of electrons or x-rays of typically only a few tens of microns diameter. We have examined several potential droplet sources, i.e. electrospray, Rayleigh droplet sources, nebulizers and airojet focusing sources, as to their suitability for this task. Experimental results indicate, that the airojet focused source is the most suitable, since clogging problems are avoided and submicron size droplets can be generated from much larger nozzles. Recently we theoretically evaluate the incident x-ray fluence required to obtain a given resolution from (1) the calculation of the count rate at the maximum scattering angle for a globular object, (2) the explicit simulation of diffraction pattern for a GroEL–GroES protein complex, and (3) the frequency cut off of the transfer function for a phase reconstruction algorithm in the projection approximation. These calculations show that at the available flux at the ALS, preliminary experiments with 2nm resolution should be possible.
Supported by NSF award IDBR 0555845 and ARO award DAAD190010500.
Reference: http://arxiv.org/abs/physics/0701129

Utilizing Coherent Diffractive Imaging Techniques with Partially Coherent Sources.

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Coherent Diffractive Imaging utilizes iterative schemes to invert a far-field diffraction pattern to recover the exit surface wave of the sample that gives rise to the pattern. These schemes make several assumptions about the nature of the experiment in order to successfully reconstruct an image of the object. One important assumption is that the illuminating radiation is fully coherent. While this is a good assumption for laser light, when using X-ray sources the degree of coherence achievable using current sources is limited. The success of several Coherent Diffractive Imaging experiments using X-rays suggests that while full coherence is not necessary[1], a relatively high level of coherence is. This can be a limiting factor.

Theoretical considerations imply that it may be possible to overcome the detrimental effects partial coherence can have on the reconstruction algorithms. By introducing significant phase curvature in the wave incident on the sample, one can effectively force the partially coherent far-field diffraction pattern to approach the fully-coherent pattern as curvature is increased[2]. The phase structure in the illuminating field also has the effect of increasing the stability of the reconstruction algorithms[3].

An alternative approach is to work with the known coherence function of the illuminating field. If the beam can be appropriately characterized[4], it should be possible to utilize this information to prepare an iterative scheme that can reconstruct an object's exit surface wave from its measured partially coherent far-field diffraction pattern.

We present a theoretical basis for the observation that curvature helps ameliorate the negative effects of partial coherence, along with the results of a coherent diffractive imaging experiment demonstrating the benefits of wavefront curvature even in the presence of relatively low levels of coherence. Also presented is a framework for building the partial coherence of the beam into an iterative scheme, with preliminary simulations of an elementary experimental setup.

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Robustness of phase-retrieval from a single x-ray image

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Retrieval of the tissue phase-map, a map of tissue projected electron densities, has great potential for improving tissue contrast and achieving quantitative imaging for tissue characterization. The robustness of a phase-retrieval approach is of critical importance for clinical imaging applications. On the one hand, the limits on permissible organ radiation doses make acquired images much noisier than that encountered in other non-clinical applications. On the other hand, the phase changes of pathological tissues are small and the diagnosis accuracy would be very much vulnerable to the phase-retrieval errors. For example, while a 4cm-thick breast tissue generates about 700 radians, the phase-shift difference between a 5mm-size invasive ductal carcinoma in the breast and surrounding breast parenchyma is of only about 5 radians for 60 keV x-rays. In this presentation the robustness of phase-retrieval from a single image based on the phase-attenuation duality is compared to its performance with that with the popular Transport of Intensity Equation (TIE) based phase-retrieval approaches for 60-keV x-rays by means of computer simulations. The imaged object is a hypothetical breast of 4 cm thick with very low tissue radiographic attenuation contrasts $\leq$ 0.83% for 60 keV x-ray, this attenuation contrast corresponds to that between a 5mm-size invasive ductal carcinoma and surrounding normal breast parenchyma. For the single phase-contrast image acquisition the source to breast distance was set to 1m and the breast-detector distance all were all set to1m. The x-ray Fresnel diffraction was simulated for the formation of the breast phase-contrast image. Moreover we assumed a quantum noise 5% associated with the image acquisitions. With this single phase-contrast image the breast phase-map was retrieved with an average relative phase-error of 0.11%. For the TIE-based phase retrieval approach, an additional image acquired at the contact mode with an anti-scatter grid was simulated. This contact-mode image is the attenuation image of the breast with a quantum noise of 5% as well. With these two images (one attenuation image and one phase-contrast image) the breast phase-map was retrieved by the TIE-based approach. Since the TIE-based phase-retrieval suffers from intrinsic instability due to the low-frequency noise-amplification, the regularization techniques such as the Tikhonov regularization was employed. The Tikhonov regularization seeks the minimum-norm, least squares solution for phase-retrieval. In spite of increased radiation dose to breast (due to two acquired images needed) TIE-based phase-retrieval still suffers from loss of phase-retrieval accuracy. The average relative phase-error with the TIE-retrieved breast phase-map is 0.91%, compared to that of 0.11% for the duality-based phase retrieval. In addition, the areas suffering from distorted tissue-contrast in the TIE-retrieved breast phase map is prominent. Hence the phase-attenuation duality-based approach is superior to TIE-based approaches for tissue phase retrieval with hard x-rays of about 60 keV or higher. Finally we discuss how this phase retrieval approach facilitates the tomographic reconstruction of a 3-D map of tissue electron densities as well.

Using Crystal Optics as Guard Apertures in Hard X-ray Coherent Diffraction Imaging

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Coherent x-ray diffraction imaging (CDI) is a rapidly evolving field of research and is becoming a useful tool for high-resolution imaging of nonperiodic specimens. In principle, the highest resolution achievable in such experiments would only be limited by the x-ray wavelength. However, in practice, the resolution is limited by radiation damage and by data quality such as the signal-to-noise ratio (SNR) of the data at high scattering angles. Preventing the contamination in the data due to the parasitic scattering from upstream elements is one of main challenges to increase the SNR of the data. In a conventional CDI experimental setup, guarding apertures are used to block the parasitic scattering. Those guarding apertures must be carefully aligned to avoid secondary scattering from the apertures themselves. This is especially true in CDI experiments where hard x-rays are used since the alignment of guarding apertures becomes more difficult.

Recently, we have developed a novel crystal guard aperture concept, in which a pair of multiple-bounce crystal optics is employed [Xiao et al, Opt. Lett. 31, 3194(2006)]. Different from the conventional guarding apertures, the crystal guard aperture does not produce secondary scattering and therefore allows a high-quality coherent incident beam. In this presentation we will present the detailed geometries of the crystal guarding apertures used in our CDI experiments. The effectiveness of the crystal guard aperture method has been verified by theoretical analysis and simulations based on Fresnel propagations of a dynamically diffracted Bragg wave. Recent coherent diffraction experiment results and phase-retrieval results have also confirmed the validity of this new guarding scheme.

The Advanced Photon Source is supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-06CH11357.

The qualitative phase retrieval by the combination of TIE and SCWP with sub 100 nanometer resolution

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In x-ray imaging, the phase contrast is generally 100 to 1000 times higher than the absorption contrast in soft material. However, the quantitative phase information is required for the phase tomography in order to fully utilize the capability of the x-ray imaging. In this work, we demonstrate that the quantitative phase retrieval with a sub-100nm resolution can be achieved from micrographs of zone plate based transmission x-ray microscope (TXM)[1,2]. In this work, a zone plate made of plastic containing objects of sizes from m down to tens of nano-meter is used as a test sample to quantify the retrieved phase. Using the focal serial images in the image plane, the phase information is retrieved quantitatively across the entire range of sizes by combining the transport intensity equation (TIE)[3] and self-consistent wave propagation methods (SCWP)[4] in this partial coherence system. The results show a good agreement with simulation. The study demonstrates a solution to retrieve the phase in the TXM and overcome the deficiency encountered in the two phase retrieval approaches of TIE and SCWP.

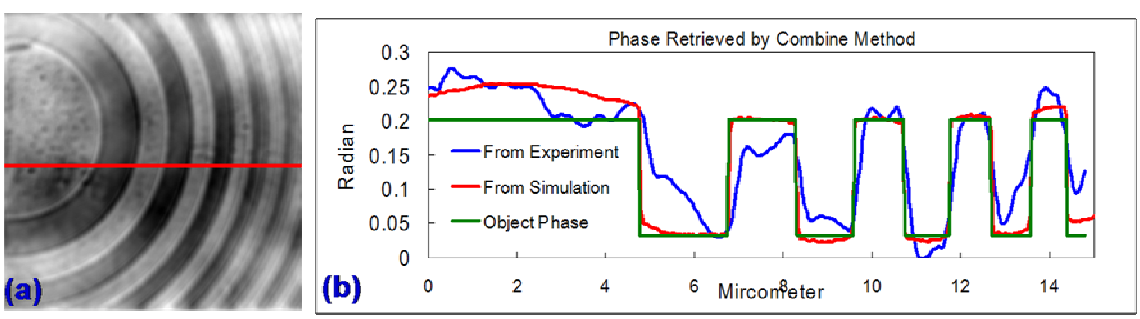

Fig1. The result phase retrieved from the plastic zone plate. (a) the phase image of zone plate. (b) the cross section plot of red line in (a).

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Sub-angstrom Resolution Electron Diffraction Imaging of Nanoclusters

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Diffraction imaging with a coherent electron beam is a promising approach to provide diffraction-limited atomic resolution. However, realizing this requires solving a number of issues, including experimental recording of high quality electron diffraction patterns, the robustness of reconstruction algorithms in the presence of experimental noises, the support and its effect on reconstruction, last but not least important, the dynamic scattering of electrons. In this presentation, we demonstrate the experimental approach for coherent diffraction of individual nanoparticles and implementation of phase retrieval algorithms to achieve atomic resolution by diffractive imaging. The results of a series of simulations will also be presented to show the effects of dynamical scattering, noise and interference with substrates on phase retrieval, and to establish effective inversion procedures. A new iterative algorithm, which uses a low-resolution image in the object domain, is proposed. With the help of the new algorithm, experimental diffraction patterns from individual nanoclusters can routinely be inverted.

Polychromatic Cone-Beam Phase-Contrast Tomography

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We have developed and experimentally implemented an algorithm for propagation-based phase-contrast cone-beam computed tomography of non-absorbing or weak monomorphous objects, which are illuminated using partially coherent radiation from a small polychromatic source. In the case of non-absorbing objects and sufficiently short propagation distances between the object and the detector, the propagation-based phase-contrast tomography (PCT) is "achromatic", thus allowing quantitative cone-beam PCT to be realized using unfiltered divergent radiation from laboratory-based X-ray microfocus sources, as well as small polyenergetic sources of cold or thermal neutrons. The method may also be applied to point-projection optical tomography using a thermal light source, and to the minimally-destructive three-dimensional imaging of cold atom clouds.

The three-dimensional distribution of complex refractive index is reconstructed from a single projection image at each view angle, on the basis of phase contrast introduced by free-space propagation from the object to the detector. A tomographic dataset for a Polyethylene Terephthalate fibre scaffold was acquired on a point-projection X-ray Ultra Microscope (XUM) based on an FEI XL-30 scanning electron microscope. Using these data, a numerical implementation of our method was used for the tomographic reconstruction.

Where applicable, our approach provides a powerful and convenient alternative to working with highly-coherent sources, such as third-generation X-ray synchrotrons or filtered reactor neutron sources. Of particular note is the stability with respect to noise of our algorithm in the case of monomorphous objects, due to the complementary nature of the phase-contrast and amplitude-contrast transfer functions.

Water-encapsulated protein source for x-ray serial crystallography

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A reliable source of micron size water droplets has been constructed for the purpose of delivering water-encapsulated protein for x-ray serial crystallography. A linear stream of droplets of negligible divergence is produced by accelerating a liquid water jet through a high pressure gradient[1] inside a converging gas nozzle. Using a coflowing gas rather than using the nozzle walls to squeeze the liquid jet to smaller diameter eliminates the problem of clogging that has thus far limited the minimum size of Rayleigh nozzle jets. The droplet stream can be either ejected from the gas nozzle into vacuum or produced within a hollow fiber optic tube to allow for further reduction in droplet size by evaporation. The source consists of two parts, an inner water nozzle and concentric outer gas nozzle of exit diameters about 20 μm and 10 μm respectively. The alignment of two parts is permanently fixed upon assembly; droplet size is controlled through water and gas pressures.
Supported by NSF award IDBR 0555845 and ARO award DAAD190010500.

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Combined projection microscopy and fluorescence micro-analysis using multilayer coated KB-optics

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Dynamically bent graded multilayers set in the KB-geometry are developed at the ESRF as an extremely efficient device to focus undulator radiation to spots below 100 nm [1]. The smallest 2D focus reached so far with this device is 80 nm in both directions, whereas it is of the order of 40 nm in one dimension. On one hand, the ultimate limits of this focusing approach are now better understood and on the other hand these systems are used as main optical element in dedicated nanofocus end-stations.

Two different fields benefit particularly from the combination small focus / high flux: projection microscopy and X-ray fluorescence mapping. In projection microscopy [2] the sample is set at a small distance downstream or upstream of the focus and a magnified Fresnel diffraction pattern is recorded on a medium resolution detector set at a large distance from the focus. For tomography, scans are acquired at different focus-sample distances. This yields variable magnifications, but also different effective propagation distances. Two approaches for phase retrieval are considered: first this configuration can be considered as a magnified version of holotomography [3], or alternatively as coherent diffraction imaging with a curved wavefront. The former approach typically yields a direct, but approximate solution, whereas the latter is iterative, but more complete. This high resolution 3D imaging technique is applied in practice using either zoom tomography. i.e. local tomography on samples exceeding the field of view [4], or using laminography, i.e. a particular scanning geometry and 3D reconstruction procedure adapted to flat samples.

In fluorescence imaging the sample is scanned through the focal plane while the spectrum of the emitted fluorescence is recorded with an energy dispersive detector. This rich probe, complementary to transmission imaging, provides element specific information and allows imaging and quantifying trace elements. First applications of this method in the field of neurochemistry at the sub-cellular level have been demonstrated.

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Phase Structure due to Refraction in Coherent X-ray Diffraction

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We have recently succeeded in obtaining full three dimensional complex density maps of a lead nanocrystal, prepared in UHV in our beamline at APS. Quantitative maps of the deformation of the crystal from its equilibrium lattice spacing have been obtained by iterative phasing and hence inversion of the coherent X-ray diffraction pattern [1]. Generally, the interpretation of the phase in such maps offers novel opportunities for mapping strain fields in nanocrystals. However the interpretation of the phase map requires careful consideration of the effects of refraction, which are considered in this presentation. The refraction effect can be calculated completely without adjustable parameters. Once corrected for refraction our image shows a distinct expansion of the surface layers at a temperature just below the melting point. Over most of the surface of the crystal there is a clear outward displacement, which decays exponentially into the bulk. The displacement is suppressed on the (111) facet itself and stronger on the surrounding regions, indicating an orietational variation in surface stress.

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