1 Newly Developed Experimental Facilities

1-1 A New Protein Crystallography Beamline BL-5

Outline

BL-5 is a new beamline at the PF 2.5 GeV ring designed for high-throughput protein crystallographic experiments. The design of the beamline is optimized for efficient multiwavelength anomalous dispersion (MAD) experiments similar to that of PF AR-NW12 constructed in the year before, the report of which is in the preceding Activity Report [1,2]. The precision of the sample axis and the size of the active area of the X-ray CCD detector are superior to those in AR-NW12, so that data collection using micro-crystals with large cell dimensions at high resolution is particularly suitable. The beamline has been constructed during FY2003. After the observation of the first beam (September 29, 2003), the commissioning of the beamline components and then test experiments by outside users were carried out. It has been opened for public use from April of FY2004.

Light source and optics

Fig.1 shows the design of the beamline from top and side views. The location and the specification of the main optical components are summarized in Table 1. Compared to AR-NW12, the main differences are (1) multipole wiggler (MPW) as the light source (insertion device) and (2) the micro-channel direct water cooling system for the monochromator crystal. The beam from the MPW is trimmed by the front-end slits to limit the divergence to 0.5 mrad (H) × 0.24 mrad (V), and passed through the collimating mirror, the double crystal monochromator and a focusing mirror before delivered at the sample position. The focal point is 250 mm downstream from the sample

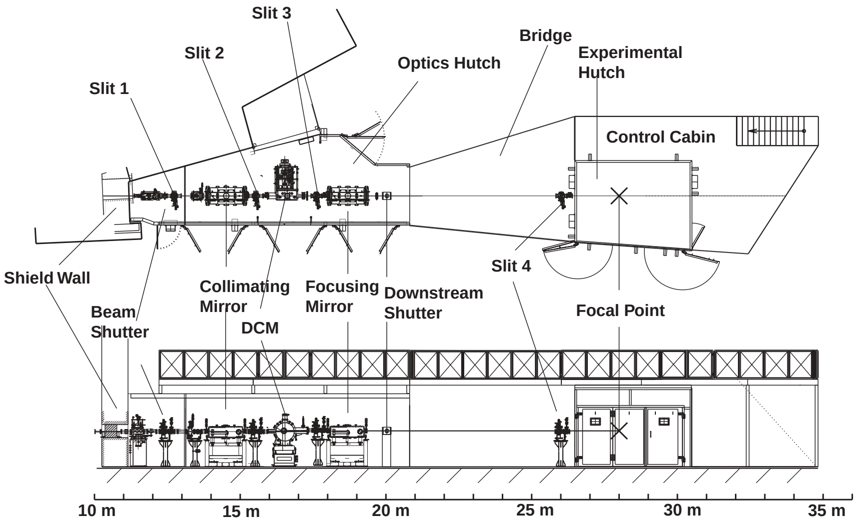

Figure 1
Design of BL-5 beamline from top (upper panel) and side (bottom panel) views. Storage ring locates at the left hand.

Table 1 Specification of the optical components.

Insertion device	Type: multipole wiggler Length of period: 120 mm Number of periods: 21 Magnetic field: max 1.4 Tesla
Collimating mirror (14.5 m from the source)	Type: flat-bent Material: Rh-coated Si single crystal Size (mm): 1000(L) × 80(W) × 70(T) Glancing angle: 3.5 mrad Radius of curvature: 8286 m
Double crystal monochromator (16.5 m)	Material: Si(111) Fixed exit: numerical link Cooling system: direct water cooling with micro-channel Energy range: 6.5 – 17 keV
Focusing mirror (18.7 m)	Type: bent-cylindrical Material: Rh-coated Si single crystal Size (mm): 1000(L) × 100(W) × 60(T) Glancing angle: 3.5 mrad Radius of curvature: 43.55 mm, 5333 m

position, and the focusing ratio is set at around 2:1.

Characteristics of the beam at the sample position are summarized in Table 2. The energy resolution of the beam was estimated from full width at half maximum (FWHM) of the rocking curve using Si(111). The beam size was measured by silt scanning. The beam intensity was estimated using PIN photo-diode at the sample position, with 0.2 × 0.2 mm slit and 0.2 mm ϕ pin-hole.

Experimental station

The basic design of the experimental station is almost the same as that of AR-NW12, with higher precision of the sample axis and larger active area of the CCD detector, which is suitable for experiments with crystals of complex proteins, whose size is often very small and cell dimensions are often large. The large active area of the detector also allows data collection with low energy X-ray at high resolution, which was difficult using the previous similar CCD detectors. Fig. 2 shows the precision of the sample axis whose sphere of confusion of the axis is less than 1 micrometer.

The CCD-type X-ray detector (ADSC Quantum 315, see Fig. 3) has the active area of 315 mm square and the pixel size of 51 µm. One image frame (6144 × 6144 × 2 byte at the full resolution mode) can be read out in almost one second; the total dead time including readout is around 2.5 seconds per frame. The exposure time for one frame is typically around 5 seconds. In total, time for collecting one data set (1 degree oscillation and 180 frames) is around 20 minutes. The speed of data acquisition is similar to the fastest beamlines in the world. In the future, this will enable fine phi-slicing experiments requiring thousands of frames in one data set. The specification of the CCD detectors and typical time for data acquisition at the structural biology beamlines are summarized in elsewhere (see pp. 71).

For error-free and high throughput operation of the beamline, various components such as an attenuator, two four-blade slits and an X-ray shutter and equipments around the sample (ex. beam stop) are all motorized for automatic data collection including exchange of protein crystals by a robot (see pp.73). The zoom and focus of the telescope are also motorized for automatic crystal alignment. All of them are controlled by the common system developed for the protein crystallography beamlines at the PF. The details are described and summarized in pp. 71.

Computing environment

We have recently installed four PCs with dual-Xeon (3.06 GHz) and two with dual-Operon, under the Linux operating system, for data processing and phase calculation at the beamline. The diffraction images are stored on the RAID file server with 2-TB storage. Two PCs are pre-

Table 2 Characteristics of the beam.

	Measured	Simulated
Energy resolution $\Delta E/E$ at 12.7 keV	2.5×10^{-4}	2.0×10^{-4}
Beam size at the focal point (mm) at 12.7 keV	1.5 (H) 0.3 (V)	0.4 (H) 0.3 (V)
Beam intensity (photons/sec) (0.2 × 0.2 mm slit + 0.2 mm ϕ pin-hole)	1.0×10^{11}	6.0×10^{11}

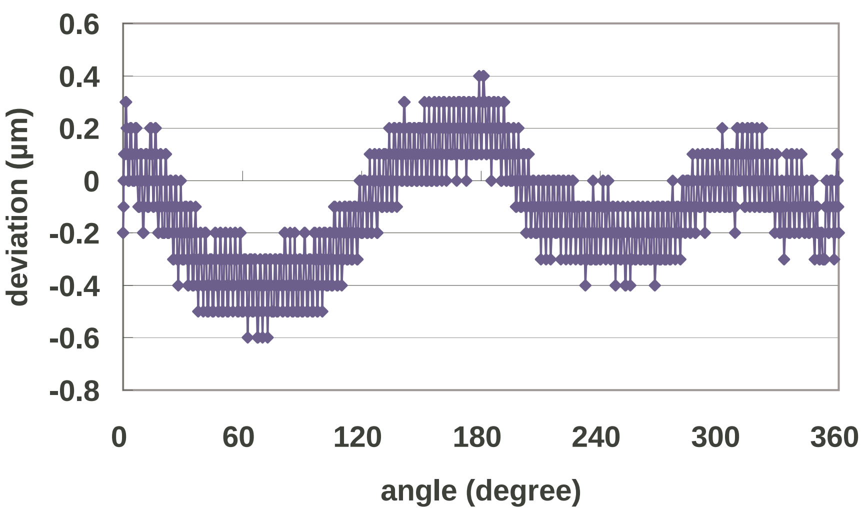

Figure 2
Deviation of the sample axis.

Figure 3
CCD detector (Quantum 315) on the experimental base.

pared for data backup, with the interfaces of IEEE1394 and USB2.0. These computers are all connected by gigabit Ethernet for rapid data transfer. In addition to the local resources, a central server for all the protein crystallography beamlines is available for data storage and processing. Details about the resources of the server and the structural biology network which all the PF structural biology beamlines belong to are described elsewhere (see pp. 72).

Acknowledgement

The construction of the BL-5 is mainly supported by Budget for Science and Technology Promotion (FY2000–2002) and in part by Protein 3000 Project, both of which are funded by the Ministry of Education, Culture, Sports, Science and Technology (MEXT).

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1-2 Construction of a New Undulator Beamline BL-28 for High-resolution ARPES

BL-28 is a helical undulator beamline which covers the photon energy of 30 to 300 eV (Table 3), and is suitable for the electronic structure study of nanomaterials, strongly correlated materials, and surfaces. We plan to restructure the beamline and construct a new end-station for high-resolution angle-resolved photoemission (ARPES) experiments. A high-resolution ARPES beamline has been strongly demanded by many users for a long time. The main purpose of this beamline is ARPES studies on high transition temperature (high-Tc) superconductors and nanomaterials fabricated by in situ combinatorial pulsed laser deposition (PLD) systems. To perform high-resolution ARPES experiments for high-Tc superconductors and nanomaterials, high energy-resolution as well as high photon flux is needed. We have chosen a varied including angle varied line spacing plane grating monochromator, which satisfies the requirement for both high energy resolution and high photon flux. A schematic diagram of the new beamline is shown in Fig. 4. The monochromator consists of three mirrors (M0, M1, M2) and one grating. M0, M1, and the grating experi-

Table 3 The specifications of the new undulator beamline BL-28.

Photon energy	30 ~ 300 eV
Energy resolution ($E/\Delta E$)	5,000 ~ 10,000
Photon flux	> 10^{12} photons/sec at the highest energy resolution
Spot size	350 μm (H) x 50 μm (V)

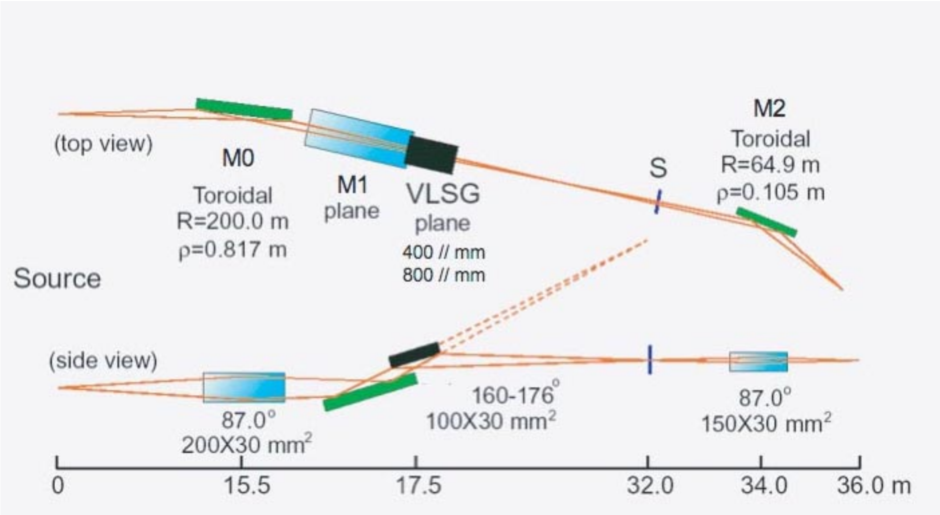

Figure 4
Schematic digram of BL-28.

ence considerable heat load from the undulator and are cooled by an indirect water-cooling system. The monochromator has no entrance slit, allowing for higher photon flux. Also high energy-resolution can be achieved over a wide energy range. The monochromator is designed to cover photon energiers of 30 to 1,000 eV and will be used after the future renewal of the undulator without any change. Precise control of the mirror and grating can be achieved by in vacuum rotary encoders.

For the high-resolution ARPES endstation, we have selected a Gammadata Scienta SES-2002 analyzer. We have also developed a new manipulator with a muti-axis goniometer to perform automated Fermi surface mapping and high-throughput ARPES experiments. The power of the newly developed manipulator has already been demonstrated at BL-1C [1]. Construction of the ARPES endstation system is in progress in collaboration with three user groups. The construction of the beamline is scheduled for the summer shutdown period of 2004. During autumn 2004 beamtime, adjustment of the new beamline and setup of the ARPES endstation will be scheduled. The new ARPES beamline will be open for users after the long-term shutdown in 2005.

Reference

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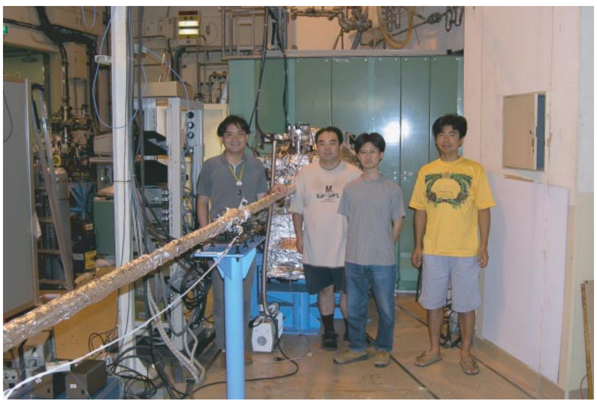


1-3 AR-NE1A2, Station for Medical Applications

We have been developing a two-dimensional imaging system for intravenous coronary angiography using synchrotron radiation monochromatic X-rays. An advantage of our system is that two-dimensional dynamic imaging of the cardiovascular system can be achieved by using the asymmetrical reflection of a silicon crystal and a two-dimensional X-ray detector.

The first four patient examinations for intravenous coronary angiography were performed in May 1996 in collaboration with the University of Tsukuba using the Two-dimensional Imaging System I at AR-NE1, a multipole wiggler (MPW) beamline [1,2]. After improvement of the imaging system and rearrangment of a conventional experimental hutch, AR-NE1A2, to the dedicated hutch for clinical examinations, system II has been employed for clinical examinations since 2000 in collaboration with the University of Tsukuba [3]. Images were obtained by X-rays above the K-edge energy of iodine (33.2 - 37 keV) using an image intensifier-television (II-TV) system. The X-ray exposure to the patient and motion blur in the heart image were minimized by inserting a high-speed X-ray shutter in front of the crystal. By the end of FY2003, forty-nine patients have been examined using the imaging systems. The advantage of a two-dimensional imaging system for intravenous coronary angiography was confirmed by the examinations. Medical doctors could observe the blood flow in a two-dimensional irradiation area to evaluate the motion of heart as well as the morphological information on the coronary arteries and the left ventricle. Schematic diagram of the Two-dimensional Imaging System II is shown in Fig. 5.

In 2003, the following two important issues were improved to facilitate more practical application of diagnostic imaging.

1. Beam size of monochromatic X-rays

The horizontal beam size had previously been limited to 73 mm at the patient's position due to the horizontal aperture of the beamline, AR-NE1, which was originally

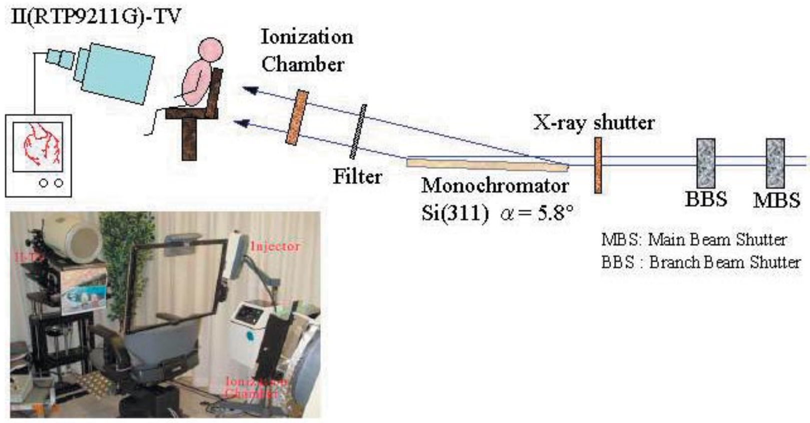
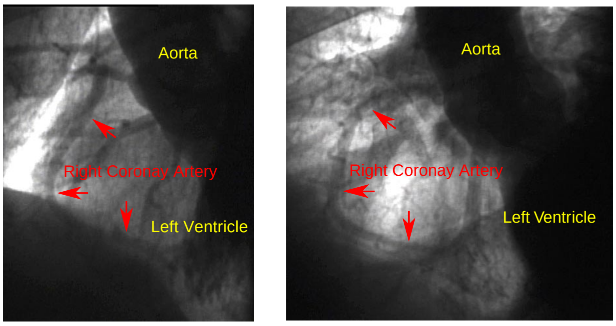


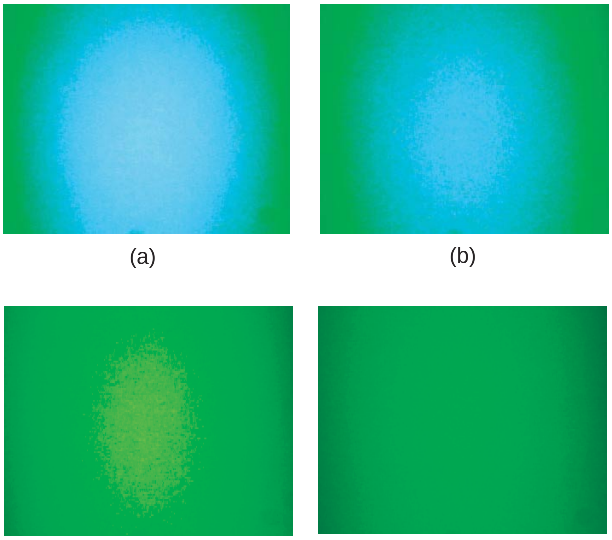
Figure 5
Schematic diagram of Two-dimensional Imaging System II with a picture photograph of inside the clinical hutch, AR-NE1A2.

designed for Compton-scattering experiments. In 2003, the horizontal size of the synchrotron radiation beam was enlarged from 73 mm to 93 mm by rearranging the beamline to make finding the optimal patient position easier and to diagnose the movement of heart in greater detail. Fig. 6(a) and (b) show monochromatic X-ray images obtained before and after the rearrangement. The right coronary artery and the left ventricle are clearly seen in both images, however, a larger part of the aorta and left ventricle can be distinguished in Fig. 6(b).



(a) **(b)**

Figure 6
Images of the right coronary artery. (a) and (b) correspond to before and after expanding the horizontal size of synchrotron radiation beam, respectively.



(c) (d)

Figure 7
Images of intensity distribution in a two-dimensional irradiation area by changing the longitudinal size of the electron beam. (a), (b), (c) and (d) correspond to the longitudinal size of 180, 230, 270, and 300 μm.

2. Intensity distribution in the two-dimensional irradiation area

The photon flux density of monochromatic X-rays transmitting the lung region is much greater than that of the mediastinum. As the dynamic range of a two-dimensional the detector (II-TV) is limited, this causes degradation of the vascular diacrisis in the mediastinal region where the coronary arteries overlap other organs, such as the aorta and the left ventricle. We prepared a 3-mm thick aluminum filter to reduce the photon flux density in the lung region, adjusting the intensity to the dynamic range of detector. The position of aluminum filters for left and right lungs were adjusted for each patient during the examination by medical doctors.

However, there were some cases where the thickness of the aluminum filter was insufficient, depending on the body thickness of the patient. We developed a new system in which the intensity distribution in a two-dimensional irradiation area can be changed by changing the longitudinal size of the electron beam. This new method was introduced in cooperation with the accelerator group [4]. Fig. 7 shows images of intensity distribution in a two-

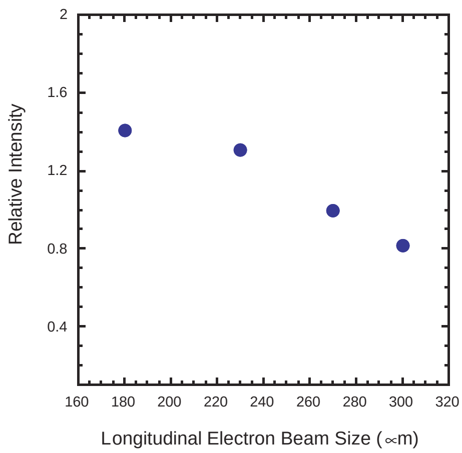


Figure 8
The relationship between the longitudinal size of electron beam and the relative intensity in the center of the irradiation area.

dimensional irradiation area by changing the longitudinal size of the electron beam. Fig. 8 shows the relationship between the longitudinal size of electron beam and the relative intensity in the center of the irradiation area. It is possible to change the intensity distribution in a two-dimensional irradiation area quickly using this method.

Those improvements, along with the high current and the long lifetime of the electron beam obtained by efforts of the accelerator group have enabled more practical examinations to be carried out. We plan to continue this project to promote application to clinical evaluation.

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[4] see "Section 3 PF-AR, Accelerators" in this volume.

1-4 AR-NW14, A Beamline for Time-Resolved Diffraction Studies

AR-NW14 is a new insertion device beamline at the PF-AR designed for time-resolved X-ray diffraction/scattering studies of condensed matter samples such as organic and inorganic materials, protein crystals and liquids. The construction of the beamline is being funded by the ERATO Koshihara Non-equilibrium Dynamics Project of the Japan Science and Technology Agency (JST), and the beamline will be operational from autumn 2005. The beamline will have two undulators with period lengths of 36 mm (U36) and 20 mm (U20). The U36 undulator covers an energy range of 5-30 keV with its 1st, 3rd, and 5th harmonics, and will be used as a tunable and intense monochromatic X-ray source by using a double-crystal monochromator and a focusing mirror. The typical photon flux of the monochromatic beam is estimated to be ~10^{12} photons/sec. The U20 undulator gives its 1st harmonics in the energy range of 13-20 keV. The energy bandwidth of the 1st harmonics is $\Delta E/E$ ~10^{-1}-10^{-2}, providing a 'narrow-bandwidth white beam' or a 'wide-bandwidth monochromatic beam' with a photon flux of ~10^{15} photons/sec. A plan view of the beamline and a virtual image of the hutches are shown in Figs. 9 and 10.

The primary scientific targets of the AR-NW14 will be condensed matter systems which can be triggered reversibly by a laser pulse. In particular, photo-induced phase transitions (PIPT) in molecular charge-transfer (CT) crystals are one of the main candidates for our research. The remarkable feature of PIPT is its cooperativity, that is, the structural relaxation of the electronic excited state of a molecule causes a large-scale photo-induced phase transformation towards a new lattice, electronic order and physical properties. Time-resolved X-ray diffraction enables direct access to the dynamics of electronic, atomic and molecular motions in such systems. In principle, one can construct 100-psec still images of the electron density at a given delay after initiation and produce a "movie" of the reaction with atomic resolution. The relative delay time between the laser and X-ray pulse is controlled by an electronic delay generator based on the radio frequency master clock (508 MHz) that drives the electron bunches in the storage ring.

In order to collect diffraction/scattering images with 2-dimensional detectors such as CCDs or imaging plates, we need to use a chopper to synchronize the X-ray and laser pulses in a 1:1 ratio, since the available detectors have no gating capabilities as yet. A high speed chopper (X-ray pulse selector, XPS) which is synchronized at 946 Hz to a subharmonic (1/537600) of the radio frequency and made by Forschungszentrum Jülich can be used for this purpose. The physical opening window of the chopper corresponds to 1.2 μsec but when it is phased to select the single bunch in the PF-AR single bunch mode, the exposure time becomes the 100 psec duration of the X-ray pulse. The novel feature of the chopper is its

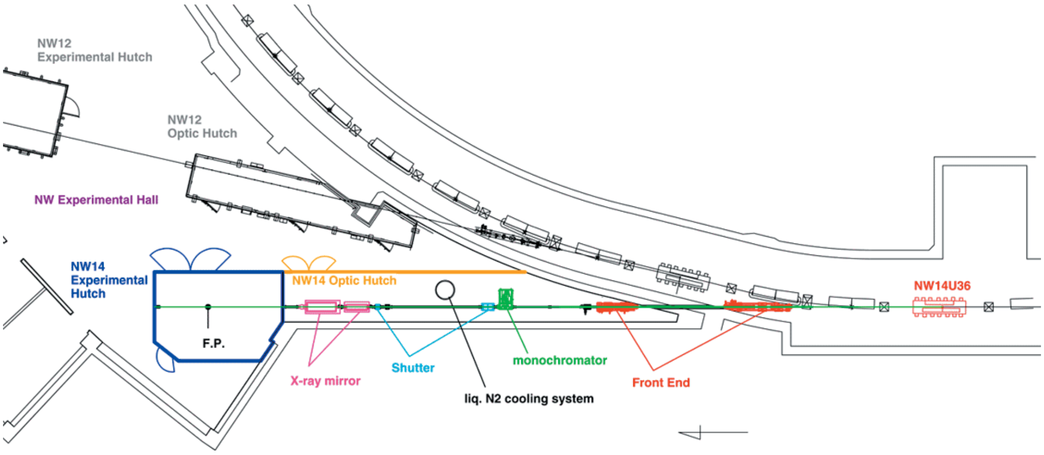

Figure 9
A plan view of the AR-NW14.

continuous phase locking with a timing jitter of less than 2 nanoseconds. One can thus produce a 946 Hz pulse train of 100 psec pulses from the X-ray pulse trains at 794 kHz emitted from PF-AR ring. If a detector with gating capabilities such as an avalanche photodiode is used, the XPS is not needed, and a gated integrator system can be used instead. If the repetition rate of the reversible reaction is slower than 946 Hz, a millisecond single-shot shutter will be available to lower the repetition rate.

A double pulse train of exciting laser pulses followed by probing X-ray pulses can be produced in which the relative delay time can be varied by the use of an optical delay line. A femtosecond regenerative amplifier system seeded by a Ti:sapphire femtosecond laser will be operational. The seeding laser pulses are phase-locked to reference signals of 1/6 of the radio frequency master clock by controlling the cavity length of the laser externally. The jitter is about a few psec. Then the laser pulses are chopped, amplified by the regenerative amplifier system and synchronized with the XPS. Finally, light pulses with 150 fsec duration and a few hundred µJ per pulse at 800 nm phase-locked to the X-ray pulses are delivered. For those reversible reactions which can run at frequencies up to 946 Hz, this instrument should give 100 psec resolution in monochromatic or Laue data collection from macromolecules and time-resolved scattering from less ordered systems.

The beamline has a double-crystal monochromator which uses flat Si(111) crystals and a liquid nitrogen cooling system. The cooling system can handle an incoming heat load of up to 450 W. The mirror system consists of three mirrors, a bent cylindrical mirror for focusing, and a double-mirror system (two parallel cut-off mirrors) to reduce contamination from higher harmonics. The detailed design of the beamline, including the hutches, diffractometers, and the laser booth is still under progress.

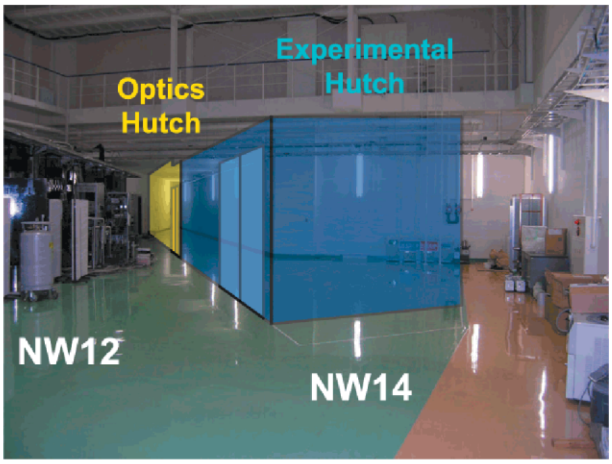

Figure 10
A virtual image of the beamline hutches.

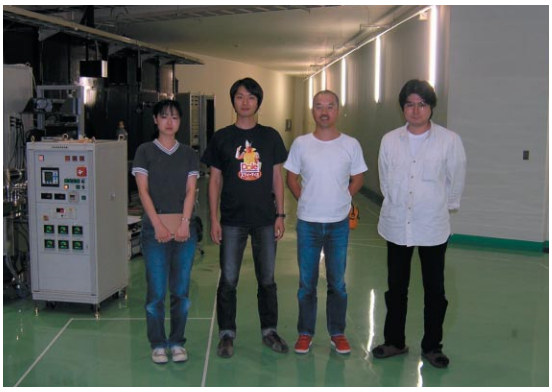

Figure 11
Part of members of ERATO/NW14 construction team. (Left to right; Ayana Tomita, Shunsaku Nozawa, Shin-ichi Adachi adn Tokushi Sato)