Tuning of X-ray phase retarder for magnetic EXAFS spectroscopy in helicity modulation mode

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A technique for polarization tuning over a wide energy range has been developed for magnetic EXAFS spectroscopies using a diamond x-ray phase retarder in helicity modulation mode. A quick rocking scan using a piezo stage enabled us to tune the angle of retarder crystal with the precision of ± 2 arcsecond within 1 s. Available energy range of circular dichroism measurements has been extended to 6–16 keV by adopting not only undulator gap scan but also the present tuning technique. This technique has been applied to magnetic EXAFS measurements in pure iron at the Fe K-edge. Higher signal-to-noise ratio was obtained in reduced data acquisition time.

Keywords: magnetic EXAFS, XMCD, x-ray phase retarder, helicity modulation, circular polarization

1. Introduction

Magnetic EXAFS (MEXAFS) spectroscopies (Schütz *et al.*, 1987; Schütz & Ahlers, 1996) are methods to investigate a local magnetic structure and magnetic short-range order. In case of K-edge absorption the relative magnetic contribution is in the order of only 10^{-3} compared to the amplitude of the normal EXAFS oscillations. Furthermore, in case of a diluted magnetic system, in which the MEXAFS method provide us with valuable information about the local spin correlation, much smaller MEXAFS signal must be detected. Applications of MEXAFS studies has been limited owing to such a week signal requiring high statistical accuracy and long acquisition time. In addition, most previous MEXAFS studies has been so far limited in materials of easily magnetized because a magnetic field reversal method was used (Schütz *et al.*, 1987; Nakamura *et al.*, 1998).

Recently developed helicity modulation (HM) technique (Suzuki *et al.*, 1998; Suzuki *et al.*, 1999) using x-ray crystal phase retarders (XPR) (Hirano *et al.*, 1991; Giles *et al.*, 1993) provides us with extremely high quality spectra of X-ray magnetic circular dichroism (XMCD) and feasibility of XMCD measurements in saturating/non-saturating system (Koizumi *et al.*, 2000) or under high magnetic field generated with a superconducting magnet (Maruyama *et al.*, 2000; Uemoto *et al.*, 2000). By applying the HM technique to the MEXAFS spectroscopy, the limitations mentioned above can be removed, so that MEXAFS studies with high precision will be realized.

Our preliminary polarization measurements in a static mode showed that diamond (111) crystals efficiently functions as a XPR in the $2\bar{2}0$ Laue transmission geometry between 6 and 16 keV. The diamond crystals of different thickness, 0.45, 0.73 and 2.7 mm, covers 6–9, 8–12 and 11–16 keV with sufficiently high rate of circular polarization, and the transmittance are 5–58, 25–73 and 20–65% in the respective energy range. In principle the HM technique can be used at the x-ray energy where the phase retarder works in the static mode. However, there is difficulty in the practical use of the HM technique in a wide energy range.

In the HM technique, fast helicity switching is made with a retarder crystal moving around the diffraction condition in bi-stable mode. To give right- (RHC) and left-handed circularly (LHC) polarization of opposite helicities, the center of oscillation must be set at the exact Bragg condition at every energy points while a spectral scan. Otherwise, the absolute value of circular polarization rates will be different for the RHC and LHC, and incorrect XMCD data can be obtained. The accuracy and reliability in controlling the XPR angle to follow the energy of monochromatic x-rays is an essential key in the HM technique.

In narrow energy range of typically 100 eV near an absorption edge, the oscillation center can be tuned only using a stepping motor-driven ω-stage. A quadratic fit function is used to correlate the pulses for rotation with the x-ray energy. XMCD spectra previously reported (Suzuki *et al.*, 1998; Suzuki *et al.*, 1999) were recorded using this method. On the other hand, in wider energy range over several keV, any polynomial fit functions or even the Bragg equation did not reproduce the practical energy dependence of the oscillation center with a sufficient accuracy.

To overcome this problem, we have adopted a method for quick and accurate tuning of oscillation center to the Bragg condition. This method is based on a quick rocking scan of the retarder crystal using a piezo translator and monitoring the Laue diffraction intensity from the used diamond $2\bar{2}0$ plane. In this paper, we present a technique of polarization tuning in wide energy range (6–16 keV) for MEXAFS measurements. High quality MEXAFS spectra at iron K-edge is presented to illustrate the performance of our technique.

2. Experiment

2.1. Instrumentation

Instruments for the present method for tuning an XPR was installed at a hard x-ray undulator beamline 39XU (Goto *et al.*, 1998; Maruyama *et al.*, 1999) of SPring-8. An experimental setup for MEXAFS was similar to that was used for XMCD experiments in the HM mode (Suzuki *et al.*, 1998). One of the improvements was electronics providing a driving voltage for a piezo-driven oscillation stage (Hirano *et al.*, 1992). A computer-controlled variable dc voltage source was added to give an offset dc voltage to a piezo translator (Physik Instrumente P-841.40) together with an ac voltage generated with a function generator. The piezo was used with a closed-loop operation and respond to sine wave at 100 Hz. Its full stroke corresponded to the angular amplitude of 380 arcsecond for the external control voltage of 0–10 V. The hysteresis was found so small as to be negligible for the present application. In addition, the x-ray intensity diffracted from a diamond crystal of XPR was monitored with an ionization chamber on a 2θ-arm of an ω-2θ rotation stage, and output signal was taken with an analog/digital converter board (National Instruments PCI-MIO-16XE-50) on a computer.

2.2. Phase retarder tuning procedure

The procedure for tuning the XPR angle is as follows: (i) after the x-ray energy is changed with a monochromator, the oscillation center of the retarder crystal is roughly moved to the Bragg angle using a stepping motor-driven ω-stage. The pulses for ω rotation is given according to the Bragg equation. (ii) The ac voltage applied

to the piezo is turned off to stop the oscillation of the crystal. (iii) The quick rocking scan is executed by applying step dc voltages to the piezo. The intensity of the Laue diffraction is monitored with an ionization chamber placed on the 2θ-arm. Quick data collection can be made within 1 sec for 20 measuring points owing to the fast response of the piezo and sufficiently intense diffracted beam. (iv) After searching the peak position of the rocking curve, the proper dc offset voltage is applied to the piezo to reproduce the Bragg condition. (v) The ac voltage is applied together with the dc offset voltage to start the oscillation of the crystal again.

2.3. Precision of the tuning

Figure 1 compares the angle of the XPR crystal measured before the quick tuning (open circles) with after that (filled triangles). Plotted in the figure are the angular deviations from the expected angle for diffraction. Before the tuning, we have made measurements of the rocking curve for the $2\bar{2}0$ Laue reflection from a diamond (111) crystal of 0.73 mm-thickness scanning the ω-rotation stage, from 6 to 16 keV. The measured dependence of the rocking curve center on the x-ray energy (wavelength) was reproduced by the Bragg equation with a lattice spacing of diamond 220 plane, 1.261 Å. However, as shown by the circles in Fig. 1 the deviations of a few tens arcsecond still remained. The deviations are too large since the precision and the reproducibility of at least a few arcseconds are required for tuning the XPR used as the quarterwaveplate (QWP) in the HM mode. The observed deviation probably came from flexing of the ω-rotation stage. Other possible reason was the energy fluctuation due to the mechanical instability in a beamline monochromator. After the quick tuning, the deviation has been successfully reduced as shown by the triangles in Fig. 1. The angle of the XPR was tuned within ± 2 arcsecond from 6 to 16 keV.

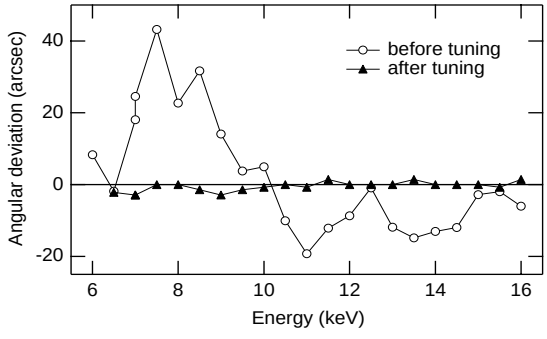

Figure 1
The angle of a diamond x-ray phase retarder measured before (open circles) and after the quick piezo tuning (filled triangles). Plotted are the angular deviations from the expected Bragg angle for the diamond $2\bar{2}0$ Laue reflection.

Polarization tunability of the quick tuning method was confirmed by linear polarization measurements with the XPRs used as a halfwaveplate (HWP). Linear polarization rates were measured by monitoring x-ray intensities scattered from a polyimide foil placed after the XPR. The scattered intensities into the horizontal and vertical directions were detected using two NaI scintillation counters, and their difference gave the linear polarization rate (Hayakawa *et al.*, 1997). This method is based on the polarization dependence of the Thomson scattering, and we could make linear polarization measurements in wide energy region with the same experimental setup. Circularly polarization measurements might be preferable to confirm the polarization tunability because the XPR is used as a QWP for MEXAFS measurements. However, more complicated setup of x-ray polarization optics is needed for the circular polarization measurements, and continuous x-ray energy scan is difficult. The difference in the tuning condition of XPR for between a QWP and a HWP is only the angle of offset amplitude. Besides, more precise tuning is required for the HWP condition than that for the QWP because variation of the phase shift is lager in the condition for HWP than that for QWP. Therefore, more sensitive measurement could be made.

Figure 2 shows the linear polarization rates, $P_{\rm L}$ measured after the XPR tuned to a HWP, where $P_{\rm L} = -1$ corresponds to perfect linear polarization in the vertical plane. Diamond crystals of different thickness, 0.45, 0.73 and 2.7 mm were tested in the $2\bar{2}0$ Laue transmission geometry. We have preliminarily determined the available vertical polarization rate for each XPR thickness typically using a 1 keV step. The XPR crystal was scanned around the $2\bar{2}0$ diffraction angle at each energy, and the minimum values of $P_{\rm L}$ were plotted by the filled makers in Fig. 2. The decrease in $|P_{\rm L}|$ for each crystal thickness at higher energies was due to the finite angular divergence of the incoming x-ray beam (Giles *et al.*, 1993). The observed vertical polarization rates were agreed with the convolution of calculated values with the measured profile of the rocking curve. At the condition for QWP the derivative of the phase shift is a quarter of that for HWP, so that the degradation of the circular polarization is less.

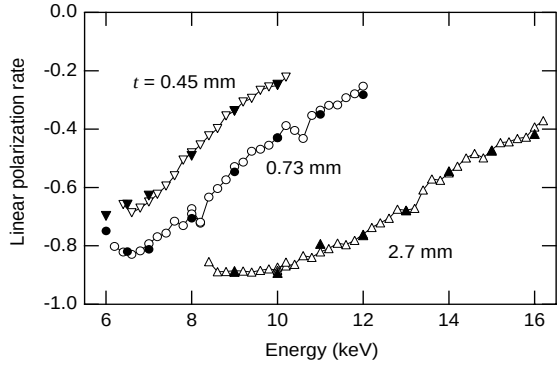

Figure 2
Linear polarization rates $P_{\rm L}$ measured after the diamond x-ray phase retarder tuned to a halfwaveplate. The $P_{\rm L}$ after the quick piezo tuning (open makers) traces the standards (closed markers) for each crystal thickness.

The open makers in Fig. 2 show that the $P_{\rm L}$ measured after the quick tuning exactly traces the standards (filled makers) for each XPR thickness. The dips observed at 8.2 and 10.6 keV for the 0.73 mm crystal came from the glitch of the retarder crystal. These were removed by changing the azimuth of the crystal. This result indicates that our method for polarization tuning worked well. The time needed for tuning was less than 1 sec at each energy point.

2.4. Undulator gap scan

In order to maximize the incoming x-ray intensity over the EXAFS region, synchronous tuning between the undulator gap and the angle of the monochromator (gap scan) (Rogalev *et al.*, 1998) is inevitably used. To achieve accurate gap scan over wide energy region, we used a function which correlates the undulator gap with the x-ray energy,

$$G = \alpha \ln\left(\frac{\beta}{E} - 1\right) + \gamma, \qquad (1)$$

where, G (mm) represents the undulator gap, E (keV) is the x-ray energy, α, β and γ are fit parameters. This equation is derived from a relation between the peak energy for on-axis radiation of a planar undulator and the deflection parameter, K, with assuming that the K is an exponential function of the undulator gap, G. The parameters, $\alpha = -4.988$, $\beta = 18.65$ and $\gamma = 13.72$ were used to fit measured variation of the x-ray energy of fundamental harmonics with the gap value.

By using the Eq. (1) with three parameters above, we could tune the undulator gap to the optimum value within ± 10 μm at 6–16 keV, and the deviation of the x-ray energy from the fundamental peak was less than 10 eV. The undulator gap was changed at every energy point while recording an MEXAFS spectrum, and we obtained smooth I_0 spectrum. The parallelism of a double-crystal monochromator was also tuned. In addition, the procedure of polarization tuning that we described in Sec. 2.2. is needed for the HM spectroscopy. We have installed instruments and computer programs that satisfy these requirements at BL39XU of SPring-8.

3. Magnetic EXAFS spectrum

We have made an MEXAFS measurement for pure iron at the Fe K-edge using the HM technique improved with the present tuning method, at the beamline. The diamond crystal of 0.45 mm thickness was used for the phase retarder. The frequency of helicity modulation was 40 Hz. Figure 3 shows a measured MEXAFS spectrum. The data were collected over the energy range from 6.8 to 8 keV totally for 300 min. The output of the lock-in amplifier was averaged for 10 s for each energy points. The measuring time for one scan (568 points) was approximately 150 min including time for moving the undulator gap, the monochromator and the XPR. Two energy scans were made for the directions of magnetic field parallel and antiparallel to the x-ray wavevector to cancel out non-magnetic backgrounds. The averaged spectrum containing no background is shown. No further smoothing was applied to presented data. The present spectrum agrees well with those reported earlier (Schütz & Ahlers, 1996; Nakamura *et al.*, 1998). The improved signal-to-noise ratio was achieved with the reduced acquisition time.

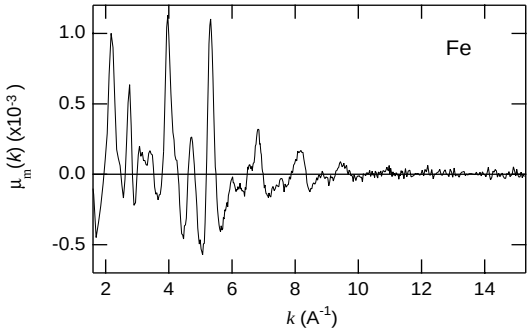

Figure 3
A magnetic EXAFS spectrum, $\mu_{\rm m}(k)$, measured for pure Fe foil at the Fe K-edge, using the helicity modulation technique.

We have extended the usable energy range for dichroism measurements in the HM mode, and MEXAFS spectroscopy of higher signal-to-noise ratio has become available. By using the high sensitivity of the HM technique, MEXAFS applications to diluted systems, e. g. investigation of a local spin pair functions of magnetic impurities, can be realized. In addition, systematic data collections of MEXAFS spectra, which have so far required long acquisition time, such as temperature or composition dependence can be executed in a realistic beam time. Since reversal of the magnetic field is unnecessary in the HM mode, a superconducting magnet can be used for MEXAFS studies in hard magnetic materials under high magnetic field.

4. Summary

We have developed a quick and precise method for polarization tuning to extend the available x-ray energy range of the HM technique. The quick rocking scan using the piezo stage provides us with the precision of ± 2 arcsecond in the XPR angle within 1 s. This polarization tuning method was combined with the HM technique, and applied to the high-precision MEXAFS measurement at the Fe K-edge.

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