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**NUCLEAR RESONANT SCATTERING BEAMLINE
AT THE ADVANCED PHOTON SOURCE (*)**

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ABSTRACT

The principal and engineering aspects of a dedicated synchrotron radiation beamline under construction at the Advanced Photon Source for nuclear resonant scattering purposes are explained. The expected performance in terms of isotopes to be studied, flux, and timing properties is discussed.

INTRODUCTION

The use of synchrotron radiation was first discussed by Ruby (1) in 1974. After a decade of many attempts (2, 3), Gerda and others performed the first successful experiment (4), then followed by many experiments (5-7). All of these early experiments were performed with very low count rates. Nevertheless, these experiments proved that this is a viable technique and made it easier to justify dedicated beamlines on the third generation synchrotron radiation sources. Today, the validity of using synchrotron radiation instead of radioactive sources has been proven for ^{57}Fe (4), ^{169}Tm (8), and ^{119}Sn (9), and the count rates exceeded 10^4 Hz (10). It now appears that dedicated nuclear resonance scattering beamlines will be available at the European Synchrotron Radiation Facility (ESRF) in Grenoble France in 1994, at the Advanced Photon Source (APS) at Argonne, Illinois, USA, in 1996, and at the new synchrotron facility in Harima Science City (Spring-8) in Japan by 1998. In addition, there is already a station at Hamburg Synchrotron Radiation Laboratory (HASYLAB) in Hamburg, Germany and at the National Synchrotron Light Source (NSLS) in Brookhaven, New York, USA, on a bending magnet source. Another station in HASYLAB on a wiggler source is being commissioned. In this paper, we will present the design aspects of a beamline at the APS.

THE ADVANCED PHOTON SOURCE

The APS construction project started in 1990, and it is scheduled to be completed in 1996 (11). The distinctive characteristics of this storage ring compared to previous ones is its low particle beam emittance and its widely tunable undulator sources. The machine parameters are tabulated in Table 1. The expected normal operating condition is 100 mA of current with positrons distributed into 20 evenly spaced sites. This filling system allows 185 ns bunch-to-bunch separation. With special fills, it is possible to fill almost any configuration. However, the current is limited to 5 mA per bunch. The general layout of the ring is given in Figure 1.

Figure 1. General layout of the Advanced Photon Source.

Figure 2. General layout of the high energy resolution x-ray scattering beamline.

Table 1. The machine parameters of APS (11)

Linear Accelerator	Storage Ring
200 MeV e^- (1.7 A)	7.0 GeV, 100-300 mA
450 MeV e^+ (8 mA)	Lattice: Chasman-Green (40 periods)
Repetition rate: 60 Hz	Radio frequency: 351.929 MHz
Length 40 m	Circumference: 1104 m
	max current per bunch: 5 mA
	Harmonic No: 1296
Accumulator Ring	Natural emittance: 8.2×10^{-9} m-rad
450 MeV DC ring	Natural bunch length: 27.5 ps (FWHM)
24 linac pulses in 0.5 sec	Max. bunch length: 72.5 ps (FWHM)
Circumference: 30.67 m	Filling time: 0.9 min (100 mA)
Natural emittance: 0.37 mm-mrad	Revolution time: 3.683 μ s
	Revolution frequency: 351.93 MHz
Booster Synchrotron	Max length of insertion device: 5.2 m
450 MeV to 7 GeV in 0.25 s	Bending field: 0.599 T
Repetition rate: 0.5 s	Bending radius: 38.9611 m
Circumference: 367 m	Injection energy: full energy
Radio frequency: 351.929 MHz	Ave. beta function: $\beta_x=10$ $\beta_y=14.2$ m
Bunch length: 61 ps	Max. beta function: $\beta_x=24.1$ $\beta_y=21.4$ m
	Beam size (rms): $\sigma_x=0.34$, $\sigma_y=0.09$ mm
	Max. dispersion : 0.4 m

Table 2. Undulator parameters

Period:	2.8 cm
Length:	2.5 m
Number of poles:	89
Positron beam emittance (m-rad)	
Horizontal, β_x :	8.2×10^{-9}
Vertical, β_y :	8.2×10^{-9}
Positron beam size (standard deviation, μ m)	
Horizontal, σ_x :	342
Vertical, σ_y :	91
Positron beam divergence (standard deviation, μ rad)	
Horizontal, σ'_x :	24
Vertical, σ'_y :	9
Total photon size (standard deviation, μ m) :	
Horizontal, Σ_x :	342
Vertical, Σ_y :	91
Total photon divergence (standard deviation, μ rad)	
Horizontal, Σ'_x :	26
Vertical, Σ'_y :	14
K _{min} - K _{max}	0.5-1.4

NUCLEAR RESONANT SCATTERING BEAMLINE

The nuclear resonant scattering beamline is being constructed as part of an effort to develop x-ray optics for high energy resolution x-ray scattering in the hard x-ray regime ($E > 4$ keV). The beamline is shared with a meV-resolution inelastic scattering studies station. The layout of the beamline is given in Figure 2. The distance between the undulator and the experimental station is 67 m. The unfocused x-ray beam at this point is about 1.8 mm high and 3.5 mm wide.

The main components for the nuclear resonant scattering beamline are a) an undulator, b) a high heat load monochromator capable of handling large power loads (up to 4 kW, with a power density of 140 W/mrad^2), c) a focusing mirror, d) a high energy resolution monochromator, e) nuclear monochromators, and f) detectors and electronics.

a) Undulator

The device tentatively chosen for the APS high energy resolution beamline is a hybrid 2.5 m long undulator with a 2.8 cm magnetic period delivering 14.4 keV radiation in the first harmonic. The calculated performance of this device is given in Figure 3 (a). It is tunable between 6 and 16 keV in the first harmonic assuming a minimum gap of 10.5 mm. The important aspect of this device is the ability to deliver 14.413 keV x-rays in the first harmonic. The vertical divergence of the x-rays emitted at this energy is $28 \mu\text{rad}$, and the horizontal divergence is $52 \mu\text{rad}$ (FWHM). It will deliver 2.4×10^{13} photons/sec/eV at 14413 eV, for 100 mA stored in the ring with a positron energy of 7 GeV. This flux corresponds to 115,000 photons per Mossbauer linewidth of ^{57}Fe isotope ($1\Gamma = 4.8 \text{ neV}$).

An important aspect of an undulator source versus a wiggler source is shown in Figure 3 (b). Here, the flux through a pinhole of $20 \times 20 \mu\text{rad}$ is calculated. This pinhole size is chosen to represent the angular acceptance of the crystal monochromators and so that diffraction takes place both in the vertical and the horizontal planes (as is the case for the polarizer/analyzer monochromator to be discussed later). The flux obtained from an undulator is about 500 times more than that from a wiggler when observed through a small pinhole.

b) High Heat Load Monochromator

The first optical component in the beamline after filters and slits is the high heat load monochromator. It is placed 29 m from the source point. The beam incident on the first crystal of this double crystal monochromator may have up to 4 kW of power, depending on the gap of the undulator. Some of this power is in the lower energy range, and can be absorbed using graphite filters. Even then, it is necessary to cool the first crystal. Water, liquid gallium, and liquid nitrogen can all be used as coolants. Each coolant has some distinct advantages. Water is the cheapest and the easiest to implement; liquid gallium is the most efficient. Liquid nitrogen, on the other hand, has a unique advantage: the thermal expansion coefficient of silicon is zero around 125 K. The monochromator planned for the APS is to be cooled using liquid Ga and specially

Figure 3 (a). The calculated tunability of an undulator with a 2.8 cm period. The vertical and horizontal acceptance depends on the experimental conditions. The ring energy is 7 GeV and the current is 100 mA.

Figure 3 (b). The calculated photon flux of undulator with 2.8 cm period, $K=0.55$, and the wiggler A with a period of 8.5 cm, $K=7.9$ through a pinhole $20 \times 20 \mu\text{rad}$ (vertical and horizontal) at APS.

Table 3. The Mossbauer isotopes within the tunability range of APS undulator, their transition energies, half-life times, and natural energy widths

Isotope	Energy (eV)	Half-life (ns)	Γ (neV)
^{181}Ta	6238.	9800.	0.067
^{169}Tm	8401	4.	114.0
^{83}Kr	9400.	147.	3.1
^{73}Ge	13263.	2953.	0.15
^{57}Fe	14413.	97.8	4.67
^{151}Eu	21532.	9.7	47.0
^{149}Sm	22490.	7.1	64.1
^{119}Sn	23870.	17.8	25.7
^{161}Dy	25655.	28.2	16.2
^{40}K	29560.	4.25	107.0

Table 4. Nested monochromators and the expected energy resolutions, ΔE , and angular acceptances, $\Delta\Theta$, for several Mossbauer isotopes.

Isotope	Energy (eV)	Crystals	Asymmetry angle (deg)	ΔE (meV)	$\Delta\Theta$ (μrad)	Reference
^{169}Tm	8410	Si (3 3 3) Si (4 4 4)	$\alpha = -40$ $\alpha = +66$	15	30	(17)
^{57}Fe	14413	Si (4 2 2) Si (10 6 4)	$\alpha = -20^\circ$ $\alpha = 0^\circ$	10	27	(15)
^{119}Sn	23870	Si (3 3 3) Si (5 5 5)	$\alpha = -12^\circ$ $\alpha = 0^\circ$	40	7	(9)

Table 5. The calculated angular acceptance and the degree of linear polarization attainable for ^{169}Tm and ^{57}Fe Mossbauer resonances.

Isotope	^{169}Tm			^{57}Fe		
Energy (eV)	8410			14413		
Reflection	Si (3 3 3)			Si (8 4 0)		
Bragg angle (degrees)	44.7			45.1		
asymmetry angle, a (deg)	1/b	$\Delta\Theta$ (μrad)	S^* (10^{-7})	1/b	$\Delta\Theta$ (μrad)	S^* (10^{-7})
0	1	8.3	1.1	1	1.8	6.19
-40	12.1	26.4	0.28	11.2	6.0	0.33
-43	33.6	45.6	0.10	27.2	9.3	0.12

designed crystals with an optimized cooling channel geometry (12). The crystals can be cut asymmetrically so that the angular acceptance of the Bragg reflections matches the angular divergence of the incident radiation. Typical losses in this monochromator are expected to be anywhere between 0.8 to 0.5, depending on many factors including the quality of the crystals and their preparation, the degree of asymmetry, and the effectiveness of the cooling.

c) Mirror

A vertical and horizontal focusing mirror is located at the 39 m point to bring the beam size close to the source size. In the case of the APS, the vertical and horizontal source sizes are 0.18 and 0.68 mm (FWHM), respectively. The focusing will be particularly useful for reducing the length of grazing incidence optics. (Thin films and multilayers operate in the range of 2-15 mrad.) When placed after the monochromator, the mirror is not subjected to high heat loads and therefore cooling is not necessary. The mirror will have an adjustable vertical focusing length, and hence the photon beam divergence can be traded with the beam size. For Mossbauer isotopes with transition energies above 20 keV, collimation proves to be more important than focusing, as the angular acceptance of Bragg reflections of Si single crystals decrease to below 10 μ rad.

d) High Energy Resolution Monochromator

The first optical component inside the hutch is a double channel-cut, nested monochromator, located about 67 m from the source point. The typical energy bandpass of the high heat load monochromator is around 2 eV for 14413 eV x-rays. Compared to the 5 neV linewidth of ^{57}Fe , this represents a signal-to-noise ratio of 2.5×10^{-9} . Further reduction in energy bandpass is needed to perform the experiments satisfactorily. A recent improvement in this area is the use of high order Bragg reflections, combined with asymmetrically cut crystals to improve the angular acceptance (13). Ishikawa and others (14) have proposed a nested monochromator consisting of asymmetrical and symmetrical channel-cut crystals. Such a monochromator has been built and successfully tested (15). The energy resolution and angular acceptance achievable with different Mossbauer isotopes are tabulated in Table 1. Here the asymmetry angle α is used to indicate the angle between the actual crystal surface and the Bragg planes, following well established conventions (16). The throughput of the monochromator is better than 60% at 14413 eV.

However, even with a high energy resolution monochromator, there is a strong limitation to forward scattering experiments (18). Typically, the detectors and the associated electronics saturate at around 10^6 Hz count rates. This corresponds to approximately 10^8 Hz per eV in a 10 meV bandpass. The available photon count rate exceeds this number by at least four or five orders of magnitude. Therefore, a different approach is necessary to increase the Mossbauer count rates. One alternative idea based on the strong optical activity of Mossbauer medium is presented below.

The angular acceptance of perfect crystals is dependent on the polarization of the incident beam. This is given by the cosine of twice the Bragg angle for π -polarized radiation and is unity for σ -polarized radiation. (In σ (π) polarization the electric field vector is perpendicular (parallel) to the scattering plane defined by the incident and the scattered beam.) For a Bragg angle near 45° , the π component will have an angular acceptance close to zero. Crystals with Bragg angles near 45° are used to design polarizers in the hard x-ray regime (19). For example, Si (333), Si (840), and Si (12 6 6) each provide a near- 45° Bragg angle for the ^{169}Tm , ^{57}Fe , and ^{119}Sn isotopes, respectively. The proposed experimental setup is shown in Figure 5. A channel-cut monochromator (P) polarizes the incident beam to 1 part in 10^4 - 10^7 , and thus an analyzer crystal (A) of the same reflection placed perpendicularly to the first one will suppress the σ -polarized beam at the same level. If a σ -to- π scattering process takes place in between the polarizer and analyzer, then the π component will pass through the analyzer.

Nuclear transitions cause strong σ -to- π resonant scattering, and this can be filtered at the expense of the nonresonant part, eliminating the overwhelming initial flux. In a recent experiment, Siddons and others have proven the principle aspects of this scheme (20). However, scattering in the horizontal plane is not desirable for a bending magnet or a wiggler source providing radiation with a horizontal divergence of few milliradians, costing more than three orders of magnitude in flux. On the other hand, with an undulator source, the vertical and horizontal angular divergences are about 28 and 52 μrad , respectively. Furthermore, the angular acceptance can be increased by asymmetrically cutting the channel-cut crystals. The results of such an exercise are given in Table 5. Here S is the square of the ratio of the area under the π -reflectivity curve to that under the σ -reflectivity curve indicating the degree of possible linear polarization using a channel-cut crystal with two bounces. The conclusion is that with undulator sources, one can provide a polarizing monochromator with a nonresonant background rejection ratio of 10^{-8} , and still obtain reasonable flux at the detector.

e) Nuclear Monochromators

The last step of monochromatization is based on the narrow energy linewidth of Mossbauer nuclear resonance. Under suitable conditions, the reflectivity of such media can be enhanced for nuclear resonant photons only at the expense of photons outside the resonance line widths. Hence, further monochromatization can be achieved either by diffraction (1,4-8), or reflection (21), or perhaps by focusing (22). Such media include single crystals and artificially synthesized multilayers, where reflectivity is based on whether the incident photon is resonant with the medium or not. For example, a single crystal of yttrium iron garnet (YIG) (13,15), Fe_2O_3 (14,16) FeBO_3 (19) or thulium iron garnet (TIG) (20) can be oriented so that the electronic reflection is forbidden, while the nuclear reflection is allowed. Such crystals have been used so far very successfully. However, these crystals are hard to grow, and they are limited in number. In addition, the nuclear levels are split by internal hyperfine fields, resulting in a beam that has time beats complicating the data analysis. In this sense, GIAR films and multilayers are very promising. A medium containing periodic and alternating layers of resonant and nonresonant material will reflect or diffract the resonant radiation with some efficiency.

Grazing Incidence Anti Reflection (GIAR) Films:

The sharp contrast between the electronic and nuclear indices of refraction allows one to design monochromators in which the electronic reflectivity is sharply reduced at a specific angle, while the nuclear reflectivity is still substantial. Such materials have been proposed (21), synthesized (23,24), and tested. The large difference in electronic and nuclear absorption cross sections provides an opportunity to adjust the thickness of the thin film layers such that x-rays reflected from electron charge interfere destructively at a given angle, while there is still appreciable reflectivity for x-rays resonantly scattered by the resonant nuclei. This method, so far, has been successfully applied to ^{57}Fe (23) and ^{119}Sn (9). It is clear that GIAR nuclear monochromators are capable of producing μeV resolution, which may prove to be very useful in inelastic scattering studies both as monochromators as well as absorbers. Currently, we have plans to implement the beamline with ^{119}Sn - and ^{57}Fe -based GIAR films.

Multilayer Structures

An alternative to perfect crystals with an electronically forbidden, nuclear-allowed reflection is the idea of multilayers alternately containing nuclear resonant/nonresonant atoms. This arrangement periodically varies the index of refraction for nuclear resonant radiation with a uniform index of refraction for the nonresonant part of the radiation (25). The Bragg angle can be adjusted by adjusting the layer thickness. The limiting factor is the interface roughness, which needs to be minimized with respect to layer thickness. The degree of suppression one may get is of a factor of 100 to 1000 depending on the relative spatial extent of the true multilayer structure. One consequence of higher incident angles is the narrower energy width (10-40 Γ) compared to that with the GIAR case. In addition, the operating angle is adjustable by changing the periodicity of the layers. One advantage of multilayers over GIAR films is that long films are not necessary due to higher operating angles. In conclusion, the ability to synthesize artificial optics for many different Mossbauer isotopes and an in-depth understanding of the response of such media will be very useful for many different applications.

APPLICATION RELATED HARDWARE

The design philosophy of this beamline is to provide as much flexibility as possible for future experiments. Therefore, the size of the experimental station is chosen to be relatively large: 4.5 m wide, about 8 m long, and 3 m high. The main instruments are :

1. high resolution monochromator,
2. energy calibration set-up
3. 6-circle diffractometer,
4. polarizer/analyzer monochromator
5. forward Scattering Experiments table

The high resolution monochromator is explained above. Currently, the construction of crystals for ^{169}Tm , ^{57}Fe , and ^{119}Sn resonance are complete. The energy calibration will be done with the Bond method, that is by accurate measurement of the angle difference between two reflections. In addition to standard a Huber 6-circle diffractometer, there will be a separate experimental table equipped with a polarizer/analyzer monochromator to facilitate forward scattering and interferometry experiments for Mossbauer spectroscopy work. A sample cryostat is planned with temperature adjustable between liquid He and room temperature equipped with a superconducting split coil to apply large magnetic fields is planned. For data analysis, the CONUSS program package (26) will be available, with standard data reduction routines. If multilayer or GIAR monochromator is used, the deconvolution programs will also be available.

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