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Anomalous magnetic hysteresis of Gd and Fe moments in a Gd/Fe multilayer measured by hard x-ray magnetic circular dichroism

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Element-specific magnetic hysteresis in a Gd/Fe multilayer has been obtained at the Gd L_3 and the Fe K edges by means of x-ray magnetic circular dichroism (XMCD) measurement with a helicity-modulation technique. Both hysteresis curves at 20 K show characteristic sharp peaks at the coercive field $H_c = \pm 50$ Oe. Above H_c, the intensities of XMCD gradually decrease with increasing magnetic fields, whereas the bulk magnetization monotonously increases. These results are explained by the theoretical prediction by Camley and co-workers.

Magnetic multilayers often show interesting phenomena such as the magnetic phase transition and the giant magnetoresistance which are closely related to the magnetic state of interface and surface, and have received considerable interest because the properties are controlled by changing the stacking sequence and the thickness of layers in sample preparation. A Gd/Fe multilayer is one of those systems and exhibits spin-flop-like behavior with an external magnetic field.[1–6] Camley and co-workers predicted a magnetic phase diagram for a Gd/Fe superlattice with a simple Hamiltonian as follows:[7–11]

$$H = \sum_{\text{layer } n} \sum_{m=0,\pm 1,\pm 2} \frac{1}{2} J_{nm} S_n \cdot S_{n+m} - \sum_{\text{layer } n} g_n \mu_B H_0 \cdot S_n , \tag{1}$$

where the first term represents the effective exchange energy, and J_{nm} is the interlayer exchange coupling between spins in atomic layers n and $n+m$. Here S_n denotes all the spins in the atomic layer n because the intralayer exchange coupling is assumed to be ferromagnetic. The second term represents the Zeeman energy in an external field H_0, and g_n is the effective Lande g factor for the spins in the atomic layer n. The competition between the exchange and the Zeeman interactions leads to a variety of field-induced phase transitions. In a low magnetic-field region, the Gd (Fe)-*aligned* phase is expected as an initial state, where Gd (Fe) moments are parallel and Fe (Gd) moments are antiparallel to the applied field. As the magnetic field increases from zero, the sample in the Gd-aligned phase is able to change to the *surface twisted* phase, where the direction of the magnetic moments in each Fe/Gd bilayer noticeably deviates from the field direction rather around the surface than in the inner part, while Gd and Fe moments keep almost antiferromagnetic coupling in the bilayer. Further increase of the magnetic field induces the *bulk twisted* phase, where both Gd and Fe moments are tilted from the field direction with different angles in each layer.

So far, magnetization measurement has been a main and effective method to observe these magnetic transitions in Gd/Fe multilayers. Recently, the study of magnetism using synchrotron radiation x rays is advancing steadily. X-ray magnetic circular dichroism (XMCD) measurement is one of such methods for investigating ferro- and ferrimagnetic materials using circularly polarized x rays. The application of XMCD measurement to multilayer systems is relevant and several works have been reported in the literature.[12–16] As XMCD is measured at the x-ray absorption edge of a specified element in a sample, this technique is feasible for the study of element-selective magnetic properties. The intensity of XMCD is proportional to the mean magnetic moment of the specified element projected onto the direction of the incident x rays, although it is difficult to evaluate the absolute value of the magnetic moment. The sign of XMCD gives the direction of the probed moment relative to that of the total magnetization, when the directions of the helicity of incident x ray and external magnetic field are known. Therefore, XMCD is useful particularly in investigating a material which consists of more than two magnetic elements, and will separately prove the relative variation of those magnetic moments caused by magnetic fields, temperature, and composition modification. The field dependence of XMCD in a Fe/Gd multilayer has been once measured, and the spin configuration of a twisted state has been suggested.[13] However, the data are not enough to elucidate the precise magnetization process in this system.

The present paper reports unusual sharp peaks on element-specific magnetic hysteresis loops of XMCD in a

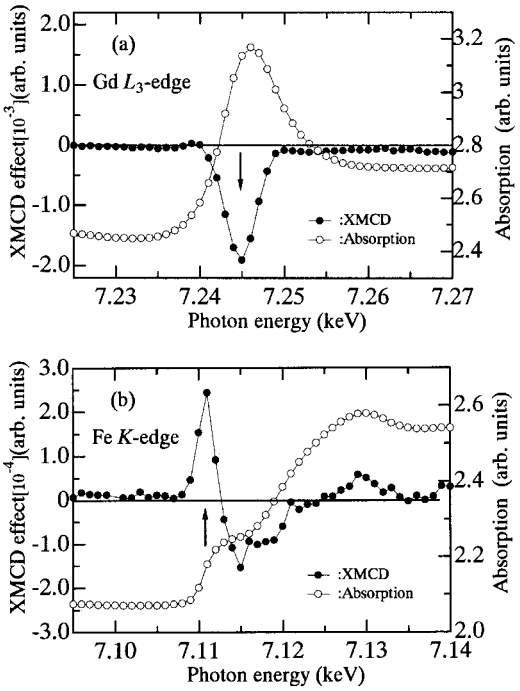

FIG. 1. XMCD and absorption spectra measured at (a) the Gd L_3 edge (7.243 keV), and (b) the Fe K edge (7.111 keV) at room temperature. The ordinate is the observed XMCD effect.

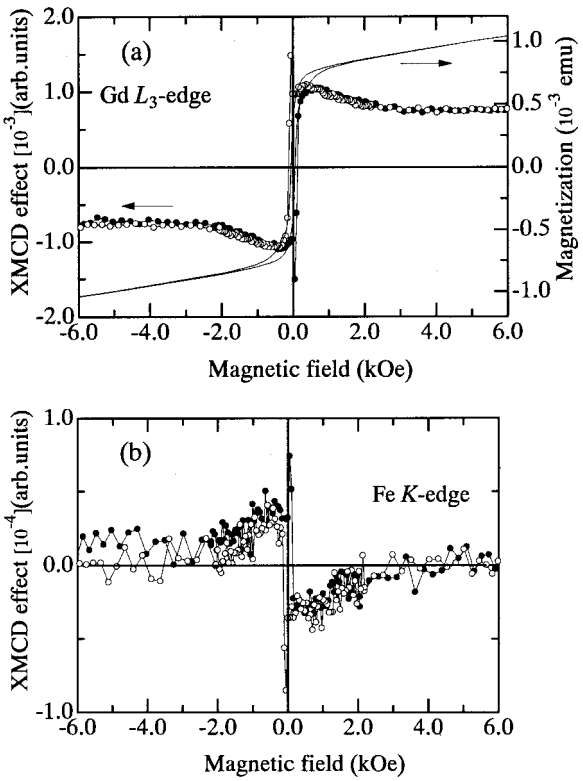

FIG. 2. (a) Magnetic hysteresis loop of XMCD measured at the Gd L_3 edge at 20 K. The energy was fixed at 7.245 keV. Magnetization curve is also drawn by a solid line. (b) Magnetic hysteresis loop of XMCD measured at the Fe K edge at 20 K, and which was obtained as the average of three measurements. The energy was fixed at 7.111 keV. Solid circles (●) indicate the XMCD effect for magnetic fields increasing from −6 up to +6 kOe and open circles (○) indicate that for magnetic fields decreasing from +6 down to −6 kOe.

Gd/Fe multilayer. For the XMCD measurement, a helicity-modulation technique in a hard x-ray region has been adopted. The results are examined by comparing with magnetization measurement and the Camley's prediction.

The sample was prepared by alternately depositing Gd (20 Å) and Fe (20 Å) layers on a polyimide film in a dc-magnetron sputtering system. The deposition was started by the Gd layer and terminated by the Fe layer, and 50 bilayers were piled up in all. The artificial periodicity of the sample was identified by x-ray-diffraction measurement.

XMCD measurements were made on BL39XU at SPring-8. The sample was 5 mm wide and 10 mm long, and three films were stacked for the measurement. Circularly polarized x rays were produced by a diamond phase retarder mounted on a piezo-driven oscillation stage. 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. This helicity-modulation technique makes it possible to keep the sample magnetization fixed during the measurement and to obtain XMCD spectra of higher signal-to-noise ratio than those with an usual field-reversal method. The details of this technique have been reported in Ref. 17.

Figures 1(a) and 1(b) show XMCD spectra measured at the Gd L_3 and the Fe K edges, respectively, with the transmission mode in the external magnetic field H of +6 kOe parallel to the sample surface at room temperature. The field direction was fixed at 45° away from the direction of the incident x rays. Hysteresis loops of XMCD shown in Fig. 2 were measured at 20 K as a function of external field H. At that time, the x-ray energy was fixed at the maximum peak of each spectrum which is marked with an arrow in Fig. 1. The origin of the ordinate is determined from the symmetry of each hysteresis loop. The signs of XMCD spectra in Figs. 1(a) and 1(b) are minus and plus, respectively, at the energy of each absorption edge. The difference in signs means that Gd and Fe moments are primarily antiferromagnetic coupling and Fe moments are dominant at room temperature. This was verified by measuring reference materials. On the contrary, the signs at 20 K are reversed in both XMCD hysteresis curves as shown in Fig. 2: they show plus and minus on the Gd and Fe XMCD curves in the positive fields, respectively. This indicates that the magnetic state of the sample changes from the Fe-dominant state to the Gd-dominant one through a compensation temperature.

The most important findings are characteristic sharp peaks appeared at about $H = \pm 50$ Oe soon after the reversal of the magnetic field, which are seen in both Figs. 3(a) and 3(b) with opposite signs. This field almost coincides with the coercive field H_c. Magnetization curve measured by a superconducting quantum interference device (SQUID) magnetometer is also drawn in Figs. 2(a) and 3(a) for comparison. The peaks are maximum in the respective XMCD hysteresis loops, while no indication associated with these peaks is observed in the magnetization curve. From Figs. 3(a) and 3(b), it is obvious that the H_c on the magnetization curve is lower than the field where the XMCD curves intersect the abscissa. Furthermore, the intersection field on the Gd-XMCD curve is different from that on Fe one: The Gd moments reverse at a

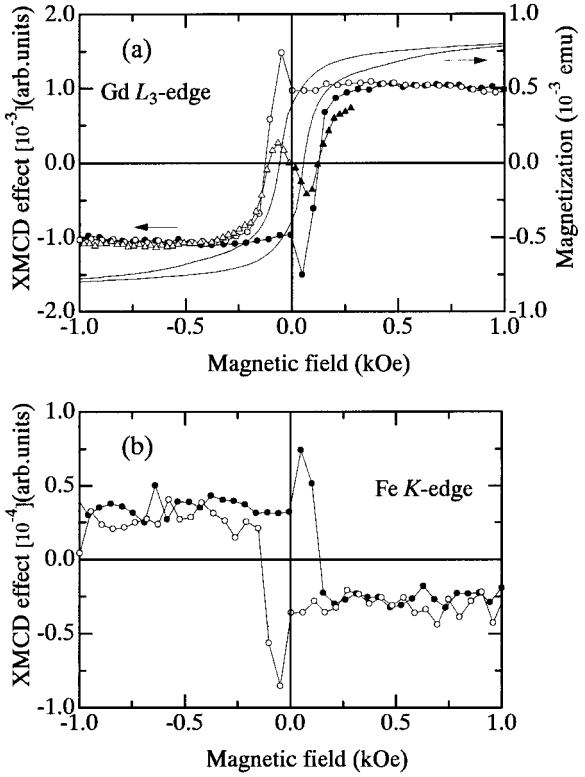

FIG. 3. Closeup of the hysteresis loops for (a) Gd-XMCD and (b) Fe-XMCD. Solid and open circles are shown in the same manner as Fig. 2. Solid and open triangles indicate the XMCD effect in the initial magnetization for magnetic fields increasing and decreasing from zero, respectively.

slightly lower field than Fe moments. Generally, bulk magnetization disappears at H_c where the symmetry of magnetic domain patterns is restored in usual magnetic materials. In this case the XMCD signal should also disappear at H_c. The XMCD peaks cannot be interpreted by the domain structure. It is therefore reasonable to consider that the individual magnetic moments of Gd and Fe exist, and they compensate each other at H_c on the magnetization curve. In such a particular case, it is expected from Eq. (1) that an aligned state lowers the energy of this system at H_c. Since the Zeeman interactions of Gd and Fe moments are completely canceled out at H_c, the antiferromagnetic exchange coupling at interfaces between Gd and Fe layers is substantial for the determination of the magnetic structure. The signs of the maximum peaks strongly suggest that this particular spin configuration is the Fe aligned state, though the sample is in the Gd-dominant state at this temperature. Although the aligned state was not predicted by Camley and co-workers, the above phenomenon is qualitatively explained by using their model. However, some magnetic anisotropy term, which is neglected in Eq. (1), will be also necessary to interpret the magnetic hysteresis.

When the magnetic field is increased above H_c, the signs of XMCD are reversed at both the Gd L_3 and the Fe K edges. This shows a transition from the Fe-aligned state to the bulk twisted state. Above H_c, the Gd and Fe moments are not fully aligned to the direction of the external field, because the intensity of XMCD is smaller than the peak intensity at H_c. The intensity of XMCD gradually decreases with increasing the magnetic field higher than 0.5 kOe, whereas the magnetization monotonously increases. In this system, a steplike behavior in the magnetization curve is usually believed to be reasonable evidence of the twisted plane transition.[1–6] As it is clear from Fig. 3(a), even though no noticeable change is observed on the magnetization curve, the present XMCD measurement obviously detects the transition from the Fe-aligned state to the bulk twisted state above H_c. The difference in slope between the XMCD curves above 0.5 kOe indicates that Gd and Fe moments gradually tilt from the field direction at different rates. A quantitative evaluation suggests that Fe moments tilt at about three times higher rate than Gd moments. Such behavior causes incomplete cancellation between Gd and Fe moments, with resultant the increase of the bulk magnetization. At high enough fields, it is expected that the Fe-XMCD changes its sign and finally all of the moments are aligned to the field direction.

XMCD in the initial magnetization process was also measured at the Gd L_3 edge, and is denoted by triangles in Fig. 3(a). In spite of the Gd-dominant state, the sign of XMCD is minus (plus) in the beginning when the positive (negative) field is applied to the demagnetized sample. This means that Gd moments point in the opposite direction of the magnetic field, therefore Fe moments probably align along the field direction. Such behavior will be related to the surface phase transition pointed out by LePage and Camley as follows.[10] When the surface is an Fe layer, the Fe moments feel the interaction from only one side of the Gd layer, so that the direction of the Fe moments are not strongly restricted by the antiferromagnetic coupling to the Gd moments. Accordingly, even in the Gd-aligned phase, the Fe moments in the outermost layer turn to the field direction at first as the external field increases from zero. This surface twisted state can be observed by soft x-ray MCD and magneto-optical Kerr effect measurements.[5] In several trilayered and thin films, element-specific hysteresis curves have been measured by using soft x-ray MCD to determine the individual magnetic moment and two- and three-dimensional magnetization behavior of each element.[18–21] The penetration depth of soft x rays is shallow, hence soft x-ray MCD is suitable for the study of thin films and surface magnetism. On the other hand, hard x rays penetrate deeply, and hard x-ray MCD will not be much sensitive to the surface condition, and probes bulk property.[22] Although the surface phase transition is triggered by a few outer layers as mentioned above, it would exert its influence on the inner layers to some extent. Then, the present hard x-ray MCD has shown the canted region in this sample permeating deeply into the inside.

In conclusion, magnetic hysteresis loops of XMCD are obtained at both Gd L_3 and Fe K edges in the Gd(20 Å)/Fe(20 Å) multilayer. Its behavior has precisely revealed the magnetization process of each element, which is impossible to be observed by magnetization measurement. The peaks appeared at H_c are interpreted as an indication of the Fe aligned state. The negative slopes of XMCD curves are clear evidence of the bulk twisted state. The measurement in the initial magnetization process suggests the surface twisted state. These striking results show the indispensable availabil-

ity of XMCD measurement utilizing the helicity-modulation technique for the study of magnetism in heteromagnetic materials.

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