Extremely Dense State

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1. Introduction

Extremely Dense State beamline is one of six public undulater-beamlines which will be available from Autumn in 1997 for condensed matter research. The beamline has two experimental stations; one is for high pressure x-ray diffraction experiments using 20 - 60 keV x-rays by the use of diamond anvil cell (DAC), and the other is for x-ray absorption fine structure (XAFS) experiments using 5 - 25 keV x-rays by the application of "tunable" undulater. Two scientific subgroups, Extremely Dense State and high brilliance XAFS, arrange these two experimental stations in tandem for sharing the monochromatized unfocused undulator radiation. Basic specifications of the beamline are as follows;

(1) Insertion Devise: In-vacuum undulater (U032V). $\lambda u = 3.2$ cm, N = 140
Tunable range: > 5 keV.
(2) Monochromator: Rotated-inclined double-crystal monochromator
Monochromatization of 5 - 60 keV x-rays
(3) Mirror for XAFS: Double-Flat Mirror system (Variable cut-off energy.)
Cut off energy: 10 - 20 keV x-rays

2. Extremely Dense State

2.1 Research subjects

The aim of the Extremely Dense State group is to investigate unusual physical and chemical properties of condensed matter in extremely dense state on the basis of the crystal structure data of good quality benefited from high brilliance x-rays.

Recent progress in high pressure technique of DAC has enabled us to reach a pressure of several hundreds GPa (1 GPa = 1000 atm). At such an high pressure, materials are brought into extremely dense state and exhibit various novel characters which can not be realized in ambient condition. The appearance of metallic property in molecular solids is a good example [1]. Obtaining at first the knowledge of crystal structures stabilized at high pressures, we can start to discuss the nature of chemical bond, elastic, electronic, magnetic, and other properties.

Our scientific programs are followings: (1) polymerization, dissociation, and metallization in molecular solids, (2) electronic transition in metals, (3) precise structure analysis of semiconductors and super conductors. X-ray measurements in our station include (a) powder x-ray diffraction, (b) single crystal x-ray diffraction, (c) XANES spectroscopy.

2.2 Experimental Facilities and techniques

The experimental station for the high pressure research will be located at the downstream of XAFS station. A monochromatized x-ray beam is shaped by an adjustable aperture and then focused by a linear Bragg-Fresnel Lens (BFL). To generate a focal spot as small as few square microns, the lens is bent in a cylindrical form.

Sample enclosed in a DAC is placed at the focal point. The DAC is mounted in a closed cycle cryostat for low-temperature experiments down to 10 K. A conventional heating technique an be used for moderate-temperature measurements to 1000 K. A laser heating system for ultrahigh-temperature experiment up to 3000 K will be installed in the future.

In the first phase of our construction schedule, we install an Imaging Plate (IP) as detectors. The IP is such a type that two or more IP sheets are equipped with it and the read-out mechanism is built-in. Thus, one of the IP sheets can be used to measure diffraction intensities while the data read-out from the other IP is being carried out. This capability provides rapid data processing to us. In the second phase of the construction, a CCD-type detector will be introduced.

In order to measure the pressure, a ruby fluorescence method is adopted. In the present system, the pressure measurements can be performed without taking the DAC away from a diffractometer. The laser beam from an Ar-gas laser located outside the hutch is transferred through thin optical fiber to the DAC mounted on the diffractometer. This beam hits a ruby chip encapsulated with a sample in the DAC and excites its fluorescence. The fluorescence is transferred back through the same optical fiber to spectrometer. Knowing the magnitude of the wave length shift from a value at 1 atm, we can calculate a value of pressure inside the DAC.

3. High brilliance XAFS

3.1 Research Subjects

The aim of high brilliance XAFS group is to apply a “tunable” x-ray undulator to XAFS research using high brilliance photon source. A tunable x-ray undulator provides highly collimated quasi-monochromatic beam with a controllable polarization characteristics. In this program, linearly polarized high brilliance x-ray beam with tenability over a wide range of photon energy (5 - 25 keV) is obtained by use of x-ray undulator. As a natural benefit of highly brilliant beam, one can deal with less number of exited atoms within a limited fraction in time and space. For dilute system such as surface and heme proteins, a fluorescence-detection technique is used for which sensitivity is limited by efficiency of fluorescence excitation and detection. In order to fully utilize the ultra-low eminence of third generation storage ring (< 10 nmrad), we plan to utilize a grazing-incidence geometry and a high density solid state detector array.

3.2 Experimental Facilities and techniques

The high brilliance XAFS station will be built in the upstream hutch of the beamline. Our program aims at the development of rapid and sensitive XAFS technique and its application to local structure studies.

Below the critical angle, x-rays are totally refracted as they cross the interface between two media reducing the extinction length by several orders of magnitude, thereby enhancing surface sensitivity. In a total reflection regime, surface XAFS experiments within a monolayer sensitivity has been achieved using a grazing-incidence fluorescence detection technique. The technique has potential surface sensitivity in the submonolayer region [2].

The total energy range of our concern is the conventional hard x-ray region, 4 - 25 keV. In general, tunability in XAFS experiments can be classed in two categories; spectroscopic tuning ($\Delta E < 1$ keV) and edge-to-edge tuning ($\Delta E = 1 - 21$ keV). We plan to move the undulator gap as we scan the monochromator over $\Delta E \sim 1$ keV which is achieved by real-time monitoring the calibrated positions of undulator gap and monochromator.

For the lower energy region, a double flat mirror is inserted behind the monochromator in order to reduce the higher harmonic radiation.

To achieve the rapid and sensitive XAFS experiments, we have developed 100-element pure Ge detector. All elements are fabricated in a single Ge wafer and assembled into an array. Each units 5 mm x 5 mm in dimension and is separated with each other by a 10-micron wide dead layer. Thus the packing ratio is almost 100 % while the conventional pure Ge detector array such as a 19-element detector has a packing ratio of 57 %. The expected average energy resolution is 190 eV at 5.9 keV.

A sample is mounted on a closed-cycle He refrigerator which is rotated by a compact Huber goniometer. Data analysis is routinely carried out at the station during data collection, which is essential for *in-situ* studies. For a data acquisition, a unix work station is used and fluorescence data is acquired via CAMAC bus.

The use of undulator and new x-ray detectors such as a monolithic array detector or two-dimensionally curved crystal will allow us the rapid measurements of more dilute specimens with less dimensions.

References

[1] Y. Akahama et.al., Phys. Rev. Lett. **74**, 4690 (1995).

[2] H. Oyanagi et al., J. Synchrotron Rad. **2**, 99 (1995).