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Polarization-modulation technique with diamond phase retarder to improve the accuracy of XMCD measurements

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A polarization-modulation technique for accurate measurements of x-ray magnetic circular dichroism (XMCD) has been developed by combination of a diamond x-ray phase retarder and a phase-sensitive (lock-in) detection system. The x-ray helicity was switched at 40 Hz by moving the phase retarder, and the resulting variation in sample absorption was detected using an amplifier locked to the helicity-modulation frequency. The measured XMCD spectrum at the Fe K-edge in $CoFe_2O_4$ agreed with that obtained using a conventional magnetic-field-inversion method. The new technique provides higher statistical accuracy in a shorter measuring time.

Keywords: x-ray magnetic circular dichroism, x-ray phase retarder, polarization modulation, x-ray circular polarization

1. Introduction

X-ray magnetic circular dichroism (XMCD) is a unique element-selective spectroscopy. The effect of XMCD is small in most materials, typically a few parts in 10^3 at the K-edge of the 3*d* transition metals. To detect such a weak signal requires a high signal-to-noise ratio. Most previous XMCD measurements (Schütz *et al.*, 1987; Maruyama *et al.*, 1993) have been made using polarized x-rays of fixed helicity and switching the direction of the magnetic field at the sample. However, in order to get good quality data using the field inversion (FI) method, a long integration time is used for each field orientation. Thus, the FI method is limited because it requires extremely good stability of the incident x-ray beam over relatively long time periods. In particular, the FI method has not provided sufficient quality data for detailed discussions of the electronic states at the K-edge of the 3*d* transition metals.

In this paper we demonstrate that extremely good quality XMCD data can be obtained quickly and reliably by combining fast switching of the incident x-ray polarization with phase-sensitive (lock-in) detection. The key element in our scheme is an x-ray phase retarder in a transmission geometry (Hirano *et al.*, 1991) which allows us to quickly and efficiently switch the photon helicity (Hirano *et al.*, 1992) so that lock-in detection becomes feasible. We note that there have been previous uses of phase plates to make XMCD measurements (Giles *et al.*, 1994; Giles *et al.*, 1995) but in that work, the polarization changes were on a time scale comparable to the FI method mentioned above, and therefore suffer from similar requirements on beam stability. We will show the XMCD spectrum measured at the Fe K-edge in $CoFe_2O_4$, using this polarization-modulation (PM) technique. The spectrum shows the advantage in higher signal-to-noise ratio over the conventional FI method.

2. Instruments for polarization-modulation technique

Instruments for the PM technique were installed in an x-ray undulator beamline, BL39XU (Goto *et al.*, 1998; Hayakawa *et al.*, 1998), at SPring-8. A piezo-driven oscillation stage (Hirano *et al.*, 1992) and electronics for the lock-in detection were incorporated into the existing apparatus for XMCD experiments at the beamline.

Figure 1 schematically shows the experimental arrangement, including an x-ray phase retarder mounted on the piezo-driven oscillation stage, and the electronics for the lock-in detection. Undulator radiation, linearly polarized in the orbital plane, was monochromated with a Si 111 monochromator, and its polarization state was modified using the phase retarder. The phase retarder was a (111) diamond plate of 0.5 mm-thickness. It was used in a transmission Laue geometry with the 220 reflection plane tilted by 45° with respect to the polarization of the incident beam. The piezo stage oscillated the phase retarder at 40 Hz around the Bragg angle θ_B with an angular amplitude $\Delta\theta$ which was tuned for alternate production of right-handed (RHC) and left-handed circular (LHC) polarization. An ω-2θ rotation stage, which held the piezo stage, was used to adjust the oscillation center to the Bragg angle. The monochromatic and circularly polarized x-ray beam whose helicity alternates at 40 Hz was incident on a sample. The sample was magnetized with an electromagnet whose magnetic field was 0.6 T and fixed in the direction of 45°-tilted from the direction of the incident beam.

In order to describe the circular dichroism, we define absorption coefficients for RHC and LHC photons as $\mu_+ t$ and $\mu_- t$, respectively, where t represents thickness of the sample. When the sample exhibits the dichroism, the absorption coefficient oscillates between $\mu_+ t$ and $\mu_- t$ according to the helicity change of the incident beam. The amplitude of the oscillation,

$$\Delta\mu t = \mu_+ t - \mu_- t \qquad (1)$$

gives an XMCD, while the average value,

$$\bar{\mu} t = (\mu_+ t + \mu_- t)/2 \qquad (2)$$

gives the absorption coefficient for linear or unpolarized x-rays.

The intensities of the incident and transmitted beam were measured with ionization chambers and converted into voltage signals $V(I_0)$ and $V(I)$ by current amplifiers (KEITHLEY428). A logarithmic converter gave a voltage signal $V(\mu t)$ which corresponded to the absorption coefficient, $\ln(I_0 / I)$, from $V(I_0)$ and $V(I)$. The ac component of the $V(\mu t)$ was proportional to the XMCD, $\Delta\mu t$, and directly measured using an amplifier locked to the helicity-modulation frequency. The dc component, corresponding to the average absorption coefficient, $\bar{\mu} t$, was measured with a digital voltmeter. Preliminary examinations confirmed that the oscillation properties of the piezo stage and the response of the logarithmic converter circuit were sufficient to give good XMCD data.

3. XMCD results and discussion

The PM technique was applied to XMCD measurements at the Fe K-edge (E_0 = 7.110 keV). Figure 2 shows an obtained XMCD spectrum of $CoFe_2O_4$ together with its XANES spectrum. These spectra were measured using a 1 eV energy step in the range from E_0 – 33 to E_0 + 87 eV. The oscillation amplitude of the phase retarder was fixed at 130 arcsecond during the energy scan, while the center of oscillation was tuned to satisfy the diffraction condition. Readings of the lock-in amplifier with a 1 s time constant were averaged for 10 s at each energy point, and it took approximately 30 min to record one spectrum. The average of two scans is shown in Fig. 2, so the total measuring time was 60 min. We have also included corrections (scale factors) for the 50% duty cycle and the measured response of the logarithmic converter.

The XMCD spectrum showed good agreement with that measured using the conventional FI method (Kawamura *et al.*, 1997). The statistical accuracy was improved and the experimental error was estimated to be a few 10^{-5}. Several fine structures located above 15 eV in the XMCD spectrum could be resolved, although the transmitted intensities through the sample were extremely low in this region. A structure due to a multi-electron excitation (MEE) was clearly observed around 62 eV in the XMCD spectrum, although the conventional FI method has not provided XMCD spectra of sufficient accuracy for detailed discussions about the MEE in a practical measuring time.

By using the PM technique, the measuring time to obtain the XMCD data of a twice signal-to-noise ratio was reduced to 1/10 of that with the FI method made at BL-28B beamline with an elliptical multipole wiggler in PF, KEK. We note that the total flux of monochromatic photons received by the sample at the BL39XU was approximately the same as that at the BL-28B (Iwazumi *et al.*, 1995), when the present experiments were carried out with the SPring-8 ring operated in a 20 mA current mode.

The XMCD spectrum shown in Fig. 2 was derived from the two spectra shown in Fig. 3. These spectra were recorded with opposite magnetic fields, parallel and anti-parallel to the x-ray wave vector respectively. All the spectral structures were reversed by inversion of the magnetic field, and an offset of about 5 x 10^{-4} appeared on each spectrum. This offset probably comes from an intensity difference between RHC and LHC polarizations. The intensity of the beam forward-diffracted by the phase retarder slightly changes on each side of the Bragg condition. The response of the logarithmic converter was not perfect, and the two ionization chambers were slightly different in sensitivity, so that the systematic offsets remained. In order to obtain the pure XMCD signal we subtracted one spectrum from the other. As shown in the Fig. 2, the parasitic signal was fully eliminated.

We have confirmed, by XMCD experiments in other materials, that the PM technique works well in the energy range from 7 to 12 keV. We must tune the flipping condition (amplitude and diffraction angle) of the phase retarder to follow the photon energy. The energy tunability of the PM technique can

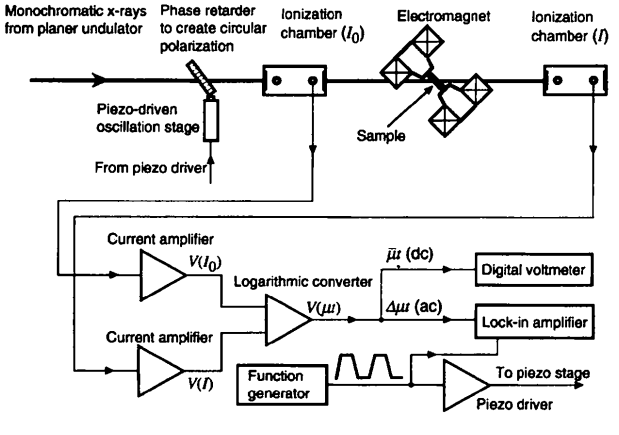

Figure 1
Experimental setup for x-ray magnetic circular dichroism measurements with a polarization-modulation technique.

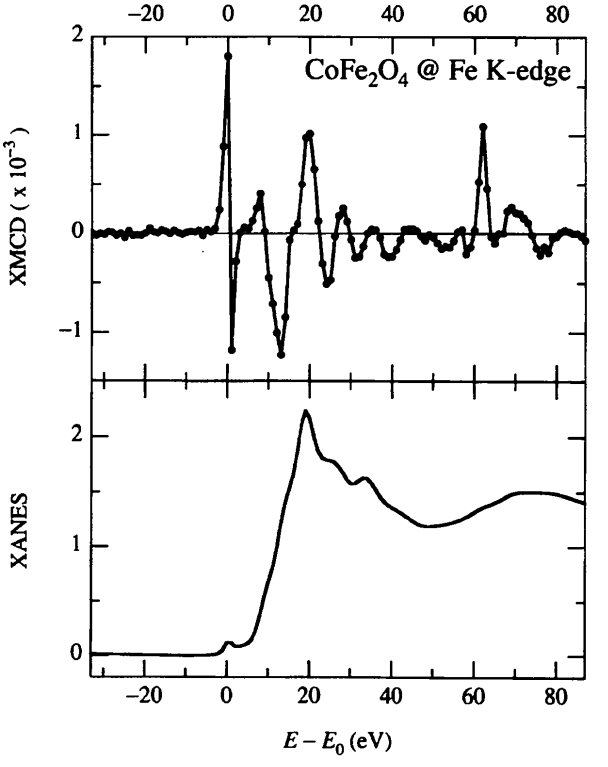

Figure 2
An x-ray magnetic circular dichroism spectrum at the Fe K-edge in $CoFe_2O_4$, measured with the polarization-modulation technique. The lower panel shows the XANES spectrum.

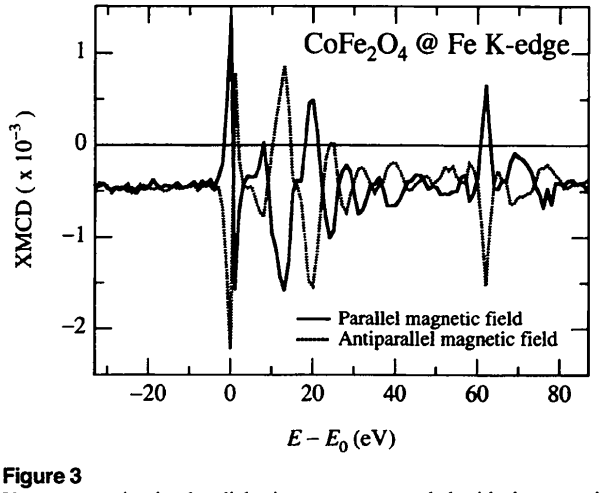

Figure 3
X-ray magnetic circular dichroism spectra recorded with the opposite magnetic field, parallel (solid curve) and antiparallel (dashed curve) to the x-ray wave vector, respectively.

lead to application to spin-polarized EXAFS studies. One can use the applied magnetic field as the degree of freedom using the PM technique; element-specific magnetic hysteresis (Chen *et al.*, 1993; Pizzini *et al.*, 1998) have been recently measured in Gd/Fe multilayer films by the XMCD at the Fe K-edge and Gd L3-edge (Koizumi *et al.*, 1998). In addition, we plan to investigate a metamagnetic transition under a high field using a superconducting magnet.

4. Conclusion

The PM technique for XMCD measurements has been developed by combining a diamond phase retarder with a piezo-driven oscillation stage and lock-in detection. The efficiency of the PM technique was confirmed with XMCD spectra obtained at the Fe K-edge. This new technique will be a powerful tool for various kinds of spin-dependent x-ray spectroscopy.

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