Investigation of helicity-dependent optical switching of ferrimagnetic Gd–Fe–Co films

Magnetization control by femtosecond light pulses has been extensively investigated for next-generation magnetic recording technologies. This approach enables the ultrafast manipulation of magnetic ordering on pico- to femtosecond timescales, potentially overcoming the speed limitations of conventional magnetic field-driven magnetization switching [1]. Since the magneto-optical effect is theoretically a non-thermal process, it offers a pathway for designing energy-efficient recording devices. Particularly, selective magnetization switching using right- and left-handed circularly polarized laser pulses, known as helicity-dependent magnetization switching (HDMS), is noteworthy both as a novel physical phenomenon and for its potential device applications (Fig. 1(a)) [2].

The origin of HDMS is explained by the inverse magneto-optical effect, where polarizations of the incident light influence the material's magnetization. Initially discovered in ferrimagnetic thin films composed of 4*f* rare-earth and 3*d* transition metals, HDMS was later observed also in ferromagnetic thin films such as CoPt. However, clear single-shot magnetization switching (monotonic HDMS), as shown in Fig. 1(a), has so far been limited to a few specific ferrimagnetic alloys such as Gd–Fe–Co. In contrast, other magnetic thin films typically exhibit only cumulative HDMS under multi-pulse irradiation, as shown in Fig. 1(b). Achieving monotonic HDMS in a broader range of material systems including rare-earth-free ferromagnetic alloys, remains a significant challenge for resource strategy and the practical development and commercialization of HDMS-based devices.

In ferrimagnetic materials, monotonic HDMS is more easily achieved owing to the difference in magnetization reversal speeds between the 4*f* and 3*d* atomic sites under femtosecond laser pulse excitation, as well as the divergence of magnetization damping near the angular momentum compensation point. These factors are believed to explain why only cumulative HDMS has been observed in ferromagnetic systems so far.

This study systematically examined both monotonic and cumulative HDMS in a series of ferrimagnetic Gd–Fe–Co thin films with varying material parameters. Furthermore, we discovered that the transition between these two types of HDMS modes was strongly influenced not only by the angular momentum compensation characteristics, but also by the thermal conductivity properties of the thin films. This study aims to provide insights into strategies for achieving clear HDMS across a broader range of material systems, including ferromagnets [3].

Figures 2(a–f) show magnetic domain images of Gd–Fe–Co thin films with different compositions after laser pulse irradiation. The magnetic domain images were acquired using a magneto-optical Kerr microscope. The $Gd_{26}Fe_{66}Co_8$ sample has an angular momentum compensation temperature (T_A) above room temperature, whereas $Gd_{22}Fe_{70}Co_8$ has a T_A below room temperature. The former exhibits stronger magnetization damping, promoting faster and clearer magnetization reversal, whereas the latter shows weaker damping, leading to diffuse magnetic relaxation [4]. For the $Gd_{26}Fe_{66}Co_8$ thin film, irradiation at the reversal threshold fluence (20.4 mJ/cm², Fig. 2(a)) induces a clear polarization dependence. Even at a slightly higher fluence (20.9 mJ/cm², Fig. 2(b)), the area of magnetization reversal remains dependent on the laser polarization. Figure 2(c) shows the magnetic domain structure when the laser irradiation spot was slowly moved (50 μm/s, 5 kHz pulse), resulting in multiple irradiations at the same location. This measurement aimed to investigate whether cumulative HDMS occurs when monotonic HDMS is not observed. For this sample, complete HDMS is clearly confirmed. Figures 2(d–f) show the results of similar experiments on $Gd_{22}Fe_{70}Co_8$. The results are nearly identical to those observed in the $Gd_{26}Fe_{66}Co_8$ case.

However, when a similar pulse irradiation was performed in vacuum, clear composition dependence was observed. Figures 2(g–i) illustrate the magnetic

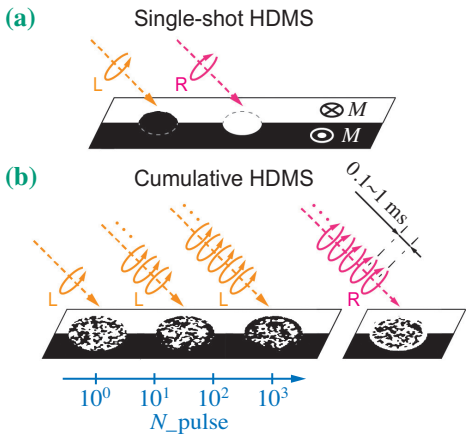

Fig. 1. Schematic illustrations of **(a)** single-shot and **(b)** cumulative HDMS. Magnetization directions of the specimens are indicated by "*M*." "L" and "R" denote the left- and right-handed circular polarizations of the laser pulse, respectively.

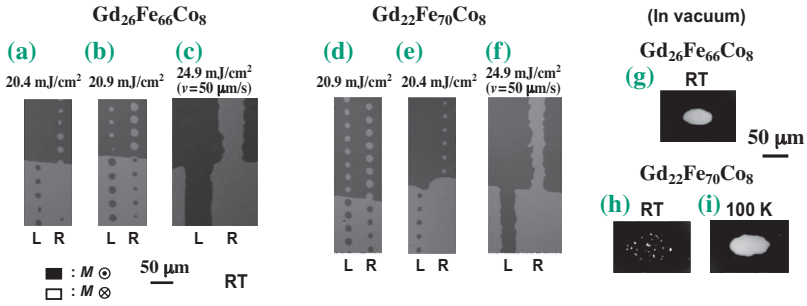

Fig. 2. **(a–f)** Magneto-optical Kerr microscopy images of Gd–Fe–Co films with different alloy compositions and pulse irradiation conditions. **(g–i)** X-ray magnetic circular dichroism (XMCD)-PEEM images of Gd–Fe–Co films with varying alloy compositions and temperatures.

domain images of the Gd–Fe–Co thin films after single laser pulse irradiation in a vacuum chamber, observed by the photoemission electron microscopy (PEEM) apparatus at SPring-8 **BL25SU**. In $Gd_{26}Fe_{66}Co_8$, a clear magnetization reversal was observed (Fig. 2(g)), whereas in $Gd_{22}Fe_{70}Co_8$, where magnetization damping was considered slower (Fig. 2(h)), random magnetic domain structure was observed, suggesting disturbances during the magnetization reversal process. All the experiments were conducted at room temperature. However, when the $Gd_{22}Fe_{70}Co_8$ sample was cooled below its T_A to 100 K (Fig. 2(i)), a clear magnetization reversal similar to that in $Gd_{26}Fe_{66}Co_8$ was observed. These results demonstrate the importance of utilizing the magnetization damping properties owing to angular momentum compensation for monotonic HDMS in ferrimagnetic thin films. Simultaneously, it also suggested that the degree of this property can be controlled by the thermal diffusion properties of the sample (note that the thermal diffusion at the sample surface is slowed down in vacuum).

For further investigation, HDMS properties were examined at room temperature and atmospheric pressure using Gd–Fe–Co thin films where a 5 nm Si_3N_4 thermal blocking layer was inserted between the film and the Ru buffer layer to slow down thermal diffusion. The film compositions were $Gd_{26}Fe_{66}Co_8$, $Gd_{24}Fe_{68}Co_8$, and $Gd_{22}Fe_{70}Co_8$, with their T_A above, near, and below room temperature, respectively. As shown in Fig. 3, the HDMS characteristics clearly changed depending on the differences in T_A of the samples. In particular, for $Gd_{22}Fe_{70}Co_8$, similar to the results in vacuum, a disordered magnetic domain structure was formed, and only a faint cumulative HDMS, as reported for ferromagnetic thin films [5], was observed (Fig. 3(i)).

Other experiments have also suggested that the magnetization reversal characteristics of Gd–Fe–Co thin films are influenced not only by ultrafast magnetization dynamics due to angular momentum compensation on a microscopic timescale but also by macroscopic magnetization dynamics, such as domain wall motion following the reversal process. This indicates that angular momentum compensation, a unique feature of ferrimagnetic materials, is not the sole factor required for achieving monotonic HDMS. Therefore, it suggests the possibility of developing promising HDMS devices even in ferromagnetic thin films, by utilizing other magnetic dynamic properties. Further studies in this direction could lead to the development of practical HDMS devices using more versatile material systems.

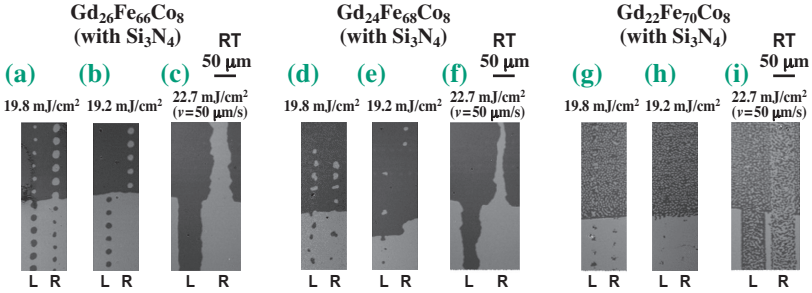

Fig. 3. Similar images to Figs. 2(a–f), but on Gd–Fe–Co films with Si_3N_4 thermal blocking layers underneath.

Takuo Ohkochi

Laboratory of Advanced Science and Technology for Industry, University of Hyogo

Email: o932t023@guh.u-hyogo.ac.jp

References

[1] A. Kirilyuk *et al.*: Rev. Mod. Phys. **82** (2010) 2731.
[2] C. D. Stanciu *et al.*: Phys. Rev. Lett. **99** (2007) 047601.
[3] *T. Ohkochi, R. Takahashi, H. Fujiwara, H. Takahashi, R. Adam, U. Parlak, K. Yamamoto, H. Osawa, M. Kotsugi, A. Tsukamoto, H. Wadati, A. Sekiyama, C. M. Schneider, M. Tsunoda, S. Suga and T. Kinoshita: J. Magn. Magn. Mater.* ***593*** *(2024) 171854.*
[4] K. Vahaplar *et al.*: Phys. Rev. Lett. **103** (2009) 117201.
[5] C.-H. Lambert *et al.*: Science **345** (2014) 1337.