By making use of the soft X-ray photoemission electron microscope (PEEM), we have observed in real space the magnetization switching driven by circularly polarized femto-second laser pulses. We have confirmed that the magnetization switching depending on the helicity of the circularly polarized laser occurs at the place where the spin temperature is raised appropriately. Comparison of local soft X-ray absorption (XAS) and magnetic circular dichroism (XMCD) spectra of GdFeCo between the areas with and without the history of laser irradiation revealed that the electronic states remain unchanged even after a number of optical magnetization reversal cycles. The results of pump-probe measurements of magnetic domains that we first demonstrated by employing PEEM are also reported.
1. Introduction

Accompanied by the dramatic improvement in recording “density” of magnetic media during past decades, the importance of “swift” magnetic writing has also been discussed within a large body of theoretical[1-4] and experimental[5-17] works. Recently, exceedingly thorough attention has been paid to pulsed photon excitation of a ferromagnet as a tool for ultrafast magnetization reversal[7,13-18] that may overcome the fundamental and practical limits of operating speed expected from the methods that employ the pulsed magnetic field[5,6], precessional switching[8-10] or spin-polarized current[11,12]. While a number of studies had been reported on quenching, instability, or small angle precession of magnetization driven by femtosecond pulsed laser [19-24], some research groups have demonstrated complete magnetization reversal[7,13-18]. To explain the underlaying physics of the light-induced magnetization reversal, we have to consider two main effects. One is the “heat driven” scenario, that is, the magnetization is once quenched by the laser pulse in the irradiation spot, and aligns in the opposite direction (or creates random magnetic domains in some cases) by the dipole-dipole interaction with nearby spins[15]. The other possible origin is the “opto-magnetic” process based on the inverse Faraday effect[25] using the left- ($\sigma^-$) or right- ($\sigma^+$) circularly polarized laser. When a laser pulse of the proper power density and spot size is applied to the sample, the spin temperature rises up to just below the Curie temperature ($T_C$) and the circular polarization of the light induces effective magnetic field, which aligns the unstable magnetization. In this case, magnetic switching occurs selectively, depending on the helicity of circularly polarized light[13,16].

Up to now, several research groups have contributed to the understanding of the mechanisms underlying the light-induced magnetization control by time resolved magnetic measurements. Ogasawara et al. have investigated the details of heat-driven magnetization reorientation process of a TbFeCo film in real space[15] by means of pump-probe magneto-optical Kerr microscope. Rasing et al. have succeeded in all-optical magnetization control[13] of GdFeCo and made time-resolved microscopic analysis of the dependence of the magnetic relaxation time on the sample composition and temperature[16]. In Ref. 24, the authors employed X-ray magnetic circular dichroism (MCD) to probe element-selective electronic and magnetic states inside the laser spot with subpicosecond time resolution[26]. The transient ferromagnetic coupling of Gd and Fe in GdFeCo during the ultrafast
magnetization reversal has been reported in Ref. 17.

In the present paper, we report on the light-induced magnetization reversal of Gd$_{23}$Fe$_{68}$Co$_9$ thin films with perpendicular ferrimagnetism investigated by the photoemission electron microscopy (PEEM). PEEM has the potential of microscopy, spectroscopy and time-resolved analysis at the same time. Therefore, it can be employed to element-selectively investigate the dynamics of the electronic and magnetic properties of the sample in real-space. For example, the analysis of spatial inhomogeneity in element-specific magnetization dynamics and the time evolution of XAS spectra in specific area is available. We believe that our PEEM study on GdFeCo helps us to comprehensively understand the mechanism of the light-induced magnetization reversal process, as well as to solve the technical issues associated with practical applications. We demonstrate that (1) helicity-dependent magnetization control is possible employing circularly polarized light of proper helicity, power, laser fluence and spot size, (2) GdFeCo keeps its electronic and magnetic properties even after the irradiation by a large number of laser pulses whose power is high enough to reverse the magnetization, suggesting that the magneto-optical read-write system may be applicable for practical use and (3) we have succeeded in time-resolved PEEM observation of light-induced magnetization reversal and found the specific area with the ultrafast magnetization reversal within the time scale of subnanosecond.

2. Experimental

In a series of experiments, we used amorphous GdFeCo thin films with perpendicular magnetization grown by magnetron sputtering onto the SiO$_2$ substrate. The detailed multilayer structure is as follows; Ta(1nm)/Ru(1nm)/Gd$_{23}$Fe$_{68}$Co$_9$(20nm)/Ru(20nm)/Ta(5nm)/SiO/Si(substrate). The sample was mounted on the manipulator installed in the PEEM apparatus (ELMITEC PEEMSPECTOR) at the endstation of the soft X-ray beamline BL25SU [27] in SPring-8. In this beamline, left (CCW) and right (CW) fully circularly polarized soft X-rays can be used as the probe beam. The helicity of the soft x-ray beam was switched by kicker magnets with a frequency of 0.1 Hz in order to minimize possible drift effects in pairs of dichroic images[28,29]. The detailed PEEM setup for pump-probe measurements is described elsewhere[30]. In this study, we apply the activating voltage (+600 V, 40 ns width) to a micro channel plate (MCP) detector with the frequency of ~5 kHz, synchronizing it with the pump laser pulses and the probing soft X-ray bunches. Since the temporal resolution of our apparatus is ~50 ps, which is limited by the pulse width of the soft X-rays, we have chosen the GdFeCo sample with ~23% Gd
composition whose magnetization switching time is considered to be relatively slow in Ref. 16 (several nanoseconds) and suitable for studying the time evolution of the spin reorientation process in detail. As an excitation source, we employed Tsunami Ti:sapphire laser combined with a regenerative amplifier, Spitfire Pro, generating $\lambda=800$ nm wavelength ultrashort pulses with a repetition rate of $\sim 5$ kHz, $\tau \sim 120$ fs. Schematic drawing of the experimental setup describing laser path to the PEEM chamber is shown in Fig. 1. First, we split the laser light into the horizontally ($\pi$) and vertically ($\sigma$) polarized beams and introduced optical path difference of $\sim 3$ m between them. Then the two beams were very accurately merged into the same optical path, reflected upward and then reflected $30^\circ$ downwards on the upper experimental deck onto the sample for the PEEM measurements. The laser beams passed also through the $\lambda/4$ phase-plate just before the PEEM chamber and were converted into two circularly polarized beams with opposite helicities. Accordingly, a pair of laser pulses with different helicities was hitting the sample with the temporal separation of $\sim 10.5$ ns. The timing chart of the pump-probe experiment is shown in the upper inset of Fig. 1. The preceding pulse (“pump” in the figure) has the helicity which prefers to reverse the magnetization, while the latter one (“reset”) has the opposite helicity and is used to initialize the magnetic domain. Note that we constructed this setup, assuming that the helicity-dependent magnetization control works as described above. In the case of static measurement, either the left or right helicity laser beam was used by blocking the path of the other beam. Due to the use of the flip-up and -down mirrors inserted into the laser path, the degree of the circular polarization was $\sim 70\%$. The laser was finally incident onto the sample at $30^\circ$ in respect to the sample plane. Continuously adjustable neutral density filters were used to obtain the optimum laser power. All experiments were done at room temperature.

3. Results and discussion

Prior to the PEEM experiment, we verified ex situ the effect of laser radiation to the GdFeCo magnetization and its dependence on the pump beam helicity. Magnetic domain images of GdFeCo taken by the magneto-optical Kerr microscope after the irradiation of laser pulses are shown in Fig. 2. Note that the laser pulses are incident normal to the sample surface plane and their degree of circular polarization is $100\%$ in these preliminary experiments. Figure 2 (a) shows the effect of circularly polarized laser pulses with power of 40–45 mJ/cm$^2$ and the spot diameter of about 150 $\mu$m. The sample was mounted on a fast-rotating disc so as to have each laser irradiation spot well separated on the sample. In this case the sample was
moved at a velocity of ~750 mm/s during the irradiation, where the pulse frequency was 5 kHz, i.e., the sample position moved by 150 µm before the irradiation by the next pulse (150 µm/pulse). The image shows random multidomains inside the spot, caused by *heat driven* demagnetization and zero-field cooling without clear helicity dependence. When the velocity was reduced to ~20 mm/s (~4 µm/pulse) to clearly see the event at the edge of the pulse spot, however, the magnetization was selectively switched into the up (black) and down (white) states in the regions for the left-(σ⁻) and right-(σ⁺) handed circularly polarized pulse excitation, respectively, as seen in Fig. 2(b). In this experiment, it seems that pure *opto-magnetic* switching takes place only at the edge of the laser spot where the circularly polarized light can induce effective magnetic field. By further fine adjustment of the laser power density and the spot size, *all-optical* magnetic recording without creation of random domain structure could be achieved, as displayed in Fig.2(c), showing the result at laser power of 11.8 mJ/cm² and the sweeping rate of the sample at 48 mm/s (~10 µm/pulse). Although the spot size was 225 µm in FWHM in this case, the switched area was only ~5 µm across at the center. In this setup, we proved that we can reproduce the results reported by Stanciu *et al.*[13]. The difference in the laser power density required for optical switching (2.8 mJ/cm² in ref.13) may be ascribed to the differences in the multilayer structure, including buffer and capping layers. Nevertheless, the window to enable one to make *all-optical* magnetic control (without a *heat-driven* reversal area) is extremely narrow (in this experiment, we had to adjust the distance of a condensing lens with accuracy of 100 µm for successful observation of domain switching). We note that the studies of material properties, multilayer designs and laser radiation conditions are extremely important to facilitate the practical use of optical magnetic control in the future.

In Fig. 3, we show the results of XMCD-PEEM domain observation during the continuous irradiation of either the σ⁺ or σ⁻ circularly polarized laser pulses of 5 kHz with 20 mJ/cm², by using the circularly polarized soft X-ray beam at the Gd M5 absorption edge (1183.5 eV). The laser spot size is estimated to be 100 µm FWHM, though it is distorted from the circular shape by the mirror reflection. In Fig. 3(a), σ⁻ laser pulses are first aimed at a black (up) magnetic domain and then moved slowly to a white (down) domain beyond an artificial domain boundary (engraved thin ditch). Here, the sample position is moved by a micrometer manipulator against the fixed laser spot. We can see the central spot of the laser (gray area) where the magnetization is thermally fluctuating under the continuous pulses. Figure 3(b) shows similar images but for σ⁺ pulses that moved from a white to black domain. In this
experiment, we clearly observe that the $\sigma^-$ or $\sigma^+$ light prefers orienting the magnetization towards the sample surface normal up (black) or down (white), respectively. We also demonstrate in Figs.3 (c) and (d) the recreation of a single domain of a given magnetization direction in a certain area defined by the engraved orthogonal ditches by using the $\sigma^-$ or $\sigma^+$ light, respectively. In this way, we can also create magnetic domains in a scale of several tens micrometers in diameter. However, in the PEEM experiment, we had not succeeded so far in all-optical magnetization control by a single shot as shown in Fig. 2(c) and the effect of heat-driven magnetization reversal was dominant (not shown), possibly due to the difference in the degree of circular polarization or the difference in the heat diffusion compared with the ex-situ experiment.

To explore the effect in detail, we compared XAS and XMCD spectra in the areas with and without the history of laser irradiation. Figures 4(a) and (b) show the spectra of GdFeCo around the Gd $M_{4,5}$ and Fe $L_{2,3}$ absorption edges, respectively. In the region encircled by the red-dashed curves in Figs.4 (a),(b) and (c), the laser pulses for a duration longer than few minutes at 5 kHz (more than 1,000,000 pulses) have been irradiated. The power of each pulse, $\sim 25$ mJ/cm$^2$, is high enough to induce the heat-driven magnetization reversal. For both absorption edges, the XAS spectra exhibit rather standard behavior typical for solid solution alloy[31], independent of the history of the irradiation and we conclude that there is no sign of the change in the electronic states. Similarly, each XMCD spectrum has almost symmetric shape. Although a slight reduction in the absolute value of the XMCD intensity (about 80%) is seen in the irradiated area, the difference is within the error caused by several reasons, mainly 1) inhomogeneous synchrotron light intensity in the field of view ($\sim 100$ µm) and 2) non-linearlity of the MCP detector. Figures 4 (c) and (d) show irradiation effects on the XAS spectra around the oxygen $K$-edge. We believe that the oxygen in this sample originates mainly from the oxidized capping layers (Ta and Ru). For comparison, we show in Fig.4 (d) the PEEM image at the same photon energy of 533 eV and position dependent XAS spectra of “damaged” GdFeCo on which a single pulse of 120 mJ/cm$^2$, about 10 times greater than the power required for the magnetization reversal ($\sim 11.8$ mJ/cm$^2$), was applied. One can see that the O $K$-edge spectra change sensitively after the radiation damage. However, as is clear in Fig. 4 (c), the O $K$-edge spectra in the same area as the PEEM images in Fig. 4 (a) and (b) show no change in electronic states of oxygen even after the pulse radiation over 1,000,000 times, as long as its power is kept around $10 \sim 25$ mJ/cm$^2$. From these results, we conclude that the power of the light pulse required for magnetization control is much lower than the
threshold power to induce noticeable damages to GdFeCo samples. Thus, we conclude that this recording technique can be applicable to practical applications in which a repetition of writing is required.

Finally, we report on the results of time-resolved magnetic domain measurement around the laser spot. Figure 5 (a) shows the pump-probe XMCD-PEEM images at the Gd $M_5$-edge observed by the system shown in Fig.1. In this experiment, we apply the $\sigma^-$ polarized pulse to reverse the magnetization to up (black) state at 0 ps, and then the $\sigma^+$ pulse is applied at 10.5 ns later to recreate the initial magnetic state (white single domain). The pulse power for both helicities is just above the threshold of magnetization reversal, namely, about 12 mJ/cm$^2$. As soon as the sample is irradiated by the $\sigma^-$ polarized laser pulse, the area of ~40 $\mu$m across is thermally demagnetized and changes into the gray color, as clearly seen at 0 ps. The magnetization direction recovers to the down (white) state gradually from the outer area of the spot in the time scale of few ns. However, there remains still an up (black) domain of about 10 $\mu$m at the center until the irradiation of the $\sigma^+$ pulse (10.5 ns later) with enough stability of the reversed magnetization direction. On the irradiation of the $\sigma^+$ reset pulse at 10.5 ns, the area of 40 $\mu$m across is again thermally demagnetized and becomes gray. Although the outer magnetizations are soon recovered to the down state, the up domains in the central area survives even after 400 ns. It is still noticed that the area A becomes to the down spin state (white) before the next $\sigma^-$ pump pulse (~100 ps). Figure 5 (b) shows the temporal evolution of the MCD intensities at the central spot (area A) and the circle area outside the central spot (area B). In the area B, magnetization is almost quenched within 50~100 ps, and then slowly recovers to down (white) state within 5 ns. In the central area A of about 10 $\mu$m, on the other hand, the magnetization reversal to up (black) state completes within 50~100 ps as well and stays reversed, at least for longer than 10 ns.

Here, we briefly review the earlier works to understand the magnetic dynamic behavior we observed. In the dipole-dipole interaction scenario expected from the heat-driven magnetization reversal[15], thermally quenched magnetizations recovered from the area surrounding the central spot within subnano seconds, followed by the magnetization reversal in the core region, taking several nanoseconds for stabilization. The results of opto-magnetic switching[16], magnetization reversal as well as the recovery in surrounding area completes within several tens of picoseconds, under the following condition. Namely, the experimental temperature ($T_{\text{exp}}$) is lower than the angular momentum compensation temperature, $T_{\text{comp}}$[32] (where the singular point of effective Gilbert damping constant, $\alpha_{\text{eff}}$ diverges) and the
temperature around the laser spot passes through $T_{\text{comp}}$ during the irradiation of a heat pulse and approaches the Curie temperature ($T_C$). Meanwhile, Hohlfeld et al. considers that there is no need for thermal assist or the precessional motion in opto-magnetic reversal process. The authors conclude that the magnetization reverses in the order of subpicoseconds, regardless of $T_{\text{comp}}$.

We can ask for the origin of relatively slow magnetization recovery (several ns) in the area B in the heat-driven scenario. If the magnetization precession is assumed to be the main factor to decide the reversal time, we can also explain the result with opto-magnetic process discussed in Ref. 16 ($T_{\text{comp}}$ of our GdFeCo sample is well below the room temperature (~200 K)). In the area A, on the other hand, the reversal time within 50~100 ps is extremely rapid. It exceeds the prediction in Ref. 16 but rather close to that in athermal optical switching senario[33]. At least it is never explained by simple heat driven process, i.e., domain wall motion after the thermal fluctuation, because the saturation of magnetization in the area A completes, not waiting for the stabilization of the area B. Nevertheless, it should be mentioned that the up (black) domain at the center keeps its direction even 400 nanoseconds after the irradiation of the $\sigma^+$ (reset) pulse which is expected to turn the magnetization toward the down (white) direction. The down (white) single domain is, however, always recreated by 200 $\mu$s after (~100 ps in the figure), probably due to some balance of dipole-dipole interaction with nearby magnetization. We were not fully successful to verify “complete” helicity dependent switching in the static PEEM observation as mentioned above. We think at present that the magnetization reversal we have seen in our experiment under the employed conditions is strongly influenced by the heat-driven effect. Still, some optical effect (i.e. ultrafast and strong effective field by inverse Faraday effect) seems also to contribute to the reversal under a critical condition. By slightly improving the experimental conditions, especially the degree of circular polarization, laser spot shape on the sample and more extensively studying the time dependence, we may be able to upgrade the research on the dynamics of all-optical magnetization reversal process, as well as its element (Gd and Fe) dependence in the near future.

4. Conclusion

In summary, we have investigated laser light induced magnetization switching in GdFeCo by using pump-probe PEEM. By utilizing the potential of PEEM, we have studied the element-specific magnetic domain structures, local electronic states and their time dependence at the same time. We observed that the magnetization reversal depending on the helicity of the
light pulse occurs only in the limited area where the light-induced field works effectively. Comparison of local XAS and XMCD spectra between the areas with and without the irradiation of a number of pulses confirmed that GdFeCo samples have not been destroyed by proper laser radiations required for magnetization control (~12 mJ/cm²). We also reported the first technical success in pump-probe measurement of light induced magnetization switching by PEEM. Although we have not yet fully succeeded in all-optical magnetization control without a heat-driven reversal area, we found that the magnetization switching finishes almost within ~100 ps in the central spot, suggesting that some opto-magnetic effect contributes to the magnetization reversal. Detailed studies on the spin reversal process under all-optical magnetization control and element specific spin dynamics are now in progress.

Acknowledgment

The experiments at the BL25SU in SPring-8 were supported with the approval of Japan Synchrotron Radiation Research Institute (JASRI) (Proposal Nos. 2009A1443, 2009B1347, 2009B2120, 2010A1645, 2011A1183 and 2011A2056). The work was partially supported by a Grant-in-Aid for Scientific Research (S, 18101004) from Japan Society for the Promotion of Science and Projects for Innovative Quantum Functional Materials (PIQFUM) of MEXT, Japan.

References

For example, $T_{\text{comp}}$ exceeds room temperature in the GdFeCo sample with the Gd concentration of more than ~25%.

Fig. 1. Schematic image of the experimental setup for PEEM. Timing chart of pump-probe measurements is also shown in the upper inset.
Fig. 2. Kerr microscope images of a GdFeCo film after irradiation of circularly polarized laser. White and black area corresponds to the down and up magnetic domains, respectively. Here, the irradiation test is performed in the air, with normal incident, and with 100% circular polarization. (a) Each pulse of 40~45 mJ/cm$^2$ landed on different areas. (b) Similar laser power with (a) but the sweeping speed is much lower than (a) so as to clearly see the helicity-dependent magnetization switching occurring at the edge of the spot. In (a) and (b), schematic trace of the laser pulses is also shown by dashed circles. The size of the laser (FWHM of Gaussian pulse profile) may be comparable to that of the magnetization reversal area. (c) Demonstration of all-optical magnetic writing by properly adjusting the power and the size of the laser spot. (d) Size and power profile of the laser used in (c). Although the FWHM of the laser pulse is ~225 µm, the effective reversal area seen in (c) is extremely small (5~10 µm).
Fig. 3. (a), (b) XMCD-PEEM images of GdFeCo at Gd $M_5$-edge during the successive 5 kHz $\sigma^-$ and $\sigma^+$ circular polarized laser excitation, respectively. (c), (d) Demonstration of single domain recreation in a rather wide region by using the $\sigma^-$ and $\sigma^+$ circularly polarized laser by moving the sample relative to the laser spot. For all figures, the spot size of the laser is comparable to the field of view ($\phi100\mu$m) and the gray areas inside each field of view are the central spots of the laser where the magnetization is thermally fluctuating by continuing pulses at 5 kHz. A single domain is recreated in a certain area defined by the orthogonal ditches engraved in advance by photolithographic patterning and sputtering.
Fig. 4. (color) (a), (b) Upper panels: XMCD-PEEM images of GdFeCo at Gd $M_{4,5}$- (1183.5 eV) and Fe $L_{2,3}$- (708 eV) edges, respectively. Lower panels: Local XAS and XMCD spectra of GdFeCo around Gd $M_{4,5}$- and Fe $L_{2,3}$-edge, respectively, of GdFeCo in the areas with (red dashed circle on the upper panel) and without (blue) the experience of laser irradiation. (c) XMCD-PEEM image at 533 eV and local XAS spectra near the O $K$-edge. Probed area is the same as (a) and (b). (d) Similar plots as (c) but with expanded scale of different area where a single pulse of $\sim$120 mJ/cm$^2$ was applied deliberately. The intensity contrast seen in the image suggests the variation of the electronic states by considerable (and inhomogeneous) damage. The spectra displayed with different colors correspond to the regions shown by the dots in the upper image with the same colors.
Fig. 5. (color) (a) Pump-probe XMCD-PEEM images after the excitation by the $\sigma^-$ circularly polarized pulse at 0 ps. Some speckles seen in the field of view are due to background noise of photoemission by laser excitation. (b) Time evolution of the magnetization at the central spot (area A) and near the edge of the magnetization reversed area (area B).