Oscillation photography applied to resonant x-ray diffraction

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Abstract. Recently, resonant x-ray diffraction has been used for detecting charge, magnetic and multipole orders. In practice, however, this technique surveys only a small portion of the reciprocal space. In this study, we applied oscillation photography to resonant x-ray diffraction. For a test sample, we explored nearly a whole Brillouin zone and successfully observed resonant peaks. This feasibility study indicates that oscillation photography is useful for expanding observable areas and enhances the capability of resonant x-ray diffraction.

1. Introduction
Resonant x-ray diffraction (RXD) is a powerful technique for detecting multipole orders in 4f and 5f electron systems. For example, the octupole order in Ce0.7La0.3B6[1] and the quadrupole order in UPd3[2] were actually verified in RXD experiments. However, this technique is limited to measuring several tiny points or narrow lines in a vast reciprocal space, because tight collimation is needed to disentangle weak resonant signals from large background noise, such as sample fluorescence and scattering from the cryostat vessel. Hence RXD is applicable only when the modulation wave vector is known.

In normal x-ray diffraction, oscillation photography is widely employed to determine crystal structures, including super-structures. Oscillation photography utilizes large two-dimensional detectors and observes almost all reciprocal space by rotating the sample. Standard two-dimensional detectors have a very wide dynamic range and thus not only strong Bragg peaks but also weak super-lattice peaks can be observed, if background noise is sufficiently suppressed. Reduction of the background is therefore of great importance in oscillation photography. This is also probably a reason why oscillation photography has not been used in RXD, in which high fluorescence background noise is inevitable.

In this work, we applied oscillation photography to RXD. We investigated a test sample of GdPd2Al3, which shows antiferromagnetic order below 17 K. Nearly the whole Brillouin zone was surveyed and resonant magnetic diffraction signals were distinctly observed. Based on the results obtained, we discuss the feasibility of this combined technique for detecting unknown multipole orders in 4f and 5f systems.
2. Experimental
The test sample GdPd$_2$Al$_3$ undergoes an antiferromagnetic transition below $T_{N1}$=17 K, followed by a second transition at $T_{N2}$=13 K[3]. The crystal structure is hexagonal ($P6/mmm$). $a$ and $c$ are 5.39 Å and 4.19 Å, respectively. The magnetic structure below $T_{N2}$ is a helical structure in the $ac$-plane with the modulation vector $(q_M,q_M,0)$, where $q_M$ ~ $\frac{1}{4}$[4].

A resonant x-ray diffraction experiment was carried out at beamline BL22XU in SPring-8. The photon energy was the Gd $L_2$ absorption edge 7.930 keV. The single crystal GdPd$_2$Al$_3$ was grown with the Czochralsky pulling method in a tetra-arc furnace. The sample was cut in a parallelepiped of dimensions 4 mm×4 mm×2 mm and the (110) surface was polished. The sample was attached to the cold head of a conventional closed-cycle refrigerator, which was mounted on a conventional four-circle diffractometer with horizontal scattering plane. Orientation of the sample was checked using 110 and 111 reflections and we set the $c$-axis of the sample perpendicular to the scattering plane. Hence the $\omega$, $\phi$ and $c$-axes were (nearly) parallel to each other. The width of the rocking curve of the 110 reflection was about 0.07°.

As a two-dimensional detector, we used the PILATUS 100K, which is a pixel array of Si detectors [5]. The number of pixels is 487×195 and the total detector size is 83.8 mm×33.5 mm. Each pixel is a complete single-photon counting device. Each pixel has its own preamplifier, discriminator and scaler. By selecting an adequate threshold for the discriminators, thermal noise and sample fluorescence can be considerably reduced.

3. Results and Discussion
First we observed resonant magnetic reflections using a scintillation detector. We measured the peak intensity of the magnetic reflection at $(1-q_M,1-q_M,0)$ as a function of photon energy and set the photon energy to the resonance energy 7.934 keV. In order to avoid beam heating at the sample surface, we reduced the incident photon flux by a factor of 1/50. In this condition, the peak intensity at $(1-q_M,1-q_M,0)$ was about 8000 counts/s. The temperature and polarization dependences were also measured and will be published elsewhere[4].

After this groundwork, we set up the two-dimensional detector. The scattering angle $2\theta$ at the center of the detector was set to be 33.72° ($2\theta$ of the 110 reflection). The camera length was 140 mm. We rotated the $\phi$-axis from 5 to 40° in 0.05° increments. When $\phi=0$, [110] was perpendicular to the incident x-ray. We took an image at each $\phi$ and obtained a total of 701 images at 8 K. The exposure time was one second. We repeated the same measurement at 20 K (above $T_{N1}$).

We now show the worst example from this experiment. Fig.1(a) is a raw image taken at $\phi=36.55^\circ$ and 8 K. The $x$-axis is horizontal and the $y$-axis is vertical. In this image, the magnetic

![Figure 1](image.png)

Figure 1. (a) Raw image taken at $\phi=36.55^\circ$ and 8 K. (b) Difference image at $\phi=36.55^\circ$ obtained by subtracting data taken at 20 K from data taken at 8 K.
Bragg peak of 700 counts at \((1-\mathbf{q}_M,2-\mathbf{q}_M,0)\) is hidden. Because of relatively high backgrounds (~1500 counts), powder lines due to scattering from the Beryllium cryostat vessels, and speck noise, it is almost impossible to find the magnetic Bragg peak. However, by subtracting data taken at 20 K from data measured at 8 K, these background noises are significantly suppressed. Eventually, the magnetic Bragg peak unequivocally emerges at \(x=138\) and \(y=118\), as shown in fig.1(b).

From difference images at all \(\phi\), we constructed the intensity map for the reciprocal space near the \(hk0\) plane shown in fig.2. In this map, data between \(y=111\) and \(y=140\) were summed. Red lines indicate Brillouin zones. Correction of the flat detector was not made, thus the coordinates are approximate. Four magnetic Bragg peaks are distinctly observed near \((\tfrac{5}{3},\tfrac{2}{3},0)\), \((\tfrac{2}{3},\tfrac{2}{3},0)\), \((\tfrac{4}{3},\tfrac{4}{3},0)\) and \((\tfrac{2}{3},\tfrac{5}{3},0)\). Although a blind area exists at the lower right, it is found that almost a whole Brillouin zone was surveyed. As for the data along the \(c^*\)-axis, an area between \(\pm\tfrac{1}{2}c^*\) is observed, if the \(hk0\) plane is set to the center of the detector.

The sensitivity of this technique is estimated as follows. Residual noise in fig.1(b) has a Gaussian distribution with \(\sigma=60\) counts. We thus estimate the lowest intensity detectable is 120 counts (2\(\sigma\)). The peak intensity of the magnetic reflection at \((1-\mathbf{q}_M,1-\mathbf{q}_M,0)\) was 2300 counts on the PILATUS detector. Hence about 1/20 of the \((1-\mathbf{q}_M,1-\mathbf{q}_M,0)\) peak is the lowest limit for

![Figure 2](image-url)

**Figure 2.** Reconstructed reciprocal-lattice layer of the (approximate) \(hk0\) plane deduced from difference images. Red lines indicate Brillouin zones. The strong peak at the center is the 110 structural reflection. Weak but clear resonant magnetic peaks are visible near \((\tfrac{5}{3},\tfrac{2}{3},0), (\tfrac{2}{3},\tfrac{2}{3},0), (\tfrac{4}{3},\tfrac{4}{3},0)\) and \((\tfrac{2}{3},\tfrac{5}{3},0)\).
this technique. In the measurements using point detectors, the peak intensity at \((1-q_M,1-q_M,0)\) was about 8000 counts and the peak intensity of the 110 reflection is estimated to be \(3 \times 10^8\) counts. The ratio of \(8000 \times \frac{1}{20}\) to \(3 \times 10^8\) is about \(10^{-6}\). Since the 110 reflection is one of the strongest structural peaks, we conclude that oscillation photography in RXD can detect peaks as small as \(10^{-6}\) of the strongest peak. On the other hand, the detection limit of the normal RXD technique using a point detector is estimated to be about \(10^{-7}\). Therefore this new technique is inferior to standard RXD methods about an order of magnitude in detection limit.

4. Summary
We have carried out a resonant x-ray diffraction experiment combined with oscillation photography. Prior to the experiment, we expected that high fluorescence background noise would prevent the detection of weak resonance peaks. However, it turned out that resonance peaks were rather easily observed. In particular, subtraction of a high-temperature image from a low-temperature image remarkably reduces background noise and significantly improves the detection limit of this technique. The evaluated detection power is about \(10^{-6}\), which is quite good. Since a wide reciprocal space is explored in a short time, this technique may be adequate for the first step of resonant x-ray diffraction experiments. All possibilities are examined at the beginning. There are some compounds whose order parameters have not been identified, such as URu$_2$Si$_2$. This technique might be applied to those “hidden order parameters”.

References
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