

# High-Energy White Synchrotron Radiation As a Source for X-ray Reflectometry: Potential Feasibility

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**Abstract.** Reflection spectra of white X-rays measured at a certain glancing angle basically correspond to reflectivity data obtained by a  $\theta/2$   $\theta$  scan with monochromatic X-rays. The present report describes how the use of white synchrotron radiation from a high-energy storage ring could make the technique fairly competitive and attractive for applications in nano sciences and nano technologies.

## INTRODUCTION

X-ray reflectometry is extremely powerful for the non-destructive investigation of the surface and buried interfaces of layered materials [1]. In ordinary measurements, monochromatic X-rays, typically Cu  $K\alpha_1$  from an X-ray tube, are employed, and data acquisition is based on a  $\theta/2$   $\theta$  scan, which is similar to powder diffraction except that the angle becomes extremely small. When a detector with a certain energy-resolution is available, one can carry out almost exactly the same experiment in a different way by using white X-rays [2]. One of the biggest advantages of energy-dispersive X-ray reflectometry is that it can be performed with a completely fixed geometry - the experiments are simply a continuation of counting by an energy-dispersive detector [3,4]. Therefore, the method can be applied to systems that are difficult to move, i.e., thin films fixed in a deposition chamber, liquid surfaces, etc [5]. Another promising and significant direction is rapid measurement to observe structural changes while controlling the environment of the sample [6,7]. However, the method has not been employed so widely, mainly because of limitations in the quality of the data obtained. The performance of the detector is of course crucial, but a number of improvements can also be expected from optimizing the X-ray source and optics. In the present report, the use of white synchrotron radiation from a high-energy storage ring is discussed.

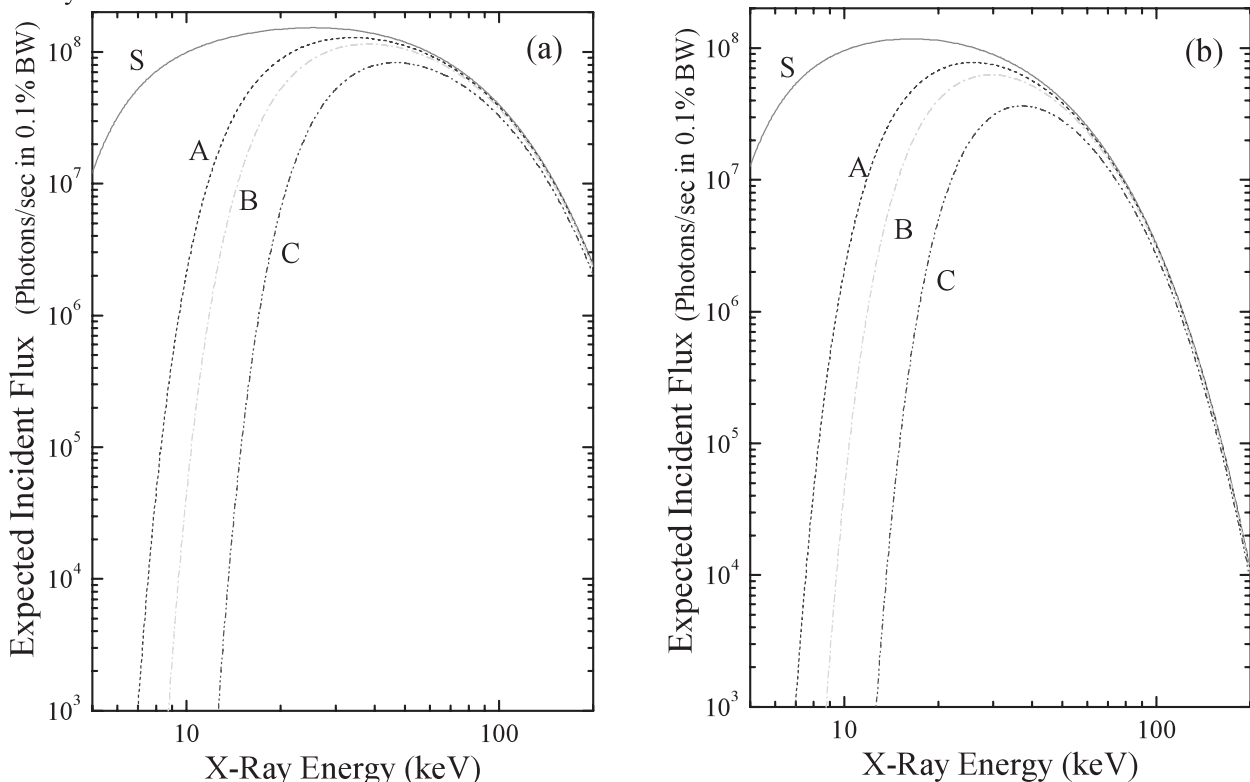
## CONSIDERATION OF WHITE X-RAY SOURCES

It is clear that smooth continuous spectral distribution is highly desirable when performing reflectivity measurements in energy-dispersive mode. The issues discussed here are the required spectral range and the intensity profile. In most cases, a Ge detector or a Si drift chamber is used as a detector. The energy range for the detector is not such a big problem unless an energy region that is extremely low is being considered. The energy resolution (typically 150 eV at 5.9 keV) is insufficient, but improves slightly at higher X-ray energy. The limit of the dynamic range of the detector is one of the biggest difficulties. The detector is saturated at a total counting rate of  $10^4 \sim 10^5$  counts/sec. This indicates that even strong flux is of no use, but a much more important issue is that the method measures reflectivity from almost 1 to  $10^{-6}$ . Even when the spectral shape is flat, the measurement of parts with low reflectivity will be fairly difficult. This is because the total counting rate of the detector is limited by the counts for the parts with high reflectivity. The spectral distribution of the white X-ray source is therefore extremely important.

How wide a spectral range is necessary for the experiments? Experimentally obtained ordinary reflectivity and energy-dispersive reflectivity are a function of  $\theta/2$   $\theta$  and X-ray energy, respectively. They are comparable when the data are plotted in terms of the wave vector,  $q_z (=4\pi\sin\theta/\lambda)$ ,  $\lambda$  is the X-ray wavelength and can be written as  $\lambda=12.3981/E$  in Å where E is X-ray energy in keV). The typical angular range for a  $\theta/2$   $\theta$  scan might be set as

something like  $0.8\theta_c \sim 8\theta_c$  (maybe more, but let's assume the minimum requirements), here  $\theta_c$  is the critical angle of the surface, for a certain X-ray energy,  $E_0$ . For instance, in the case of silicon and 8 keV X-rays,  $\theta_c$  is ca. 4 mrad. A  $\theta/2 \sim \theta$  scan range from 3.2  $\sim$  32 mrad is therefore realistic. We need to choose a fixed glancing angle  $\theta_0$  for energy-dispersive experiments. For small angles, from the simple relation,  $E\theta_0 = E_0\theta$ , the X-ray energy corresponding to the critical angle would be expressed as  $E_c = E_0\theta_0/\theta$ . The energy range can be written as  $0.8E_c \sim 8E_c$ . When  $\theta_0$  is set as 5 mrad for silicon,  $E_c$  is 6.4 keV, and the required energy range becomes 5.12  $\sim$  51.2 keV. This apparently depends on the choice of  $\theta_0$ , but the lower-edge energy is usually in the 4 $\sim$ 10 keV range and the higher end will be 10 times that range. Thus, energy-dispersive X-ray reflectometry needs quite a wide X-ray spectral range. So far, synchrotron radiation sources have not been used so frequently for this type of experiment, because such high energy X-rays are not available at low energy storage ring facilities.

Figures 1 (a) and (b) show some calculations for the spectral distribution of the white beam at the sample position (44m from the source) at BL28B2, SPring-8 [8]. Note that the original spectra are fairly wide and even reach 200 keV. The use of a mirror is effective for removing unnecessarily high energy X-rays, but another possible way is the use of a beam out of the orbital plane as shown in Fig.1 (b). The spectral distribution of the beam can be optimized by attenuating the low-energy parts preferentially. The adjustment can be done by inserting an absorber plate that has a certain designed thickness. For example, if a 1.5 mm thick aluminum plate is used, the spectra become curve A. The peak of the spectra is around 30 keV (for both Figs.1 (a) and (b)), but the decrease of the high-energy region is quite small. In this case,  $E_c$  can be chosen at around 8 keV, and the measuring energy range will be 6.5 $\sim$ 65 keV. Primary X-rays at 30 $\sim$ 60 keV are ca.  $10^4\sim 10^5$  times more intense than at 8 keV. This would be of quite a lot of help for improving the limitation on obtaining reflectivity in the high  $q_z$  region. Moving to even higher energy is promising as well. On the other hand, the synchrotron source is so brilliant that the detector is easily saturated. The calculation assumes a  $50 \mu\text{m} \times 50 \mu\text{m}$  beam, and the intensity at  $0.5 \mu\text{m} \times 0.5 \mu\text{m}$  (i.e.,  $10^{-4}$ ) is probably still sufficient. This indicates that the technique is suitable for other attractive applications, such as sub-micron area analysis, in contrast to conventional reflectometry, which normally on average give results in the  $\text{mm}^2\sim\text{cm}^2$  area. It would be significant to develop a device that can form such a small beam for high-energy white X-rays.

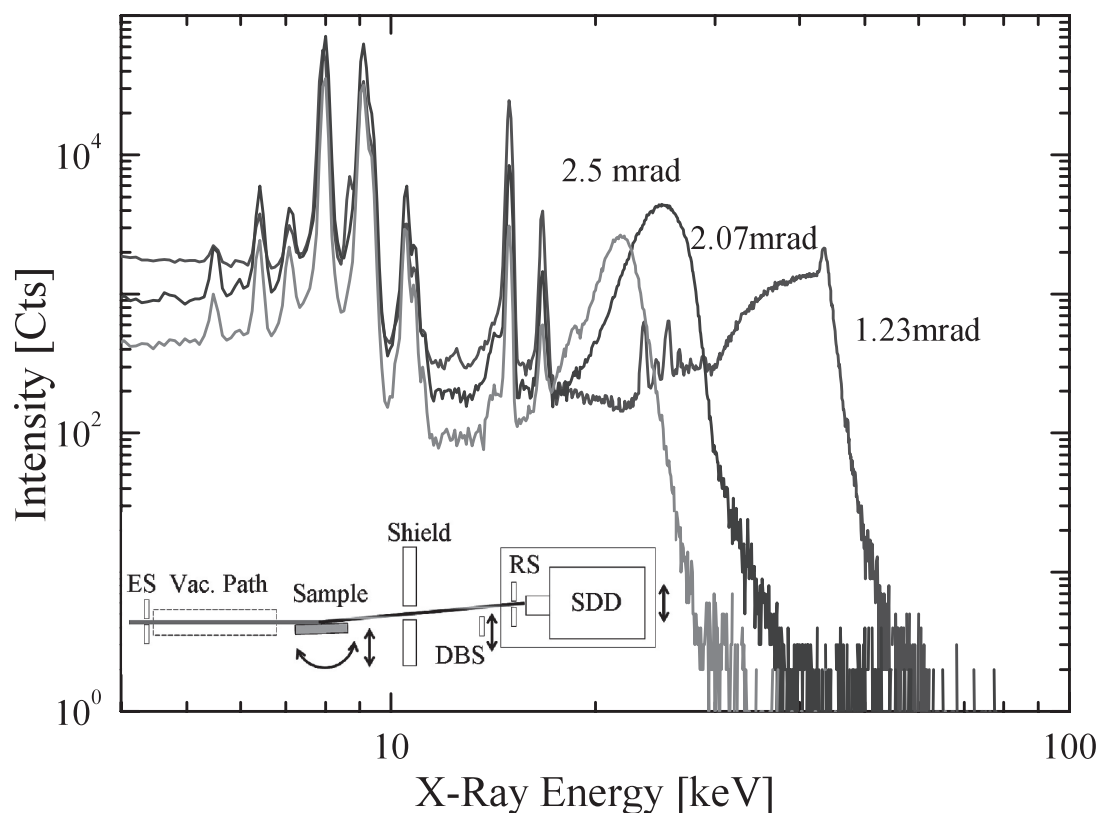


**FIGURE 1.** Expected X-ray flux for  $50 \mu\text{m} \times 50 \mu\text{m}$  beam at 44m position from the source point at BL28B2, SPring-8; (a) in the orbital plane (beam center), (b) out of the orbital plane (2mm off center). S: original spectra with standard condition considering absorption of Be (0.75mm), He (3m) and air (30cm), A: additional absorption of Al (1.5mm), B: additional absorption of Al (3mm), C: additional absorption of Al (9mm),

## PRELIMINARY EXPERIMENTAL RESULTS

Energy-dispersive reflectivity measurements with a high-energy white synchrotron beam were performed at BL28B2, SPring-8 [9]. Figure 2 shows one of the preliminary results. The sample measured here is a single crystal molybdenum mirror, which had been previously studied in detail by monochromatic X-rays [10]. The setup used is schematically shown in the inset. A Si drift chamber (Röntec, XFlash, 5mm<sup>2</sup> effective area, Peltier cooling -10°C) and a silicon photo-diode were used for measurements and alignment, respectively. The data are processed by a digital processing unit (Seiko, MCA7600). The beam size used was 50 μm(H) × 25 μm(V), and the flux was even too much for the detector. To suppress high background, a beam out of the orbital plane was used. Since the entrance slit was placed 3 mm off (above) center, the expected spectra should move more to the lower energy side, compared with the calculation shown in Fig.1(b) (2mm off). During alignment, the maximum reflected X-ray intensity (measured by a photo-diode) was obtained at 1.23 mrad. As an attenuator for the beam, a 7 mm thick aluminum plate was employed. In addition, a 0.3 mm thick copper foil and a 1mm thick iron plate were used for  $\theta_0=1.23$  and 2.5 mrad, respectively. One can see several features in Fig.2; (i) a lot of sharp spikes are found in the low energy side, (ii) besides such spikes, the baseline of the reflectivity curve has a peak, (iii) the peak position depends on  $\theta_0$ , (iv) at the higher energy side of the peak, the intensity decays quickly.

The strong low energy peaks were from the receiving slit (made of tantalum), the direct beam stopper (made of nickel-tungsten alloy) and other parts (mainly made of stainless steel). As their characteristic X-rays and the sum peaks degraded X-ray reflection data, apparently, it is necessary to remove such background. However, one should carefully choose the method so that the shape of the X-ray spectra does not change. Unfortunately, the present experiments were not successful in this point. As  $\theta_C$  for 16 keV is 3.7 mrad [10],  $E_C$  for 1.23, 2.07 and 2.5 mrad are 48.1, 28.6 and 23.7 keV, respectively. As seen in Fig.2, the reflection peak is lower than  $E_C$  for all three cases; in particular, for  $\theta_0=1.23$  mrad, the peak was far from  $E_C$ . The intensity at the energy above  $E_C$  was very weak as well. That is, low reflectivity in high energy range was not compensated by the shape of incident X-ray spectra. This suggests that in the present experiments we lost too much intensity at the higher energy side, and possibly we increased continuum background in low energy region as well.



**FIGURE 2.** Reflection spectra of single crystal molybdenum mirror. White synchrotron beam emitted out of plane (3 mm off at 44m position at BL28B2, SPring-8). Schematics of experimental setup are shown as an inset.

On the other hand, in the energy-dispersive experiments, the observed spectra should be normalized by incident X-ray spectra. This has been one of big problems for this type of experiments so far. When it is not easy to obtain that experimentally, instead, measuring reference samples at several specific angles would be required. To use such indirect data properly, the calibration process needs to be as simple as possible.

## CONCLUSIONS

The use of high-energy white synchrotron X-rays as a source for energy-dispersive X-ray reflectometry seems promising. The method can open up novel applications which have not been possible by the conventional X-ray reflectometry based on a  $\theta/2$   $\theta$  scan. During the preliminary experiments, we found the following: (i) Designing a device to prepare a small beam around  $\mu\text{m}$  is extremely important for future experiments. The beam intensity is strong enough even when only a small part is used for the present experiments. (ii) The use of a beam out of the orbital plane is effective for removing those unnecessary parts where the energy is too high. The attenuator can reshape the incident X-ray spectra so that higher intensity is available for high  $q_z$  in reflectivity measurements. The parameters are sensitive, and therefore need to be carefully chosen. Unfortunately, the present trial was not satisfactory. (iii) Shielding the detector from the background is crucial. It is necessary to deal with the background from any of the slits, in particular, the receiving slit and also from the direct beam stopper. Background correction will be practically important, compared with a normal  $\theta/2$   $\theta$  scanning measurement. (vi) Some good methods for the normalization should be developed.

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