X-ray imaging based on small-angle X-ray scattering using spatial coherence of parametric X-ray radiation

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Abstract. X-ray imaging based on small-angle X-ray scattering (SAXS) was carried out using the parametric X-ray radiation (PXR) source at the Laboratory for Electron Beam Research and Application (LEBRA) of Nihon University. The experimental setup employed in this novel imaging approach is the same as that employed in diffraction-enhanced imaging (DEI), a kind of X-ray phase-contrast imaging method. In SAXS-based imaging, the image contrast is correlated with the broadening of the rocking curve peak due to the scattering from micron- or sub-micron-sized grains in the sample material. An experiment using the 25.5-keV PXR beam demonstrated that SAXS-based imaging with PXR provides a substantially strong contrast for granular materials despite the extremely low density of the material.

1. Introduction

A tunable and monochromatic X-ray source based on parametric X-ray radiation (PXR), a phenomenon caused by the interaction between a relativistic electron and a crystal medium, was developed at the Laboratory for Electron Beam Research and Application (LEBRA), Nihon University [1, 2]. The PXR beam was obtained using a 100-MeV electron beam from the linac of LEBRA, and the LEBRA-PXR source has a double-crystal system for a wide tunability [3]. A PXR beam from 4 to 34 keV is available using Si(111) or Si(220) planes as the radiator. The current specifications of the LEBRA-PXR source are listed in Table 1. Because the PXR irradiation field is large (100 mm in diameter), X-ray imaging experiments have been the most popular application of the PXR source [4, 5]. In particular, the results of diffraction-enhanced imaging (DEI), which is a sort of X-ray phase-contrast imaging, have demonstrated the high performance of PXR with respect to spatial coherence [6].

Recently, a new imaging technique based on small-angle X-ray scattering (SAXS) has been of special interest for next-generation X-ray imaging. This technique is derived from phase-contrast imaging, and the same setup can be used for measurement. For instance, SAXS-based imaging using a Talbot interferometer has successfully been demonstrated [7]. In principle, information...
on the SAXS effect due to an object material can be obtained using a DEI measurement system. Therefore, the DEI experiment was carried out to demonstrate the applicability of PXR to SAXS-based imaging.

Table 1. Typical parameters of the linac and the PXR source of LEBRA.

| Parameter                                | Value                        |
|------------------------------------------|------------------------------|
| Electron energy                          | 100 MeV (typ.)              |
| Accelerating frequency                   | 2856 MHz (S-band)           |
| Macropulse beam current                  | 120 – 135 mA (typ.)         |
| Macropulse duration                      | 4 – 5 μs (typ.)             |
| Macropulse repetition rate               | 2-3.5 pps                    |
| Average beam current                     | 1 – 5 μA                    |
| Beam size on the target                  | 0.5 – 2 mm in dia. (FWHM)   |
| PXR energy range                         | Si(111) target: 4.0 – 20.5 keV |
|                                          | Si(220) target: 6.5 – 34 keV |
| Irradiation field at the X-ray exit      | 100 mm in diameter          |
| Total X-ray photon rate                  | ~ 10^7 /s @17.5 keV         |

2. Diffraction-enhanced imaging using PXR
The radiation cone of PXR depends on the energy of the incident electron, as is the case with other radiation phenomena. In the case of the LEBRA-PXR source, the actual aperture angle of the PXR cone-beam extracted from the X-ray exit is determined by the size of the exit window 7.3 m distant from the radiator crystal irradiated with the electron beam, and is estimated to be 13.7 mrad. On the other hand, a PXR beam has spatial chirp, which means a slight energy shift according to the change in the X-ray emitting direction, along the horizontal plane. Combining the cone like divergence and spatial chirp of PXR allows the diffraction of the whole PXR beam when an analyzer crystal for DEI (third crystal) is set up in the (+, −, +) arrangement as shown in Fig. 1. The rocking curves observed at the third crystal have almost the same shapes as in the case of plane-wave-like X-ray beams. The narrow diffraction widths for the PXR beam mean that the DEI measurement system using the LEBRA-PXR source has an angular resolution of 10^{-6} rad despite the cone-beam aperture of the mrad order [8].

![Figure 1. Schematic top view of the setup of the DEI experiment using the LEBRA-PXR source. The same crystal plane as the PXR double-crystal system is used for the analyzer crystal.](image)

There are three considerable interactions between X-rays and materials as the rays pass through the sample: absorption corresponding to X-ray amplitude attenuation; refraction resulting from X-ray phase-shift; and, when the sample material is granular, small-angle scattering due to micro structures of the sample materials. These effects can be evaluated using the result of DEI measurement, which corresponds to the rocking-curve measurement for each pixel of the image detector. Here, we refer to an angular distribution of the X-rays diffracted by the analyzer including the influence of samples as a rocking curve. The X-ray
absorption effect is estimated from the reduction of the rocking-curve peak area. The angle of X-ray refraction is measured as the angular shift of the peak center of the rocking curve. Finally, the peak broadening of the rocking curve reflects the effect of SAXS [9]. The angular resolution for the refraction and the scattering depends on the diffraction width of the analyzer crystal.

3. Experimental results of DEI and SAXS-based imaging

To demonstrate SAXS-based imaging using PXR, a DEI experiment for samples of light materials was carried out in the setup shown in Fig. 1. Since the rocking curves for higher-energy X-rays are narrower, the higher energy of X-rays is advantageous in detecting slight refraction and/or scattering of X-rays in DEI experiments. Thus, 25.5-keV PXR from the Si(220) double-crystal system was used, though the photon rate is estimated to be one order of magnitude less than that of the 17-keV PXR from the Si(111) double-crystal system. The X-ray photon density at the sample was roughly estimated to be on the order of $10^2$ mm$^{-2}$ s$^{-1}$.

![Figure 2. Image of the samples for the DEI experiment using a 25.5-keV PXR beam. From left to right, an acrylic rod, a styrene-foam rod, and a polystyrene rod with respective diameters of 3 mm, 6 mm, and 3 mm and respective densities are 1.17, 0.16, and 0.986 g/cm$^3$.](image)

Acrylic-resin, styrene-foam, and polystyrene cylindrical rods were selected as samples and are shown in Fig. 2. The X-ray absorption in such materials is extremely small, especially in styrene-foam. The DEI analyzer crystal was a 5-mm thick Si(220) plate having a dimension of 160 mm $\times$ 35 mm and was mounted on a gonio stage having an angular step of 0.4625 $\mu$rad. Since the Bragg angle is 7.25$^\circ$ for 25.5 keV, the imaging area was limited to 20 mm $\times$ 35 mm.

![Figure 3. Results of the DEI experiment for the samples as shown in Fig. 2. The DEI contrast varies according to the analyzer angle $\theta$.](image)

In the DEI experiment, 13 DEI images were obtained as a function of the analyzer angle $\theta$, where the peak of the rocking-curve for the direct PXR beam is defined as $\theta = 0$. Figure 3 shows several examples of DEI images taken by an X-ray CCD with a resolution of 24 $\mu$m $\times$ 24 $\mu$m pixels. Because the quantum efficiency of the detector is 10% at most for 25.5-keV X-rays, 15-min exposure was necessary for each image measurement, and the total measurement time was longer than 3 hours. The contrast of the DEI images strongly depends on the analyzer angle. The rocking curves at the direct beam region and at the styrene-foam region in the DEI images are plotted as shown in Fig. 4. The rocking curve of the styrene-foam region is an order of micro-radian broader than that of the direct beam region. The peak broadening effect seems to be caused by SAXS due to the micro-structure of styrene-foam.
3.1. Absorption-contrast image
The absorption-contrast image is obtained by an integral of the DEI image $I(x, y, \theta)$ with respect to the analyzer angle $\theta$, 

$$I_{abs}(x, y) = \int I(x, y, \theta) \, d\theta = \sum_i I(x, y, \theta_i).$$

(1)

The logarithmic function $\ln(I_{abs}/I_0)$ means the absorption coefficient, where $I_0(x, y)$ denotes the intensity map of the direct X-ray beam. Figure 5 shows the absorption-coefficient map calculated from the DEI images. In general, a complex refraction index $n(x, y)$ at the position $(x, y)$ in a two-dimensional coordinate system is expressed as $n(x, y) = 1 - \delta(x, y) + i\beta(x, y)$, where $\delta$ and $\beta$ denote the phase-shift coefficient and the extinction coefficient, respectively. Except in the anomalous dispersion region, both $\delta(x, y)$ and $\beta(x, y)$ are almost proportional to the material density $\rho(x, y)$. Since $\beta$ is proportional to the absorption coefficient, the absorption-contrast image contains information about the density map $\rho(x, y)$. The contrast, however, is very weak for such light materials, especially styrene-foam.

3.2. Phase-contrast image
The phase-gradient map obtained by the calculation 

$$I_{grad}(x, y) = \int \theta I(x, y, \theta) \, d\theta / \int I(x, y, \theta) \, d\theta = \sum_i \theta_i I(x, y, \theta_i) / \sum_i I(x, y, \theta_i),$$

(2)

corresponds to the partial differential of the phase-shift $\delta(x, y)$ with respect to the $x$ direction, and is also referred to as a refraction-contrast or differential phase-contrast image [10]. The result of the process for the phase-gradient map is shown in Fig. 6. The phase-shift itself is obtained by the integral of the phase-gradient map along $x$, 

$$\delta(x, y) = \int \partial \delta(x, y) / \partial x \, dx \propto \int I_{grad}(x, y) \, dx,$$

(3)

and Fig. 7 shows the result of the image calculation. The phase map essentially has the same information as the absorption-contrast image, but the sensitivity is drastically improved.
3.3. SAXS-based imaging

When the X-ray attenuation in the sample is negligible, the peak-height reduction of the rocking curve is almost equivalent with the peak broadening due to the SAXS effect. Thus, there should be several methods to quantify the effect as the image contrast. For simplicity, we defined the contrast due to the SAXS effect as the difference between the image of the analyzer angle $\theta = 0$ and the image of $\theta = 2\sigma$, namely, $I_{\text{saxs}}(x, y) = I(x, y, 0) - I(x, y, 2\sigma)$, where $\sigma$ denotes the standard deviation of the rocking curve for the direct PXR beam and is estimated to be 3.93 $\mu$rad in this case. This definition can provide a comparatively strong contrast without complicated calculation. Figure 8 shows the result of this process. Here, the average of $I(x, y, -7.93 \mu$rad) and $I(x, y, +8.27 \mu$rad) was substituted for $I(x, y, 2\sigma)$, since the angle $\theta$ was a discrete parameter. The SAXS-based contrast is extremely sensitive to the styrene-foam region independently of the material density and the sample macro-shape. The contrast is thought to be attributable to the micrometer-ordered micro structures in the sample material, and is essentially equivalent to the visibility contrast in the case of the Talbot-interferometer method [11].

Here, the distribution of small-angle scattering contains information about the particle-
domain size. Figure 9 shows another expression of the rocking curves in Fig. 4, using the variable conversion \( q = \left( \frac{4\pi}{\lambda} \sin(\theta/2) \right) \), where \( \lambda \) is the X-ray wavelength. Such a logarithmic graph as a function of \( q^2 \) is referred to as a Guinier plot in the field of typical SAXS analysis. At the region of small \( q^2 \), the SAXS distribution can be approximated by \( \exp(-R_g^2 q^2 / 3) \), where \( R_g \) means the inertial radius of particles in the sample material. Thus, the average of \( R_g \) is estimated from the slope of the Guinier plot [12]. When DEI data are used, however, the intrinsic diffraction width of the analyzer-crystal itself restricts the detection of the effect due to large particles. The slope of the Guinier plot for the direct beam (red line) corresponds to \( R_g = 1.57 \) \( \mu m \), which is the upper limit of the detectable structure size under this condition. From the slope for the styrene-foam region (blue line), \( R_g \) is estimated to be 1.00 \( \mu m \). Since this value contains the effect of the analyzer diffraction width, the actual inertial radius of the styrene-foam structure may take on a value between 1.00 and 1.57 \( \mu m \). Nevertheless, we can conclude that the SAXS effect in DEI is sensitive to structures much smaller than the 24 \( \mu m \) pixel size of the image sensor. For a more exact estimation, the sample thickness has to be optimized and a lot of measurement points are necessary.

4. Summary
Combining the cone-like divergence and the spatial chirp of PXR allows DEI using a PXR beam, which is sensitive to the refraction and the small-angle scattering of X-rays passing through the sample materials with an angular resolution of \( 10^{-6} \) rad order. The DEI experiment using PXR successfully demonstrated that SAXS-based imaging is sensitive to micro-structures of sample materials smaller than the pixel size of the image sensor. The results proved that the PXR beam has a sufficient spatial coherence to detect scattering angles in the micro-radian range.

In addition to the sensitivity to granular materials with low densities, SAXS-based imaging could be used for the quantitative analysis of material structures of the sub-micron order. Thus, the novel imaging technique would be a useful method for material science and fundamental medical research.

Acknowledgments
This work was partly supported by MEXT. KAKENHI (24560069, 24651105 and 25286087) and by Grants-in-Aid for Scientific Research, Nihon University (S12-19, Takashi Kaneda).

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