Zone Plate Microscopy to sub-15 nm Spatial Resolution with XM-1 at the ALS

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With many advances in the last decade, soft x-ray zone plate microscopy has come to be a valuable imaging technique that complements conventional nano-analytic techniques such as electron and scanning probe microscopies. It offers large elemental, chemical and magnetic sensitivities and a myriad of permissible in-situ sample conditions at sub-50 nm spatial resolution. The highest measured resolution was 20 nm [1], achieved with a 25 nm outermost zone width zone plate at the full-field microscope XM-1, at the Advanced Light Source (ALS), Berkeley. The zone plate was fabricated using e-beam lithography. As spatial resolution is roughly equal to the outermost zone width of zone plate, zone width reduction has been the center focus of x-ray microscopy. While e-beam lithography has been providing a path to fabricate high resolution zone plates, shrinkage of outer zone width from 25 nm to 20 nm and below, however, was extremely difficult due to fabrication process limitations in dense line fabrication, such as electron scattering, low e-beam resist contrast and development issues. Isolated lines on the other hand, do not suffer from these problems, and lines of around 10 nm wide can be routinely fabricated. This fact led us to develop a new zone plate fabrication technique, in which a dense zone plate pattern is divided into two (or more) semi-isolated, complementary patterns, and each pattern is sequentially fabricated and overlaid to the other patterns. The key of success to this overlay technique is pattern alignment accuracy, which for zone plate is needed to be better than one third of the smallest zone width. Using the technique, we have successfully realized zone plates of 15 nm outer zone width, with alignment accuracy of 1.7 nm, at the LBNL's Nanofabrication Laboratory. Experiments showed that these zone plates have significantly improved the resolution of the XM-1 microscope, allowing the instrument to resolve 15 nm half-period test pattern [Fig. 1], which has yielded no contrast with our previous 25 nm zone plates. The results indicate that sub-15 nm resolution has been achieved. In the talk, the details of the overlay technique, and its extensibility towards fabrication of 10 nm zone plates will be presented.

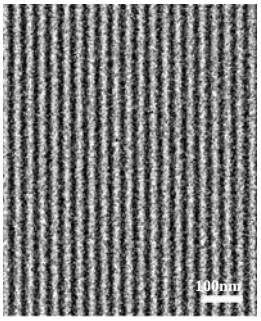

Figure 1. A soft x-ray image of 15 nm test pattern obtained with a 15 nm zone plate at 1.52 nm wavelength (815 eV).

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X-ray micro-analysis activities at the ESRF

J. Susini, M. Salomé, R. Tucoulou, G. Martinez-Criado, S. Bohic, D. Eichert, P. Bleuet, I. Letard, M. Cotte, J. Cauzid, B. Fayard, R. Baker and S. Labouré

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The X-ray microscopy activities at the ESRF have to be considered in the broad context of the development of synchrotron based multi-keV micro-analysis techniques. This evolution aims not only at following the evident trend of nano-technologies by pushing spatial resolution to new limits, but also at providing novel and complementary analysis techniques: considering the concomitant developments of high performance laboratory instruments and the construction of new dedicated synchrotron beam lines worldwide, a highly competitive context can be anticipated for the coming years. Synchrotron based X-ray micro-analytical techniques (diffraction, imaging and spectro-microscopies) will play a crucial role by offering unique capabilities (3D information, time-resolved, chemical selectivity,…).

Among the 40 beamlines in operation at the European Synchrotron Radiation Facility, three are fully dedicated to X-ray microscopy and micro-spectroscopy techniques in the multi-keV range (2-30keV). In the past three years our R&D activities have been focussed on three major programmes:

i) *In-situ experiments*: non-uniform systems in the three broad categories of earth/environmental, material/archaeological and chemical/biological sciences, in which spatial inhomogeneity of the order of the micron or below is omnipresent. X-ray microprobe applications should evolve towards two dimensional mapping of valence state / local structure on the (sub)micron scale in both stable and time dependant inhomogeneous systems under controlled conditions (temperature and pressure).

ii) *New detection techniques*: the solid-state energy dispersive detectors used until recently at ID21 and ID22 offer limited solid angle coverage of a few 10^{-2} sterad. On one hand, significant efforts are being made to enhance fluorescence collection by optimising detector geometry, while on the other, new detection techniques such as phase contrast and XEOL are proposed.

iii) *A new FTIR spectro-microscopy endstation*: most of our scientific cases require a multi-modal approach, consisting of a combination of analytical techniques providing chemical as well as structural information. There is therefore a strong interest to perform different measurements on the same sample under optimal conditions. The availability of a synchrotron based IR spectro-microscopy instrument, complementing hard (ID22) and soft (ID21) X-ray spectro-microscopy beamlines will constitute a unique micro - characterisation facility.

An overview of these activities will be given. New developments will be presented and illustrated by examples of scientific results.

Phase Contrast Hard X- ray Microscopy with the Spatial Resolution better than 100 nm

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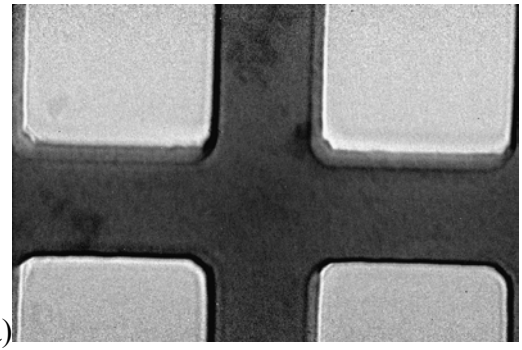
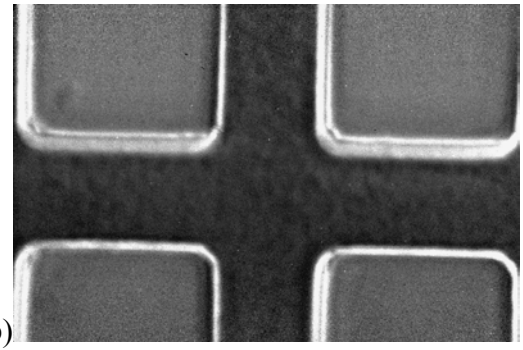
The present status of the hard x-ray full field microscopy[1] at 1B2 beam line at PLS (Pohang Light Source) will be presented.

This is an analog of an optical microscope, consisting of a condenser and an objective zone plates. We further magnify the visual image on the scintillation crystal with a microscope objective. Zernike phase contrast was lately implemented by adding an annular aperture on the condenser zone plate and phase plates at the back focal point of the objective zone plate.

Below are images of a Cu #2000 mesh taken a) in amplitude and b) in Zernike phase contrast at 6.95 keV. In figure b) the edges of the square holes look brighter than the background because it was shot in negative phase contrast.

fig. a)  fig. b) 

Figures a) and b). Cu #2000 mesh imaged with 1B2 transmission x-ray microscope in bright field and in negative phase contrast, respectively. The width of a bar and a square in the Cu # 2000 is 5 and 7.5 microns, respectively. The field of view is about 20 × 13 micron^2.

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High Spatial Resolution Scanning Transmission X-ray Microscopes at the Advanced Light Source

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The main advantage of scanning transmission x-ray microscopes (STXM) over other types of microscopes is the ability to record high quality images from samples of widely varying sizes and to complement these images with high resolution spectroscopy, time resolved, polarization, and magnetization measurements.

The Advanced Light Source is the home of two high performance STXMs. One is located at a bending magnet on beamline 5.3.2 and is dedicated to work mainly in the 250 eV to 600 eV energy range covering all important C, Ca, K, N, and O absorption edges. The other STXM is located at the ALS Molecular Environmental Science beamline 11.0.2. This beamline has as a source a 1.5 m elliptically polarizing undulator (EPU) and a high quality entrance slit-less variable included angle plane grating monochromator providing x-rays from 80 eV to 2100 eV with high energy resolution, excellent beam stability and a high degree of coherence. Both microscopes have high spatial resolution that is currently limited by zone plate performance. Current zoneplates made by LBNL-CXRO have 25 nm outer zones which focuses the x-ray beam to a spot size of about 30 nm. With such a zoneplate, features of 15 nm -20 nm in size can be resolved.

While the 5.3.2 STXM is mainly dedicated to routine measurements, the 11.0.2 STXM is constantly modified to meet the requirements of many novel and challenging experiments. The current performance characteristics and capabilities of the ALS-MES beamline 11.0.2 STXM follow:

- Energy range: 80 eV – 2100 eV with energy resolution > 7500.
- Spot size: 30 nm (theoretical). Can resolve smaller structures -15 nm (practical).
- Photon flux: Up to 10^9 ph/s with full spatial resolution and $E/\Delta E$ > 3000. 4 basic zone plates: 25, 35, 40 and 45 nm.
- Minimum dwell time per pixel: 0.05 ms, maximum scanning rate: 12 Hz.
- Scanning range: 4000 x 2000 pixels – up to 20 x 4 mm.
- Minimum step size: 2.5 nm.
- Positional stability of staying at the same spot for spectra acquisition: < 50 nm (laser interferometry).
- EPU – polarization dependence, circular dichroism + electromagnetic studies.
- Possibility to scan sample at 60 deg relative to the photon beam for out of plane linear polarization and in plane magnetization.
- Single photon timing (50 ps resolution).

Those STXM capabilities and performance will be demonstrated with results of measurements of environmental, polymer, magnetic and biological samples.

TwinMic – A European Twin X-ray Spectromicroscopy Station

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Two types of transmission X-ray microscopes are worldwide in operation - scanning and full-field imaging - with different but complementary imaging capabilities. A novel approach used in a RT&D project of the European Commission (HPRI-CT-2001-50024) is aiming at integrating both microscope types in a single instrument with easy switch between the two modes. For the first time, a X-ray microscope is constructed by the united effort of European groups that have mastered in X-ray instrumentation, optics and detectors, nanotechnology, imaging and X-ray spectroscopy using different contrast mechanisms.

The expected potential and capability of such a twin microscope station is the combination of complementary microscope modes with versatile contrast techniques into a *single* instrument to perform: (i) X-ray imaging for morphological characterization combined with dynamical studies and tomography; (ii) spectromicroscopic analysis including elemental mapping and determination of specimen's chemistry on microscopic scale; and (iii) specimen characterization in their natural, solid or liquid environment.

Essential strength of the instrument is its wide working energy range from 250 - 2500 eV preserving the performance of individual microscopes. The photon energy range covers the water window between the C, N and O absorption edges and L-edges of Fe, Ni, Co with particular importance for characterization of magnetic materials. Access to K-absorption edges of elements opens up the opportunity for advanced studies in biology, medicine, pharmacology, geochemistry, environmental and earth sciences, and material research.

The TwinMic station is temporary hosted by the ELETTRA BACH beamline, where both scanning and full-field imaging modes past successfully first commissioning experiments.

A full-field KB-FZP microscope for hard X-ray imaging with sub-100 nm resolution

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A full-field X-ray microscope for sub-100 nm imaging and tomography has been built at the UNICAT-beamline 34 ID-C at the Advanced Photon Source (APS). The instrument works with a Kirkpatrick-Baez mirror (KB) as condenser and a micro-Fresnel-zone plate (FZP) as objective lens. 80 nm-features in a Nickel structure have been imaged, operating the microscope at a photon energy of about 9keV.

In sector 34 of the APS, the beam is shared between two hutches. A platinum-coated silicon single-crystal mirror deflects the main cone of the beam into the C hutch, and cuts off the higher undulator harmonics. The double-crystal fixed-exit Si-111 monochromator has an energy bandwidth of $\Delta E/E \sim 10^{-4}$. At 9 keV photon energy, the flux is in the order of 10^{13} photons/s. The intensity drops to a third of this value when the microscope is operated in the so-called "parasitic mode", that means when the undulator is tapered for the experiments in the 34 ID-E hutch. The cross section of the beam is about 1mm^2 at 55 m distance from the source. At this location a Kirkpatrick-Baez (KB) system used as a condenser [1] focuses approximately 63% of the incoming intensity onto the sample spot, matching the aperture of the objective lens. For the latter we have a choice of gold micro-Fresnel-zone plates (FZP)[2] having outer zone widths from 40 to 70 nm. Under these conditions the X-ray microscope provides 50-85 nm resolution and short exposure times due to the high efficiency of the KB-system. We will also discuss phase-contrast techniques, applicable with this microscope.

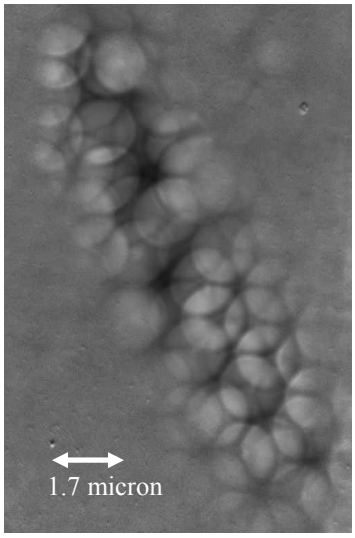

Figure 1: Image of hollow spheres in a Nickel structure, taken with the KB-FZP microscope

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Development of a soft X-ray microscope with Wolter mirrors for the observation of biological specimens in the atmospheric state

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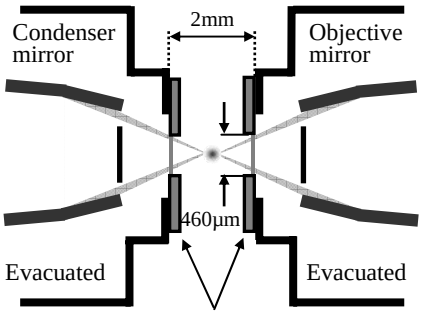
Soft X-ray microscopes using water window X-rays are powerful tools for the observation of biological specimens in the natural state. Most of them are constructed at the synchrotron radiation facilities with zone plates and approximately 20nm resolution is achieved[1]. In the laboratory, a laser plasma soft X-ray microscope with Wolter mirrors has been developed as one method of these microscopic techniques. Relatively narrow band spectrum around 3.2nm soft X-rays is obtained using a tantalum target and a titanium filter. In our previous microscope, specimens were put in a vacuum to avoid the absorption of soft X-rays by the air[2]. However, this method was very laborious because the sample preparation was difficult to preserve the natural state of the specimens in a vacuum. So, a new environmental sample chamber was developed so that biological specimens can be set in the atmospheric state. The schematic diagram around a specimen is shown in Fig.1. A silicon nitride membrane (460×460μm, 100nm thickness) on the silicon substrate was used as a transparent window between a vacuum chamber and the atmosphere. As the window is also transparent for visible rays, the alignment of a Wolter mirror is possible with visible rays before using soft X-rays. The atmospheric layer is approximately 2mm, so the working distance of a sample is 1.2mm. Therefore, the application to various sample holders can be expected. The transmission of 3.2nm soft X-rays of a silicon nitride membrane and the atmospheric layer are 67% and 60%, respectively. So, the net transmission except a specimen is 25% compared with a previous microscope. The resolution of this microscope is estimated to be better than 100nm and biological specimens are observed using a renewed microscope. The X-ray micrograph of a diatom and red blood cells of a cow are shown in Fig.2 (a) and (b), respectively. A photographic plate was used as a high resolution detector. In the X-ray image of a diatom, approximately 100nm fine structures are observed with relatively high contrast. In the case of red blood cells, each cell and crystal like structures are clearly observed. Exposure was less than 10seconds.



SiN membrane on the Si substrate

Fig.1 The schematic diagram of a new sample chamber

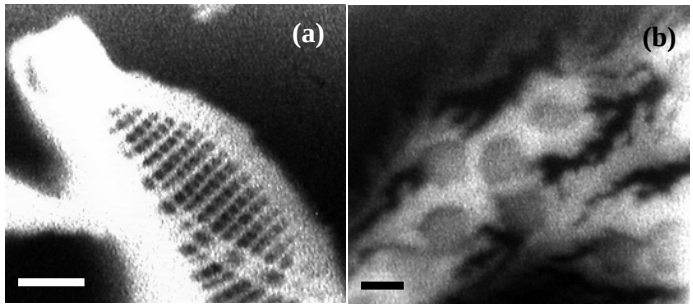


Fig.2 X-ray micrographs of biological specimens: bar 2μm
(a) A diatom (b) red blood cells of a cow

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Table-Top X-ray Microscopy

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Abstract

X-ray microscopy in the water-window region (λ = 2.3-4.4 nm) is an attractive technique for high-resolution imaging. In this wavelength region state-of-the-art optics has demonstrated sub-20 nm resolution and the sample preparation techniques are maturing. Unfortunately present operational x-ray microscopes are based on synchrotron radiation sources, which limit their accessibility. Many biological investigators would benefit from having the x-ray microscope as a tool among other tools in their own laboratory. For this purpose we recently demonstrated the first compact x-ray microscope with sub-visible resolution.[1]

In this presentation we will describe a recently developed, flexible, compact x-ray microscope featuring operation at λ = 3.37 nm and λ = 2.48 nm. The microscope is based on a 100 Hz liquid-jet-target laser-plasma x-ray source, condenser optics, diffractive zone plate optics and CCD detection. The microscope can be operated at λ = 3.37 nm with a methanol-liquid-jet laser plasma[2] and a normal-incidence multilayer condenser[4]. With this arrangement we have resolved 40 nm L/S Ni gratings with good contrast at exposure times of ~1 min, a significant improvement compared to previous compact microscopes. This operation mode provides good contrast for thin carbon-containing objects. However, imaging of thick biological objects require operation in the lower part of the water window. Here we employ a liquid-nitrogen-jet laser plasma (λ = 2.48 nm)[3]. Since present normal-incidence multilayer mirrors neither have the required reflectivity nor the necessary uniformity at this wavelength instead a zone plate condenser[5] will be used. The sample holder will initially be positioned in a helium atmosphere with silicon nitride membranes separating it from the vacuum in the condenser and imaging module. The holder enables both dry and wet sample handling with simple sample replacement. Future systems will include cryo sample stages.

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Hard X-Ray Nanoprobe with Refractive X-Ray Lenses

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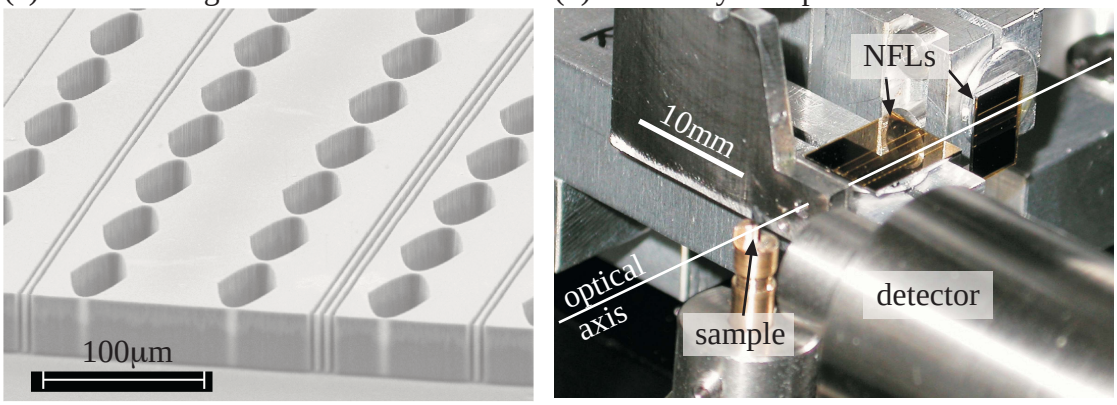
At synchrotron radiation sources, parabolic refractive x-ray lenses allow one to built both full field and scanning microscopes in the hard x-ray range. For the latter microscope, a small and intensive microbeam is required. Parabolic refractive x-ray lenses with a focal distance in the centimeter range, so-called nanofocusing lenses (NFLs), can generate hard x-ray nanobeams in the range of 100nm and below, even at short distances, i. e., 40 to 70m from the source [1]. Recently, a $50 \times 50\mathrm{nm}^2$ beam with $1.6 \cdot 10^8 \mathrm{ph/s}$ at $21\mathrm{keV}$ (monochromatic, Si 111) was generated using silicon NFLs in crossed geometry (cf. Figure) at a distance of 47m from an undulator source (ID13) at the European Synchrotron Radiation Facility. This beam is not diffraction limited, and smaller beams may become available in the future. Lenses made of more transparent materials, such as boron or diamond, could yield an increase in flux of one order of magnitude and have a larger numerical aperture. The fundamental limit for focusing with refractive lenses lies below 5nm [2].

(a) nanofocusing refractive lenses (b) hard x-ray nanoprobe



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Practical use of quasi-kinoform zone plate : Towards high-efficiency microbeam for hard /high-energy x-rays

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The sputtered-sliced zone plates (ss-FZPs) with various aspect ratios (thickness) are possible to use in wide x-ray energy ranges (8 ~ over100 keV). For more practical use of ss-FZP, the higher focusing efficiency is indispensable as well as the high spatial resolution. We have planned to develop a kinoform type zone plate (K-ZP). The ideal focusing efficiency of the K-ZP is 100% assuming that there is no absorption. As a first step we have tried to fabricate a quasi-kinoform zone plate (QK-ZP) using the sputtered-sliced method to obtain high-efficiency. The QK-ZP was fabricated by depositing four kind layers of different Al/Cu compositions in one cycle, a transparent layer (Al), two half transparent layers (Al:66% and Cu:33%; Al:33% and Cu:66%) and an opaque layer (Cu), thus, 60 layers in whole zone area on a rotating fine gold wire core having a diameter of 50 micron. The zone plate thickness and the film thickness of the outermost zone were ~55 micron and 0.145 micron respectively.

The focusing experiment was performed at the end station of the 250m-long beamline (BL-20XU) of SPring-8. At first, the experiment was made at 30 keV without OSA (order sorting aperture) to observe the whole diffraction order lights from the QK-ZP in the CCD detector located downstream of the focusing point. We, then, compared the intensities of the 1st and the -1st order diffraction light. It was found that the intensity of the 1st order light was apparently increased while that of the -1st order was almost disappeared. The ZP was, then, proven to be worked as a kinoform-like ZP. The focusing efficiencies of the 1st order focus were ~50% and ~10% at 50keV and 23keV respectively, while that of the 2nd order was determined to be the maximum value of 21% at 23keV. We, then, made a scanning x-ray microscopy experiment using the 2nd order beam without using OSA at 23keV. The transmission image of Ta test pattern (thickness: 0.5 micron) was obtained. The fine pattern up to 0.5 micron was resolved.

Multilayer Laue lens for hard x-ray nano-focusing optics

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The Multilayer Laue Lens (MLL) is a new concept for hard x-ray nano-focusing optics, technically challenging to fabricate, but having theoretically promising performance [1,2]. A MLL is a diffractive x-ray focusing optic operated in transmission Laue geometry, and can be fabricated by depositing a thick graded-spacing multilayer on a flat substrate and then sectioning it to produce a high-aspect-ratio optical structure. It differs from a "sputtered-sliced" zone plate [3] in that the thinnest layers can be deposited first and the two halves of the structure can be tilted to the optimum diffraction angle for high efficiency. We have successfully fabricated MLL structures by employing deposition and sectioning techniques. The methods used to fabricate the MLL structures will be presented [4], as well as the measured x-ray diffraction and focusing properties of these MLL structures. X-rays at an energy of 19.5 keV were focused to <60 nm FWHM in a one-dimensional line focus. This is close to the expected theoretical focus size, indicating that the MLL is a promising candidate for future hard x-ray nano-focusing optics.

This work is supported by the U. S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. W-31-109-ENG-38.

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Hard X-ray diffraction-limited nanofocusing with unprecedentedly accurate mirrors

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Recently, at grand-scale synchrotron radiation facilities, modern X-ray microscopy with highly brilliant and coherent X-rays have been used to make detailed investigations on material characteristics, such as element distributions and chemical bonding conditions, of a variety of samples, including biological and medical samples. Nanofocused X-ray beams are necessary for nanometer-scale spatial microscopy analysis. X-ray focusing using a Kirkpatrick-Baez setup with two total reflection mirrors is a promising method, allowing highly efficient and energy-tuneable focusing.

In this presentation, we will report the development of ultraprecise mirror optics and the realization of a nanofocused hard-X-ray beam. The designed elliptically curved shapes were fabricated by a computer-controlled figuring system using plasma chemical vaporization machining (PCVM)[1] and elastic emission machining (EEM)[1], on the basis of surface profiles accurately measured by combining microstitching interferometry (MSI)[2] with relative angle determinable stitching interferometry (RADSI)[3]. Platinum-coated surfaces were employed for hard X-ray focusing with a large numerical aperture (NA). Focusing tests were carried out at the 1-km-long beamline (BL29XUL) of SPring-8. Fabricated mirrors having a figure accuracy of 2 nm peak to valley height give ideal diffraction-limited focusing at the hard X-ray region. The focal size, defined as the full width at half maximum in the intensity profile, was 36 nm × 48 nm at an X-ray energy of 15 keV[4]. We anticipate that nanofocused X-ray beams will bring about a marked improvement from the micrometer to nanometer range in the spatial resolution of all X-ray microscopy techniques.

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Status and recent developments of microfocusing optics in IMT RAS

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In this report a summary is given giving light on recent achievements of X-ray optic with submicron resolution in IMT RAS. Main topics of microfocusing optics that have get a significant evolution, namely phase zone plates, refractive X-ray planar lenses, X-ray waveguides are reviewed. A majority of developed devices is based as firstly proposed Bragg-Fresnel zone plates on silicon and associated high aspect ratio deep etching techniques.

Planar refractive lenses formed from silicon and mentioned above etching technique become well suited for synchrotron radiation sources. Real planar lenses suffer from undercut of refracting profile edges. Now this bottleneck is overcome by computer simulation methods giving feasibility to calculate necessary precorrections. Demands for profile precision have been stated allow reaching perfect parabolic profile.

Minimized absorption and kinoform lenses have been developed firstly by this planar approach. At last time they are reproduced in some works in different versions verifying this direction in refractive optic. Nickel kinoform lenses for hard X-ray 212 keV focusing may be mentioned as a most exciting example.

Recent experimental results of lenses and zone plates testing on ESRF (Grenoble, France), Spring8 (Japan) and Kurchatov synchrotron source (Moscow, Russia) are overviewed. Step from single lens toward many lens systems is illustrated for planar refractive lenses and phase zone plates. Combined focusing system consisted from X-ray planar refractive lens and planar X-ray waveguide is considered.

An overview of applications of developed microfocusing devices is given including next topics:

- Achievements of planar silicon lenses as refractive collimators;
- Holographic imaging using phase zone plates;
- Microfluorescence tomography.

The main problems and goals of X-ray microfocusing optics are considered in connection with future applications for nanotechnologies and life science.

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High-brightness liquid-metal-jet-anode electron-impact hard x-ray source

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Abstract
Hard x-ray (multi keV) synchrotron sources with high brightness have enabled novel and important imaging techniques, from x-ray microscopy to medical imaging with improved and new contrast. Unfortunately the brightness of conventional compact hard x-ray sources has not improved significantly since the invention of the rotating anode, making it difficult to envision compact systems for these new imaging modalities based on present sources. In this paper we describe a novel compact electron-impact hard x-ray source with potential for much higher brightness than present state-of-the-art rotating-anode sources. The source can enable improved resolution and contrast in medical imaging and non-destructive evaluation, and has applicability for compact hard x-ray microscopy and certain diffraction applications.

For electron-impact sources the brightness is proportional to the electron-beam power density at the anode. Present rotating-anode and micro-focus technology show little potential for further improvement due to intrinsic thermal limitations. We have introduced a new anode concept, the liquid-metal jet. Calculations show that this new anode allows a 100-1000× increase in source brightness compared to today's compact hard x-ray sources[1,2]. We presently operate it with a 20-50-µm diameter liquid-tin jet at ~70 m/s with a 50 kV, 600 W electron beam focused to 40-50 µm FWHM. The resulting electron-beam power density is 200-400 kW/mm^2, which is approx 10× higher than on present sources. The emitted spectrum exhibits two tin emission peaks around 25 keV and a broad bremsstrahlung background. Conventional absorption imaging shows excellent spatial resolution (tens of microns). Furthermore, the small source allows phase imaging for improved contrast. Initial results compare well with theoretical modelling.

This paper will describe the source system properties as well as show early high-resolution imaging results, with and without phase effects. We will also discuss the source's extension to lower (10 keV) and higher (>50 keV) energies. Finally, we will elaborate on the new source's applicability for hard x-ray microscopy and medical imaging.

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Development of high-average power extreme ultra violet source by laser produced plasmas

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Extreme ultraviolet (EUV) radiation from laser-produced plasma has recently attracted particular attention for use in production of the next generation semiconductor devices of node-size below 40 nm. It is expected to generate over 115 W at 13.5 nm within 2% bandwidth at the repetition rate of 7-10 kHz. Since 2003 a new project of MEXT has started to provide experimental and theoretical databases for clean and efficient EUV generation, and technical guidelines to build-up an EUV-source system for practical use in industry. The main tasks of the project are

(1) **Acquirement of databases**: Comprehensive experimental databases are provided for a wide range of parameters of lasers and targets. These experimental data will be utilized to benchmark a radiation hydrodynamic code including equation-of-state solvers and advanced atomic kinetic models dedicated for EUV plasma prediction.

(2) **Target design and fabrication**: Various types of targets have been proposed such as gas, liquid, low-density solid or clusters. In addition, innovative targets are desired in order to attain acceptably high conversion efficiency and mitigate debris from targets.

(3) **Establishment of laser technology**: A 5-kW laser at a repletion rate of 5-kHz system is ready to demonstrate efficient EUV source generation under optimized plasma conditions.

Present status of the laser produced EUV source development and future prospects for various applications will be discussed.

Hard X-ray Microscopy at the Advanced Photon Source

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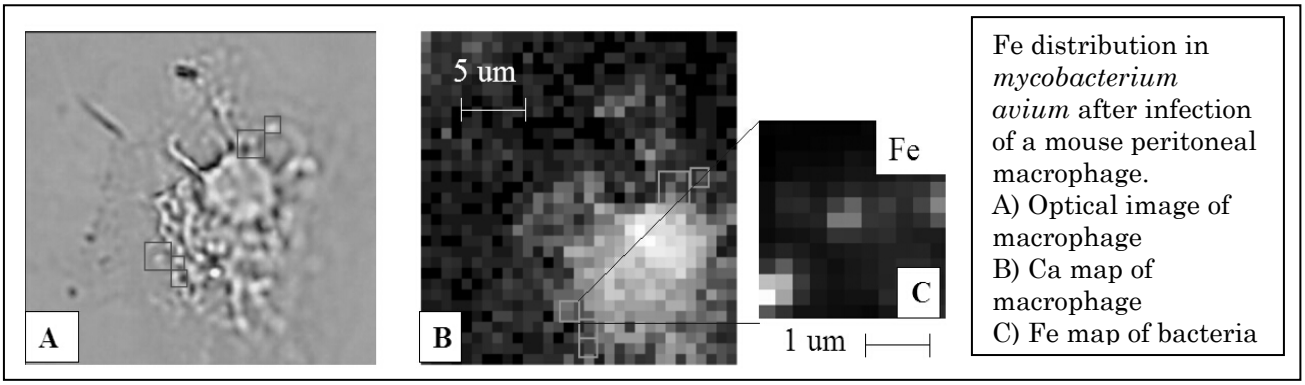
X-ray microscopy at the Advanced Photon Source has, since its beginning in 1997, developed into a powerful, versatile tool that has been implemented at various APS beamlines and is being applied to a large variety of scientific topics. Due to its high brilliance in the hard x-ray range, the Advanced Photon Source lends itself in particular to scanning probe applications that utilize x-ray fluorescence and x-ray diffraction as contrast forming mechanisms. Hard x-ray microprobes using both Fresnel zone plates and Kirckpatrick-Baez mirrors are in operation, with a spatial resolution typically in the range of 100-300 nm. With the development of x-ray optics of higher numerical aperture and concurrent improvements of the microscopy facilities, a spatial resolution of below 100 nm has been obtained more recently.

Microprobes using x-ray fluorescence are being applied in particular to biological, medical and environmental sciences, where they are used to map the distribution of trace metals in tissues, cells and bacteria.[1,2,3] Microprobes using x-ray diffraction are typically used in materials sciences, both for direct study of the crystalline structure of materials,[4] and to map strain, e.g. in systems showing ferroelectric and magnetic order.[5,6,7] An emerging application is the quest for novel materials and their applications, namely the study of nanoparticles and their interaction with their environment.[8] We will present an overview over current x-ray microscopy activities at the Advanced Photon Source, and provide an outlook for future plans. We will focus in particular or the hard x-ray nanoprobe beamline that is under construction as part of Argonne's Center for Nanoscale Materials, and aimed to provide a spatial resolution of 30 nm in the hard x-ray range.



Fe distribution in *mycobacterium avium* after infection of a mouse peritoneal macrophage.
A) Optical image of macrophage
B) Ca map of macrophage
C) Fe map of bacteria

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Recent Developments on the X-ray Phase Contrast Imaging and CT in BSRF

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Abstract

Since 2001 hard x-ray phase contrast imaging and computer tomography (CT) methods have been developed at the Beijing synchrotron radiation facility (BSRF). The most recent advancements of these imaging methods are reported below:

1. A model discussing the mechanism of imaging of cylinder-like structures in inline phase contrast imaging has been proposed. Cylinder-like structures are present in many biological and medical samples, and the correlation among contrast, radius, refraction index difference, distance between sample and detector and spatial coherence of the source have been included in this model.
2. The main parameters of the diffraction enhanced imaging (DEI) equation were investigated, the influence of small angle scattering accepted by the analyzer crystal on imaging is discussed and the term to describe the influence was introduced into DEI equation, some results were given in this contribution.
3. The condition for phase contrast CT has been carefully investigated. In particular we demonstrated that the projection of the function to be reconstructed can not be described as a simple line integral along the x-ray path, and in addition this function has to be rotational invariant. An example of the refraction index gradient of CT is discussed in this manuscript.

The new instrumentation is operative at BSRF for users for both phase contrast imaging and CT experiments. Recent experimental results will be presented and discussed.

Volume zone plate development at BESSY

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State-of-the-art Fresnel zone plates can be described by scalar diffraction theory neglecting the three-dimensional shape of the zone structures. According to this theory their diffraction efficiency scales as $1/m^2$ where m is the diffraction order. While keeping the zone height constant, the aspect ratio of the zones increases inversely with decreasing outermost zone width. For photon energies below one keV, it is shown by applying electrodynamic theory that scalar theory is no longer suited to describe zone plates with outermost zone width below 20 nm and aspect ratios of about 10:1 [1,2].
Full electrodynamic theory - which includes forward and backward diffracted as well as evanescent waves - predicts that the diffraction efficiency decreases continuously if the lateral dimensions of the zone width approach the wavelength used for imaging. This result is obtained for zone structures parallel to the optical axis. Unlike the diffraction properties of parallel zone structures, rigorous coupled wave theory (RCWT) predicts for zone structures tilted to the optical axis according to the local Bragg condition that the diffraction efficiency can be up to 50 % [3]. In addition, RCTW calculations show that similar diffraction efficiency values can be obtained in any high order of diffraction $m > 1$.
The resolving power of zone plates scales with the order of diffraction m. By applying high orders of diffraction, it is possible to increase the resolution without the need for manufacturing increasingly smaller outermost zone width far below 20 nm. Applying high orders for imaging requires manufacturing tilted zone structures with aspect ratios of about 20:1 [2]. To overcome the extremely difficult problem of manufacturing tilted zones with high aspect ratios of 20:1, we propose to manufacture zone plates on top of each other with slightly decreasing zone radii [3]. In good approximation – depending only on the number of layers – the zones can be tilted according to the local Bragg condition and each single layer requires only moderate aspect ratio structures (see Fig. 1C). However, the overlay accuracy for e-beam writing is in the nanometer range. Theoretical results on the dependency of the number of layers and their required overlay accuracy will be presented. We will also present the current status of the zone plate development at BESSY.

Fig. 1: A) Zone plate with zones parallel to the optical axis, B) Zone plate with tilted zones, C) Zone plate where the local tilt angle of the zones is approximated by 5 layers.

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Hard X-ray Microscopy with sub-30 nm Spatial Resolution in Taiwan

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With the advances in synchrotron radiation and the mature in fabricating high performance X-ray focusing optics, X-ray microscopy has been realized recently as a nano-scale probe that can compete with light and electron microscope on the common ground of a nondestructive manner. In 2004, under the *NSRRC X-ray Microscopy Project*, we have installed a full-field transmission X-ray microscope (TXM) to the BL01B end station of an advanced X-ray source generated by a superconducting wavelength shifter. The X-ray microscope equipped with capillary-type condensers and objective Fresnel zone-plates (outermost width 50nm) can provide 2D imaging and 3D tomography at X-ray energy 8-11 keV with spatial resolution 60 nm, and with the Zernike-phase contrast capability for imaging light materials such as biological specimens. While operating the Fresnel zone-plate in third order diffraction, the microscope has almost reached the theoretical resolution limit sub-30nm (Fig.1). In this presentation, we would demonstrate the actual setup of the beamline and microscope. Commissioning results, including the characterization of the microscope, preliminary studies in IC failure, fault rocks, and biological specimens will be presented.

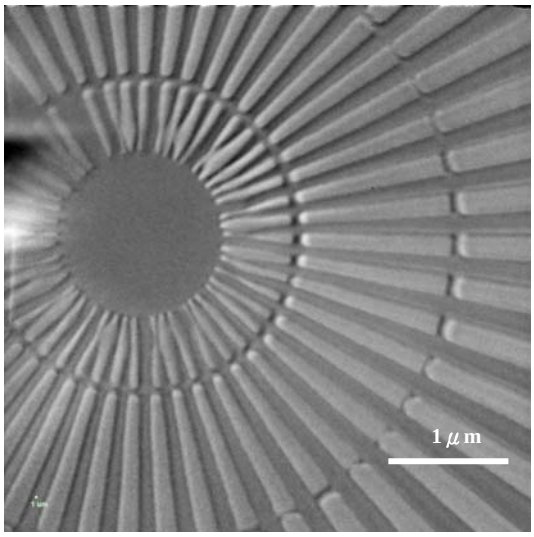

Figure 1: The gold spoke-pattern imaged at BL01B at NSRRC by using 3rd order diffraction of Fresnel zone plate at X-ray energy 8 keV. The spatial resolution is estimated better than 30nm. Exposure time was 10 minutes.

PORTABLE SYNCHROTRON HARD X-RAY SOURCE "MIRRORCLE-6X" FOR X-RAY IMAGINGS

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MIRRORCLE-6X is a portable synchrotron composed of a 6-MeV microtron injector, and a 60cm outer diameter exactly circular synchrotron ring made of a normal conducting magnet. The injection is performed at 400 Hz repetitions by 100 mA injector peaks current that lead to 3A initially accumulated current. X-rays are generated by a collision of the relativistic electron beam and a small target placed inside the circulating beam. The generated X-ray energy is dominated at around 30-300keV, and the total flux in the 0.1% bandwidth and ±85-mrad spreads is 1000 times higher than a conventional synchrotron light source. A few μm wide target, which defines the X-ray emitter size, produces extremely fine resolution edge enhanced images. Due to the small emitter size the brilliance of MIRRORCLE reaches 10^{16} photons/[s, mrad^2, mm^2, 0.1% band width] at any point within ±85-mrad. The image field can be more than 30 cm wide at 2 m distances from the source point. This machine provides highest quality non-destructive testing of heavy constructions. Due to the phase contrast effect this machine also enables imaging of soft tissues. When the imaging device is set at the distance from the sample we can take magnified images. We have already obtained 10 times magnified fine resolution images, and are challenging to 100 times magnification that forms a novel X-ray microscope without optical elements. MIRRORCLE-6X opens up new frontiers of X-ray imaging in medical, biological, commercial and industrial uses.

The observed X-ray beam quality, brilliance, coherence, and challenge to the sub-micron size target will be discussed.

A fast-readout CCD system for configured-detector imaging in STXM

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The use of a transmitted x-ray detector in scanning transmission x-ray microscopy that has a software configurable response function makes possible the use of a number of simultaneous imaging modes, allowing both absorption and phase contrast to be derived from a single scan of the specimen [1].

This paper describes a practical system based around an electron-multiplying CCD system (iXon DV860 from Andor Technology) that combines fast frame-transfer readout with very high sensitivity. The detector consists of a Peltier-cooled CCD array of 128 by 128 sensors, and makes use of visible light coupling to a phosphor screen, to make it easy to operate over a wide range of photon energies, from the oxygen K edge upwards. It has been tested successfully on the Twinmic end-station at the Elettra synchrotron.

In essence, such a detector system records, for every pixel in the STXM raster, a map of the intensity distribution in and around the brightfield cone of illumination produced by the focusing optic in the STXM, resulting in a large volume of data from a single raster scan.. Simple real-time processing of these data yields absorption and differential phase contrast image signals, but more elaborate processing can subsequently be applied to the full 3-D dataset generated by the STXM scan. In combination with through-focal series, and the acquisition of image data on either side of an x-ray absorption edge, a configured detector system can provide a wealth of information about the complex transmittance of the sample.

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QUANTITATIVE X-RAY PHASE-CONTRAST MICROSCOPY AND MICROTOMOGRAPHY USING AN SEM

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The projection method for X-ray microscopy is very simple in principle and enables X-ray imaging at sub-optical resolution without the need for any focusing X-ray optics. The use of an SEM to provide an electron beam focussed on a target as a source of X-rays for imaging was described as early as 1945[1]. In this and subsequent works, the mechanism for contrast formation and interpretation was based on absorption.

Our approach to projection X-ray microscopy is to take advantage of the phase distortions imposed on an X-ray wavefront passing through an object, which, in combination with Fresnel diffraction leads to phase contrast in the resulting X-ray image.[2,3] We have developed an X-Ray ultra-microscope (XuM) based on an SEM as host (in the present case an *FEI XL-30 S-FEG SEM*) which is designed to take advantage of both phase and absorption contrast and has the capability for X-ray imaging at around 50 nm line pair resolution with X-rays from 0.8-15keV in energy. This system uses a scientific grade CCD in direct mode to give high signal to noise and optionally also an energy resolving capability for the transmitted beam. Images acquired using this system can be post-processed using combined phase-retrieval and deconvolution algorithms to produce very high resolution quantitative information about the sample[4-6].

This technique has particular strengths which are applicable to a wide range of microscopy- and microtomography-based studies of materials and devices. A number of examples of microtomography studies using the XuM will be presented including studies of manufactured devices, nanocomposites based on polymers, corrosion effects and biological samples.

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Contribution of X-ray Microscopy to Bone Mineral Studies

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Bone, which is formed by the infusion of an organic matrix, principally collagen, with calcium phosphate, performs two major functions in the body. This combination of about 20wt% collagen and 80wt% calcium phosphate provides the biomechanical properties needed for body support and movement. In addition, bone mineral is in metabolic interrelation with body fluids, serving principally as a reservoir for body minerals, storing or releasing them as the need arise, changing in size, distortion and chemical perfection with age, disease and chemical/medical treatment [1-2]. Trace elements have been shown to have a profound influence on the chemistry and solubility of bone as this latter is the accumulation "target" organ for many heavy metals. Consequently, many human diseases and pathological conditions result in changes in bone tissue that affect the rate of bone turnover and its physico-chemical properties. Bone mineral presents then a complex composition, and appears as a very sensitive, reactive and sophisticated material.

Several techniques may be used for the study of bone mineral but often allow a global measurement only without spatial information. However the study of the mineral content of bone at a microscopic scale is of particular interest, since it can give new insights into remodelling activities, mineralization processes, effect of drugs and related mechanical properties.

We report here applications of X-ray microprobe techniques to the analysis of bone and biomaterials (analogous to bone mineral and implants). In particular, we will present the studies carried out on bone mineralization and maturation processes using X-ray micro-fluorescence, micro-XANES and micro-diffraction and micro-infrared [3-4]. Secondly, examples of applications to more specific topics will be shown, namely the study of bone mineral in osteogenesis imperfecta pathology [5], the effect of Sr based drugs against osteoporosis and the integration of titanium implants in bone [6]. Last, recent developments in x-ray micro-tomography allowing in-situ dynamic follow up of the formation and propagation of micro cracks in bone under strain will be presented [7].

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A deep look into polycrystals: X-ray diffraction contrast tomography

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We present here two possible extensions of X-ray microtomography, capable to characterize the shape and the orientation of individual grains in the bulk of *undeformed* polycrystalline materials: (i) diffraction contrast tomography in "topo-tomography" alignment [1] and (ii) diffraction contrast tomography in conventional alignment. In both cases, Bragg diffraction (transmission case) gives rise to an additional contribution to the local attenuation coefficient, which in turn can be exploited by means of analytic or algebraic tomographic reconstruction techniques in order to reconstruct the three dimensional grain outlines. The second approach offers in addition the possibility to determine grain orientations.

The imaging principle and related data analysis strategies will be illustrated for both methods with the help of first experimental data, obtained from a coarse grained Al (1050) multicrystal. A comparison of diffraction contrast tomography with respect to the more widely applicable 3DXRD approach [2] will be given. Based on this, we will discuss possible future developments and applications for combined tomographic imaging and diffraction experiments.

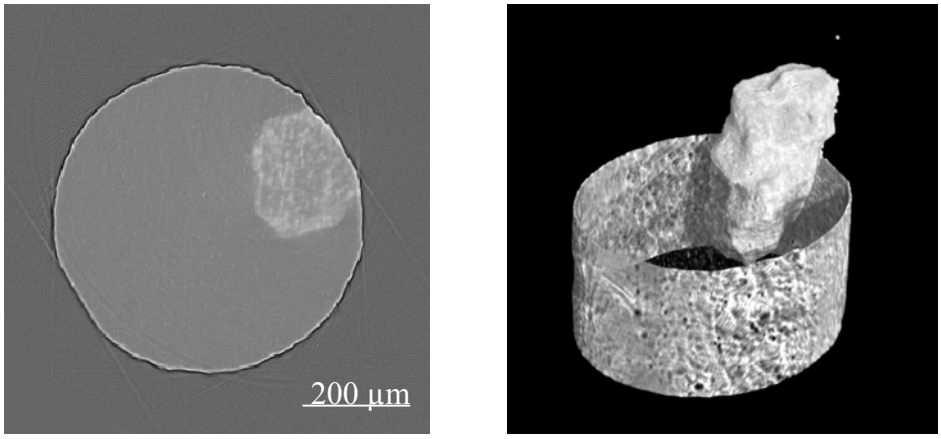

Fig.1: Diffraction contrast tomography (topo-tomo alignment) of an undeformed, coarse grained Al (1050) multicrystal. (a) tomographic slice showing the outline of the cylindrical sample as well as the shape of the diffracting grain with some substructure (inclusions). (b) 3D rendition of the diffracting grain, the Al matrix has been set to transparent.

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Observation of Anisotropic Disorientation Grain Boundaries in Sn_2O_3 Nanobelts Using X-Ray Nanodiffraction*

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Molecules in a thermodynamic system have a spontaneous tendency to seek maximum interactions among themselves for lowering the free energy of the system via variation of density distribution. It is well known that the intensive parameters like temperature and pressure often limit the extent of the interactions and are responsible for the richness of phases that a system exhibits. However, the exploration of the extent of interactions due to the extensive parameters, such as the volume and the number of molecules in a system, and the effect on the density distribution is still in its infancy, due to the limited availability of such systems. A wire-like nanostructure synthesized through one-dimensional growth exhibits the effect due to its condensed phase, which enables molecules to significantly interact, while the small cross-section size limits the extent of the interactions. For instance, the energy needed to form a point defect at the growth front of a nanowire is much less than the energy needed to form the same defect in the bulk, since the lattice elastic deformation and associated strain field quickly die out when the boundary of the nanomaterial is reached. Once the energy can no longer counter the energy due to the change of the entropy associated with the formation of the point defect at the growth temperature, disorder starts to become the favorable configuration. This imposes a fundamental limitation on the smallest cross-section size of a perfect nanowire crystal that can be synthesized through one-dimensional growth. In order to observe the limitation, the internal structures of tin oxide (Sn_2O_3) nanobelts of various cross-section sizes, from 200 nm x 40 nm down to 22 nm x 8 nm, were individually investigated using x-ray diffraction with a focused beam (7 keV) obtained from advanced zone-plate optics. By mapping the diffraction intensity of the (030) reflection with a CCD area detector along the full length of the nanobelts, we observed disorientation boundaries in the belts of cross-section sizes of 100 nm x 30 nm and smaller. The lattice disorientation of grains inversely relates to the cross-section size of the belts. Moreover, the lattice disorientation boundaries are confined only in the plane of growth front and none of that out of the plane has been observed. Given the fact that a tin oxide nanobelt is bounded by faces of high density and low surface energy, all the observations obtained from the tin oxide nanobelts can be explained consistently with the limited extent of interactions among molecules at the growth front.

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3D Internal Strain Mapping by Tracking Microstructural Features in Tomographic Volumes of Structural Materials

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Synchrotron X-ray microtomography has been utilized for the 3D characterisation of microstructures in several aluminium alloys. Tomographs, consisting of isotropic voxels with a maximum of 0.474 μm edge, were collected mainly at the X-ray imaging beamlines BL20B2 and BL47XU of the SPring-8. A combination of the high-resolution deflection contrast imaging technique and several state-of–the-art application techniques have enabled the quantitative image analyses of internal microstructure, such as micro-pore, intermetallic compound particles and grain boundary as well as the assessment of their effects on deformation and fracture behaviours of the aluminium alloys. The application techniques include liquid metal wetting which enhances nanoscopic microstructural features in term of the absorption contrast, microstructural tracking which enables large scale strain mapping, in-situ observation technique using a material test rig specially designed by the present authors and local area observation for samples larger than available fields of view. 3D finite-element meshes were also generated from the tomographic volumes to monitor local stress and strain distributions, then being used to verify the image analyses.

In terms of the microstructural tracking, in order to evaluate microstructural effects quantitatively, the tomographic dataset was thresholded and labelled utilizing a grey value for each 3D feature of interest. Volume, surface area and centre of gravity of each feature were automatically measured at sub-voxel accuracy using software developed by the present authors. Centre of gravity of each microstructural feature was then utilized as a displacement gauge marker to calculate three-dimensional mechanical parameters of the underlying aluminium, such as strain, stress and crack driving force. Variations during loading in the centroid spacings of the pairs of neighbouring microstructural features may then be separated into three directions by orthogonal decomposition.

The tracking technique has provided a highly effective way of assessing microstructure/property relationships in the structural materials, together with supplementary ways of verifying and interpreting them by visualising and quantifying various mechanical behaviours. The proposed technique has been clearly advantageous compared to the very limited procedures for such measurements available in the current literature, where detailed internal information can only be accessible for limited types of material, such as transparent materials.

Ancient cosmetics and painting analysed by combination of complementary microanalysis techniques

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Our research is focused on two different archaeological issues: the study of the **cosmetics** used in the Mediterranean geographic area along a period of time (going from the Egyptian antiquity to the Greco-Roman time). Cosmetics were of primary importance in the everyday life, not only in aesthetic context, but also in religious, ritual and medical practises. Second, the study of the pigments used in Northern European **paintings** at the beginning of the Renaissance (Cranach, Dürer, Grünewald, Holbein, etc.). The physical-chemical signatures of the pigments, grounds and binding media are necessarily related with relevant practises and art expertise in Europe at that time.

Several similarities exist between these two subjects. First, the chemical nature is quite the same: products are made of pigments, with the possible addition of a binding medium (e.g. oil). Second, the analytical problematics are common: we search to determine the chemical composition of the products (are they natural or artificial compounds? Where they come from?) and the manufacturing process (chemical/mechanical transformation, heating…). More generally, our aim is to identify some characteristic "paint-pot", ingredients and practices used in a limited time and space. Besides, in addition to supply information on ancient know-how, knowledge about degradation process can be useful for conservators.

Due to the samples complexity and preciousness, a network of various micro-analytical techniques was employed. Experiments were performed at the ESRF, on the beam lines ID21, ID22 and ID18F, which provide a panel of micro-imaging techniques, with high spatial resolution and high sensitivity. **Micro X-ray fluorescence** was used to identify the trace elements, which enable sometimes to distinguish between natural ores and synthesised products and to determine geographic provenances of minerals. **Micro X-ray diffraction** is a relevant method to analyse crystallised compounds and identify phases. **Micro XANES** analyses were performed at the sulphur and manganese K-edge to identify the chemical composition of some cosmetics and paintings. **Infrared micro-spectroscopy** gives access simultaneously to organic and mineral phases. It was particularly useful to study the interaction products between oil and pigments. More generally, each technique can generate images. This fact is very important since it enables elemental and chemical co-localisations, hence an easier identification of components. In addition, inter-technique correlations are also very fruitful. Finally, the combination of complementary and quantitative synchrotron based techniques is a powerful way to study complex ancient chemicals.

Biological Nano-Tomography

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Soft X-ray microscopy is an emerging new imaging technique that can examine whole, hydrated, biological specimens up to 10 microns thick with a spatial resolution up to 15 times higher than that obtained with light microscopy. In the energy range of X-rays used to examine cells, organic material absorbs approximately an order of magnitude more strongly than water. This produces a quantifiable natural contrast in fully hydrated cells and eliminates the need for chemical fixatives or contrast enhancement reagents to visualize cellular structures. We have used this imaging approach to reveal remarkable details of the nuclear and cytoplasmic architecture of fully hydrated whole cells. We have also localized molecules in the nucleus and cytoplasm of whole, hydrated cells using immunogold labeling protocols. Using cryo X-ray tomography of cells held in micro capillaries, we have obtained three-dimensional reconstructions of cells in their native state at better than 50 nm isotropic resolution. With X-ray imaging, the internal structures are not masked by ice and the resulting images are inherently of greater contrast. In addition the proteins, lipids and nucleic acids are detected by the amount of carbon and nitrogen they contain, generating quantifiable data based on their absorption coefficient. Three-dimensional tomographic reconstructions of the yeast, *Saccharomyces cerevisiae*, reveal high fidelity views of the internal architecture of these eukaryotic cells. Tomographic reconstructions of several bacteria reveal unprecedented details of their internal structural organization. All data obtained using this unique imaging approach are quantifiable by calculating the x-ray linear absorption coefficient. Using immunolabeling combined with tomography, we will be able to obtain 3-D information about the spatial distribution of proteins throughout the entire cell. Data collection is extremely fast, with a complete data set for tomographic reconstruction requiring less than 3 minutes. Consequently, X-ray tomography is an exciting new high-throughput approach for obtaining 3-D, quantifiable information from whole, hydrated cells.

Optimizing Organic Thin Films from Microspectroscopic Analysis

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High-resolution near-edge x-ray absorption fine structure (NEXAFS) spectroscopy is ideally suited to investigate the electronic properties of organic molecules. The development of new instrumentation at 3rd generation synchrotron light sources offers both, superior spectral quality in the soft x-ray regime as well as high lateral resolution for spectromicroscopy experiments. Both are without doubt required for detailed insight into the properties of ultrathin organic films (from the submonolayer regime to several 100 monolayers).

In-situ experiments with the presently installed SMART spectromicroscope at BESSY-II will be presented to discuss the influence of the adsorption kinetics on the growth of organic films and their effect on the film morphology. The electric moments (dipole or quadrupole moments) of the molecules may induce metastable (structural, orientational) phases, which can easily be followed during the in-situ observation in PEEM. The influence of steps is evident from the spatially resolved in-situ observation of the film growth. The molecular film grows best under conditions far from thermal equilibrium as will be demonstrated for PTCDA and NTCDA multilayer films adsorbed on Ag(111).

Whereas XPEEM easily allows in-situ detection of organic film growth, scanning transmission x-ray microspectroscopy (STXM) is limited to ex-situ prepared films due to spatial and vacuum restrictions. Deposition from wet cells has not yet been performed. STXM experiments were performed at the ALS concentrated on the morphology, crystallization and spectroscopic properties in ex-situ prepared organic films. The molecular substances were ranging from heteroaromates to magnetic supramolecules. The latter consist of a well-defined number (4 – 8) of paramagnetic ions (Ni, Mn, Co) which are stabilized by an organic ligand shell. Spatially resolved NEXAFS experiments focussed on the oriented growth of these molecules and on their electronic properties. The metal L-edges of the respective transition metals were found to be extremely sensitive to radiation damage. The damage leads to a reduction of the metal ions most likely from electrons excited in the ligand shell.

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Micro XANES Study on Vanadium in Living Blood Cells of Ascidians by Fluorescence Scanning X-Ray Microscopy at ESRF ID21 Beamline

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Blood cells of ascidians (tunicates) are known to accumulate vanadium selectively from sea water [1]. Several questions have been already addressed and discussed at length: how much is vanadium accumulated? which organ does participate in its accumulation?, and what is the route of accumulation process?. Michibata *et al*., by using combination of density gradient centrifugation and neutron activation analysis [2], showed that signet ring cells were assigned as true vanadocytes. However, it is not yet known where in the blood cells vanadium is accumulated, and what the accumulation mechanism is.

Micro-XANES is a powerful tool to investigate micro-distribution of chemical species of a given element. Fluorescence scanning X-ray microscope at the ESRF (ID21), covering an energy range from 2 to 7 keV, has the capability of investigating chemistry of vanadium, sulfur, magnesium and calcium. XANES spectra along with fluorescence imaging revealed the spatial distribution and chemical states of vanadium in blood cells derived from two vanadium rich ascidian species (*Phallusia mammillata* and *Ascidia sydneiensis samea).* In particular, used of micro-XRF imaging associated with high pressure cryo-fixation technique demonstrated that vanadium is distributed uniformly in the vacuole of a signet ring cell. Vanadium is selectively accumulated in the intravacuolar granule with a diameter of about 3 micron. Based on pre-edge XANES analysis, vanadium in vacuole and in granule was identified as V(III) and V(IV), respectively. The specific issue of radiation damages on bio-specimens will be also addressed.

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In situ STXM: Studies of Wet Electrochemical Systems under Potential Control

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Scanning transmission X-ray microscopy (STXM) has great potential for studies of *in situ* modification of samples, since the photon in / photon out character provides the capability to penetrate through complex structures such as electrodes, electrolytic solutions, and support layers. Here we report the first *in situ* measurements with scanning transmission X-ray microscopy (STXM) of an active electrochemical cell. The system used to develop *in situ* electrochemical STXM is an electrochromic layer of polyaniline The electrochemical STXM wet cell (Fig. 1) consists of an electrodeposited polyaniline (PANI) thin film on a thin Au film covered by an overlayer of 1 M HCl solution sitting between two X-ray transparent silicon nitride windows. X-ray absorption images and spectra of the PANI film under potential control were acquired using the beamline 5.3.2 STXM at the Advanced Light Source. The chemical state of the polyaniline film was reversibly converted between reduced (leucoemeraldine) and oxidized (emeraldine chloride) states by changing the applied potential. The electrochemical changes were monitored by spatially resolved C 1s and N 1s X-ray absorption spectroscopy (Fig. 2). Electrochemical state imaging (differences between images at two energies at different potentials) monitors the electrochemical changes of the polyaniline film. Kinematic studies in the sub second regime are demonstrated (Fig. 2). Extension of *in situ* electrochemical STXM to other electrochromic systems, and to situations such as displays where transverse fields are required, will be discussed.

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2) Research supported by NSERC, the Canada Research Chair program. ALS is supported by DoE (DE-AC03-76SF00098).

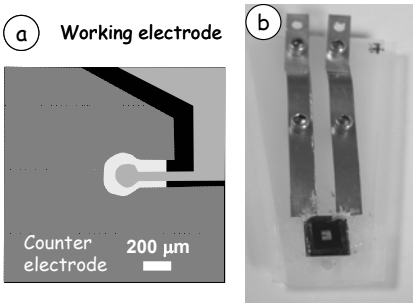

Fig 1 (a) Details of the 2-electrode cell used for STXM with *in situ* potential control. The chemical state of polyaniline electro-deposited on the working electrode is varied by potentials applied via the spring contacts (b)

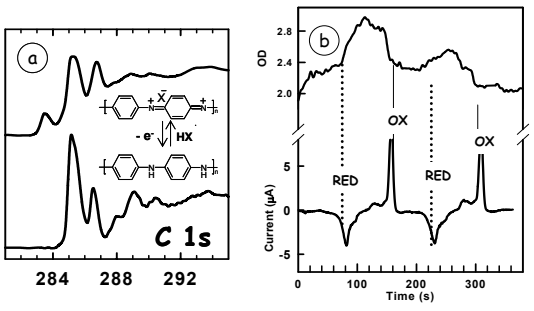

Fig. 2 (a) C 1s spectra of the oxidized (upper) and reduced (lower) states of polyaniline. (b) (upper) Optical density changes at 283.6 eV (quinoid pi*) tracked in concert with cyclic voltammetry (CV). (lower) current flows in CV.

Diffraction Microscopy of biological specimens: imaging of a freeze-dried yeast cell

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Diffraction microscopy provides an alternative to lens-based methods of imaging non-crystalline objects. Thus, one can potentially reach resolutions beyond the limit imposed by optics. Image reconstruction in this case is a simple inverse Fourier transform of the correctly phased diffraction pattern. Phasing diffraction data of non-crystalline objects is achieved by iterative algorithms: the two most widely used are Fienup's HIO algorithm and Elser's Difference map. [1, 2] Both of these algorithms utilize available information about a sample as constraints. The important constraint in the real image space is called the finite support, which is guaranteed by oversampling diffraction pattern. [3] The Fourier space constraint is the measured diffraction pattern. Several groups have reported successful reconstructions of biological or material science samples by this technique. [4, 5, 6, 7, 8]

In our experiment, the X-ray diffraction pattern of a freeze-dried yeast cell was collected at 750 eV and phasing was performed using the Difference map. The freeze-dried yeast cell was 3 microns in diameter and its exit wave became complex-valued at 750 eV. The reconstruction of complex valued objects has been found to be particularly challenging [9,10], and the success of the reconstruction illustrates a step forward in this technique. The reconstructed image shows the nucleus and cell membrane clearly in 30 nm resolution. The reconstruction of a freeze-dried yeast cells gives us confidence that the phasing algorithm would work when one has diffraction data from frozen hydrated biological samples, which most resemble the living biological state.

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X-ray microscopy studies of electromigration in integrated circuits

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State-of-the-art high-performance microprocessors contain more than 100 million transistors which are connected using metal on-chip wires (interconnects). Further increased clock rates while continuing to down-scale transistor feature sizes requires a highly sophisticated interconnect design in combination with new technologies and materials, in particular to minimize the so called RC (Resistance x Capacitance) delay time. This task includes development and implementation of interconnect materials with lower resistivity and of isolating interlayer material with low permittivity. The aluminum-based interconnects have been replaced by inlaid copper providing reduced electrical resistivity and improved electromigration behaviour [1]. Currently, technology development is focussing on insulator materials with lower dielectric constant than silicon dioxide. Electromigration, stress-induced degradation and mechanical weakness in case of low-k materials are reliability concerns for inlaid copper interconnects. Formation of voids in copper lines induced by electromigration during microprocessor operation will cause an interconnect open or high resistance resulting in malfunction or speed degradation [1]. Stress-induced degradation phenomena are not well understood so far. Particularly, fast diffusion paths have to be identified, and failure mechanisms based on the directed transport of atoms have to be understood.

Optical microscopy does not provide the necessary spatial resolution. Transmission electron microscopy can only image thin layers with significantly less than 1 micron thickness. Scanning electron microscopy is more surface sensitive, and requires a very precise sample preparation [2]. Additionally, the structures of the Cu/low-k structures shrink extremely during observation in a state-of-the-art scanning electron microscope which is usually operated in the several-kV voltage range for achieving the required spatial resolution. X-rays have the advantage that they penetrate samples which are several micrometers thick without significant sample damage, and that they provide a chemical image contrast between different dielectric layers of the Cu/low-k on-chip interconnect stack.

In this report, results of x-ray microscopy studies on state-of-the-art microprocessors are presented to demonstrate that x-ray microscopy has the potential for a new powerful analytical technique in semiconductor industry.

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Dichroism Soft X-ray Absorption Spectromicroscopy and Antiferromagnetic Surface and Interfaces

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Synchrotron based dichroism x-ray absorption spectro-microscopy (dichroism XAS) is an excellent tool for the investigation of magnetic heterostructures, because of its ability to address antiferromagnetic, ferromagnetic, chemical and structural order of different elements in an unknown sample. Even more important, dichroism XAS can be used as a contrast mechanism in a photoemission electron microscope (PEEM) to characterize surfaces, buried interfaces and nanostructures with high spatial resolution (~50nm).

In my talk I will present results of our research focusing on *exchange bias* systems. These are systems in which a ferromagnetic and an antiferromagnetic material are in close contact with each other and hence the magnetic properties of the ferromagnet are changed in a very distinct way. In this area dichroism XAS has helped to improve our insight tremendously over recent years because conventional magnetic imaging techniques are not able to address the antiferromagnetic order which is a key ingredient in these systems. For the first time we were able to correlate the antiferromagnetic, ferromagnetic and interfacial domain structure. Furthermore we could identify and analyze the structure of lateral antiferromagnetic domain walls and extract quantitative information about the relevant magnetic anisotropy energies in NiO.

More recently we investigated the sensitivity of x-ray absorption to deviations of the crystal lattice from the ideal symmetry and we employed this sensitivity to image crystallographic domains in NiO using polarized soft x-rays. For example, in antiferromagnetic NiO each magnetic domain is suspected to correspond to a crystallographic domain which exhibits a distinct lattice distortion. By comparing the magnetic XAS at the nickel absorption edge with the polarization dependence of the oxygen XAS which is related to the crystal structure the correlation between magnetic and crystallographic domains could be unambiguously identified at the (100) surface of NiO.

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Time-resolved imaging of magnetic excitations in micro-particles using X-Ray microscopy

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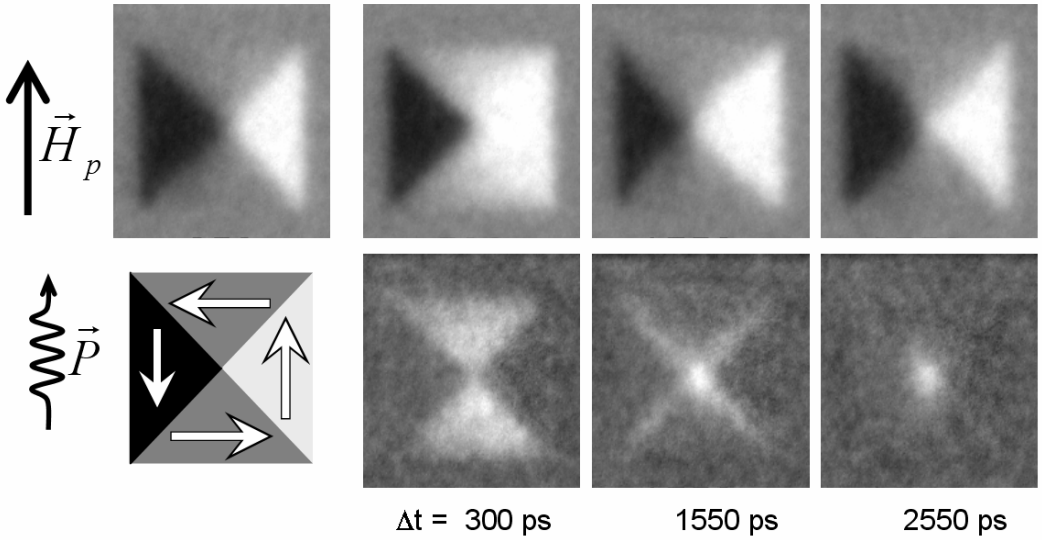
Synchrotrons are pulsed x-ray sources allowing experiments with time resolutions of <100ps. We combine this pulsed source with a Photoemission Electronmicroscope (PEEM) to image the magnetic state of micron sized permalloy objects following the excitation by a pulsed magnetic field.

The equilibrium state of permalloy squares is a Landau flux-closure pattern consisting of three substructures: the domains, the domain walls and the vortex core. The PEEM allows imaging all of them and following their evolution after excitation by a magnetic field pulse (H_p ~ 50Oe, 200ps rise time). After the excitation we first observe coherent precession of the magnetization (~300ps) in two of the domains (M perp. H_p). This precession is quickly (~1ns) damped transferring the energy into oscillations of the domain walls (1550ps). The domain walls oscillate analogous to an excited chord. The slowest process observed is the motion of the vortex core. This motion is perpendicular to the applied field pulse and strongly damped (2550ps). We observe no gyrotropic oscillation of the vortex.



Dichroic images (upper half) of a permalloy square (6x6 mum^2) following the excitation by a field pulse H_p. Sketch and difference images (lower half) highlighting the changes.

We compare excitations in permalloy squares to those in disks which do not contain domain walls. We also discuss how this technique can be extended to more complex materials like ferri-magnets and dilute magnetic semiconductors.

Hard x-ray spectromicroscopy using photoelectron emission microscope

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We have demonstrated an x-ray spectromicroscopy using photoelectron emission microscopy (PEEM) with hard x-rays. The advantages of the use of hard x-rays as an excitation source are the large proving depth, clear chemical contrast, and XAFS capability. The hard x-ray spectromicroscopy measurements were performed at the circularly polarized hard x-ray undulator beamline BL39XU of the SPring-8.

The spatial resolution of the hard x-ray PEEM is estimated to be 40 nm. The magnetic image of ultra high-density recording media CoCrPt are obtained at the Pt L-edge utilizing x-ray magnetic circular dichroism. The written magnetic patterns are clearly observed with a 130 nm spatial resolution.

We have also achieved a visualization of the buried interface of Au nanostructures buried by a 200 nm Co capping layer. The buried nanostructures are clearly imaged. The chemical mapping and nano-XAFS of iron meteorite are also shown. Nano-XAFS from a sub-micron area is obtained.

X-Ray Absorption Spectromicroscopic Analysis of Functionalized Pattern Surface

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The ability to manipulate matter in the nanometer scale attracts great attention in the nanoscience and technology. Nanofabrication of functionalized patterns is one of the most interest issues in the emerging field of nanotechnology. It is because of their potential applications in the biosensors, optoelectronics, or resist templates, etc. The functionalized nanopatterns afford the chemical defined surface is hence a key feature in the development of nonotechonology. Self-assembled monolayers (SAMs) have been the promising functionalized materials because of the well-ordered and functionality-controlled properties at the molecular level. It provides a platform for investigating biomolecules adsorption and bio-sensing with its unique selectivity and flexibility. Soft X-ray spectromicroscopy provides more chemical mapping capability at a relevant spatial scale than the conventional high resolution microscopy (e.g. SEM or AFM). Inheriting the nature of x-ray absorption, photoemission electron microscope (PEEM) has superior surface and chemical sensitivity to characterize the functionalized nanopatterns. We present the fabrication of SAMs-patterned surface with terminal COOH- and NH_2- on gold and silicon substrates. By utilizing the X-PEEM, near-edge X-ray absorption fine structure (NEXAFS) and attenuated total reflection infrared spectroscopy (ATR-IR), the composition and structure of functionalized patterns are explored.

X-ray Linear Dichroism Microscopy of Crystalline Short Chain Alkanes and Semi-crystalline Polyethylene Thin Films

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Scanning transmission x-ray microscopy (STXM) has been used to determine the two Near Edge X-ray Absorption Fine Structure (NEXAFS) spectra of *n*-tetracontane ($C_{40}H_{82}$) crystallites corresponding to the ***a*** and ***b*** directions of the orthorhombic unit cell of crystalline polyethylene. The two anisotropic lateral crystalline axis ***a*** and ***b***, *i.e.* <100> and <010> directions, respectively, can be distinguished by intensity changes in the spectral doublet near 287.5 eV. Prior NEXAFS dichroism data of PE and alkanes integrated over the <100> and <010> directions and only the dichroisms between the C-C chain along the ***c***-axis and the average for the C-H bonds has been known [1]. The spectra of *n*-tetracontane crystals can only be explained by a "further breakdown" of the building block model [2] and interactions between individual molecules.

This new information allows us to understand details of the growth orientation of polyethylene (PE) in ultrathin films. Overall, the same average spectra are found for PE thin film as in the bulk for PE of the same density [3, 4], clearly indicating that PE thin films are just as crystalline in thin films as they are in the bulk. In linear low density PE films, STXM shows large dichroic signals between various sample features for photon energies of ~287.4 eV and ~294 eV, irrespective of film thickness. This suggests that in these thin films, the average C-C bonds are in the plane of the thin film with predominantly edge-on lamellae orientation. In very thin films, the averaged NEXAFS spectrum switches from a slight dominance of ***b***-axis signal to one of slight ***a***-axis signal dominance. This suggests that the interfacial constraints alter the average orientations of crystallites. Linear medium density PE thin films showed a transition from edge-on to flat-on lamellae (C-C backbone parallel to the surface normal) in films thinner than 30 nm.

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Organic Analysis of Extraterrestrial Materials at the Sub-Micron Scale

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Chondritic meteorites, hydrated and anhydrous interplanetary dust particles (IDPs) all contain organic matter. The detailed characterization of this organic matter, including its abundance and type(s), is important in inferring the origin of the pre-biotic organic matter of the Solar System and in determining the extent of the contribution organic matter delivered to Earth by asteroids, comets, and IDPs makes to the origin of life on our planet. Organic matter, which is a minor component (≤4% of the mass) in meteorites, is generally studied by first concentrating this material by either water extraction or acid dissolution of the major silicate phases. The organic-rich concentrate is then analyzed with conventional instruments. Concentration allows the detection of rare organic compounds, some present at the ppm level. However, the concentration techniques mix together organic matter from all locations, thus all geological context is lost.

We employ a Scanning Transmission X-ray Microscope (STXM) on beamline X1A of the National Synchrotron Light Source (Brookhaven National Lab) to map carbon and determine the functional groups by X-ray Absorption Near-Edge Structure (XANES) spectroscopy on meteorites and IDPs. This technique allows us to determine the abundance and types of C, N, and O in ultramicrotome sections, typically ~80 to 150 nm thick, on particles as small as a few micrometers in size. STXM analysis preserves the geological context of each C-rich region, allowing us to separately analyze the carbon at different sites in the sample. In a single ~20 micrometer sample of the Tagish Lake meteorite we detected four spectroscopically distinct types of carbon: two areas of organic carbon with different C=C to C=O ratios, carbonate, and amorphous carbon (Flynn et al., Lunar & Planet. Sci. XXXII, #1593). In 10 micrometer IDPs we identified several different morphologies of carbon – thin coatings on minerals, sub-micron C-rich units, and larger organic regions (Flynn et. al., Geochim. Cosmochim. Acta, 67, 4791-4806).

Organic analysis using the STXM will be important for two extraterrestrial sample return missions – STARDUST and HAYABUSA. NASA's STARDUST spacecraft collected small particles, most <15 micrometers in size, from Comet Wild-2. These samples will be delivered to Earth on January 15th, 2006. Because of the small size, the traditional organic concentration techniques cannot be used. High-resolution analytical techniques, such as the STXM, will be critical to the identification and analysis of the very primitive organic matter expected from this comet. The Japanese Space Agency's HAYABUSA spacecraft is enroute to the asteroid Itokawa, where it is expected to collect a few grams of material. The organic content of Itokowa is not known, but meteorites with similar reflection spectra have low carbon. Thus sample size and carbon content suggest the STXM will be vital to characterization of this asteroid sample.

Speciation of sulphur in soils

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Sulphur moves freely within lithosphere, hydrosphere and atmosphere. It is an important element in soils, highly reactive, existing in various oxidation states, and an indispensable nutrient for plants and microorganisms. The soil environment is the primary component of the biogeochemical sulphur cycle. The speciation of sulphur is intimately related to the physico-chemical conditions of the soil environment, such as Eh and pH. Changes are caused by pedogenic and anthropogenic processes and result in changes of soil properties.

In strong contrast to this enormous relevance of sulphur in soils, the status of current methods of chemical analysis for its speciation until recently was highly unsatisfactory. Wet chemical methods can only distinguish total soil sulphur into operationally defined fractions, which only in rare cases can be exactly assigned to a specific S species. Moreover, with conventional methods different sulphur forms can only be assessed for bulk soil samples, but not with spatial resolution. The solution is X-ray spectromicroscopy as a combination of high spectral and high spatial resolution. It allows for a characterization of associations of soil colloids and soil micro habitats regarding structure, sulphur speciation and differences within and between associations.

The aim of the studies presented here is a clear determination of specific sulphur species in soils under varying chemical conditions. A podsol-gleysol-histosol catena representing a hydrological gradient within a small forested watershed has been studied. Here the groundwater level is found in different distances from the soil surface, so the relevance of anaerobic sulphur species strongly increases from podsol to histosol. The intermediate energy scanning X-ray microscope at beamline 2-ID-B of the Advanced Photon Source in the Argonne National Laboratory has been used for these spectromicroscopy studies. Four different sites and four different depths per site have been probed resulting in sixteen sample positions. This documents the entire soil profile including all horizons from the forest floor down to the solid bedrock. A matrix showing the resulting spectra visualizes clearly differences and similarities and will be presented and discussed in detail as well as spatially resolved data. The results presented here prove that it is possible to successfully apply spectromicroscopy even to samples with low total sulphur content. Moreover, spatially resolved studies allow for characterization of associations of soil colloids and soil micro habitats regarding structure and sulphur speciation.

Distribution and XANES of Arsenic in Root of Hyperaccumulator Fern (*Pteris vittata* L.) measured by μ-SR-XRF analysis

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Pteris vittata L. is known as an arsenic hyperaccumulating fern and expected as a strong candidate plant for a technology of Phytoremediation in order to remedy a polluted environment. Some researchers reported that a reduction of arsenic, arsenate ion (As^{5+}) to arsenite ion (As^{3+}), took place in a plant body[1][2][3]. These results are strange and interesting phenomenon, because arsenite is more toxic than arsenate.

In this study, we attempted to observe the distribution of arsenic in a horizontal cross-section of root by XRF imaging and to obtain the XANES spectra for the determination of a valence change from surface to centre of the root. XANES spectra were collected following the XRF imaging, these measurements were carried out using the μ-SR-XRF system at BL37XU in SPring-8 (JASRI, Japan). X-rays from the undulator were focused into a microbeam with beam-size of $1.5\mu m^2$ by the K-B mirror optics. Ferns were cultivated in hydroponics, and were transferred into the arsenate containing nutritional solution with concentration of 10mg As/L as KH_2AsO_4. Each samples cut from a root system of fern were frozen rapidly and sliced. And then sliced root samples were freeze-dried and were subjected to the X-ray microbeam analysis.

In our experiment, the arsenic distributions of the cross-sections were successfully measured by the μ-SR-XRF analysis. A comparison of XANES spectra of the 6 points in the root tissue at high As level with those of the standard compounds(As_2O_3, H_3AsO_4) has revealed that arsenic in root tissue exist as a mixture of As^{5+} and As^{3+} and their ratios changed markedly with reductive direction at the boundary area between a cortex and central cylinder.

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Combined Differential Phase Contrast Imaging and Fluorescence Trace Element Mapping at the Advanced Photon Source

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Hard x-ray microprobes excel at fluorescence mapping and quantification of trace metals in biological cells. They are less good at putting these elements in structural context: where are they in the cell? The reason is that biological ultrastructure is absorbing only weakly at the multi-keV energies required for the excitation of fluorescence from biologically interesting metals. We describe here a solution involving the use of phase contrast imaging integrated into an x-ray microprobe.

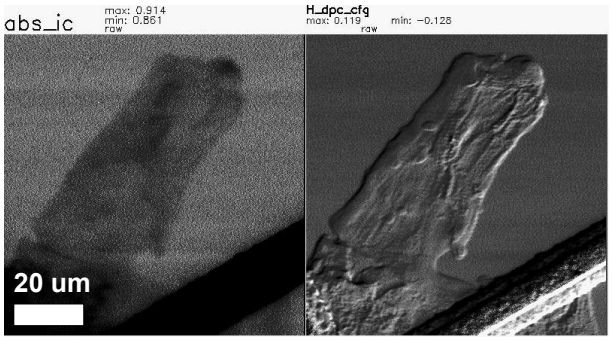

Fig. 1: Cardiac myocyte (heart muscle cell). Collaboration with B. Palmer, U. Vermont. While absorption contrast shows only an outline of the cell, differential phase contrast shows subcellular structures.

Phase effects in the specimen lead to a modification of the intensity distribution following the specimen. We have previously used a segmented silicon detector with rapid analog signal readout, and a Fourier filtering image reconstruction approach, to obtain quantitative soft x-ray phase contrast images of microfabricated test patterns [1]. We have modified this detector for differential phase contrast imaging at multi-keV x rays. While a second generation detector optimized for operation in this energy range is now under construction, experiments with the existing detector at beamline 2-ID-E of the Advanced Photon Source (APS) show the utility of phase contrast. Fig. 1 shows images of a cardiac myocyte investigated by Palmer *et al*. While these cells are thick enough to show some absorption contrast at 10 keV incident x-ray energy, a simple difference signal between opposing segments of our detector shows ultrastructural components of the cell to provide the information necessary to understand the physiological role of trace elemental concentration variations.

We describe the use of phase contrast in a variety of microprobe studies, and the characteristics expected for the new detectors optimized for present APS zone plate microprobes, as well as the Nanoprobe system presently being built for installation at the APS. When the new detectors become available, we also hope to measure phase shift quantitatively to obtain specimen thickness and therefore concentrations rather than absolute amounts of trace elements.

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The Scanning Photoemission Microscope at Elettra: recent results and developments

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With respect to the other photoelectron microscopy techniques a Scanning PhotoEmission Microscope (SPEM) uses the most direct approach to photoelectron spectromicroscopy which is the use of a small focused photon probe to illuminate the surface. The SPEM at the Elettra synchrotron light source can operate in two modes: imaging and spectroscopy. In the first mode the sample surface is mapped by synchronized-scanning the sample with respect to the focused photon beam and collecting photoelectrons with a selected kinetic energy. The second mode is photoelectron spectroscopy from a microspot. The SPEM on the ESCAmicroscopy beamline at Elettra has a lateral resolution of 150 nm; and an overall energy resolution which is now better than 200 meV [1]. Samples can heated and cooled (liquid N2) during the measurements. More than 70% of the available beamtime is dedicated to national and international users; two call for proposals of experiment per year are available. In order to offer the maximum flexibility for the preparation of the experiments three sample preparation chambers are available. One is equipped with standard surface sensitive analysis techniques (AES, LEED, PEEM) and all needed for the preparation of surfaces (heating, sputtering, atomic gas plasma deposition). A second chamber is dedicated to gaseous exposures up to 1 bar. The most recent fields of investigation deal with the oxidation and reduction processes of Rh thin films and single crystal, mass transport studies on multiwall carbon nanotubes, spatially resolved chemical analysis of Manganites. Industrial collaborations are encouraged aiming to enlarge the potential user community of the photoemission microscopy techniques. The formation of dark spots in commercial emitting OLED responsible for the degradation of these devices has been recently investigated [2].

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X-ray projection microscopy to investigate liquid Ga penetration in Al bicrystals

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The penetration of liquid Ga along the grain boundaries of Al bicrystals is analysed by synchrotron X-ray projection microscopy. Using Kirkpatrick-Baez focussing optics, a state of the art secondary X-ray source size of 90×90 nm^2 is produced with typical divergences of a few milli-radians [1]. The present investigation deals with one of the very first applications of such a microscope, currently under commissioning at the ID19 beamline of the European Synchrotron Radiation Facility (ESRF, Grenoble, France).

In situ observations of the Ga penetration process reveal linear propagation of the penetration front accompanied by a continuous thickening of the intergranular Ga wetting layer [2,3]. By combining absorption measurements and image correlation techniques it has been possible to characterize simultaneously the presence of nanometric penetration layers and, for the first time, associated continuous relative movement of the Al bicrystal grains of same amplitude [4]. The measured deformation of the bicrystal is compared to the predictions of elasto-plastic crack propagation under mode I loading conditions.

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Achievements and perspectives of magnetic soft X- ray transmission microscopy

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A fundamental understanding of magnetism on the nm length and sub-ns time scale is currently the focus of both basic and applied solid state physics research. The origin of interlayer exchange coupling, perpendicular magnetic anisotropies, unusual magneto resistance effects, precessional and relaxation phenomena are just a few examples. Laterally patterned magnetic systems are also promising candidates in future magnetic storage and sensor technologies, where both the miniaturisation and the speeding up of switching processes are crucial.

The X-ray magnetic circular dichroism (X-MCD) effect, in which the absorption coefficient of right and left handed circularly polarized x-rays is dependent on the magnetization state of the sample, has stimulated the development of numerous powerful analysis techniques to investigate the magnetism of solids, surfaces and thin films with inherent energy tuneable element-specificity. The combination of a high-resolution full field transmission soft X-ray microscope with lateral resolution, demonstrated down to 15nm, provided by Fresnel zone plate optical elements, with X-MCD which provides a large and element specific contrast mechanism creates a ideal instrument to study the magnetic behaviour in low dimensional magnetic systems in detail, e.g. layer resolved magnetisation reversal and nucleation processes on an granular length scale. Moreover, the complexity of an elliptically polarized undulator is not required for this technique; the off-axis radiation of a simple bending magnet produces elliptically polarized x-rays either above or below the electron orbital plane. The environment of the sample, which can be at atmospheric pressure, can be controlled by the application of external fields and temperature conditioning.

In addition to high spatial resolution, the inherent sub-ns time structure of current synchrotron radiation facilities can be used in a stroboscopic pump-and-probe experimental arrangement to image the fast magnetization dynamics in microstructured elements. An electronic pulse launched into a microcoil or stripline (pump) excites the magnetization in the sample under test, which is imaged at a variable delay time by the flash of the synchtrotron pulse (probe) [2]. Hence local spin dynamics, e.g. vortex motions on a ns time scale can be addressed.

We will review the current status of the full-field soft X-ray microscopy endstation XM-1 at the ALS in Berkeley CA with respect to technical improvements [3]. Recent experimental results to study the microscopic origin of magnetization reversal will be reported providing detailed insight into the reproducibility and stochasticity of local switching phenomena [4]. Perspectives of MTXM aiming at ultrafast (ps) magnetization dynamics will be discussed.

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Sub-10 nm X-ray Microscopy: Status and Pathways

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X-ray microscopy offers important and desirable visualization and characterization capabilities of great importance to broad range of scientific disciplines, including for example biology and emerging nanoscience and nanotechnology. Its short wavelength permits nanometer resolution imaging without the limitation of wavelength. Its high penetration power allows nondestructive imaging of internal structures of an object. It also has many contrast mechanisms that can be employed beyond simple structural imaging, such as chemical state imaging or elemental specific imaging.

Over the last decade, the resolution of x-ray microscopy has improved significantly. Thanks to mainly the advancement in developing high resolution x-ray focusing optics, x-ray imaging with a spatial resolution better than 15 nm and 20 nm has been demonstrated with soft and hard x-rays, respectively. Sub-10 nm resolution x-ray imaging is expected to be realized within the next few years. My talk will present the current status of the state-of-the-art x-ray imaging in terms of spatial resolution and discuss pathways toward achieving sub-10 nm x-ray imaging.

100ps Time-Resolved Magnetic X-ray Microscopy – Techniques and Applications

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Fast magnetization dynamics of ferromagnetic elements on short length scales is currently attracting substantial scientific interests for both technological and fundamental reasons. Measurements with a time resolution of 70-100 ps combined with a lateral resolution of 20-40 nm were performed using two different sample geometries (magnetic 'in-plane' excitation by a microcoil and 'out-of-plane' excitation by a stripline) at two different microscopes: a full-field soft X-ray microscope (XM-1, ALS beamline 6.1.2) and a scanning transmission X-ray microscope (STXM, ALS beamline 11.0.2). The scanning microscope equipped with a fast avalanche photo diode (APD) detector allowed us to speed up time-dependent measurements by about a factor of 10.

Spin precession [1] and gyrotropic vortex motion [2,3] in micron-sized ferromagnetic patterns have been studied. Complementary to the time-domain 'pump-and-probe' measurements a frequency-domain 'sine excitation' technique [2] was implemented into X-ray microscopy. In this way, the dynamics of specific eigenmodes of ferromagnetic patterns could be imaged. A novel highly non-linear effect has been found in micron-sized magnetic Landau structures for the first time [3]: a clear change of the sense of rotation of the magnetic vortex is observed by increasing the amplitude of an exciting alternating magnetic field over a well pronounced threshold level (about 2 mT in the present experiments). This unambiguously leads to the conclusion that the direction of magnetization of the magnetic vortex core could be repeatable switched by 180°, allowing a deliberate and reproducible switching of the vortex core and consequently of the chirality (handedness) of the vortex structure. A model for this surprising effect is given based on transitions from deterministic to chaotic behaviour of magnetic vortex structures.

In contrast to static experiments, where vortex core switching only occurs with external magnetic fields of about 100 mT or higher, the novel dynamic vortex core switching needs only a few mT. This might open paths for new technological applications.

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Coherent x-ray diffraction microscopy: fundamental and technical limits

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In this paper we restate the basics of a coherent x-ray diffraction microscopy experiment with two goals in mind. The first is to allow the ultimate limits of the technique to be evaluated. We consider the limitation of imaging speed due to both currently-available and projected-future x-ray beam lines and the limitation of resolution due to radiation damage in the case of biological samples. The second goal is to determine the technical requirements for spatial and temporal coherence, x-ray wavelength, motion control and stability requirements etc that must be met if our future goals for resolution, statistical accuracy and 3D imaging speed are to be reached. This study is motivated in part by plans at the Advanced Light Source in Berkeley USA to build a new undulator and beam line for x-ray diffraction microscopy. In light of the fundamentals considered above we discuss the technical choices that we propose for the new diffraction microscopy facility.

Characterization of medium-range order in noncrystalline systems by fluctuation x-ray microscopy

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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 can not 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. 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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Coherent X-ray Diffraction on Nano-size Objects

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Coherent x-ray diffraction (CXD) is a new experimental method for studying perfect and imperfect crystals. The method has become available by the recent development of high-brilliance third generation sources of synchrotron radiation (ESRF, APS, SPRING-8). The beams coherence volume being of the order of few microns can entirely enclose a nano-size object. Instead of incoherent averaging, a coherent sum of amplitudes produces a coherent diffraction pattern originating from the real space arrangement of the sample. If high-quality x-ray lenses were available as they are for electrons, such diffraction patterns could be transformed to magnified images directly. However such x-ray microscopes still suffer a lot from optics aberration and resolution is often limited to the pixel size of the CCD.

In this talk we will show how the objective lens of the microscope can be replaced by a special iterative phase reconstruction procedure that inverts intensity measurements of the CXD pattern to real space image. The method is based on the fact that the diffraction pattern can be oversampled relative to its spatial Nyquist frequency so that the Fourier transform can be overdetermined in spite of missing phase information. In principle this method does not have any limitations on the available resolution.

Several applications of the method will be given in the talk. It will be shown how 3D images of the interiors of Au nanocrystals that show 50 nm wide bands of contrast with {111} orientation can be obtained applying this technique [1]. The size of the objects can be further reduced to the size of the quantum dots samples if repetitive motive in the form of 2D crystal is used. It will be demonstrated that in the case of coherent illumination of these samples the correct shape and orientation of individual island can be obtained. In the case of partially coherent illumination the correct shape of the particle can be obtained only when the coherence of the incoming beam is reduced to match the size of the island [2]. In the last example experimental results of CXD scattering on the sample of specially fabricated GeSi islands of nanometer size and in a regular array embedded to Si substrate will be shown [3]. Two geometries of scattering that is grazing incidence diffraction (GID) and grazing incidence small angle x-ray scattering (GISAXS) were used. Applying a microfocuse coherent beam on our sample give rise to coherent diffraction pattern with Bragg spots and broad diffuse maxima in GID geometry. The GISAXS pattern has a typical shape resulting from the periodic array of identical islands. This diffraction pattern was used to reconstruct the average shape of the islands using a model independent phase retrieval algorithms.

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Hard X-ray Diffraction Microscopy at SPring-8

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X-ray diffraction microscopy is an innovative method to reconstruct high-spatial-resolution images from oversampled Fraunhofer diffraction intensities of non-crystalline samples. As the ultimate spatial resolution is limited by the x-ray wavelength and for biological samples by radiation damage, shorter-wavelength hard x-rays are beneficial to achieve higher resolution. Using the hard-x-ray beamline BL29XUL at SPring-8, we have been developing x-ray diffraction microscopes, and have shown the usefulness of the method [1-3].

In the x-ray diffraction microscopy experiment, it is desirable to reconstruct the sample image in parallel with conducting experiment in order to get quick feedback from the measurement. In early dates, the problem of missing central data due to a beamstop hindered it, because a supplemental experiment has been required to restore low-resolution information. To solve the problem, we developed a new reconstruction algorithm which is effective even with relatively-large missing central data [3]. In addition, we are developing a dynamic reconfigurable processor to quickly perform fast Fourier transform, which will be used to perform faster image-reconstruction.

To achieve higher spatial resolution, the development of larger-area detectors is essential. We are developing a large-area in-vacuum imaging plate detector and a vacuum chamber for it, and are planning to evaluate their performance in the immediate future.

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Spectromicroscopy analysis: clustering, error-finding, and interpreting

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Soft x-ray spectromicroscopy provides the means for studying chemical speciation at the 30-50 nm resolution scale, and it is finding wide use in studies in biology, environmental science, astrobiology, polymer research, and other fields. For a specimen that can be characterized in terms of a set of known spectra, a variety of approaches[1,2] can be used for compositional mapping. However, this is rarely the case in biology or environmental science where the complexity of the specimen and reactivity of components precludes advance knowledge of all signature spectra.

Cluster analysis provides a way to find the signature spectra that exist in a specimen, and form compositional maps based on these "discovered" spectra. Following preliminary work[3], we have carried out a systematic development[4] of this approach to soft x-ray spectromicroscopy analysis, and have extended it with methodologies aimed at classifying only on compositional variations rather than specimen thickness[5]. New developments include the recovery of the "true" incident flux spectrum from a dataset, and new methods for the analysis of mixtures. These and other developments will be reviewed and illustrated with examples from studies in biology and environmental science.

We gratefully acknowledge support from the NIH under contract R01 EB00479-01A1, and the National Science Foundation under grants CHE-0221934 and OCE-0221029.

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Quantitative phase imaging and tomography with polychromatic x-rays

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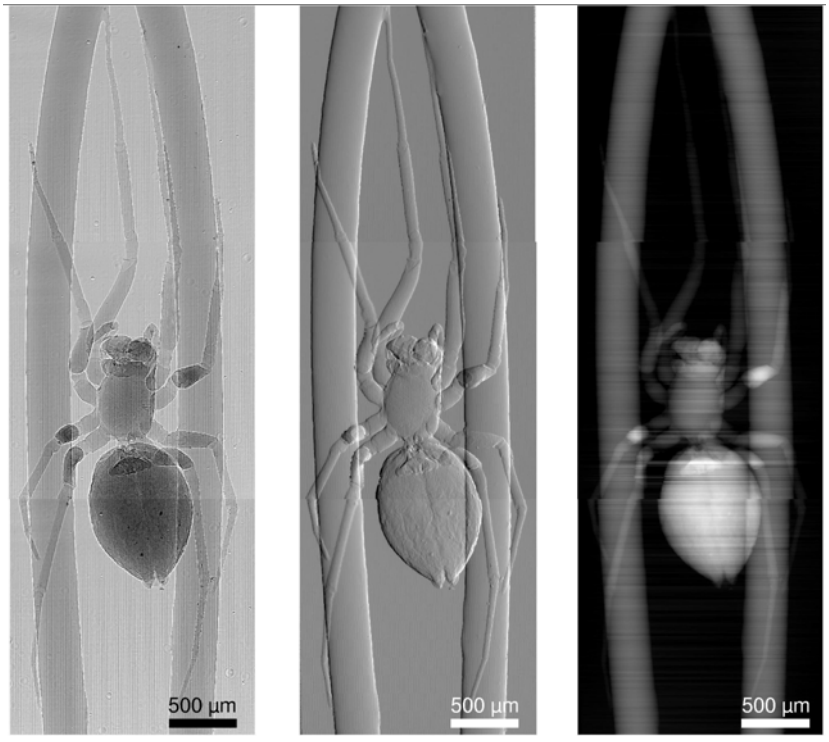
We have developed a two-grating interferometer for hard x-rays that can be used for phase imaging and tomography. The instrument consists of a phase grating acting as a beam splitter and an absorption grating. The recorded signal on the detector is essentially the first derivative of the projected x-ray refractive index in the direction perpendicular to the grating lines. The refractive index can be reconstructed by simple integration. For absorbing samples, the contribution of absorption contrast to the signal can be completely separated from the phase signal by applying a phase-stepping technique during data acquisition. We have used the instrument with a lateral resolution of a few microns and a field of view of 3mm, but it can be scaled up to large fields of view of many centimeters, and correspondingly large pixel sizes. Since the entire setup is quasi-achromatic, the device can be operated with broadband radiation, such as the filtered spectrum of a laboratory x-ay source, or a "pink" synchrotron beam, and is not restricted to monochromatic radiation. We have used synchrotron radiation with a bandwidth of 5 per cent around a mean energy of 17.5keV to quantitatively reconstruct the refractive index distribution in a sample from a tomographic dataset. It was possible to distinguish materials with differences in refractive index of only a few percent. The method is expected to have wide applications in imaging of low absorbing samples such as biological and medical tissue or fibre reinforced polymers.



X-ray micrographs of a spider in absorption contrast (left), interferometric differential phase contrast (middle), and phase contrast (right).

Nanometer-scale x-ray holography with 1-2 keV x-rays

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ABSTRACT

X-ray holography is a promising means for imaging biological and materials science specimens with nanometer-scale resolution. Fourier transform holography offers intrinsic phase as well absorption contrast, at a resolution limited only by the precision with which the reference wave is known. It has also been proposed as a starting point for iterative phase retrieval in lenseless coherent diffraction imaging experiments with potentially atomic scale resolution.

Using 1-2 keV x-rays at the 2-ID-B undulator beamline at the Advanced Photon Source, a zone plate lens to form the spherical reference wave, and a direct-detection CCD camera, we recorded Fourier transform holograms of lead micro- and nano-crystals. The zone plate had an finest zone width of 50 nm. The third diffraction order of the zone plate was used for its potentially higher resolution. Hologram exposures were typically 10 s; series of 100-200 exposures were acquired to improve the hologram signal to background. Numerical reconstructions of the individual and summed series of holograms required only a few seconds on a PC-class computer. Fine features in the lead crystal sample were resolved to ~50 nm in the reconstructed holograms. These results give impetus to ongoing efforts to develop high resolution flash coherent x-ray imaging methods using existing synchrotron radiation and future x-ray laser sources.

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Soft x-ray phase-sensitive imaging with diffractive optical elements

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In the x-ray region, for nearly all elements the real part delta of the complex index of refraction n (n = 1 – delta – i·beta) is larger than the imaginary part beta. It follows that the phase shift of any object is stronger than the absorption. Thus, phase sensitive x-ray imaging techniques like Zernike phase contrast x-ray microscopy [1] were developed to access the phase shifting, real part of the refractive index of the object in order to examine thin and weakly absorbing samples with sufficient contrast. Another alternative approach was the use of segmented detectors in scanning x-ray microscopes [2]. Recently, the differential interference contrast (DIC) method – first introduced by Nomarski [3] for visible light – was demonstrated with so call twin zone plate optics (TZP) at 4 keV photon energy [4]. However, these elements are difficult to fabricate. A possible solution to this problem is the combination of the two zone plates into a single diffractive optical element (DOE). First proof of principle experiments with this kind of optics for DIC-microscopy at 4 keV photon energy were successful [5] and showed the strong potential of DOE x-ray optics.
Obviously, x-ray microscopy in the water window region would also benefit from the DIC contrast mechanism. In this contribution we present the first diffractive optical elements for soft x-ray DIC microscopy. Due to an improved calculation method the nanofabrication accuracy is the same as for a comparable normal zone plate optic with the same outermost zone width. Different DOEs were fabricated with outermost zone width of 100 nm and 50 nm, respectively, different spot separation directions and different phase relations between the two spots. The optics were successfully used in DIC experiments both at the synchrotron radiation based TWINMIC microscope and at the Stockholm compact liquid-nitrogen laser-plasma source based microscope.

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Hard X-ray Micro-Interferometer for High-Spatial-Resolution Phase Measurement

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Owing to the highly brilliant undulators in SPring-8, a large spatially coherent region is available even in the hard X-ray region, and therefore many kinds of coherent optics have been developed. We proposed and constructed a novel hard X-ray micro-interferometer using an imaging microscope for high-spatial-resolution phase measurement at Hyogo-BL of SPring-8 [1]. We report about the latest results of this work.

The optical system is shown in Fig. 1(a). This hard X-ray micro-interferometer is formed as a wavefront-division-type, therefore both object and reference waves are necessary. Two zone plates (ZP-A and ZP-B) are arranged closely in the same plane perpendicular to the beam axis. If the two zone plates are illuminated coherently, the corresponding two secondary point sources are produced in their back focal positions. Two spherical waves diverging from these two point sources overlap each other and interference fringes are formed at an image plane. In order to prevent the −1st order diffracted waves from being mixed on the interference region, ZP-A was designed to have a half-moon shape. We call this optical element consisting of two zone plates "twin zone plate". The circumstantial parameters are shown in Fig. 1(b). Photon energy was tuned to 9 keV. An X-ray zooming tube was employed to observe interference patterns. To convert the interference pattern into the quantitative phase map, the fringe scanning method was applied. A 125-μm-thick kapton film was used as a rotatable phase plate.

Quantitative phase map of polystyrene microparticles with a diameter of 7 μm was imaged clearly. Spatial resolution of the phase map was 160 nm, estimated by edge response of phase retrieved image of a copper #2000 mesh. Furthermore, by putting a sample on a high-precision rotating stage, tomographic image was also obtained. Spatial resolution of the reconstructed image was achieved to be 250 nm, which was estimated by edge response of tomographic slice of a grass capillary. From these results, we have succeeded in high-spatial-resolution phase measurement using the X-ray micro-interferometer.

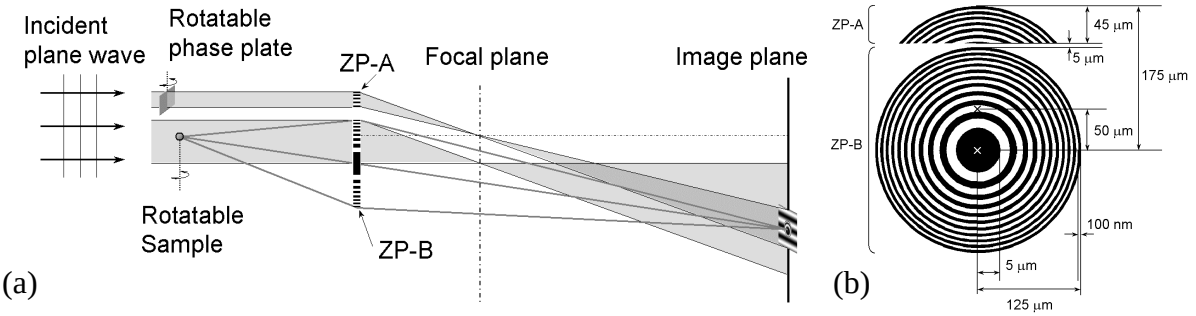

Fig. 1. (a) Schematic drawing of the hard X-ray interferometer. (b) Design of the "twin zone plate" made of tantalum, fabricated by NTT Advanced Technology Corporation.

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Novel X-ray Microscopes for 3-D and fs-imaging at BESSY

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The full-field x-ray microscope installed at the 3rd generation electron storage ring BESSY II is dedicated for applications in life, environmental and materials sciences. It covers the photon energy range between 250 - 750 eV. Currently, the spatial resolution is about 20 nm. Due to the small numerical aperture of zone plates, X-ray objectives have a depth of focus on the order of several microns. By treating the X-ray microscopy images as projections of the sample absorption, computed tomography can be performed.
3-D x-ray microscopy - pioneered at the BESSY I electron storage ring using a full-field TXM and at the NSLS using a scanning TXM - has found numerous applications worldwide. To further improve 3-D x-ray imaging towards 10 nm spatial resolution and to increase the usable photon energy range into the hard x-ray region, progress has to be made in nanotechnology of the x-ray optics, the instrumentation and the theory for recovering the full 3-D information of an object at this resolution level. In the talk, the current status at synchrotron sources and future aspects of x-ray imaging with fs-pulses from Free Electron Lasers will be discussed. In Fig. 1, a bird´s eye view of the BESSY site with the planned BESSY HGHG FEL is shown. It will provide 20 fs pulses for x-ray imaging in the photon energy range between 20 – 1000 eV.

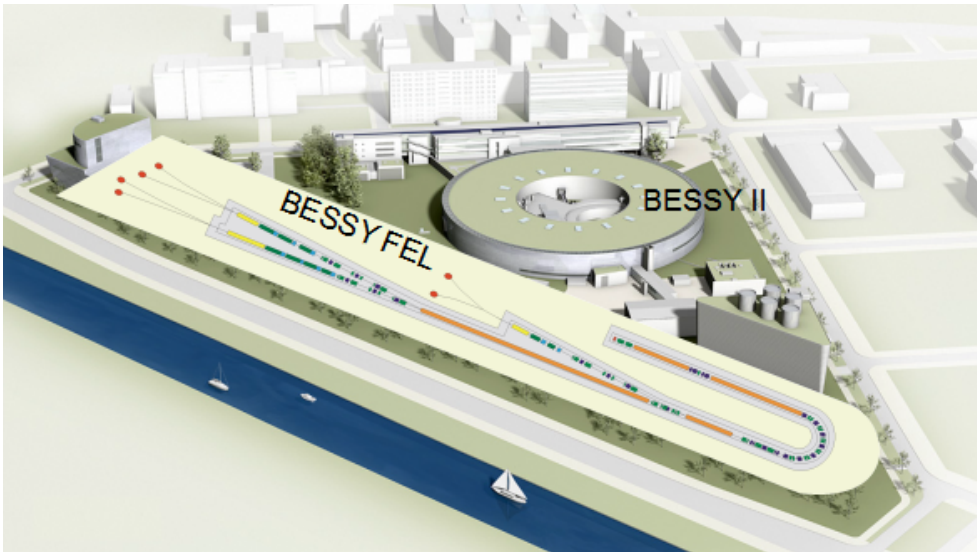

Fig. 1: The planned BESSY HGHG Free Electron Laser will be installed next to the BESSY II storage ring (see also www.BESSY.de).

X-ray Image Reconstruction using the Transport of Intensity Equation

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The transport of intensity equation (ToI) describes the flow of energy in a wavefield and applies quite generally to all forms of waves, including quantum mechanical waves such as neutrons, atoms, electrons, as well as light and x-rays. The only assumption in its derivation is that the flow of light is paraxial, meaning that the flow of light is predominantly along a given direction. This is almost always true for synchrotron-based imaging. The ToI can be used to create a phase image by measuring the intensity in the plane of interest and the rate at which the intensity is changing along the optical axis. It has been used to reconstruct phase images for all of the types of wave mentioned above.

For longer wavelength waves, such as visible light, a number of quantitative phase measurement tools are available. Interferometry is the most familiar of these and it requires waves with a high degree of spatial coherence. This level of coherence is harder to achieve with x-ray sources. Phase recovery using the ToI is very forgiving in terms of spatial coherence and can be used to obtain phase images even for almost completely incoherent light. Moreover, the method of image acquisition is inherently linear and so it has the unique property that image formation can be properly described using optical transfer functions even for partially coherent illumination.

In this talk I will review the ideas underlying imaging using the ToI, present experimental results and then discuss how the ideas may be extended to very high resolution coherent diffractive imaging.

Coherent Imaging: Materials Science

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We are assessing X-ray diffraction microscopy by phase retrieval as a means to perform high-resolution three-dimensional characterisation of non-periodic isolated objects (particles). Several recent experimental and computational developments have enabled us to perform full 3D X-ray diffraction imaging, with high resolution in all three dimensions. These 3D reconstructions were performed from the diffraction data alone. These developments include the Stony Brook diffraction apparatus [1], which allows 3D diffraction datasets to be quickly acquired; the *Shrinkwrap-Hybrid Input Output* phase-retrieval algorithm [2], which allows images to be reconstructed *ab initio* from incomplete diffraction datasets; a fast distributed FFT [3] and reconstruction software implemented on a computer cluster, which allows 1024^3 diffraction datasets to be phased in several hours. We have achieved high-resolution 3D reconstructions of both well-characterized test objects and of mesoporous foams that cannot be otherwise characterized. We find that high resolution imaging of thick objects can only be attained in the context of 3D measurement and reconstruction. Reconstruction from diffraction data acquired over many sample orientations allows one to avoid defocus (depth of field) artifacts as well perform a quantitative measurement of refractive index that is not possible from single-view diffraction data. A recent development is the use of reference points deposited near the object to provide an x-ray hologram. The hologram can be used to rapidly identify the specimen, and help the phase-retrieval algorithm.

Resolution of X-ray diffraction imaging will ultimately be limited by radiation damage. One eventual goal is to surpass damage resolution limits of individual particles by using streams of identical particles, such as protein macromolecules, using flash-imaging by X-ray free-electron lasers (XFELs) [4]. Models show that these methods should allow close to atomic resolution imaging.

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