SPring-8 BL04B2 Review Committee Report on High-Energy X-ray Diffraction Beamline (BL04B2)

Report for Director General of
Japan Synchrotron Radiation Research Institute

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1. Introduction

The review committee meeting was held at the SPring-8 site on December 2-3, 2004. All of the committee members had received two written reports, "Beamline Report BL04B2 (High-Energy X-ray Diffraction)" and "SPring-8 Overview 2004 for Beamline Review Committees", and submitted individual reports to the committee chair prior to the meeting. Four local members attended the meeting. The members heard the guideline on the beamline review and the overview of the SPring-8 facility and organization from the SPring-8 side, and then received an explanation of the beamline facilities from beamline scientists and visited the beamline. After the beamline tour, the members heard the details of research results, future plans and subjects, and current problems, and had a question-and-answer session. This report was put together based on the discussion among the local members and the individual reports from Professor Poulsen and Professor Price.

2. Current Performances and Achievements of Beamline and Experimental Facilities

*Evaluation

This beamline was constructed in 1998 as one of the bending magnetic beamlines. It has a horizontally-curved monochromator that delivers high-energy mono-chromatic X-rays to the experimental station using a single reflection. The X-ray wavelengths (or energies) available are discrete because of a fixed Bragg angle. In accordance with the purpose of an user's experiment, Be compound X-ray refractive lens (CRLs) are used for vertical focusing. The beamline scientists have come up with various ideas to make the best use of the available high-energy X-rays, and the BL04B2 beamline has achieved a world-class performance. Since 1999, the following instruments have been accommodated by the experimental stations and have been operating satisfactorily:

(1) Two-axis Diffractometer for Disordered Materials
(2) Imaging-plate Diffractometer for Diamond Anvil Cell (DAC)
(3) Small-angle Diffractometer for Metallic Fluids
(4) Weissenberg Camera for Single Crystals

With the current setup, 37.8 keV (monochromatized by Si 111) and 61.7 keV (Si 220) X-rays are delivered to the experimental stations. With the start of the top-up operation of the storage ring in 2004, the beamline scientists tried to install an assembly of CRLs to obtain a more stable and intense incident X-ray beam for measuring a smaller sample of disordered materials. However, it still has not achieved it's ideal performance.

The four instruments are installed for measuring special samples of microcrystals, disordered materials and high-temperature liquids, and every instrument is operating satisfactorily by taking full advantage of the SPring-8 storage ring, which can produce high-energy X-rays higher than 50 keV. The two-axis diffractometer is designed to make the most of high-energy X-rays, and is operating satisfactorily. In particular, the routine top-up operation has largely stabilized the intensity of incident X-rays, leading to a substantial improvement in the reproducibility of measured data in a high-Q region, which is indispensable for the study of disordered materials. In addition, the two-axis diffractometer is appropriately equipped with a high-temperature and high-pressure device to make the best use of the world-class apparatus on the beamline. The imaging plate diffractometer with a DAC for high-energy X-rays and the high-pressure vessel up to 2000 bar under 1700^{o}C for the small-angle scattering study of expanded fluid Hg have been successfully developed. On the other hand, one of the drawbacks is that it takes approximately one day to change the experimental setup among the four instruments. The beamline scientists have been scheduling users' experiments so as to minimize the time loss.

Each instrument is installed into the BL04B2 beamline to make the most of the features of high-energy X-rays such as (1) small corrections (on absorption and extinction effects), and (2) access to high-order reflections and acquisition of high-Q diffraction data. However, we admit that the four instruments with different purposes are just put in the experimental stations. This situation has been acceptable to users in the first phase of beamline operation. However, in the second phase, in which the general-purpose beamline is changed to a specific-purpose beamline, the JASRI should plan a strategy for further improving the beamline performance.

*Recommendation

It is required to rearrange the four instruments so as to make the most of the features of the BL04B2 beamline.

3. Research Activities

*Evaluation

The most fundamental parameters that describe the structure of liquids and amorphous solids are interatomic distance and coordination number. The accurate determination of interatomic distance and coordination number requires the measurement of the structure factor S(Q) up to a high-Q value. The successful installation of the two-axis diffractometer in BL04B2 enables us to measure the data up to a high-Q value at SPring-8, which has been only accessible to pulsed neutron total scattering experiments. This development has enabled the combined research on X-ray S(Q) and neutron S(Q) for determing the partial structures of a multicomponent glass. In addition, the real-space structure obtained by the high-Q measurement is so accurate that its comparison with that obtained by nano-scale simulation provides the clear physical interpretation of an experimental data. The use of highly brilliant, high-energy X-ray beams at SPring-8 has facilitated the corrections for absorption, polarization, multiple-scattering, extinction and so on, and has reduced the technical difficulties in measuring the intensity of diffracted X-rays. The structural research on glassy materials has produced notable results at BL04B2. The demonstrated value of Q_{max}=35 Å is almost equal to the value of Q_{max} for pulsed neutron measurements. The complementary use of both techniques will expand in the future. We expect that the demonstration will develop research covering all types of glassy material, together with other research methods (including research on dynamics by neutron scattering, in addition to the use of X- rays) and theoretical research.

(1) The following excellent results are obtained; (a) elucidation of the slight difference in the medium range structure between SiO_2 and GeO_2 glasses, (b) modeling the network configuration of SiO_4 or MgO_n (n=4,5,6) polyhedra in noncrystalline Mg_2SiO_4 that is a prototypical compound found in magma in the upper mantle.
(2) The observation of the density fluctuation in fluid Hg for studying the metal-insulator transition is a unique result, although the high-energy X-rays are not suitable for the small-angle scattering. The conclusion that the atomic configuration of expanded fluid Hg fluctuates between insulating liquid and dense vapor at the critical point is highly important valued since it has settled the long-standing issue in this field. However, we are concerned with the small impact of this result to other fields when we consider the future of this research. The research group and the JASRI should promote the use of this research technique in other fields.
(3) Pressure-induced structural changes are reported in amorphous SnI_4, liquid oxygen and nitrogen, and $SrCu_2O_3$. These successes are due to the use of high-energy X-rays. The research into crystal, and noncrystalline and liquid properties under a high-pressure region using a DAC is unique.
(4) The study of modeling the structure of Al-Pd-Re using an LDEM (low-density elimination method) is very important valued in the field of quasicrystals. On the other hand, we think that the Weissenberg camera does not take full advantage of the features of the beamline since a commercially available apparatus for laboratory use is only installed in the experimental station. We admit that the apparatus is not suitable for the beamline although the research on the structure of complexes containing heavy elements is important in related research fields.

* Recommendations

(1) Both the noncrystalline and liquid research using the two-axis diffractometer and the high-pressure structure research using a DAC should be further developed in the future since their results are highly evaluated.
(2) Research studies on the structure of complexes should be carried out at another suitable beamline.

4. User Support

*Evaluation

<u>Numbers of Submitted and Selected Proposals</u>

The number of submitted proposals increased by two fold in 2001B. The sharp rise decreased the selection rate from ~90 percent to ~60 percent, although we do not know whether the rise occurred in the four fields. The selection rate of 60 percent is low, but it is reasonable for maintaining a high level of proposals by encouraging competitiveness among users. The number of proposed studies performed (performed proposals hereafter) was almost constant at approximately 20 per research term for the past five years. There has been no significant change over the last five years in the number of performed proposals for each research field. The percentages are roughly 40 percent for Disordered Materials, 30 percent for Single Crystals, 25 percent for High Pressure, and 5 percent for Supercritical Fluids. However, the number of performed proposals for Disordered Materials suddenly increased in 2004A. The number of performed proposals for Supercritical Fluids is one each term, showing that this research is a special case in the beamline. The JASRI should support this research because of its uniqueness, and should develop a new research field using the apparatus.

We do not know whether the number of submitted proposals is in proportion to the activity of each research field. However, we feel that the users might hesitate to submit proposals with a large impact since the beamtime for each apparatus is limited.

Beamtime

The assigned beamtime (number of shifts) for each selected proposal has been converging to 80~90 percent of requested beamtine except for 111 percent in 2001A. This is a favorable trend.

The share of beamtine for each research field is not constant, and largely varies with year. The beamtime for Disordered Materials has increased by three fold from 1999B to 2003B. The beamtime for Single Crystal is almost constant. On the contrary, the beamtimes for Supercritical Fluids and High Pressure have been relatively decreasing annually. The assigned beamtime is 8~10 shifts per proposal on average, and there is no larger gap among the research fields. In particular, 20~30 shifts were assigned to Supercritical Fluids in every research term because of its requirements.

Publication of Results

Since 2001, many of the results obtained in the beamline have been published in refereed journals. The number of publications has reached approximately 60 including reviews and proceedings in 2004. About ten articles are published annually, while 18 articles were published in 2004. However, there has only been one publication of a review in English over the past five years. We also admit that the number of presentations at international conferences is less. The results of some of the performed proposals have not been published. These situations must be improved.

The average numbers of shifts and performed proposals used to communicate one article are given below for each research field.

(1) Disordered Materials : 38 shifts and 4 proposals,
(2) Single Crystal : 74 shifts and 6 proposals,
(3) High Pressure : 25 shifts and 3 proposals,
(4) Supercritical Fluids : 86 shifts and 3 proposals.

The research field of Single Crystal has a low productivity since it has used up many shifts and has required many proposals to produce an article. The field of Supercritical Fluids has used up the highest number of shifts, while the number of proposals is the same as that of the other two fields. The research field of High Pressure is most productive. We admit that publication largely varies with research field.

We judge that the research fields of Disordered Materials and Supercritical Fluids are maintaining high activities since they have more Invited Talks and Review

Articles. The number of publications peaked in 2002 and then decreased or saturated in 2003 and 2004. The JASRI should look into the cause of this decrease or saturation.

Five proposals from overseas users and two from industrial companies have been performed over the past five years. The number of submitted proposals from overseas users has been increasing after 2004. More effort is required so that the BL04B2 beamline gains international recognition.

*Recommendations

(1) The BL04B2 beamline has gained recognition in Japan. In the future, more overseas users should be invited to the beamline.

(2) Some of the performed proposals have no publications. The JASRI should look into the cause of this and to increase the number of publications on the beamline.

5. Instrumentation and Research Direction in Future

*Evaluation

The six plans proposed in Beamline Report BL04B2 are appropriate for the development of the beamline in the future. We require that all of the plans should be implemented. An important point to be considered when drawing up the plans in detail is to develop the facilities from the hardware and software sides to make the most use of the highly brilliant and high-energy X-ray source and attract more users from every field of research.

The Weissenberg camera has not attained its ideal resolution since the camera radius is short. This apparatus is not suitable for synchrotron radiation use since it is a commercially available product for laboratory use. The JASRI has proposed that the research studies using the Weissenberg camera will be moved to the BL02B1 beamline in which a vacuum camera is available and the other research studies will be carried out using the two-axis x-ray diffractometer. This strategy is quite appropriate for the second phase of this beamline. The JASRI should move the Weissenberg camera to another suitable beamline to gather same research studies into a beamline. When rearranging the activities, the JASRI should reflect users' opinions.

This beamline should focus on its original researche studies using a world-class apparatus for high-energy X-rays. A plan that involves the installation of a levitation apparatus for the structural study of liquids and supercooled liquids is challenging and promising. The installation of Si 511 monochromator for higher-energy X-rays and the addition of a new function to the two-axis diffractometer for vertical-plane measurement (including possible use of a position-sensitive detector) are appropriate for the future development.

The assignment of only two in-house staff members is not appropriate for such a world-class facility. We strongly suggest that the JASRI should employ additionally one or two staff members for this beamline. New members should support users, maintain the beamline facility and conduct their own research.

* Recommendations

(1) The proposed strategy is appropriate for the development of the beamline over the next five years.
(2) The JASRI should increase the beamline staff with a broad outlook.

6. Summary

The BL04B2 beamline has produced sufficient results. For further development of the beamilne, the JASRI should focus on research into disordered materials using high-energy X-ray diffraction. In addition to metallic and inorganic glasses, the target should be broadened to water solutions, melted salts, and organic compounds. The implementation of supercritical fluid research, which takes up considerable beamtime, is regarded as an outstanding policy of the JASRI. However, such a special project should be reviewed in terms of requested beamtime and related publications in the proposal review process. We request that the JASRI should expand the target of supercritical liquid research from metals to other substances for further development of this research field.

The BL04B2 beamline currently accommodates four apparatuses for different research studies using high-energy X-rays. We believe that this is a temporary situation in a trial phase which is unavoidable in such a large-scale facility. The JASRI should prepare for the second phase of the beamline, prioritizing the use of an apparatus which can perform world-class research at SPring-8. We suggest that BL04B2 should shift from a general-purpose beamline to a specific-purpose beamline. Along this line, the JASRI should promote researche studies with an impact such as structural study on glassy and non-crystalline materials. Therefore, it is important to develop BL04B2 to be a dedicated beamline for research into the structure of random systems (including non-equilibrium systems). We think that the proposed future plans agree with our perspective.

The SPring-8 facility should develop an experimental apparatus that is not available in other laboratories since it has more financial support than universities. In particular, it is difficult to develop a researcher's ability on instrumentation in universities. We hope that young scientists who investigate experimental apparatuses and techniques will develop in SPring-8.