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X-ray linear birefringence and linear dichroism in a cobalt crystal measured with a tunable X-ray polarimeter

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X-ray linear birefringence and linear dichroism spectra have been measured simultaneously and quantitatively for the first time. The sample was a hexagonal cobalt single-crystal foil. The apparatus used was an energy-tunable X-ray polarimeter, consisting of a polarizer, a phase retarder and an analyser. The X-ray energy was scanned over a range of 350 eV near the *K*-absorption edge. The Kramers–Kronig relation between the refraction and absorption anisotropy has been confirmed on an absolute scale. The phase-difference sensitivity of the polarimeter as a polarization inferometer is as small as $2\pi/10000$, which is about 100 times smaller than that of the Bonse–Hart inferometer.

Keywords: X-ray polarimeters; polarization interferometry; polarized XAFS; X-ray birefringence; X-ray dichroism.

1. Introduction

Though polarized X-ray beams have been available since the advent of synchrotron radiation, there are still many new areas for X-ray polarization experiments. It is our long-term aim to explore areas such as linear birefringence, linear dichroism and optical activities (circular birefringence, circular dichroism), including electro- and magneto-optical effects, in the X-ray region. With this in mind, we have developed a new type of X-ray polarimeter which consists of a polarizer, a phase retarder and an analyser. The present work is concerned with (i) simultaneous and quantitative measurements of linear birefringence and linear dichroism spectra of a uniaxial cobalt crystal with this apparatus and (ii) confirmation of the Kramers–Kronig relation in the X-ray region.

Historically, X-ray polarization-dependent absorption of an anisotropic crystal was observed for the first time by Templeton & Templeton (1980) at the vanadium *K*-edge using a technique nowadays known as polarized XAFS. They pointed out that the dielectric constants of crystals near X-ray absorption edges should be dealt with not as scalar quantities, but as tensor quantities (Templeton & Templeton, 1982, 1985). Subsequently, Petcov *et al.* (1990) reported not only linear dichroism but also linear birefringence of a lithium niobate crystal using an X-ray polarimeter. According to their results, the real and imaginary parts of the dielectric anisotropy did not satisfy the Kramers–Kronig relation. Though their work was challenging, their results are open to question.

An energy-tunable X-ray polarimeter consisting of a polarizer and an analyser equipped with an offset channel-cut mechanism (Hart & Rodrigues, 1979) has been developed (Siddons *et al.*, 1990; Hart *et al.*, 1991). With this apparatus, the Faraday effect and optical activity in the X-ray region were measured for the first time. Based on this X-ray polarimeter, we have constructed a new type of X-ray energy-tunable polarimeter, in which a transmission-type X-ray phase retarder (Hirano *et al.*, 1993, 1994, 1995) has been introduced as a third optical device in addition to the polarizer and analyser. Using this apparatus (Okitsu *et al.*, 1996), we have quantitatively measured the X-ray linear birefringence and linear dichroism spectra simultaneouly in a cobalt crystal.

2. Experimental

Fig. 1 shows the experimental set-up of the new type of X-ray energy-tunable polarimeter. The synchrotron white X-ray radiation was monochromated and highly polarized by a silicon-crystal polarizer giving four-bounce 422 symmetric Bragg reflections. The intensity ratio of the horizontal to the vertical polarization in the outgoing X-ray beam from the polarizer is over 10^{10}, according to calculations from dynamical diffraction theory. Well defined elliptically polarized X-rays were obtained from the highly horizontally polarized X-rays by the transmission-type phase retarder (Hirano *et al.*, 1993, 1994, 1995) and were incident on the sample crystal. The sample was a hexagonal close-packed (h.c.p.) cobalt single-crystal foil of 12 μm thickness, the normal of which was $11\bar{2}0$. The optical *c* axis of the sample crystal, which lay on the sample surface, was inclined by 45° from the direction of the major axis of the elliptical polarization. The polarization state of X-rays transmitted through the sample was analysed by rotating the analyser around the beam direction over a range of ±2° in the vicinity of the crossed-nicol position. The data were least-squares-fitted using

$$I(\chi) = \{(a^2 + b^2) - (a^2 - b^2)\cos[2(\chi - \alpha)]\}/2, \qquad (1)$$

where $I(\chi)$ is the number of X-ray photons detected by the

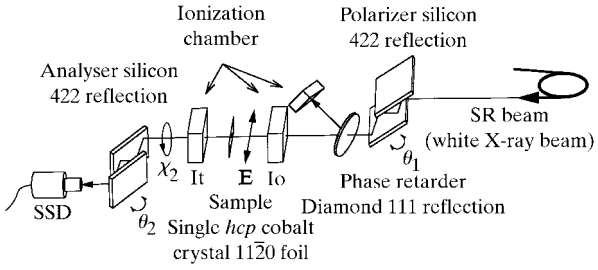

Figure 1
Experimental arrangement of the X-ray energy-tunable polarimeter with a phase retarder. The polarizer and analyser crystals are similar channel-cut crystals giving four-bounce 422 symmetric Bragg reflections and are equipped with the Hart–Rodrigues offset mechanism. The phase retarder is a diamond (001)-oriented plate of 313 μm thickness giving 111 asymmetric Laue reflection, the plane of incidence of which is inclined by 45° from the horizontal plane. Two ion chambers were placed upstream and downstream of the sample to measure the absorption spectrum.

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germanium SSD placed behind the analyser, χ is the rotation angle of the analyser around the beam direction from the crossed-nicol position, α is the rotation angle of the major axis of elliptical polarization and a and b are the lengths of the major and minor axes of the elliptical polarization, respectively. The ellipticity of polarization R is given by $R = b/a$. $I(\chi)$ was measured at 17 χ positions with a step of 900 arcsec at each photon energy. Each $I(\chi)$ was measured for 20 s. The X-ray energy was scanned by changing the Bragg angles of the polarizer, analyser and phase retarder crystals simultaneously with an energy step of about 1 eV. The energy resolution was estimated to be about 1.5 eV from the vertical size (2 mm) of the X-ray beam and the distance (10 m) from the synchrotron radiation light source.

3. Results and discussion

Fig. 2 shows spectra of the ellipticity of the X-ray polarization due to birefringence with the h.c.p. cobalt crystal; the ellipticity of the incident X-ray polarization was set to be (*a*) −0.04, (*b*) −0.02, (*c*) +0.02 and (*d*) +0.04 using the phase retarder, based on calculations from dynamical diffraction theory. The absorption spectrum is also plotted to provide a convenient calibration of the energy axis. The ellipticity R is related to the phase difference of polarization between the a and c axes by

$$R = \tan(\Delta\varphi/2), \tag{2}$$

where $\Delta\varphi$ is the phase difference of polarization between the a and c axes. The vertical bar in Fig. 2(*a*) corresponds to the detectable phase difference ($2\pi/100$) of the Bonse–Hart X-ray interferometer (Bonse & Hart, 1965*a*,*b*,*c*) as calculated with equation (2). Note that this X-ray linear birefringence would be difficult to detect with the Bonse–Hart X-ray interferometer, which is regarded as a unique apparatus for directly obtaining X-ray phase information.

Figs. 3(*a*) and 3(*b*) are expansions of the boxed regions in Figs. 2(*b*) and 2(*c*). A slight change in ellipticity observed at +2 eV from the cobalt K-absorption edge is evidently significant since it is clearly reversed when the helicity of the incident polarization is reversed. The half-lengths of the error bars below the absorption edge correspond to a phase difference of about $2\pi/10\,000$ (a

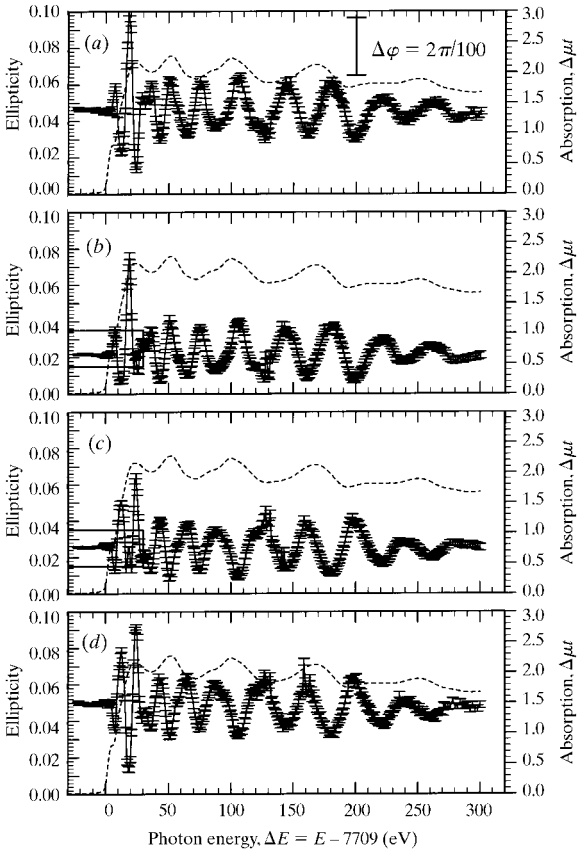

Figure 2
Spectra of X-ray polarization ellipticity due to birefringence in the cobalt sample. The ellipticity of incident X-rays from the phase retarder was controlled so as to be (*a*) −0.04, (*b*) −0.02, (*c*) +0.02 and (*d*) +0.04 during the energy scan, based on calculations from dynamical diffraction theory. The vertical bar drawn in (*a*) corresponds to a polarization phase difference of $2\pi/100$ between the a and c axes in the cobalt crystal. $2\pi/100$ is the smallest phase difference practically detectable by the Bonse–Hart X-ray interferometer.

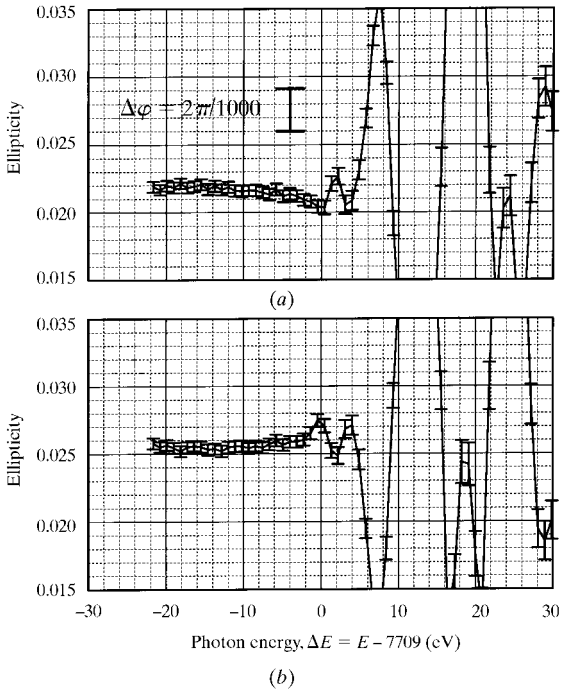

Figure 3
Spectra of X-ray polarization ellipticity. (*a*) and (*b*) are expansions of the squared regions in Figs. 2(*b*) and 2(*c*), respectively. The vertical bar drawn in (*a*) corresponds to a phase difference of polarization of $2\pi/1000$ between the a and c axes. A signal observed at +2 eV from the cobalt K-absorption edge is evidently significant since a reversal of the signal is clearly observed when the helicity of the incident polarization is reversed.

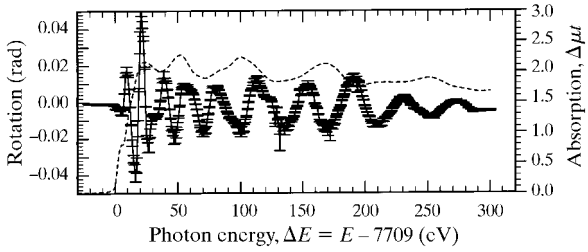

Figure 4
Spectrum of polarization rotation due to dichroism in the cobalt crystal; identical spectra were obtained for incident X-rays with polarization ellipticity of −0.04, −0.02, +0.02 and +0.04.

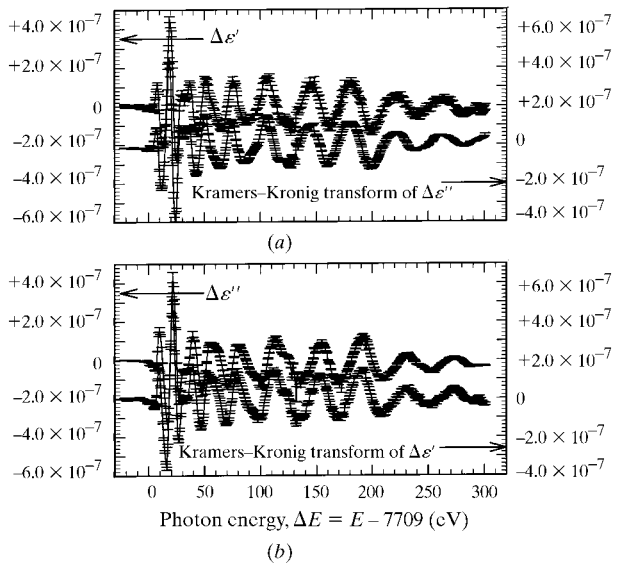

Figure 5
The upper spectra in (*a*) and (*b*) are $\Delta\varepsilon^{(r)}$ and $\Delta\varepsilon^{(i)}$ calculated from the ellipticity and rotation of polarization. The lower spectra in (*a*) and (*b*) are the Kramers–Kronig transforms of $\Delta\varepsilon^{(i)}$ and $\Delta\varepsilon^{(r)}$. The upper and lower spectra are shifted for clarity. Here $\Delta\varepsilon^{(r)} = \Delta\varepsilon_c^{(r)} - \Delta\varepsilon_a^{(r)}$ and $\Delta\varepsilon^{(i)} = \Delta\varepsilon_c^{(i)} - \Delta\varepsilon_a^{(i)}$, where $\Delta\varepsilon_a^{(r)}$, $\Delta\varepsilon_a^{(i)}$, $\Delta\varepsilon_c^{(r)}$ and $\Delta\varepsilon_c^{(i)}$ are the real and imaginary parts of the relative dielectric constants of the cobalt crystal in the *a* and *c* axis directions. No procedure for scaling was made for either (*a*) or (*b*).

wavefront shift of 1.6×10^{-4} Å). The phase-difference sensitivity of the polarimeter as an X-ray polarization interferometer (Okitsu, 1993), in which *a*- and *c*-axis polarizations interfere with each other by being reflected by the analyser, is 100 times more sensitive than that of the Bonse–Hart interferometer.

Fig. 4 shows a spectrum of rotation of polarization due to dichroism in the h.c.p. cobalt crystal. The upper spectra in Figs. 5(*a*) and 5(*b*) are the real and imaginary parts of the dielectric constant anisotropy, respectively. These are calculated from the ellipticity and polarization-rotation spectra. The procedure for the calculation is described in a separate paper (Okitsu *et al.*, 1998). The Kramers–Kronig transforms of the imaginary and real parts of the dielectric anisotropy are also plotted in Figs. 5(*a*) and 5(*b*) (the lower spectra), respectively. The left and right ordinates of Figs. 5(*a*) and 5(*b*) are shifted for clarity but are drawn on an absolute scale of relative dielectric constant. It can be concluded that the Kramers–Kronig relation holds between the real and imaginary parts of dielectric anisotropy of the cobalt crystal. This is the first quantitative confirmation of the Kramers–Kronig relation in the X-ray frequency region.

4. Conclusions

X-ray linear birefringence and linear dichroism have been detected and measured quantitatively for the first time by using a new type of X-ray energy-tunable polarimeter with a phase retarder. The Kramers–Kronig relation was confirmed quantitatively between the real and imaginary parts of the dielectric anisotropy.

The detectable phase difference of the polarimeter as a polarization interferometer has been estimated to be $2\pi/10000$, 100 times smaller than that of the Bonse–Hart X-ray interferometer.

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