

Co-containing Spinel Ferrite Thin-Film Perpendicular Magnetic Recording Media with Mn-Zn Ferrite Backlayer

Setsuo YAMAMOTO^{†a)}, Hirofumi KUNIKI[†], *Regular Members*, Hiroki KURISU[†],
and Mitsuru MATSUURA[†], *Nonmembers*

SUMMARY Co-containing ferrite thin-film/Mn-Zn ferrite thin-film double-layered perpendicular media were prepared using reactive ECR sputtering and magnetron sputtering methods, and their magnetic and structural properties and recording characteristics were studied. The Mn-Zn ferrite thin-film backlayer had saturation magnetization of 3.5 kG and coercivity of 60 Oe. Reproduced voltage for the Co-containing ferrite thin-film/Mn-Zn ferrite thin-film double-layered medium was about twice of that for the Co-containing ferrite single-layer medium.

key words: *perpendicular magnetic recording, ferrite thin-film, recording media, backlayer, Mn-Zn ferrite*

1. Introduction

Ferrite thin-film recording media is promising as high density perpendicular recording media because a protective overcoat layer is not necessary [1], [2]. We have already reported that a Co-containing ferrite thin-film single-layer perpendicular medium prepared by reactive sputtering method utilizing ECR (Electron-Cyclotron-Resonance) microwave plasma without post-oxidation showed high coercivity and perpendicular magnetic anisotropy which are available as perpendicular magnetic recording medium [3]–[6]. The introduction of a soft magnetic layer as a backlayer is effective to improve recording and reproducing performance when media are used in combination with a single-pole head [7].

In this study, Co-containing ferrite thin-film/Mn-Zn ferrite thin-film double-layered perpendicular recording media were prepared and their magnetic and structural properties and recording characteristics were studied following survey of the optimal preparation condition of Mn-Zn ferrite thin-film.

2. Experimental

2.1 Deposition of Mn-Zn Ferrite Thin-Films

Mn-Zn ferrite thin-films used as a soft magnetic underlayer were prepared using an RF magnetron sputtering apparatus. A Mn-Zn ferrite sintered target and pure argon gas were used. RF input power was fixed at 3.1 W/cm² and substrate temperature were varied from

room temperature to 300 degrees Celsius. Mn-Zn ferrite thin-films were deposited directly onto SiO₂/Si substrate or 2.5-in. glass disk substrates (OHARA Corp.: TS-10SX) without seed layers.

2.2 Deposition of Co-containing Ferrite Thin-Films

In the deposition of Co-containing ferrite thin-films, reactive sputtering was employed using an ECR sputtering apparatus (AFTI Corp.: AFTEX-3400U). A cylindrical Fe-Co alloy target with a Co content of 6 at.% was used. Microwave input power, target voltage and substrate temperature were set at 2.4 W/cm², –250 V and 150 degrees Celsius, respectively. The Co-containing ferrite thin-film was deposited directly onto 2.5-in. glass disk substrate (OHARA Corp.: TS-10SX) or the Mn-Zn ferrite thin-film backlayers.

Measurements of magnetic and crystallographic properties and surface morphology of the media were performed using a vibrating sample magnetometer (VSM), X-ray diffraction (XRD (Cu-K α)) and an atomic force microscopy (AFM). Recording characteristics was measured using a contact sliding type MIG head designed for 8 mm VCR.

3. Results and Discussion

3.1 Mn-Zn Ferrite Thin-Films Deposited on SiO₂/Si Substrate

Large saturation magnetization, low coercivity, and high permeability are required for a soft magnetic backlayer.

Figure 1 shows the dependence of magnetic properties of as-deposited Mn-Zn ferrite thin-films on a substrate temperature during RF sputter-deposition. The Mn-Zn ferrite thin-films were directly deposited onto SiO₂/Si substrate without seed layer. In-plane coercivity increased with increasing substrate temperature. Saturation magnetization of the Mn-Zn thin-film prepared at room temperature was 3.5 kG. The saturation magnetization decreased with increasing substrate temperature to 2.5 kG at 300 degree Celsius.

X-ray diffraction diagrams of Mn-Zn ferrite thin-films deposited at various substrate temperature are shown in Fig. 2. In all samples, X-ray diffraction peak

Manuscript received February 3, 2003.

[†]The authors are with the Faculty of Engineering, Yamaguchi University, Ube-shi, 755-8611 Japan.

a) E-mail: yamamoto@yamaguchi-u.ac.jp

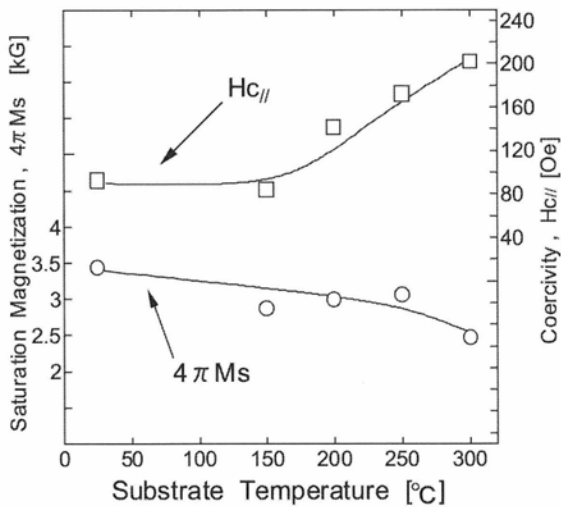


Fig. 1 Substrate temperature dependence of in-plane coercivity and saturation magnetization of Mn-Zn ferrite thin-films deposited onto SiO₂/Si substrate.

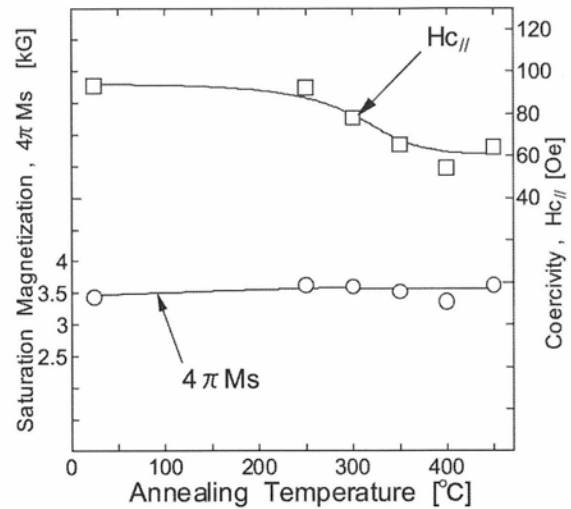


Fig. 3 Annealing temperature dependence of in-plane coercivity and saturation magnetization of Mn-Zn ferrite thin-films.

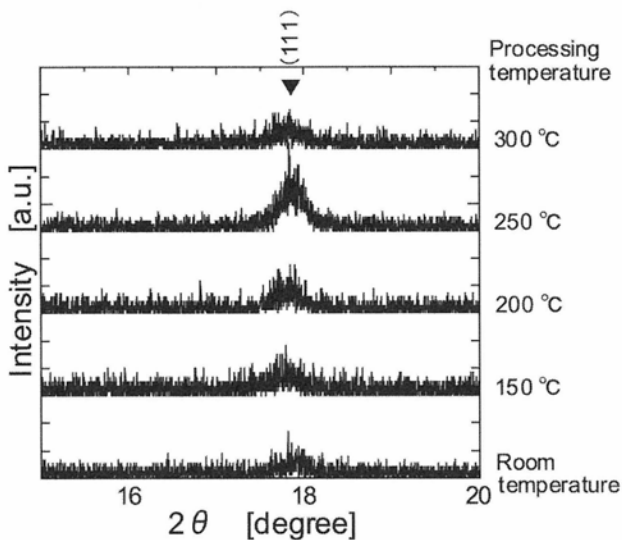


Fig. 2 X-ray diffraction diagrams of Mn-Zn ferrite thin-films deposited onto SiO₂/Si substrate at various temperatures.

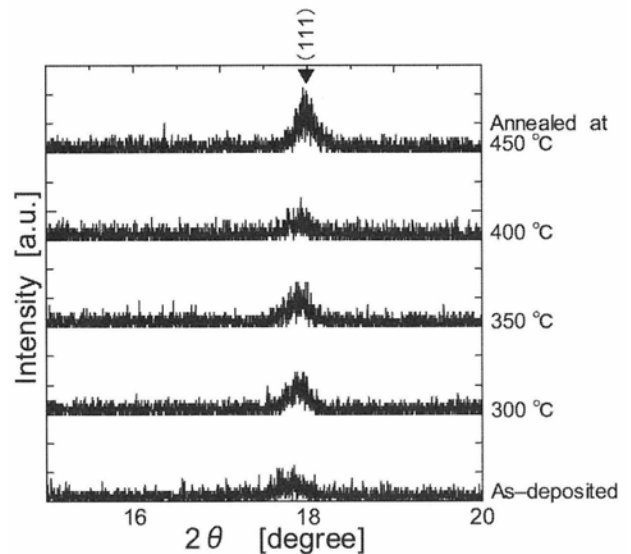


Fig. 4 X-ray diffraction diagrams of Mn-Zn ferrite thin-films post-annealed at various temperatures.

only from spinel ferrite (111) plane was observed. The highest amplitude of the diffraction peak was observed for the Mn-Zn ferrite thin-film deposited at 250 degrees Celsius with suggesting good crystal orientation and good crystallinity.

Post-annealing effect was investigated. The Mn-Zn ferrite thin-films deposited onto SiO₂/Si substrate were heated up, and kept at temperatures from 250 to 450 degrees Celsius for 60 minutes in air. Annealing temperature dependence of magnetic properties of the Mn-Zn ferrite thin-films is shown in Fig. 3. In-plane coercivity gradually decreased at annealing temperatures higher than 250 degrees Celsius. Coercivity of the film annealed at 350 degree Celsius showed the lowest value

of 60 Oe. No remarkable change in saturation magnetization was not observed. It was supposed that the decrease in coercivity after annealing was due to the relaxation of internal stress accumulated during film deposition by being packed with oxygen atoms in vacancy.

Figure 4 shows the X-ray diffraction diagrams of the Mn-Zn ferrite thin-films post-annealed at various temperature. The peak intensity from (111) plane of the Mn-Zn ferrite thin-films annealed at over 300 degrees Celsius became higher than that for the as-deposited sample. These results suggest that the crystallinity of the ferrite thin-films annealed at over 300 degrees Celsius was improved by post-annealing.

Figure 5 shows the AFM surface images of the

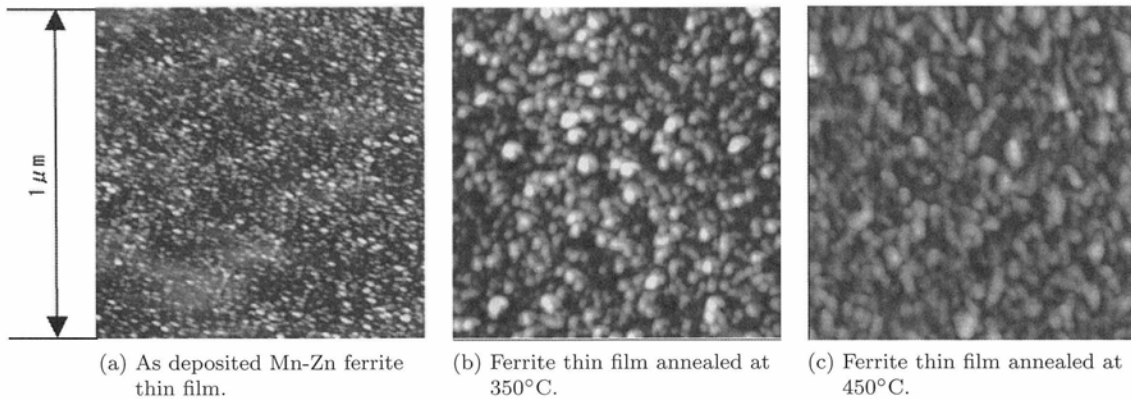


Fig. 5 Surface AFM images of Mn-Zn ferrite thin-films deposited on glass substrate.

Table 1 Magnetic properties, grain size, R_a and R_{max} of Mn-Zn ferrite thin-films.

	Saturation magnetization $4\pi M_s$ [kG]	Coercivity H_{c1} [Oe]	Grain size [nm]	R_a [nm]	R_{max} [nm]	Relative permeability μ_i
As-deposited Mn-Zn ferrite thin-film	3.49	109	16.9	0.38	3.09	1
Ferrite thin-film annealed at 350 °C	3.43	60	34.5	1.10	6.03	11
Ferrite thin-film annealed at 450 °C	3.53	59	70.3	1.97	13.8	8

200 nm thick Mn-Zn ferrite thin-films on glass substrate (OHARA Corp.: TS-10SX). The as-deposited film has a small grain size of 16.9 nm and a smooth surface roughness (R_a) of 0.38 nm. However, the Mn-Zn ferrite thin-film annealed at 350 degrees Celsius had a relatively large grain size of 34.5 nm and large R_a of 1.10 nm. The thin-film annealed at 450 degrees Celsius showed a larger grain size of 70.3 nm and R_a of 1.97 nm. At high annealing temperatures, the grains in Mn-Zn ferrite thin-film grew resulting in considerable increase in the surface roughness.

From the view point of application to magnetic recording media, there is a trade-off relationship between soft magnetic properties and smooth surface with respect to annealing treatment as seen in Figs. 4 and 5. We chose the annealing temperature of 350 degree Celsius as optimum condition because coercivity is substantially decreased without large sacrifice of smooth surface and small grain.

Thus far, Mn-Zn ferrite thin-film were deposited on SiO_2/Si substrate prior to survey the optimal preparation condition. Based on these results, deposition of Mn-Zn ferrite thin-films was carried out on glass substrate in the following study.

3.2 Mn-Zn Ferrite Thin-Films Deposited on Glass Substrate

Table 1 shows the magnetic properties, grain size and surface roughness (R_a and R_{max}) of Mn-Zn ferrite thin-film deposited on glass substrate for recording disk. As can be seen from Table 1, saturation magnetization, in-

plane coercivity and relative initial permeability, μ_i , of as-deposited film were 3.49 kG, 109 Oe and 1. The in-plane coercivity decreased from 109 to 60 and to 59 Oe and initial permeability, μ_i , increased from 1 to 11 and to 8 by annealing in air at 350 and 450 degrees Celsius, respectively. Although remarkable changes in saturation magnetization were not observed, coercivity was decreased by post-annealing treatment.

The X-ray diffraction diagrams of the Mn-Zn ferrite thin-films deposited on glass substrate before and after post-annealing are shown in Fig. 6. All the films showed the preferential orientation of (111), and that the crystallinity was improved by post-annealing.

These results shown in Table 1 and Fig. 6 are consistent with the experimental results described above with respect to the Mn-Zn ferrite thin-films deposited on SiO_2/Si substrate.

Post-annealing time dependence of magnetic properties of the Mn-Zn ferrite thin-films deposited onto glass substrate are shown in Fig. 7. Annealing temperature was fixed at 350 degrees Celsius. In-plane coercivity decreased from 109 Oe to the steady value of about 60 Oe after 60 minutes annealing.

Figure 8 shows the X-ray diffraction diagrams for the Mn-Zn ferrite thin-films as-deposited and annealed for various annealing time from 20 to 90 minutes. The diffraction peak intensity from (111) plane of the Mn-Zn ferrite thin-films showed an increased value when samples were annealed within 60 minutes.

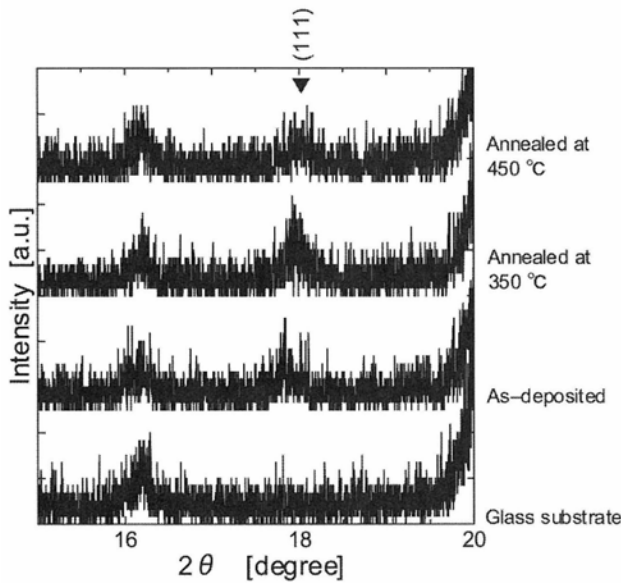


Fig. 6 X-ray diffraction diagrams of Mn-Zn ferrite thin-films deposited onto glass substrate.

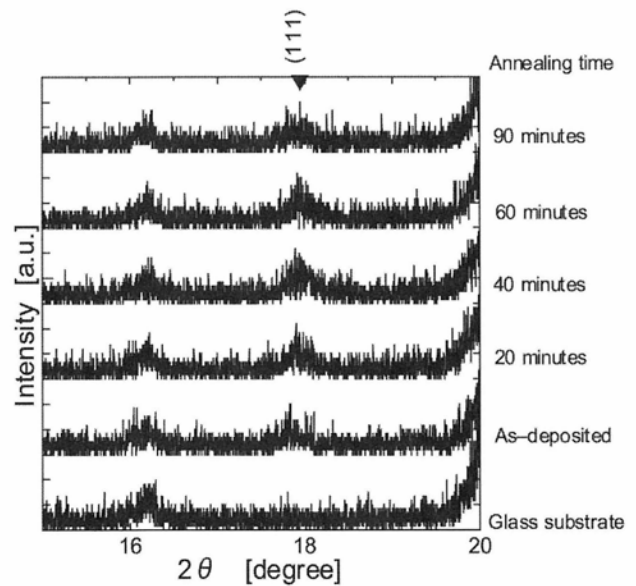


Fig. 8 X-ray diffraction diagrams of Mn-Zn ferrite thin-films deposited onto glass substrate and post-annealed. (annealed at 350°C)

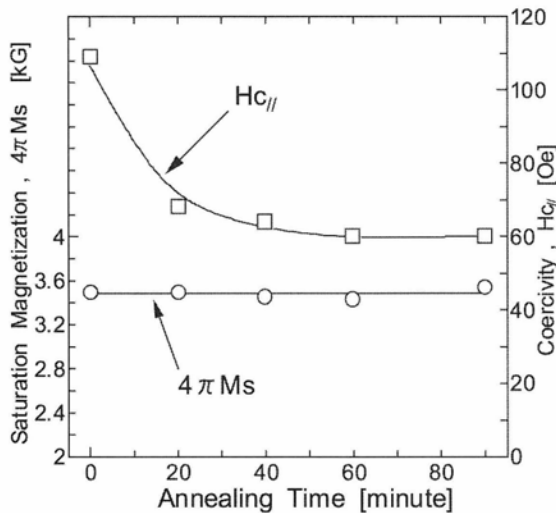


Fig. 7 Post-annealing time dependence of in-plane coercivity and saturation magnetization of Mn-Zn ferrite thin-films deposited onto glass substrate.

3.3 Co-containing Spinel Thin-Film/Mn-Zn Ferrite Thin-Film Double-Layer

Based on the experimental results described above, Mn-Zn spinel ferrite thin-film was deposited on a 2.5 inch glass substrate by RF magnetron sputtering and annealed in air at 350 degrees Celsius for 60 minutes to reduce coercivity. After that, a 55 nm thick Co-containing spinel ferrite thin-film was deposited on the Mn-Zn ferrite thin-film by reactive ECR sputtering. Co-containing spinel substrate as a reference exhibited saturation magnetization of 2.9 kG, perpendicular

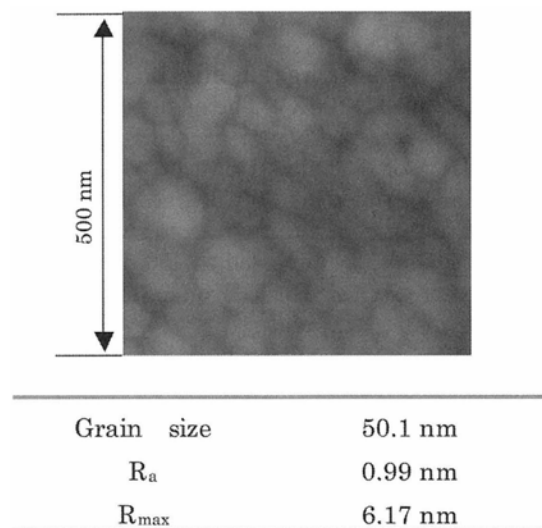


Fig. 9 AFM surface image of Co-containing ferrite thin-film/Mn-Zn ferrite thin-film double-layer medium.

lar coercivity of 2.8 kOe, and perpendicular anisotropy. Figure 9 shows the AFM surface image of the Co-containing ferrite thin-film/the Mn-Zn ferrite thin-film (annealed at 350 degrees Celsius) double-layered medium.

The Co-containing ferrite thin-film single-layer film had a small grain size of 14.9 nm and a smooth surface R_a of 0.60 nm. The double-layered film exhibited a grain size of 50.1 nm and R_a of 0.99 nm. These values were larger than that for Co-containing ferrite thin-film single-layer. However, their increase was kept at a minimum by setting the annealing temperature of the Mn-Zn ferrite backlayer to 350 degrees Celsius.

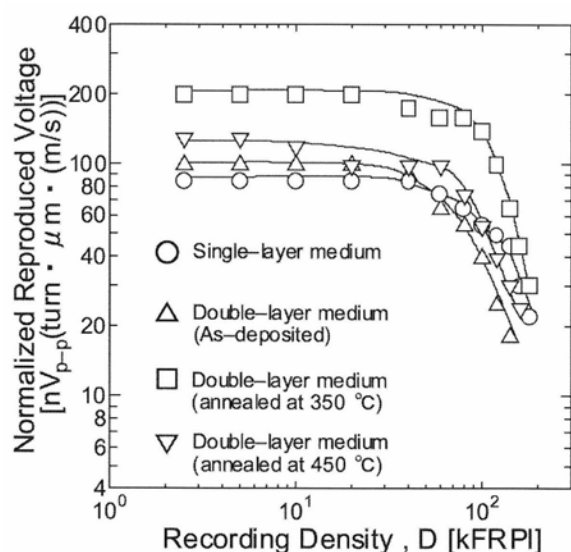


Fig. 10 Roll-off curves for Co-containing ferrite thin-film single-layer medium and Co-containing ferrite thin-film/Mn-Zn ferrite thin-film double-layer media.

3.4 Recording Characteristics of Co-containing Ferrite Thin-Film/Mn-Zn Ferrite Thin-Film Double-Layered Media

Recording and reproducing experiment was performed on the Co-containing ferrite thin-film/Mn-Zn ferrite thin-film double-layered perpendicular recording media and the Co-containing ferrite thin-film single-layer medium as a reference. Figure 10 shows the roll-off curves for these media. A MIG head with a gap length of about 200 nm was employed in contact sliding operation. The D_{50} values were 150, 90, 120 and 90 kFRPI for the single-layer medium and double-layered media as-deposited and annealed at 350 and 450 degrees Celsius, respectively. The reproduced voltages for the double-layered media in which the Mn-Zn ferrite thin-film was not post-annealed or annealed at 450 degrees Celsius were only slightly higher than that of the Co-containing ferrite thin-film single-layer medium. This was due to the insufficient soft magnetic properties for the former medium and large surface roughness for the latter medium. The reproduced voltage for the double-layered medium in which Mn-Zn ferrite thin-film was annealed at 350 degrees Celsius was approximately double the amplitude of the Co-containing ferrite thin-film single-layer medium.

4. Conclusions

The following results were obtained.

(a) The as-deposited Mn-Zn ferrite thin-films prepared by RF sputtering method with the Mn-Zn ferrite sintered target exhibited (111) spinel preferential orientation.

(b) Post-oxidation in air at 350 degrees Celsius was effective to decrease in-plane coercivity of the Mn-Zn ferrite thin-films to about 60 Oe without causing severe increase in grain size and surface roughness.

(c) The reproduced voltage for the Co-containing ferrite thin-film/Mn-Zn ferrite thin-film double-layered medium was about double of that for the Co-containing ferrite thin-film single-layer medium.

The optimal preparation of the Mn-Zn ferrite thin-film backlayer and the advantage of ferrite thin-film double-layered perpendicular recording media over the single layer medium was proved in this study.

References

- [1] S. Yamamoto, T. Andou, H. Kurisu, M. Matsuura, T. Doi, and K. Tamari, "Recording characteristics of Co- γ Fe₂O₃ perpendicular magnetic recording media," *J. Appl. Phys.*, vol.79, no.8, Part 2, pp.4884-4886, 1996.
- [2] K. Tamari, T. Doi, and N. Horiishi, "Magnetic and magneto-optical properties of the Co- γ Fe₂O₃ perpendicular magnetic films," *Appl. Phys. Lett.*, vol.63, no.23, pp.3227-3229, 1993.
- [3] S. Yamamoto, T. Santoki, H. Kurisu, M. Matsuura, J. Kojima, K. Nakata, Y. Kakihara, T. Doi, and K. Tamari, "Co- γ Fe₂O₃ thin film disks fabricated by plasma oxidation," *J. Appl. Phys.*, vol.22, no.S3, pp.23-25, 1998.
- [4] S. Yamamoto, H. Kurisu, M. Matsuura, T. Doi, and K. Tamari, "Co- γ Fe₂O₃/NiO thin-film media for perpendicular and longitudinal magnetic recording," *J. Magn. Soc. Jpn.*, vol.23, no.5, pp.1739-1745, 1999.
- [5] K. Hirata, S. Yamamoto, H. Kurisu, M. Matsuura, T. Doi, and K. Tamari, "Co-containing ferrite thin-film disk prepared by reactive sputtering method," *J. Jpn. Soc. Powder Powder Metall.*, vol.48, no.8, pp.742-747, 2001.
- [6] S. Yamamoto, K. Hirata, H. Kurisu, M. Matsuura, T. Doi, and K. Tamari, "Preparation and magnetic properties of ferrite thin-film media," *IEICE Trans. Electron.*, vol.E84-C, no.9, pp.1142-1146, Sept. 2001.
- [7] S. Iwasaki, Y. Nakamura, and K. Ouchi, "Perpendicular magnetic recording with a composite anisotropy film," *IEEE Trans. Magn.*, vol.MAG-15, no.6, pp.1456-1458, 1979.



Setsuo Yamamoto received the B.S. degree in physics, the M.S. degrees and Ph.D. degree in applied physics from Tohoku University in 1980, 1982 and 1985, respectively. In 1985, he joined Tohoku University as a research associate of Research Institute of Electrical Communication, where he engaged in research and development on perpendicular magnetic recording. Since 1988, he has been an associate professor of department of Advanced Materials Science and Engineering, Faculty of Engineering, Yamaguchi University. His current research interest includes R&M on magnetic materials, micro magnetic devices, and high density information storage technology. Dr. Yamamoto is a member of IEEE Magnetic Society, the Institute of Electrical Engineering of Japan, the Magnetics Society of Japan, the Materials Research Society of Japan (MRS-J), the Japan Society of Powder and Powder Metallurgy, and Committee 144 (Magnetic Recording) of Japan Society for the Promotion of Science.



Hirofumi Kuniki received B.E. degree in Advanced Materials Science and Engineering from Yamaguchi University, Japan in 2002. He is now on the graduate school of Yamaguchi University where he engaged in research on perpendicular magnetic recording.



Hiroki Kurisu received the B.Ed. in Physics from Nara University of Education in 1987 and D.Sc. from Osaka City University in 1993. Shortly after that, he joined Faculty of Engineering, Yamaguchi University as a Research Associate. His current interest is Electronic and Optical Properties of Condensed Matter, especially Dynamical Process in Mesoscopic Materials.



Mitsuru Matsuura received the D.Sc. from Tohoku University in 1970. Then he joined Department of Physics, Faculty of Science, Waterloo University in Canada, and also joined National Research Council of Canada as a research fellow. He was an associate professor in Faculty of Engineering, Yamaguchi University in 1975. He's been Professor in Faculty of Engineering, Yamaguchi University since 1981. He's majored in Quantum Theory of Condensed Matter, Electronic Properties of Materials, especially in Semiconductors.

He's majored in Quantum Theory of Condensed Matter, Electronic Properties of Materials, especially in Semiconductors.