SESSION III

Present and Future Synchrotron Facilities for Radionuclide Studies

Chair: L. Soderholm

CURRENT AND FUTURE STUDIES OF ACTINIDE MATERIALS AT THE ADVANCED LIGHT SOURCE

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Abstract

The vacuum ultraviolet (VUV)/soft X-ray region of the synchrotron radiation (SR) spectrum has revolutionised the approach to surface materials chemistry/physics. This spectral region has long been recognised for its potential to afford investigations of fundamental actinide science and to provide an improved analytical surface characterisation capabilities for actinide materials. Previously, the actinide science community was unable to take advantage of the opportunities afforded by VUV/soft X-ray SR methodologies because of radiological safety concerns and the need to prepare pristine actinide materials compatible with experiments in ultra-high vacuum end stations. With the advent of third-generation light sources operating in the VUV/soft X-ray region, such as the Advanced Light Source (ALS) at LBNL, experiments with small amounts of actinide materials are possible. Actinide investigations at the ALS are based on a graded approach to radioactive materials safety and are conducted with the assistance of LBNL Environment, Health and Safety (EH&S) personnel during the experiments. LBNL EH&S personnel have many years of experience supporting experiments utilising radioactive materials at LBNL user-based accelerator facilities.

The SR techniques and technologies used in this energy region have continued to mature, permitting more complicated investigations and enabling new spectroscopic approaches. Of particular importance for work with actinides is the development of the micro-spectroscopy and spectro-microscopy capabilities on several beamlines at the ALS. These capabilities allow the use of small amounts of actinide material. In addition, these techniques can provide information on the spatial distribution of actinide species that complements the spectral information. Early demonstrations of the ability to safely collect data from actinide micro-samples and the active support of the ALS for such experiments has led to the investigations of actinide materials on several different ALS beamlines.

Results from current studies, pending scientific plans and future opportunities for the investigation of actinide materials at the ALS will be presented. The status and capabilities of the ALS Molecular Environmental Science (ALS-MES) Phase I Beamline, an important component for actinide research at the ALS in the near future, will also be discussed.

Acknowledgements

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SYNCHROTRON STUDIES ON ACTINIDE-CONTAINING SAMPLES AT THE ADVANCED PHOTON SOURCE

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Abstract

The Advanced Photon Source (APS) is a third-generation synchrotron source located at Argonne National Laboratory in Chicago, IL USA. The APS currently provides a wide range of opportunities for synchrotron studies, including standard spectroscopy techniques as well as beam lines dedicated to micro-focusing and scattering. Of particular interest to the actinide community is the availability of high-energy (> 60 keV) photons that provide high-quality diffraction or scattering data with high resolution and that are not vitiated by absorption or extinction problems. The location of the APS on a DOE site with existing facilities for handling radioactive samples provides a unique opportunity for the experimenter wishing to perform *in situ* studies on actinide-containing samples. Access to these hot laboratory facilities and user support for experiments at the APS may be obtained through the Actinide Facility. Examples of experimental capabilities will be outlined, as will the current availability of the APS for experiments involving radioactivity.

Acknowledgements

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OPPORTUNITIES FOR XAFS SPECTROSCOPY ON ACTINIDES AT THE ROSSENDORF BEAMLINE (ROBL) AT THE EUROPEAN SYNCHROTRON RADIATION FACILITY

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Abstract

Since January 1999 two different experimental stations have been fully operational at the Rossendorf Beamline (ROBL): a radiochemistry laboratory for X-ray absorption spectroscopy of non-sealed radioactive samples and a materials-research station for X-ray diffraction and reflectometry. Both experimental stations have been utilised by external users on the basis of peer-reviewed proposals submitted to the ESRF or in collaboration with the Forschungszentrum Rossendorf (FZR). Since spring 2000, ROBL has become a European Large Scale Facility. This gives external research groups an additional opportunity to obtain beamtime for their experiments at ROBL, which is financially supported in part by the European Commission.

The radiochemistry end station has been designed to study radionuclides of environmental importance. The current regulations agreed upon the ESRR and the French authorities make it possible to study the following actinides at ROBL: ^{nat}Th, ^{231}Pa, ^{nat}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{244}Cm, ^{246}Cm, ^{248}Cm as well as the radionuclides ^{226}Ra, ^{208}Po, ^{209}Po and ^{99}Tc. The maximum equivalent activity of all samples present at ROBL at any given time is 185 MBq (5 mCi). The main characteristics of the optics of ROBL and its radiochemistry end station will be presented [1]. Selected XAFS results obtained on Tc, U, Np and Pu samples will illustrate the performance and the experimental opportunities available at ROBL.

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A NEW HARD X-RAY ABSORPTION SPECTROSCOPY MOLECULAR ENVIRONMENTAL SCIENCES BEAMLINE AT SSRL

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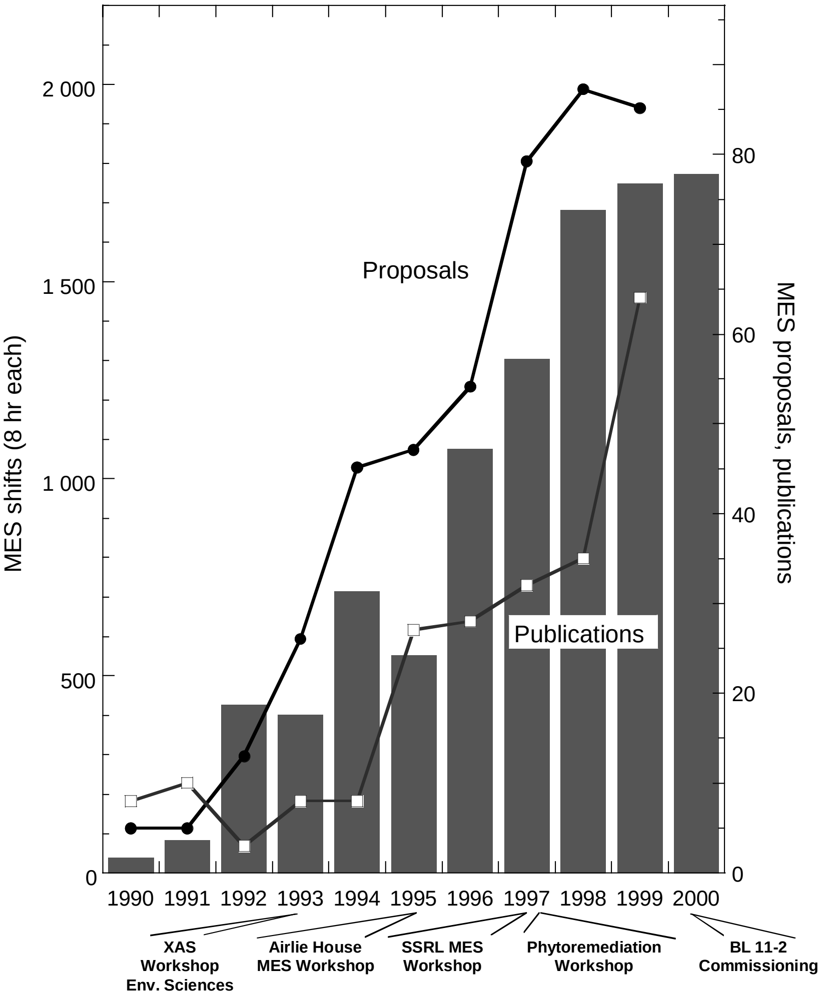
Abstract

The growth of X-ray absorption spectroscopy (XAS) as an important tool in molecular environmental sciences (MES) has generated substantial demand for beamline facilities optimised for MES XAS experiments. Chief among these is a beamline designed to accommodate the analysis and safe handling of radioactive materials such as soils contaminated with actinides. In response to this demand, the Stanford Synchrotron Radiation Laboratory (SSRL) has built a new hard X-ray beamline optimised for XAS experiments on MES samples of all types and designed for the safe handling of radioactive samples. A description of the facility concept and commissioning results to date are provided. General user operation of this facility is anticipated to begin in the summer of 2001.

Introduction: Growth of XAS and synchrotron-based molecular environmental sciences

XAS spectroscopy has grown rapidly during the past decade to become one of the essential techniques in MES. Evidence for this growth at SSRL is shown in Figure 1, which includes information on the number of MES beam time proposals submitted, the number of eight-hour shifts awarded to MES users, the number of MES publications and important milestones over the past 10 years. This increased MES activity derives from the unique chemical and local atomic structural information for cations and anions provided by XAS studies, and the ability to probe complex natural materials, solutes, amorphous solids and mixtures of phases that may be inaccessible to other techniques. Recently, XAS studies have been widely used to characterise the speciation and chemical forms of metal contaminants, including actinides and other radionuclides [1-3], in environmental samples. Similar measurements have been performed to optimise chemical separation technologies for high-level waste (HLW) [4-6], to develop stable waste forms for HLW disposal [7-9], and to study the effectiveness of *in situ* remediation technologies for contaminated groundwater such as permeable reactive barriers (PRBs) [10]. The utility of the XAS technique has led to demand for beam time by the MES community in excess of the available capacity of SSRL and other US synchrotron facilities.

Figure 1. Growth of molecular environmental science activity at SSRL over the past 10 years

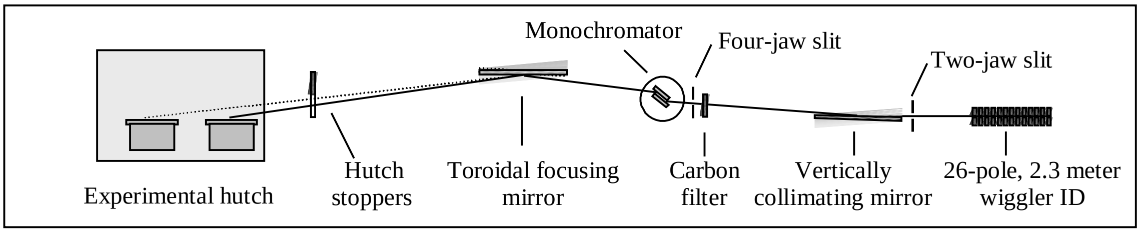


Extension of the XAS technique to demanding problems such as *in situ* characterisation of radioactive and/or nanoscale environmental materials is necessary to fully realise its role in technology and science-based policy development. Meeting these challenges has required development of beamline facilities possessing engineered controls for the safe handling of radioactive samples and optimised specifically for the needs of MES experiments. Chief among these needs are (1) high X-ray fluxes over the range 5-23 keV to provide access to transition-metal K- and heavy-metal and actinide L-edges, (2) high-resolution/high-throughput detector systems for dilute sample fluorescence measurements on chemically heterogeneous samples and (3) high spatial resolution to probe structural and chemical heterogeneities on the micron scale, which are ubiquitous in environmental materials.

Beamline 11-2 concept

Beamline 11-2 was conceived in the context of these issues and designed to provide a dedicated facility for MES XAS measurements. Key aspects of beamline 11-2 are illustrated in Figure 2 and include the following items:

Figure 2. Schematic layout of principle optical components of SSRL BL 11-2



Source

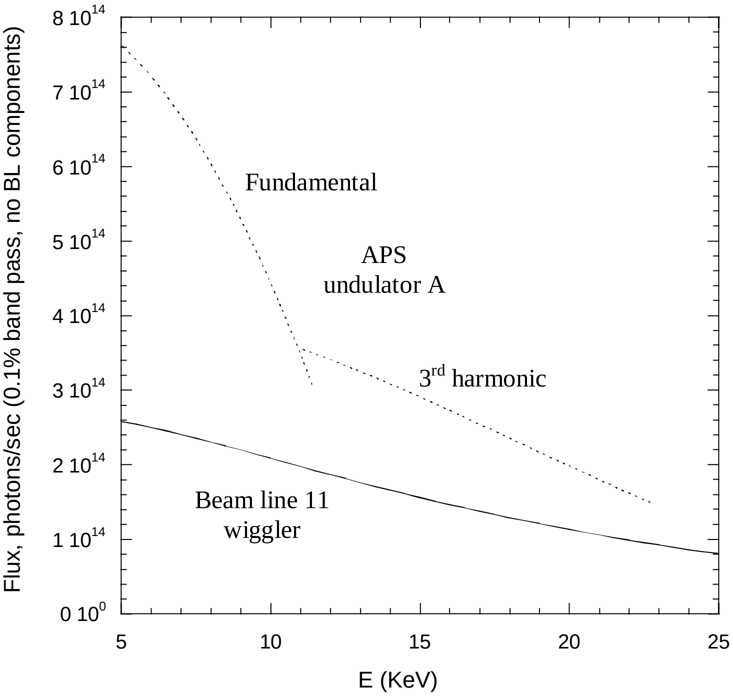
The 26-pole, 2 Tesla hybrid magnet wiggler has a critical energy of 12 keV, providing continuous high flux throughout the 5-23 keV energy range. Figure 3 shows a comparison of the flux from this source on the current SPEAR2 storage ring at SSRL versus an APS undulator A insertion device. When SPEAR3 is completed in late 2003 and is operating at the design current of 500 mA, the flux curve for the BL 11-2 wiggler will be similar to that of an APS undulator A insertion device over the energy range 5-15 keV.

Monochromator

The necessity of stable beam during scanned energy (XAS) measurements drove the design of a mechanically robust, pseudo-channel cut (variable exit geometry), double-crystal monochromator. Intensive cooling, provided by a pressurised liquid nitrogen (LN) closed-loop re-circulating chiller system, is required to minimise thermal deformation of the monochromator crystals under the high power loadings produced by the beamline 11 wiggler. The monochromator chamber contains two pairs of Si(220) crystals (0° and 90° azimuthal angle orientations). These crystals were chosen because of their accessible energy range, energy resolution and user familiarity with their glitch curves. Both pairs of crystals are maintained in vacuum on a rotating turret and can be interchanged via computer control, dramatically improving the efficiency of crystal changes. Commissioning tests performed in June 2000 indicate the LN-cooled monochromator to be remarkably stable; I_0 noise attributable to the monochromator was measured to be $\leq 0.05\%$ during scanned measurements at LN flow rates of 500 L/hr. and 2 atm pressure.

Figure 3. Beamline 11-2 calculated flux at 2 T wiggler field, SPEAR 2 (100 mA), 1.5 mrad horizontal fan and full vertical acceptance

Flux curves for an APS undulator A are shown for comparison, calculated at 7.0 GeV, 100 mA, full central cone. To facilitate a general comparison, calculations do not account for X-ray optic efficiencies or carbon and beryllium windows/ filters. (Beamline 11-2 contains 50 µm carbon and 660 µm beryllium windows.)



Rh-coated reflecting optics

A collimating mirror is located upstream of the monochromator to optimise monochromator energy resolution and provide high energy power filtering. Low-energy power filtering is provided by an insertable graphite filter located between the collimating mirror and the monochromator. A toroidally bent cylindrical mirror located between the monochromator and experimental hutch provides vertically and horizontally focused beam into a spot of dimensions ca 0.4 × 2 mm at 5 to 10 KeV, reducing to about 0.25 × 1 mm at 19 KeV (SPEAR2 storage ring). These beam dimensions should reduce by approximately a factor of 2 when SPEAR3 is fully operational in late 2003. Quasi-focused beam is available from 19 to 23 KeV. A mirror pitch feedback system integrated with the M1 mirror provides automatic correction of vertical beam motion at frequencies ≤ 1Hz. Table 1 lists X-ray fluxes and accessible energy ranges for operation of BL 11-2, together with focused beam size and monochromator energy resolution.

Table 1. Beamline 11-2 characteristics at 2 T wiggler field

Flux (10 KeV)	2.0×10^{13} sec^{-1}
Focused beam size (10 KeV)	0.4 × 2 mm
Energy resolution (10 KeV)	0.65 eV (FWHM)
Focused energy range	4.5-38 KeV
Unfocused energy range	4.5-19 KeV

Hutch instrumentation

As shown in Figure 4, two hutch tables (forward and rear, centred at 27 and 29 m from the source, respectively) are provided for experiments at beamline 11-2. Measurements on radioisotopes will be carried out on the rear table, which is optimally located for focused beam measurements in the 15 to 19 keV range. The hutch is also equipped with a high-resolution/high-throughput 30-element Ge array detector, equipped with digital signal processors under full computer control. The detector is supported by a variable-angle stand that can be positioned in horizontal side-observing and vertical down-observing attitudes. Commissioning tests conducted in June 2000 indicate that the 30-element detector performs well. Detector energy resolution at 0.25 μsec shaping time is *ca* 250 eV FWHM (Zn Kα), and high quality XAS data have been collected from dilute samples at incoming count rates up to 400 KHz. As a feasibility test, U L_{III}-edge EXAFS spectra were measured from a dilute sample (190 ppm U and *ca* 5 000 ppm Sr) collected from an aquifer remediation field site (Figure 5). Due to the low U concentration and overlap between the Sr Kα and U Lα fluorescence lines, it was previously not possible to analyse this sample beyond 6 A^{-1} at SSRL wiggler beamlines used for MES measurements. On beamline 11-2, EXAFS data were collected from this sample (using the 30 element array) up to *ca* k = 12 A^{-1}.

Figure 4. Layout of beamline 11-2 hutch area and radiological controls

Radioactive samples that require continuous monitoring will be analysed on the rear hutch table within Zone 1, which is defined by a small enclosure of approximately 0.5 m^3 volume surrounding the sample space.

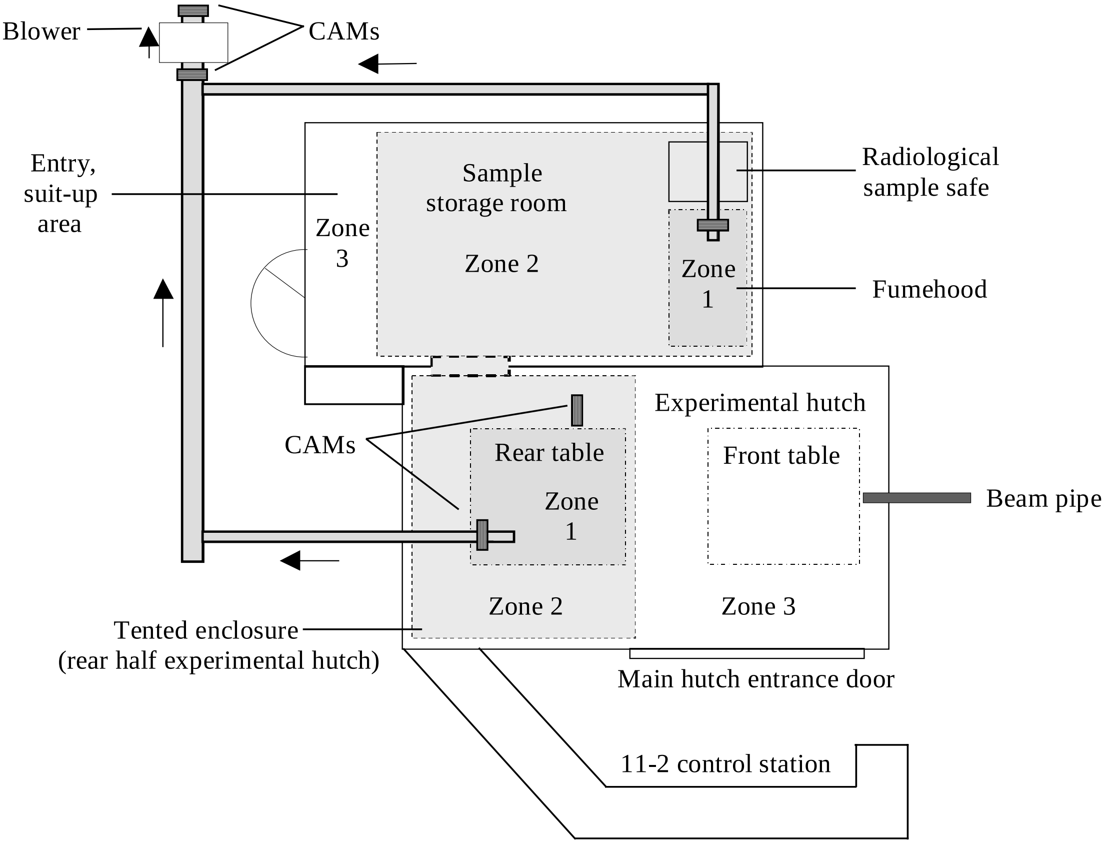
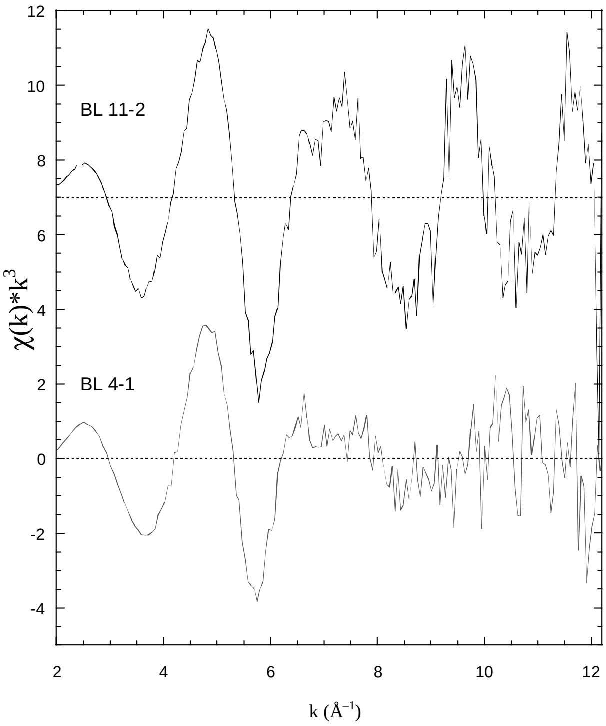


Figure 5. Comparison of background-subtracted, k^3-weighted U L_{III}-EXAFS spectra collected on SSRL wiggler BL 11-2 and wiggler BL 4-1 (10 scans each) from a field sample collected at Fry Canyon, Utah and containing 190 ppm uranium [10]



The beamline 11-2 hutch is also equipped with a spectrometer designed for grazing incidence XAS, reflectivity, and specular XSW measurements. Metal micro-capillary optics that can focus 10^{11} photons/sec into a 5 µM beam spot size using SPEAR2 are currently under design, and will be installed on the forward hutch table. Other detectors and instrumentation available include a liquid-helium cryostat modified to accommodate high X-ray power loadings, a Lytle-type fluorescence detector, transmission ionisation chambers and computer-controlled slits and sample positioners.

Radiological controls at beamline 11-2

A series of radiological controls have been implemented at beamline 11-2 to permit analysis of radioactive samples, while providing an acceptable level of safety to the SSRL community. Primary elements of the radiological controls include engineered barriers (cf. Figure 4), personal protective equipment and administrative measures. Radioactive samples are required to be contained within one to three layers of containment material, depending on isotope class and toxicity. In addition, samples undergoing analysis at beamline 11-2 will be separated from the general laboratory air space by three containment zones defined by physical enclosures, through which air flow is controlled by negative pressure gradients. Continuous air monitoring (CAM) of the ventilation system and use of high efficiency particulate air (HEPA) filters insure that an accidental release of radioactivity in the hutch can be detected in real time and radioactive material entrapped, thereby minimising the impact to other SSRL operations. Activity levels are monitored at multiple points along the ventilation system. Critical monitoring points include the entry point to the system (i.e. above the sample) and immediately upstream and downstream of the HEPA filters.

A sample inventory/storage room is located immediately adjacent to the beamline 11-2 hutch to accommodate sample unpacking/repacking, inspection, containment preparation and activity surveying in a controlled, monitored environment. Samples can be transported into the beamline 11-2 experimental hutch via a direct doorway, obviating the need to enter the communal SSRL air space with prepared samples. In addition to the alarmed CAM system, the beamline is equipped with hand-held radiation monitors, swipe counters, hand friskers and a variety of accessory probes to accommodate detection of α, β and γ radiation.

Isotope quantity limits and administrative controls

Radiological controls required for a given experiment are based primarily on the isotope to be studied, its physical form and the total isotope quantity required. Upper limits on isotope quantities have been established by SLAC, based on the US Department of Energy standards (document DOE-ST-1027-92). These limits generally exceed the isotope quantity necessary to conduct an experiment by one or more orders of magnitude. The total radiation levels allowed at SSRL at any particular time are limited to half the limit of a DOE Category 3 Non-nuclear Facility (e.g. up to 260 mCi (4.2 g) of ^{239}Pu or up to 210 mCi (300 g) of ^{237}Np). Users are urged to plan experiments based on the minimum isotope quantities required. This procedure is key to minimising the total on-site quantity of isotopes and associated risks to the SSRL community and facility. Based on such an analysis and in conjunction with the experimenter, SSRL establishes isotope quantity limits specific to the individual experiment. Forms and guidelines relevant to this process can be found on the web at http://www-ssrl.slac.stanford.edu/safety/.

Acknowledgements

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INSTRUMENTAL DEVELOPMENTS FOR XAFS ANALYSIS OF DILUTE ENVIRONMENTAL SAMPLES AT HASYLAB

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Abstract

In many environmental samples the detection limit of XAFS analyses is less determined by the flux of the X-ray source than by the detector concept. Two major problems restrict the detection limits on the detector side. First, the fact that a detector must initially count all photons, including those originating from the matrix, meaning that the detector might be in overflow although the number of photons from the analyte is still small. Second, the limited energy resolution of conventional energy-dispersive detectors causes overlaps between neighbouring emission lines. Both problems can be overcome by use of a secondary monochromator which filters out unwanted photons before they reach the actual detector. The design and projected abilities of a planned secondary detector for analyses in dilute (environmental) samples at HASYLAB will be described in the following.

Introduction

Over the past few years XAFS spectroscopy has turned out to be a valuable instrument for the determination of the chemical form of metals in environmental samples. The chemical form of metals, radioactive and non-radioactive, has been investigated in real environmental samples, mainly by fingerprint methods [1,2] and in synthetic environmental samples. The latter investigations aim at the exploration of the interaction of metals of interest with single important constituents of environmental samples like clay, iron and manganese minerals or different organic compounds. In either case the concentration of the analyte is normally small. That means that XAFS spectra from these samples have to be measured in diluted samples. The most often used technique to do this is to measure the spectra using the fluorescence yield technique (Fl-XAFS). The detectors used are (multiple) energy-dispersive semi-conductor detectors.

Conventional energy dispersive X-ray detectors must measure all photons coming from the sample, including the elastically scattered photons and fluorescence photons from matrix elements. This fact results in three major problems: First, the elastically scattered photons produce a background which, to make it worse, changes its value during an energy scan (see Figure 1, right). Second, the detector is in overflow although the number of the interesting photons is still small (Figure 1). Third, it is sometimes impossible to separate neighbouring emission lines (see Figure 2), because the FWHM of an emission line is between ~130 and 1 000 eV, depending on the integral count rate and the resulting necessary shaping time.

As a result of the described problems is the detection limit in many cases more strongly determined by the detector than by flux from source. A better detector should avoid the mentioned problems by excluding the background photons from the actual counting device.

Secondary monochromator

A secondary monochromator for XAFS analysis of diluted samples should fulfil two major tasks. First it must offer an energy resolution which is large enough to cut off the emission lines from neighbouring matrix elements and from the elastically scattered photons. The bandwidth on the other hand should not be smaller than the width of the interesting fluorescence peak. So typically a bandwidth of roughly 10 eV seems suited. The second task a secondary monochromator should fulfil is that it should increase the used solid angle, meaning that the analysator design has to focus the emitted photons on the detector.

Figure 1. Pb *L3* Fl-XAFS measured in a sample from the cinder disposal of a metal smelter facility

c(Zn) = 116.0 g/kg, c(Pb) = 42.5 g /kg, c(Cu) = 8.2 g/kg
Left total count rate spectra, right energy-filtered spectra

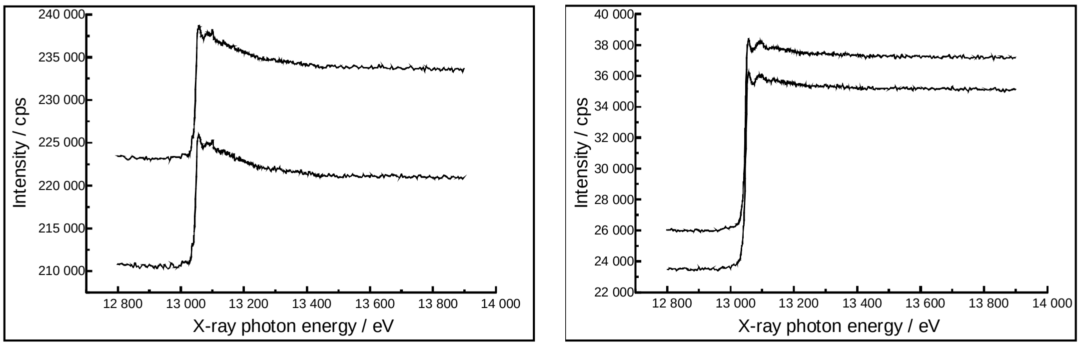
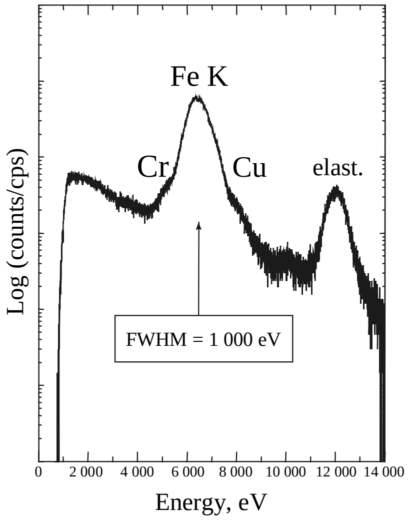


Figure 2. Fluorescence spectrum of a sample which contained small amounts of Cu and Cr in a Fe matrix, measured with a "conventional" energy dispersive detector (Ge 5 Pixel) at high count rates (200 kcps)

Note that the energy of the incident beam (elast.) is much higher than it would be during a XAFS measurement



An often-used design that fulfils both tasks mentioned above is the Johann Geometry. The Johann Geometry is based on the Rowland circle, meaning that sample, analysator crystal and detector are positioned on a circle with radius R. In the Johann Geometry a bent analysator crystal with a bending radius of 2R is employed. This leads to focusing on the detector.

In contrary to the more common design with sample, analysator crystal and detector on the circumference of the Rowland circle [3], the sample will be positioned inside the circumference of the Rowland circle (see Figure 3). This enables to work with an illuminated area on the sample, thus minimising the risk of radiation damage at high flux beamlines and the need for homogenous samples [4]. In contrast, the "classical" design works with a spot source on the Rowland circle and the energy resolution depends strongly on the spot size. Both designs are currently used at synchrotron radiation sources in several different fields of science, mostly for X-ray Raman spectroscopy [5-10]. If for one reason or another spot measurements are necessary, it is possible to mount the sample on the circumference of the Rowland circle. The large flexibility of the design can only be used with a 2-D detector, therefore a CCD chip will be used as detector.

Figure 4 shows the broadness of the energy band that can be mapped on a 3 cm wide CCD detector over the Rowland radius (R_{row}). Together with the chosen crystal and the resulting Bragg angle, R_{row} determines the achievable energy resolution and limits the achievable bandwidth. However even with a Si(111) crystal working at an incident angle of roughly 20° the energy resolution is better than 1 eV/pixel (for a detector with 2 000 pixels in a row and an R_{row} of 0.5 m).

Figure 6 shows a schematic top view of the proposed detector system. Four or five crystals are mounted on a crystal changer, because it is necessary to use different crystals for different resolutions and energies. The whole secondary monochromator is installed in an elliptical-shaped steel tank to enable working under UHV conditions if necessary. The Rowland circle, which is defined by the analysator crystal and the detector, can be moved along the long axis to adjust the distance between the fixed sample and the analysator crystal. This enables measurements with different solid angles and energy resolutions. The analysator crystals used are either spherically (for large solid angle) or cylindrically (for high-energy resolution) bent.

Figure 3. Ray paths for the proposed secondary monochromator

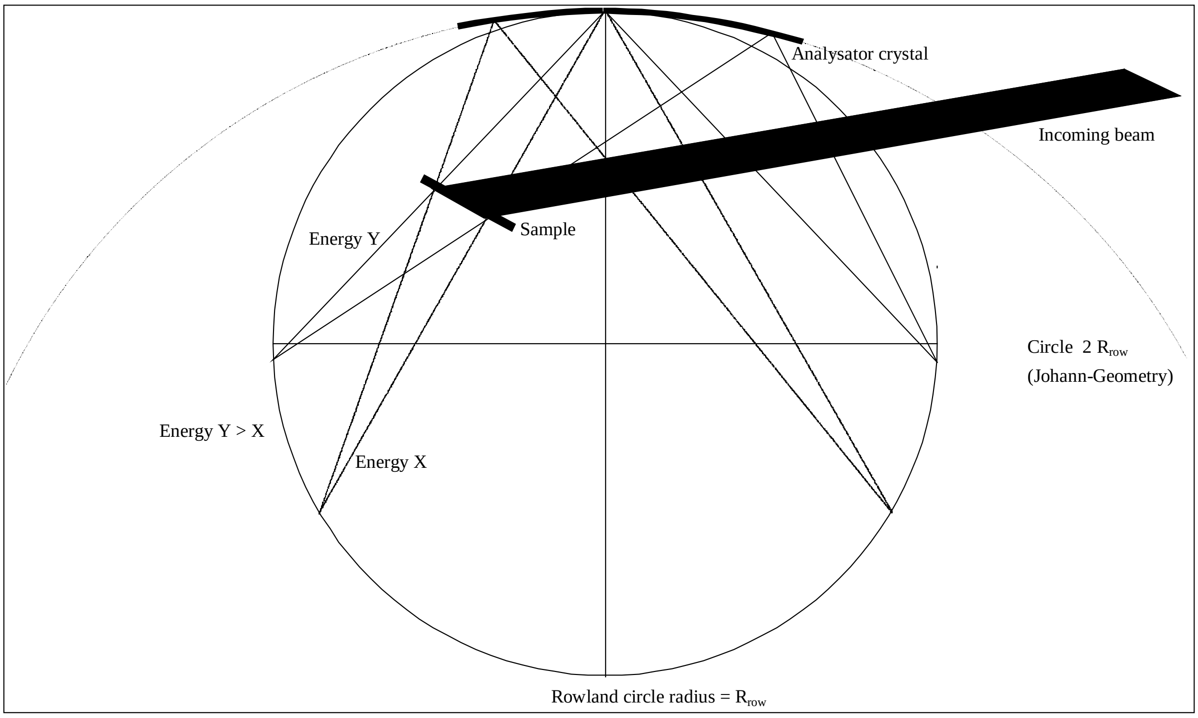


Figure 4. Energy range mapped on a 3 cm wide CCD detector vs. Rowland radius, Si(111), 6 000 eV, θ = 19.2°

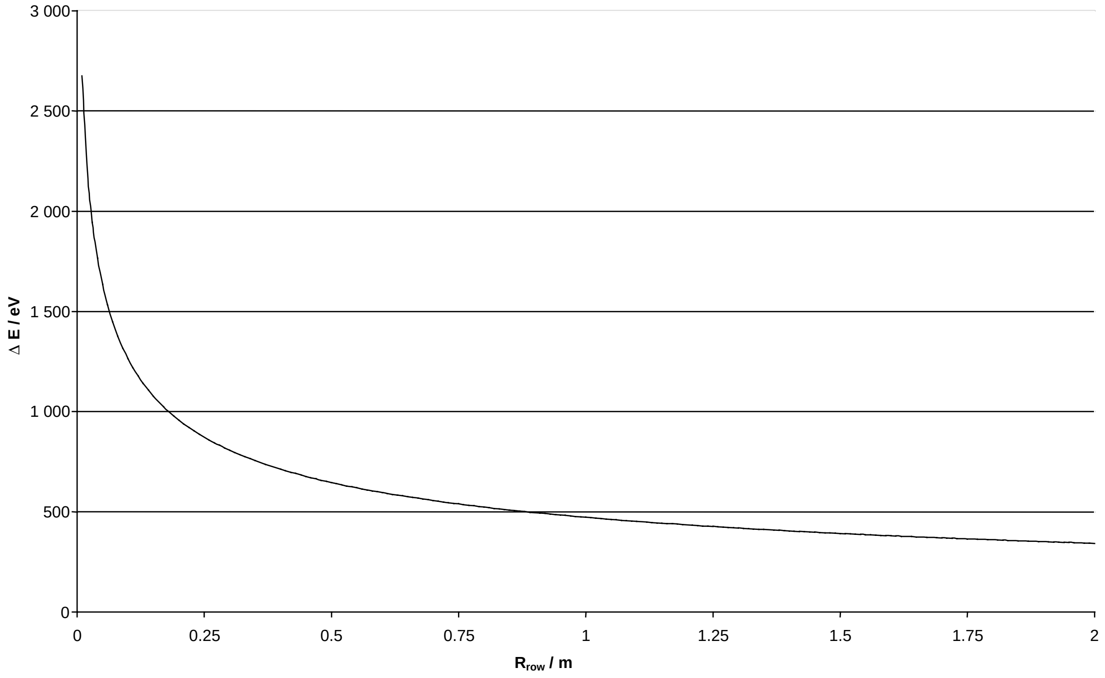


Figure 5. Energy range mapped on a 3 cm wide CCD detector vs. Rowland radius, Si(111), 6 000 eV, θ = 19.2°

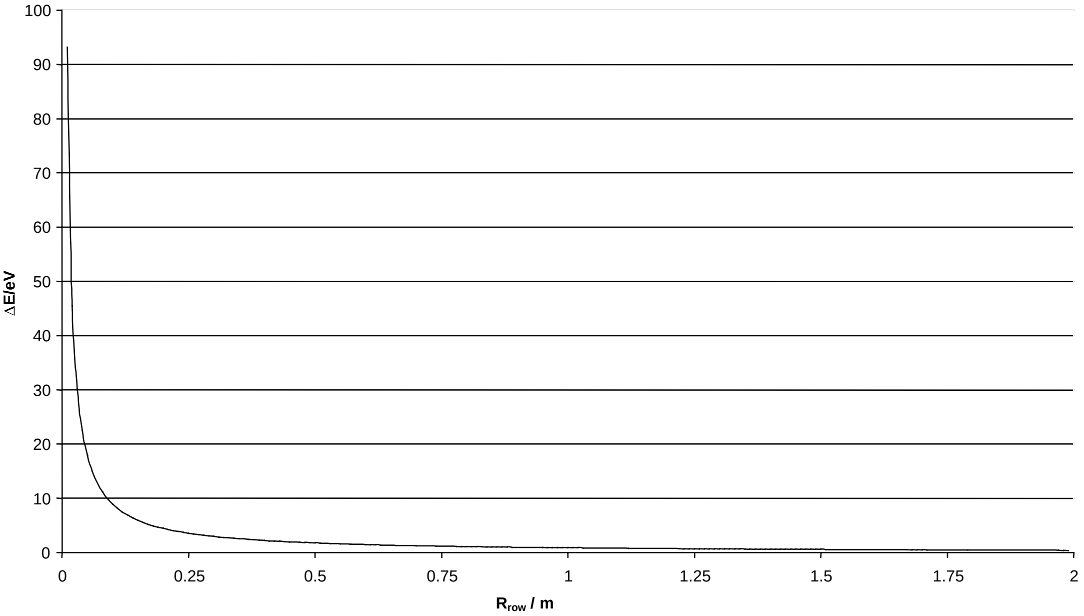
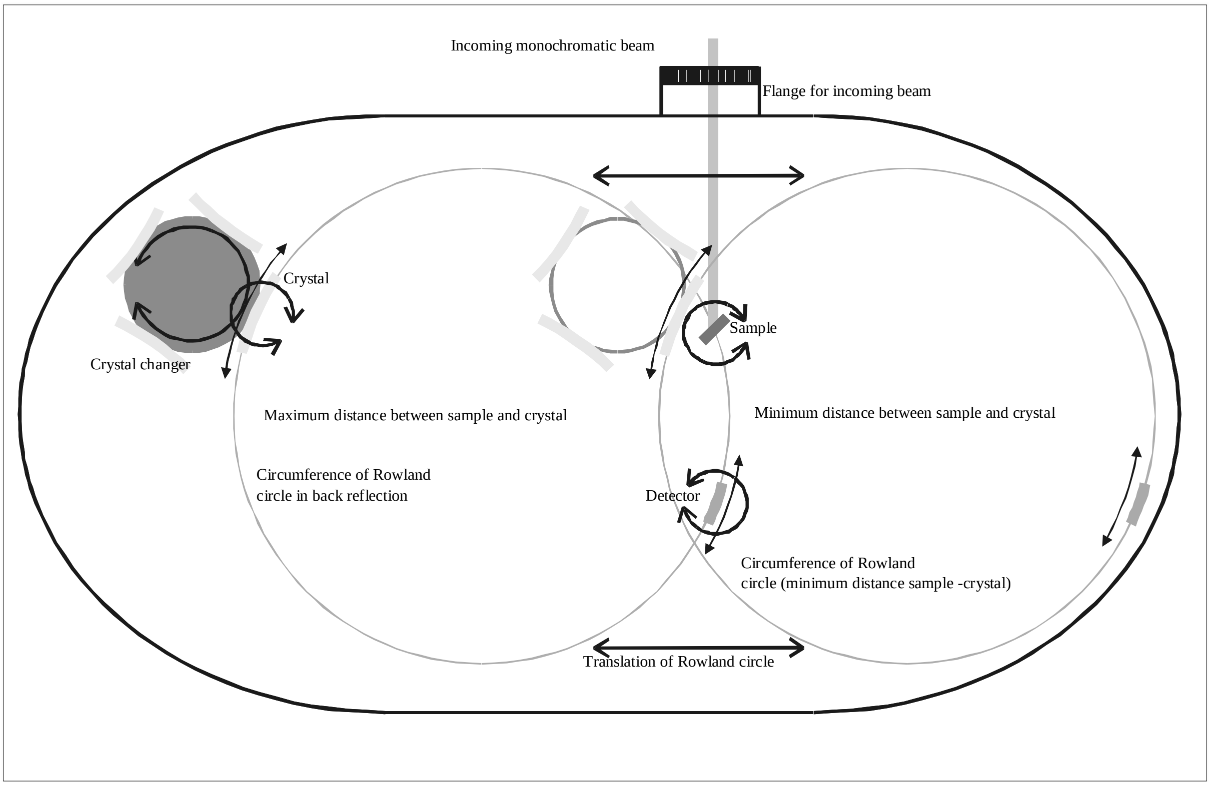


Figure 6. Top view of the principal design of the proposed second monochromator



Possible use in other research fields

Due to the large flexibility of parameters such as energy resolution, band width, illuminated area, etc., the detector can be used in several other scientific fields. Examples are methods like non-resonant inelastic X-ray scattering (X-ray Raman spectroscopy) or resonant inelastic X-ray scattering (RIXS). This might offer some additional fields of work with environmental samples. X-ray Raman spectroscopy for instance enables the measurement of the K-edge "absorbance spectra" of light elements like C, N and O in the hard X-ray region. That means under ambient conditions (pressure, humidity, etc.) [10].

Conclusion

A secondary monochromator which uses a modified Johann geometry seems suited to overcome the problems which arise in the measurement of Fl-XAFS in highly diluted samples. This detector design offers, aside from the expected considerable decrease of the detection limits in speciation analysis, some potential use in different areas of physics.

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SYNCHROTRON ENVIRONMENTAL LABORATORY (SUL) AT ANKA

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Abstract

A research facility dedicated to environmental/geochemical research, the Synchrotron Environmental Laboratory (SUL), is planned to be installed and operated at ANKA. ANKA is the new synchrotron facility at the Research Centre Karlsruhe (FZK), Karlsruhe, Germany. ANKA is now in commissioning and planning operations for the fall of 2000.

As the Institute for Nuclear Waste Disposal (INE) at FZK conducts a vigorous synchrotron-based research programme, INE was instrumental in the original impetus for installing such a facility at ANKA. These research activities at INE concentrate on actinide speciation in nuclear waste forms, geological media and geochemical model systems. In order for INE to direct their synchrotron research activities to ANKA, equipment and licensing required for performing experiments on actinide-containing samples is required. One great advantage of performing experiments on actinide-containing samples at ANKA is that the INE radiological laboratories lie in the near vicinity of the facility. This will minimise transport hazards and costs and allow experiments to be performed on samples whose characteristics may change with time.

Experiments on radioactive samples with activities below the exemption level, according to German regulations, will be possible at ANKA at the start of operations. Licensing for work on higher levels of activity will be applied for in the future. The decades of experience in radiological work at FZK will facilitate development of procedure and equipment as prerequisites to licensing.

A consortium of synchrotron radiation-user groups with environmental research interests has specified their requirements and needs for this facility. This scientific case serves as the foundation for the SUL design and is the basis for an application for federal funding. The SUL design reflects the heterogeneity and complexity of challenges facing researchers in the environmental/geochemical sciences. X-ray absorption fine structure (XAFS) speciation methods, applications of micro-focusing for studies on heterogeneous systems and imaging techniques as well as synchrotron infrared spectroscopy are planned to be available.

BEAMLINE PROJECT TO STUDY RADIOACTIVE MATERIALS AT SOLEIL SYNCHROTRON RADIATION SOURCE

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Abstract

In the perspective of the construction of the new French synchrotron radiation facility SOLEIL, the scientific council decided that a beamline dedicated to study radioactive materials would be erected. Based on collaboration between different scientific groups, a multi-purpose beamline has been chosen regarding the needs of each. Either XAS or diffraction experiments could be performed. Special attention will be paid to the focusing beam in order to reduce the radioactivity by studying small samples or to probe local atomic arrangements.

This presentation will report on the conceptual design of the station including ray-tracing studies and layout.