The Idea of Gradient-monochromator for High Energy X-ray Structure Analysis

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Diffraction experiments with high energy X-ray radiation will give new frontier in structural investigations. Now a day, structural studies are well reproducible for different crystals and various temperatures under the same procedure of structural analysis. However, still there are several difficulties to get reliable results. One of the most difficult problem is the correction of the extinction effects. Recent trend to avoid the extinction problem is the usage of modern technique of powder diffraction. However, the application of a powder diffraction experiment to the phase transition phenomena is still difficult since the atomic displacement is very small and the relevant intensity is considerably weak. Another possibility to reduce the extinction effects is to use high energy X-ray radiation. High energy X-ray diffraction technique also gives another advantage since the transmission for the apparatus to achieve extreme condition is very high and the area of reciprocal lattice expands considerably wide. Application to high pressure apparatus is one of the most important case. For the laboratory experiments, Ag targets (0.559Å 22.2keV) are sometimes used. A W target will give a shorter wave length 0.209Å (59.3keV) for laboratory structural analysis, but technical development is still required [1].

We performed X-ray diffraction experiments to test the validity of the structural analysis with high energy X-ray radiation by using synchrotron radiation of the Photon Factory[2] with a test sample of $K_3H(SO_4)_2$. Experiments were carried out at BL14A in the Photon Factory with 2.5GeV ring energy and 340-200mA ring current. X-ray radiation coming from a Wiggler beam line is monochromatized with a double-crystal monochromator of Si(553). About 80 hours and 44 hours are elapsed for the accumulation of data on 50keV and 25keV experiments, respectively. The maximum scattering angles $2\theta_{max}$ are 70, 80 and 40 degrees, and the observed number of reflections are 1953, 7874 and 9438, for 17.4, 25 and 50keV X-ray radiation, respectively. Here, the 17.4keV radiation corresponds to a characteristic radiation of MoKα performed at laboratory equipment. For the 50keV radiation, we limited $2\theta_{max}$ to 40 degrees since the Bragg intensity beyond this angle completely vanished. Extinction parameters obtained by the structure analyses given in ref. 2 are apparently reduced for the results of the high energy X-ray radiation, as is expected.

Concerning Bragg intensities for synchrotron radiation experiments, the observed intensities were considerably weak: the peak intensity of Bragg reflection observed in the 50keV experiment was 50 times weaker than that of the 17.4keV experiments, and the width of diffraction profile observed in the 50keV experiments was 10 times sharper than that of the 17.4keV experiments. Examples of the diffraction profile are shown in Fig.1. The energy resolution of the Si(553) monochromator was too high for the purpose of the present experiments, and currently available beam lines are not optimized for the conventional structural analysis with high energy X-ray, since a significant part of beam time is used for other purpose such as XAFS experiments requiring high energy resolution. For the structural analysis with such purpose, we will propose a more optimized beam line to get much more intensity and proper resolution. Reduced energy resolution and wider beam divergence are necessary to use all of photons more effectively. One of the method to achieve the optimized energy resolution for structural analysis is to use a gradient monochromator of Ge_xSi_{1-x} with the continuous variation of the concentration. We plan to fabricate it by MBE. The d-spacing is gradually changed by changing the concentration of Ge and Si, at a level of about 3%. We expect that the energy width ΔE/E increases more than one order of magnitude, and then the flux increases considerably.

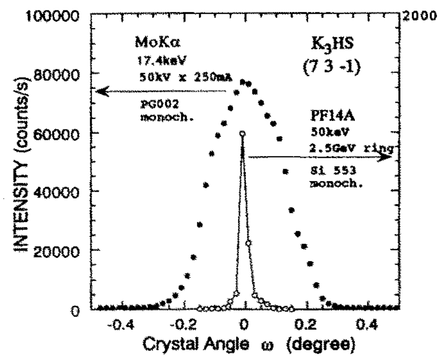

Fig.1 Observed diffraction profiles of (7 3 -1) Bragg reflections.

References
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