Observation of Magnetic Circularly Polarized Emission in the X-ray Region

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A new magneto-optical effect is reported in the hard X-ray region. The new effect is an X-ray analog of magnetic circularly polarized emission (MCPE) in the visible region, which is a phenomenon whereby photons emitted from a magnetized material are circularly polarized. The degree of circular polarization is measured for the $K\alpha_1$ emission of metallic iron and is found to be large (~12%). It is also found that the sense of the circular polarization is reversed when the magnetization of the sample is reversed. These results constitute evidence that MCPE exists in the hard X-ray region. The observed large dichroic effect is also a feature suggesting that the new effect can be applied to a new measurement technique for practical materials.

The circular polarization of light is related to the magnetization of materials through symmetry and this fact gives rise to a magneto-optical effect. An obvious example is magnetic circular dichroism (MCD), which is the difference between absorption cross-sections when the circular polarization of incident light is parallel and antiparallel to the magnetization of a sample. Although the Faraday and magneto-optical Kerr effects are phenomena whereby the polarization plane of linearly polarized light rotates after transmission through and reflection from a magnetized sample, respectively, they are consequences of a difference between refractive indices of right and left circularly polarized light in a magnetized medium. Magneto-optical effects are very basic phenomena involving light and magnetism and now play vital roles in applications such as optical isolators and magneto-optical Kerr microscopes.

Because X-rays are a form of light, magneto-optical effects also exist in the soft and hard X-ray regions. The advent of synchrotron X-ray facilities has provided intense, highly polarized, and energy-tunable X-rays and has enabled us to observe magneto-optical effects in the X-ray region. Following the discovery at DESY of MCD in the hard X-ray region [1], major X-ray magneto-optical effects were reported in 1990s [2–4]. In particular, X-ray MCD is now a standard technique in research on magnetism because of several advantages, namely (i) element selectivity due to core-level spectroscopy, (ii) applicability of magneto-optical sum rules at spin-orbit split edges, and (iii) modestly good lateral resolution due to well-collimated synchrotron X-rays.

However, a problem with X-ray MCD is the very small dichroic effect (~0.5%) in the hard X-ray region for 3*d* transition metal (TM) elements. The use of hard X-rays is indispensable for bulk-sensitive measurements because of the long penetration length of hard X-rays into the materials. In addition, 3*d* TMs, which include Mn, Fe, Co, and Ni, are crucial elements in magnetic materials. Accordingly, there has been strong demand for finding a new principle or technique that would allow element-selective X-ray measurements with a large dichroic effect for 3*d* TM elements in the hard X-ray region.

Magnetic circularly polarized emission (MCPE) is a magneto-optical effect and a phenomenon whereby photons emitted from a magnetized material are circularly polarized. In 1971, Marrone and Kabler reported that the luminescence of excitons in alkali halides is markedly circularly polarized under magnetic fields [5]. Surprisingly, MCPE is yet to be reported in the X-ray region. For instance, the $K\alpha$ emissions of 3*d* TMs are well-known characteristic X-rays in the hard X-ray region and correspond to transitions from the 2*p* level to the 1*s* level. The final state of $K\alpha$ emission is the $2p^5$ state and has large spin-orbit coupling, which is an essential requisite for a magneto-optical effect. A considerable interaction between the 3*d* magnetic moments and the 2*p* hole spin is also known [6]. A large dichroic effect is thus expected in $K\alpha$ emission. To address the aforementioned challenging issue, a measurement of circular polarization in $K\alpha$ emission is therefore planned here for metallic iron [7].

Experiments were carried out at beamline BL22XU at SPring-8. The experimental setup is described below and illustrated in Fig. 1. The sample was an iron single crystal that was inserted between permanent magnets to saturate the magnetization. The sample was illuminated by intense incident X-rays generated from an undulator. Fluorescence X-rays emitted from the sample were collimated by an exit slit (slit 1) down to 120 μrad in divergence. This is most important because the optical elements mentioned below are based on crystal optics. The quarter-wave plate (QWP) was a diamond single crystal that mutually converts circularly and linearly polarized X-rays. The polarization analyzer that reflects the vertical component of the incoming fluorescence X-rays was a Ge single crystal, which also functioned as an energy analyzer. The combination of a QWP and a polarization analyzer is a standard device for detecting the circular polarization of a beam of photons. Ideally, the intensity at the detector equals $I_0(1+P_C)/2$ ($I_0(1-P_C)/2$) when the QWP generates a $\pi/2$ ($-\pi/2$) phase shift, where I_0 is the total intensity and P_C is the degree of circular polarization.

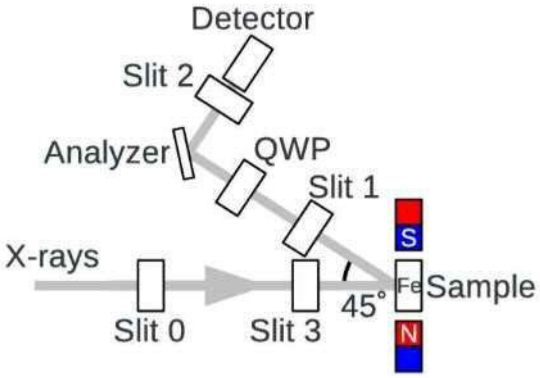

Fig. 1. Top view of experimental layout. QWP: a diamond phase retarder that acts as a quarter-wave plate. Analyzer: a Ge(400) single crystal that is both an energy and a polarization analyzer.

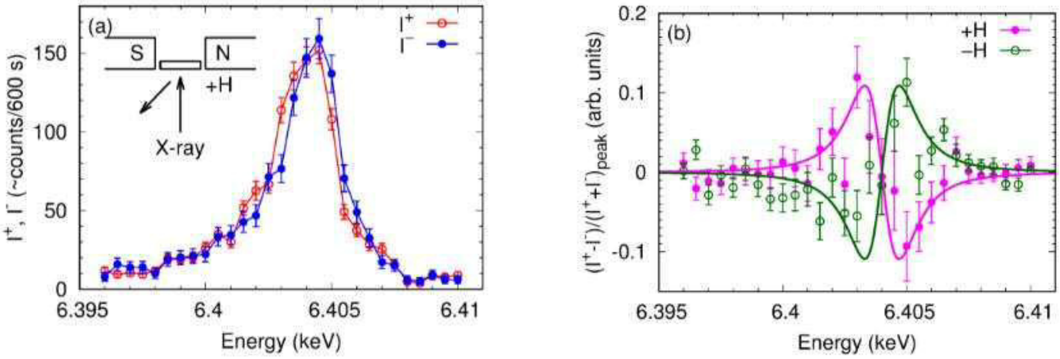

Fig. 2. (a) Fe $K\alpha_1$ emission spectra I^+ and I^- for a phase shift of π/2 (red open circles) and −π/2 (blue solid circles), respectively. The lines connect the data points to guide the eye. The magnetic field is applied as shown in the inset and the direction is defined as positive. It is obvious that the I^+ spectrum is shifted to the low-energy side compared to the I^- spectrum. (b) Difference spectra between I^+ and I^- normalized by the peak intensity. Magenta solid circles and green open circles are measured when the magnetic field is directed along the positive and negative directions, respectively. Solid lines are a guide to the eye.

The obtained Fe $K\alpha_1$ emission spectra are shown in Fig. 2(a). I^+ (red open circles) and I^- (blue closed circles) are data observed when the π/2 and −π/2 phase shifts, respectively, are introduced by the QWP. The magnetic field is applied as shown in the inset and is defined as the positive direction. It is obvious that the I^+ and I^- spectra do not coincide. The I^+ spectrum is shifted to the low-energy side by approximately 0.3 eV compared to the I^- spectrum. This difference between the two spectra is direct evidence that the $K\alpha_1$ emission is circularly polarized. The difference spectrum I^+-I^- normalized by the peak intensity of the sum spectrum is shown in Fig. 2(b) as magenta solid circles. When the magnetic field (and therefore the magnetization of the sample) is reversed, the sense of the circular polarization is reversed, as shown by the green open circles, which are data measured when the magnetic field is applied along the negative direction.

These results clearly illustrate that (i) the energy-resolved $K\alpha_1$ spectrum of ferromagnetic Fe indicates finite circular polarization and (ii) the circular polarization is inverted when the magnetization of the sample is inverted. These two features unambiguously indicate the existence of MCPE in Fe $K\alpha_1$ emission. The flipping ratio $(I^+-I^-)/(I^++I^-)$ is a measure of the size of the dichroic effect and was 12 ± 4% at 6.405 keV. If the scattering-angle correction and QWP efficiency are considered, the value would amount to 18 ± 6%. Because the flipping ratio of metallic iron in soft X-ray MCD is around 30%, a dichroic effect comparable to that in the soft X-ray region is obtained in the hard X-ray region.

To summarize, it is confirmed experimentally that MCPE exists in X-ray core-level emission and that the dichroic effect is quite large even in the *K*-edge of 3*d* TMs. Hence, this magneto-optical effect may open a new way to perform element-selective and truly bulk-sensitive measurements of the magnetization of 3*d* TMs.

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