

Large magneto-optical effects in a non-collinear antiferromagnet and its application for antiferromagnetic spintronics

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Spintronics has attracted considerable attention because of potential applications in non-volatile data storage and low-power information processing. These applications are traditionally based on the control of magnetization in ferromagnetic (FM) materials. On the other hand, in the last few years, there also has been a surge of interest in antiferromagnetic (AF) materials as an active material for next-generation spintronics devices, with the prospect of providing higher density and much faster operation speed for the devices than their ferromagnetic counterparts [1]. However, it is also true that the absence of magnetization has made the AF state notoriously difficult to electrically detect and externally manipulate, unlike in the case of FM systems. Motivated by these intriguing properties, several breakthroughs have been made relatively recently. For example, anisotropic magnetoresistance and spin Hall magnetoresistance, an even-function response under time-reversal (TR), have been found useful for detecting the electrical switching of the collinear AF ordering [2,3]. Another breakthrough is the discovery of an odd-function response under TR in the non-collinear antiferromagnets Mn_3Sn [4]. As the first case in AF metals, Mn_3Sn has been experimentally found to exhibit a large transverse response such as an anomalous Hall effect (AHE) [4] and an anomalous Nernst effect (ANE) [5] which had been considered to be restricted to FMs.

Mn_3Sn is the hexagonal D_{019} system (space group $\text{P6}_3/\text{mmc}$) and has the ABAB stacking sequence of the (0001) kagome layer of Mn atoms. Below the Néel temperature $T_N \sim 430$ K, the geometrical frustration leads to a non-collinear AF ordering called the inverse triangular spin (ITS) structure. This AF state can be viewed as a ferroic ordering of cluster magnetic octupoles [6]. Interestingly, in Mn_3Sn , the octupole ordering, not the magnetization, plays a role of the major order parameter breaking the TR symmetry macroscopically. The polarization direction of the TR-odd order parameter (magnetic octupole in Mn_3Sn) determines the distribution of the Berry curvature in momentum space, which acts as an effective magnetic field and gives rise to the TR-odd transverse responses [4,7]

In this presentation, I will talk about magneto-optical properties of Mn_3Sn [8]. The magneto-optical Kerr effect (MOKE) has been intensively studied in various ferro- and ferrimagnetic materials because it provides a powerful non-contact and non-destructive probe for electronic and magnetic properties as well as for various applications, including magneto-optical recording. Although MOKE had been believed to be absent in the fully compensated collinear AFMs, recent theoretical and experimental progress has revealed that AHE has the same symmetry requirements as MOKE [9], suggesting the potentially large MOKE in Mn_3Sn . In fact, we have found that despite a negligibly small magnetization, Mn_3Sn exhibits a large zero-field MOKE (~ 20 mdeg) at room temperature, comparable to that in ferromagnets. Our first-principles calculation has clarified that the ferroic ordering of the cluster magnetic octupoles causes MOKE even in its fully compensated AF state. This large MOKE further allows imaging of the octupole domains (Fig. 1), which are closely related to the large transverse responses (e.g., AHE, ANE) induced by the Berry curvature, as discussed above. I will also report recent progress of development of the Mn_3Sn thin films [10] and newly observed optical responses from the terahertz waves [11]. These findings have provided an important avenue for further studying the AF dynamics [12] and developing AF devices such as memory [13] and heat-flux sensor [14].

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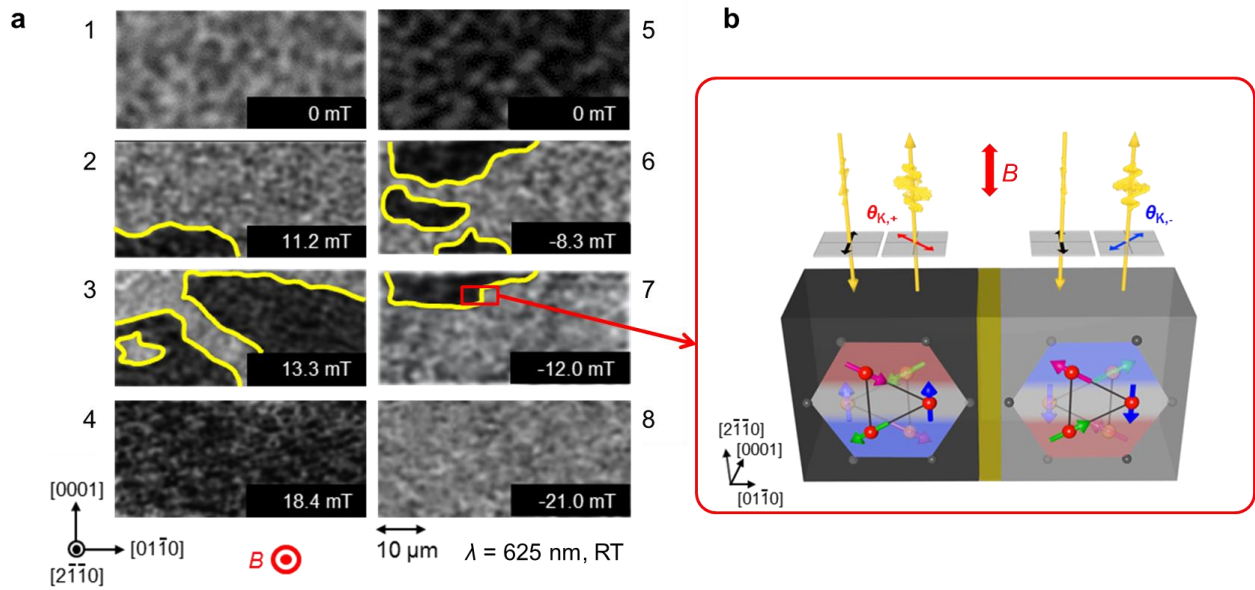


Fig.1 (a) Evolution of the antiferromagnetic domains of the Mn_3Sn (2-1-10) plane as a function of a field B (1-4: $-21 \rightarrow 21$ mT, 5-8: $21 \rightarrow -21$ mT). (b) Schematic illustration of two regions with different MOKE image contrasts due to opposite signs of the Kerr angles, corresponding to two types of cluster magnetic octupole domains that have inverse triangular spin structures with opposite spin directions within the (0001) plane.

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Theoretical proposal for control of spin textures and vortices with topological light waves

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Topological light waves such as vortex beams, vector beams, etc., have attracted much attention in the field of optics and laser science. Their basic natures and efficient methods of generating these waves have actively explored. These spatially structured lights have high potential to create various novel photo-induced phenomena when we apply them to materials. However, the study of such possibilities has been finally activated very recently. Motivated by this situation, we have theoretically studied several ways of application of topological light waves to materials. Particularly, we have focused on ways of controlling electron states in solid materials with topological lights.¹⁾⁻⁴⁾ In this symposium, I would like to mainly report two topics among our results.

The first topic is the application of vortex beam to magnets (Fig.1).^{1),2)} Vortex beam is the laser beam carrying orbital angular momentum (OAM). This wave has two characteristics. The first feature is that the intensity profile of the vortex beam is a doughnut type, i.e., the center of the beam focused plane is always the position of zero intensity irrespective of the value of OAM. The second is that electric- and magnetic-field directions has an angular dependence around the zero-intensity center in the focused plane. Based on these two natures, we have proposed ultrafast ways of creating topological magnetic defects such as skyrmion and skyrmionium (Fig.2), spin waves with a spiral wavefront (Fig. 3), and so on.

The second topic is the application of vector beam.^{3),4)} The vector beams also have unique spatial features. A simple way of creating a vector beam is to prepare a superposition of two vortex beams with OAM $+m$ and $-m$. Strongly focusing such a vector beam with lens, at the center of the focal plane we have the small singular area where only the AC magnetic (electric) field is dominant, while AC electric (magnetic) field is negligible. Namely, we can create a strong high-frequency magnetic (or electric) field without its dual field. Using this property, we have proposed several applications of vector beam to materials: Detecting magnetic resonance in magnetic semiconductors and multiferroic magnets, observing the edge current in topological phases (topological insulators, topological superconductors, etc.), estimating Fermi surface of metallic magnets, Floquet engineering, etc.

In this symposium, I would like to report these results, concentrating on their essential aspects. Moreover, if possible, I will also report our on-going result of application of vortex beam to superconductors.⁵⁾

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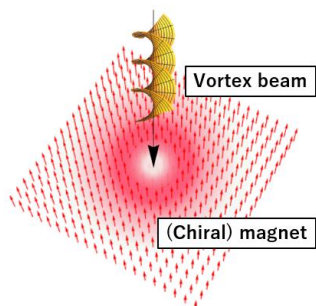


Fig. 1: Image of application of vortex beam to a magnet.

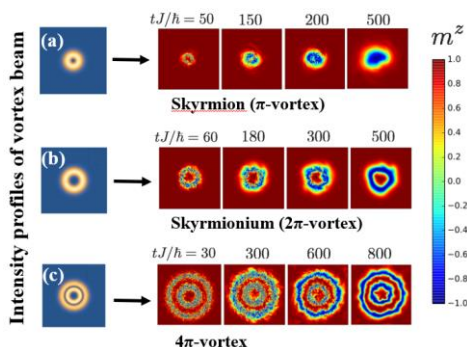


Fig. 2: Processes of creating topological defects in a magnet with (a) small-ring, (b) wide-ring, and (c) double-ring vortex beams.

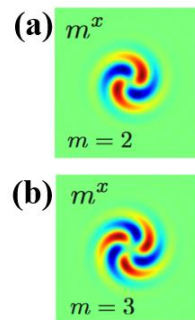


Fig. 3: Spin-wave resonances driven by vortex beams with QAM (a) $m=2$ and (b) 3.

Metamaterials and metasurfaces with broken symmetries

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In this invited talk, I am going to present intriguing optical responses of structured metamaterials and metasurfaces with broken symmetries [1,2,3]. Mostly, my talk focuses on directional birefringence independent of light waves polarization, referred to as optical magneto-chiral (MCh) and magneto-electric (ME) effects. I would like to convince the audience that metamaterials and metasurfaces can boost optical MCh and ME effects by several orders of magnitude compared to natural materials.

The optical MCh effect is a combination of the magneto-optical (MO) effect and optical activity (OA). While the effect is usually very small in natural materials, it can be enhanced using metamaterials [1]. We embody experimentally a metamolecule using a Cu chiral structure (chiral meta-atom) for OA and YIG ferrite rod/cylinder (magnetic meta-atom) for the MO effect. The metamolecule is studied at the X-band microwave frequency at room temperature. At a chiral resonance frequency around 10 GHz, a large non-reciprocal refractive index difference around $\sim 10^{-3}$ enhanced by MCh effects is observed [4]. Numerical calculations have successfully reproduced this experimental observation and revealed that the enhancement is traced back to the hybridization of ferromagnetic resonance (FMR) in the magnetic meta-atom and chiral resonances in the chiral meta-atom. Furthermore, numerical simulation predicts a giant MCh effect with a much larger index difference [5]. Notably, our concept of enhanced MCh effects using metamaterials is applicable to other regions of the spectrum including, the THz, infrared, and visible region.

For a higher frequency operation than microwave, MCh metamolecules are miniaturized using a strain-driven self-coiling technique. A micrometer-sized free-standing Py chiral metamolecule is studied by cavity-FMR and coplanar-waveguide FMR [6]. This miniaturized metamolecule is very promising for obtaining optical MCh effects at millimeter wave and THz frequencies. However, many challenges remain within the optical region. To reach the optical region, the MCh metamolecule must be miniaturized to the nanometer-scale, and losses need to be reduced significantly. The former issue can be addressed by using supramolecules or biomolecules; for example, viruses, proteins, and coiled peptide.

The enhanced and giant MCh effects open a door toward the realization of synthetic gauge fields, in other words, effective magnetic fields for electromagnetic waves. However, the values of non-reciprocal refractive index differences obtained in this study are not yet large. Additionally, although numerical simulation predicted the giant MCh effect, it is relevant to a characteristic electromagnetic mode in the waveguide. Hence, implementation of the giant MCh effect by assembled metamaterials and metasurfaces in free-space is of great importance.

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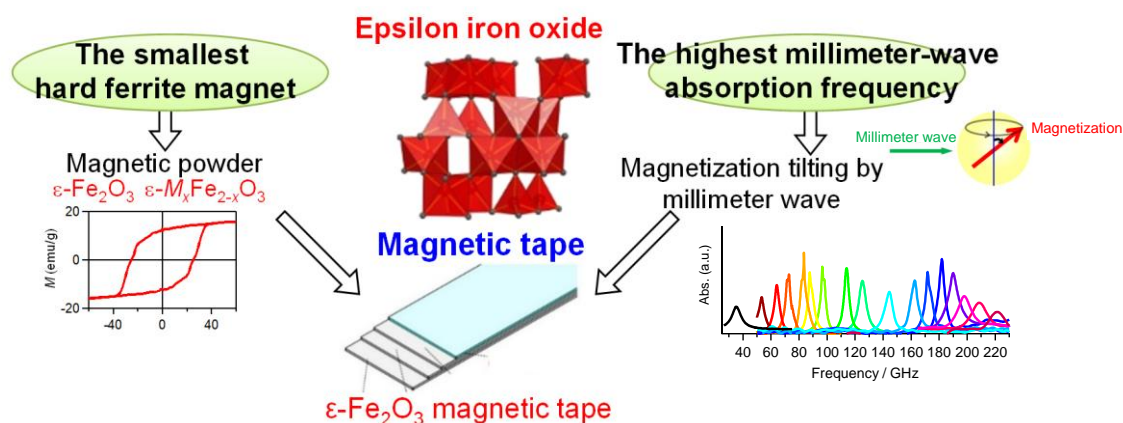
Focused-millimeter-wave-assisted magnetic recording based on epsilon iron oxide

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We reported the first example of a single-phase epsilon iron oxide ($\epsilon\text{-Fe}_2\text{O}_3$) exhibiting the strongest coercivity among all known ferrite magnets in 2004. Metal-substituted epsilon iron oxide exhibits a coercivity of up to 37 kOe. $\epsilon\text{-Fe}_2\text{O}_3$ is the world's smallest hard ferrite magnet with a ferromagnetic order of up to 7.5 nm, which can be applied to high-density magnetic recording tapes. Moreover, $\epsilon\text{-Fe}_2\text{O}_3$ and metal-substituted epsilon iron oxide have the highest millimeter wave absorption frequency in the range from 35 to 222 GHz.

In the era of Big Data and the Internet of Things, data archiving is an essential technology, and therefore magnetic recordings are drawing attention because they guarantee long-term data storage. To archive a huge amount of data, further increase of the recording density is required. Herein a new magnetic recording methodology, "focused-millimeter-wave-assisted magnetic recording (F-MIMR)," is proposed. To examine this methodology, magnetic films based on epsilon iron oxide nanoparticles are prepared and a focused-millimeter-wave generator is built using terahertz (THz) light. Irradiating the focused millimeter wave to the epsilon iron oxide instantly switches its magnetic pole direction. Additionally, the spin dynamics of F-MIMR are studied using the calculation based on stochastic Landau–Lifshitz–Gilbert model considering all of the spins in an epsilon iron oxide nanoparticle. In F-MIMR, the heat-up effect of the recording media is expected to be suppressed. Thus, F-MIMR can be useful for high-density magnetic recordings. Millimeter wave magnetic recording technique enables to reduce the particle size of the magnetic material and solve the magnetic recording trilemma, leading to the increase of the recording capacity. The transition energy of the millimeter wave is *ca.* 1/5000 compared to that visible light. Therefore, heat-up is avoided in millimeter wave-assisted magnetic recordings, which is very important for magnetic recording tapes that use organic resin for the base film.



Focused-millimeter wave assisted magnetic recording (F-MIMR)

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Development of magnetic holographic memory using artificial magnetic lattice media

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Holographic memory has been attracting attention as a data-storage technology with high recording density and data transfer rates because two-dimensional (2D) page data can be recorded and read selectively from a single position. In holographic memory, 2D data is recorded as an interference pattern in recording media. Most holographic memories use photopolymers as a write once recording media, while the magnetic hologram is a candidate for rewritable holograms in which the interference patterns of light can be recorded as differences in the direction of magnetization on the magneto-optic recording materials. A magnetic hologram is recorded using the thermomagnetic recording technique. When focused signal and reference beams are incident on a perpendicularly magnetized media, the light interference produces a temperature distribution in the film corresponding to the interference pattern through the absorption of light. As a result, the magnetization of the region above the Curie temperature is reversed during cooling, and the interference pattern can be recorded as differences in the direction of magnetization of the magnetic material in the magnetic hologram. A written hologram can be reconstructed by a magneto-optical effect such as the Faraday effect, and a large Faraday rotation angle results in a bright reconstruction image.

We have studied to realize magnetic hologram memory using polycrystalline magnetic garnet (Bi:RIG) films using the collinear interference method. We succeeded recording and reconstruction of magnetic hologram but this first reported reconstruction image of the collinear magnetic holography was dark and noisy with large background noise¹⁾. On the other hand, we have also developed magnetic materials that introduce artificial structures with a scale of several nm to several hundred nm showing new magnetism and functions depending on its structure, which is called artificial magnetic lattice (AML). Based on this knowledge, we investigated the recording conditions including the AML recording media to improve the reconstruction image of magnetic hologram. In this report, the improvement of recording media is reported to achieve a large magneto-optical effect to achieve bright reconstruction images.

The diffraction efficiency of magnetic hologram, which is an index of the brightness of reconstructed image, is described as $\eta = \sin^2(\theta_F) \approx \theta_F^2 = (Ft)^2$, where θ_F is Faraday rotation angle of magnetic hologram, F is the Faraday rotation coefficient representing the rotation angle per unit thickness, and t is the depth of magnetic hologram, respectively. So, the recording of deep magnetic fringe in the recording media with large Faraday rotation coefficient is required to achieve bright reconstruction images. We have taken two ways to solve this issue. One is the use of magneto-photon microcavity (MPM) media for increasing the rotation angle per unit thickness^{2,3)}, and the other is the introduction of heat dissipation layers (HDLs) for controlling the heat generated during writing to form deep magnetic fringes⁴⁻⁶⁾. The MPM media, in which Bi:RIG layer is sandwiched between dielectric mirrors, can increase the Faraday rotation angle of a design wavelength by Fabry-Pérot resonance. We proved that the diffraction efficiency can be increased by the use of MPM media depending on the structure²⁾, and that the brightness of reconstruction image of magnetic hologram recorded on MPM medium with 1 μm Bi:RIG layer was also increased compared to that of the same thickness single layer film as shown in Fig. 1³⁾. On the other hand, the recording thickness of magnetic fringe in single layer films is limited by the heat diffusion during thermomagnetic recording although the formation of deep magnetic fringe increases the diffraction efficiency. That is, the excess heat by the absorption of light during recording diffuses laterally and merges the regions above Curie temperature near the surface of recording medium. As a result, the interference information is disappeared in such the region. To overcome this issue, the multilayer media with transparent HDLs without light absorption is proposed to control the heat diffusion in magnetic layer. In this HDL media, the most of excess heat is diffused into the HDLs, so the deep magnetic fringe can be formed. The effectiveness of this HDL multilayer film was first shown by numerical simulation⁴⁾. The HDL medium using $\text{Tb}_3\text{Ga}_5\text{O}_{12}$ (TGG) for HDL layers was designed and fabricated as shown in Fig. 2(a), and the diffraction efficiency of the HDL and single layer media was evaluated using magnetic assist recording. As a result, the diffraction efficiency of HDL medium was about 1.6 times higher than that of single layer film as shown in Fig. 2⁵⁾. In addition, the error-free recording and

reconstruction was also achieved using this HDL medium. These results suggest that the use of artificial magnetic lattice media such as the MPM and HDL media is effective to achieve bright reconstruction image without error for magnetic holographic memory.

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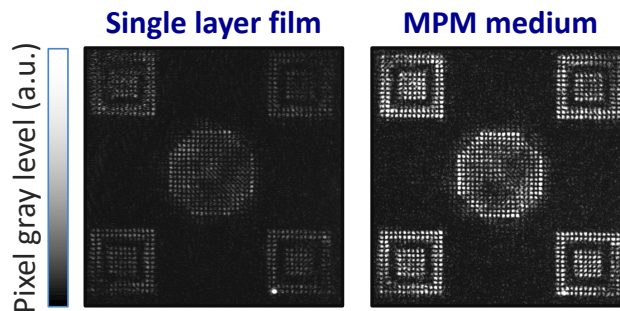


Fig. 1 Reconstruction images from Bi:RIG single layer film and MPM medium. Both media have the 1 μm thick Bi:RIG recording layer. The bright and high contrast image was obtained from the MPM medium.

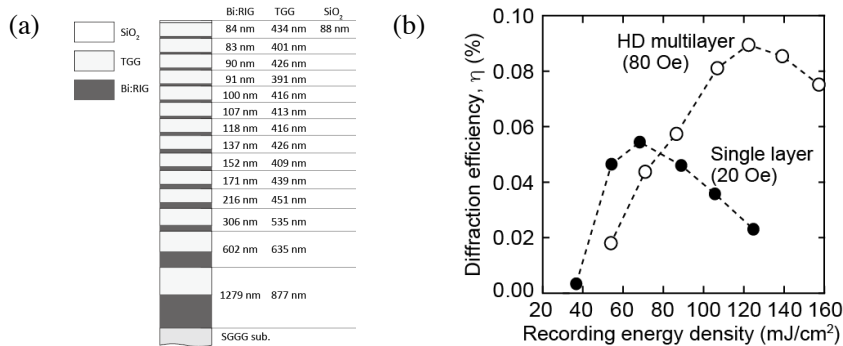


Fig. 2 (a) The structure of the fabricated HDL medium and (b) the diffraction efficiency of HDL and single layer media using magnetic assist recording.

Near-Infrared Magneto-Refractive Effect for Magnetic Multilayer; GMR film and Ferro./Antiferro. stacked film with Surface Plasmon Resonance

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1. Introduction

Much attention has been paid for magneto-refractive effect with the reflection configuration (R-MRE), because this effect simply brings light intensity change according with the arrangement of magnetizations. The magnitude of this change is small of about several %, which makes an application of R-MRE to real device difficult in this stage. In general, for a metallic magnetic multilayer film, a dominant factor of MRE is thought to be the spin-dependent scattering of conduction electrons and its optical properties are expressed by expanded Drude model corresponding to the magnetization state [1]. Therefore, evaluation of scattering time and spin-dependent scattering coefficient of metallic magnetic multilayer film are of importance as material physical properties to enhance R-MRE. However, materials examination to evaluate these properties in optical frequency region is not enough. In this study, the measurement of magneto-optical properties in IR region for antiferro-magnetically exchange coupled Co/Ru multilayers was carried out.

2. Experimental procedure

The multilayer films were fabricated by dc magnetron sputtering on glass substrates. A Ti(2 nm)/ Ru(3 nm) layer was adopted as an interlayer for the purpose of adhesion of the film to the substrate and the control of crystalline sheet texture with atomic closed packed plane parallel to the film plane. A SiN with 10 nm thick was used as a capping layer to avoid oxidation. The stacking structure of the magnetic multilayer was [Co or CoB(4 nm)/ Ru(d_{Ru} nm)]_N with the repetition number *N* of 10 with changing d_{Ru} from 0 to 1.2 nm. Here, Ru was selected as a nonmagnetic layer material to stabilize anti-parallel magnetically coupling state under zero field with a thinner film thickness than that of Cu by RKKY-like interlayer exchange coupling.

Magnetic properties were evaluated by the vibrating sample magnetometer with the maximum applied field (H_{max}) of 14 kOe. Optical properties were spectroscopically measured by the ellipsometer (M-2000, J. A. Woollam) with the wavelength region from 250 to 1700 nm. R-MRE was spectrometrically measured by hand-made system with an electromagnet applying magnetic field (H) along the film plane direction [2]. Here, the measurement wavelength (λ) range was 550–1650 nm limited by monochromator (USB-2000 and NIR-QUEST, Ocean Optics) and H_{max} was 14 kOe.

3. Results and discussion

Figure 1 shows color maps of experimental reflectance with the incident angle 70 deg. plotted against wavelength and magnetic field for a (a) [Co(4)/ Ru(0.7)]₁₀ film, (b) [Co₈₈B₁₂(4)/ Ru(0.7)]₁₀ film, and (c) [Co₈₀B₂₀(4)/ Ru(0.7)]₁₀ film, respectively. Here, magnetic field was applied in the film plane and along the normal direction of the reflection plane. For reader's understanding, reflectance curves normalized by the magnitude of reflectance of $H = 14$ kOe for the sample (a) are shown with the wavelength of (a-1) 1550 nm, (a-2) 1200, and (a-3) 900 nm, respectively. As shown in (a), in the longer wavelength region, reflectance changes corresponding to the arrangement of magnetizations such as parallel and anti-parallel, whereas in the shorter wavelength region, corresponding to the direction of magnetizations. This means that both R-MRE and transvers Kerr effect (TKE) are appeared in the same sample but different wavelength region due to intra-band and inter-band transition. On the other hand alternating crystalline Co layers with amorphous CoB layers as shown in (b) and (c), R-MRE is completely vanished, though antiferromagnetically interlayer coupling is realized in the all films. This fact clearly shows that crystalline symmetry is necessary condition for MRE due to suppression of

scattering of conduction electrons in the ferromagnetic layer. Magnetic multilayer for GMR film and Ferro./Antiferro. stacked film with Surface Plasmon Resonance will be also introduced in the presentation.

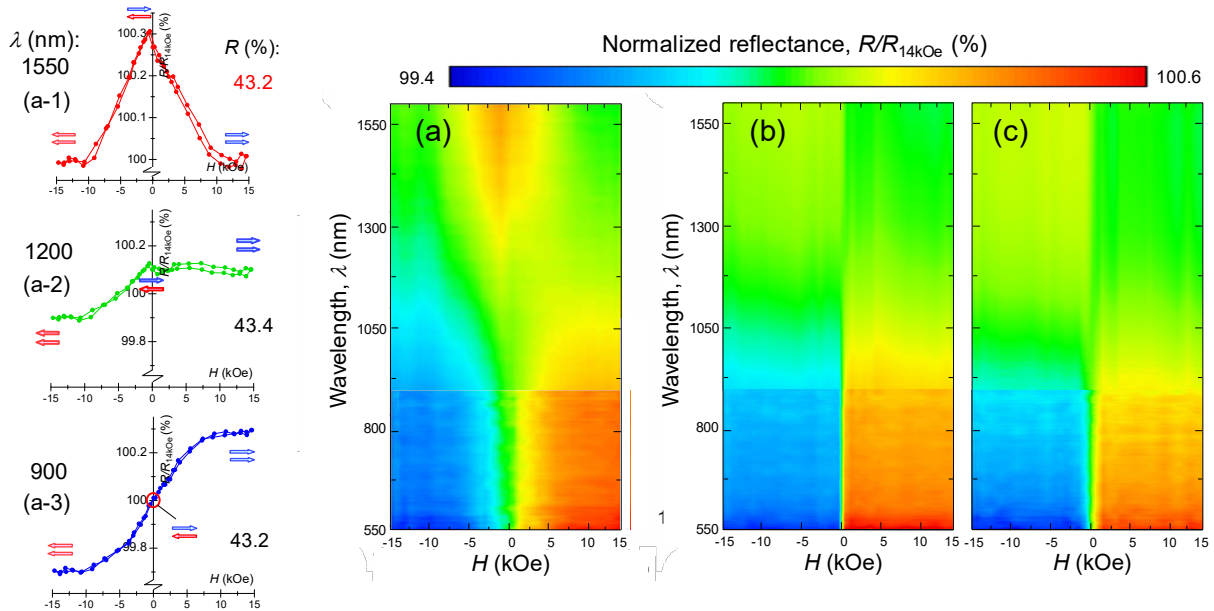


Fig. 1 Color maps of reflectance normalized by the magnitude of reflectance of $H = 14$ kOe plotted against wavelength and magnetic field for $[\text{Co or CoB}(4 \text{ nm})/\text{Ru}(0.7 \text{ nm})]_{10}$ multilayer films for (a) Co, (b) $\text{Co}_{88}\text{B}_{12}$, and (c) $\text{Co}_{80}\text{B}_{20}$, respectively. Reflectance curves for the sample (a) are also shown with the wavelength of (a-1) 1550 nm, (a-2) 1200, and (a-3) 900 nm, respectively.

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Faraday effect of nanogranular films

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Optical isolators utilizing Faraday effect are used in many applications, such as quality assurance of optical amplifier, optical ring laser, optical communication system, in the commercial, industrial and laboratory. They are highly reliable and important tools to support the advanced information society. However, since the discovery of the bismuth garnet (Bi substituted YIG, Bi-YIG) in 1972, no material with Faraday effect beyond Bi-YIG has been found. Furthermore, to respond of miniaturization of the optical devices, thin films with large Faraday effect have been studied, but their properties are inferior to the bulk Bi-YIG. Since new Faraday materials are not discovered, proposals for new optical devices are restricted. On the other hand, we have proposed a new magnetic transparent material of FeCo-fluoride nanogranular films. Nanogranular films consisting of nanometer-sized magnetic metal (Fe, Co or FeCo alloy) granules and a ceramic insulating matrix (nitride, oxide or fluoride) exhibit various functional properties such as high frequency permeability, tunneling magneto resistance, magneto-dielectric effect and magneto-optical effect depending on the composition ratio of granules and matrix. In addition, these films have significant practical advantages (e.g., they are easily fabricated and are thermally stable, and have been applied in magnetic sensors). Here, we introduce the giant Faraday effect of nanogranular films. Faraday rotation angles of these films are 40 times larger than the Bi-YIG at a wavelength of optical communication band (1500 nm)⁽¹⁾.

Thin films were fabricated using a RF tandem sputtering method. With the tandem method, a thin film is obtained by rotating a substrate holder so that the substrate alternately moves over a magnetic metal target and ceramics matrix target. The film composition was adjusted via power input to respective targets. The substrate used was heated quartz glass and heat treatment was performed after the film deposition. Thickness of, thus, obtained film materials was 0.5–1.0 μm . Films composition were determined with a wavelength - dispersive x - ray spectrometer (WDS), and film structure was evaluated using x - ray diffraction (XRD). Faraday effect was evaluated at six wavelengths (405, 515, 650, 830, 1310, and 1550 nm) using Faraday effect measuring equipment (BH - 600LD2M by NEOARK Corp.). Optical transmittance of thin films was measured with a spectrophotometer (UV - 3150 by Shimadzu Corp.); refractive index was determined from results of measurement in a wavelength range of 200–2000 nm using a spectroscopic ellipsometer. Values of optical transmittance and Faraday rotation angle in samples with different thickness were compared by reduction to thickness (optical path length) of 1 μm . All the mentioned evaluation procedures were conducted at room temperature.

Fig.1 shows the wavelength dependence of the Faraday rotation angle of $\text{Fe}_{21}\text{Co}_{14}\text{Y}_{24}\text{F}_{41}$, $\text{Fe}_{25}\text{Y}_{23}\text{F}_{52}$ and $\text{Fe}_{13}\text{Co}_{10}\text{Al}_{22}\text{F}_{55}$ films and bulk Bi-YIG. All the nanogranular films depicted in Fig.1 have much larger absolute Faraday rotation angles than Bi-YIG. The angle in the wavelength of 1310 to 1550 nm, which corresponds to the bands of optical communication, is small in Bi-YIG. In contrast, the nanogranular films exhibit large values in optical communication bands. In this presentation, we will also introduce the recent data.

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Reference

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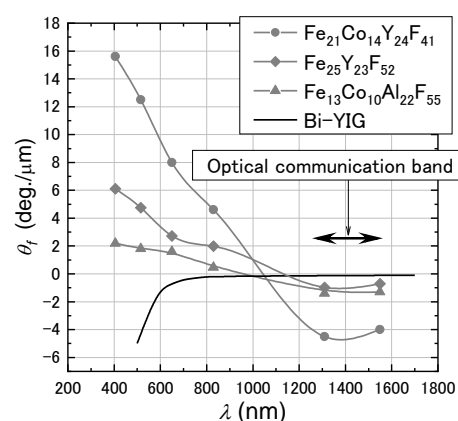


Fig.1 Relationship between the wavelength of incident light and the Faraday rotation angle of $\text{Fe}_{21}\text{Co}_{14}\text{Y}_{24}\text{F}_{41}$, $\text{Fe}_{25}\text{Y}_{23}\text{F}_{52}$ and $\text{Fe}_{13}\text{Co}_{10}\text{Al}_{22}\text{F}_{55}$ films. The films were deposited on substrates of 600 $^{\circ}\text{C}$, 550 $^{\circ}\text{C}$ and 680 $^{\circ}\text{C}$.