# **Journal of the Magnetics Society of Japan**

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# **Journal**

# **Magnetic Recording**

Design Parameter and Anisotropy Constant Ratio for Heat-Assisted Magnetic Recording

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# **Hard and Soft Magnetic Materials**

The hysteresis properties analysis of Fe-4 wt% Si prepared by high purity metallurgy

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# Thin Films, Fine Particles, Multilayers, and Superlattices

Structure and Magnetic Properties of (001) Oriented CoPt-Ag and CoPd-Ag Alloy Films

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学術講演会論文特集号発行に伴うタイトル募集について

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# Design Parameter and Anisotropy Constant Ratio for Heat-Assisted Magnetic Recording

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A design guideline for heat-assisted magnetic recording (HAMR) is presented with a view to obtaining a minimized anisotropy constant ratio  $K_{\rm u}$  /  $K_{\rm bulk}$  for media by using a model calculation. We investigated the relationship between the design parameters and the limiting factors of HAMR design. The parameters, namely the writing temperature  $T_{\rm w}$  and recording layer thickness h, are related to the medium thermal stability factor of the limiting factor. In addition, inter-grain exchange coupling J is related to the statistical thermal stability factor. HAMR combined with shingled magnetic recording (SHAMR) has the advantage of reducing the medium thermal gradient, and increasing  $T_{\rm w}$  is also effective in increasing the heat-transfer thermal gradient of the limiting factor. The combination of increasing h and SHAMR or increasing h and SHAMR can successfully reduce h0 / h1 / h2 / h3 / h3 / h4 / h4 / h5 / h4 / h5 / h5 / h5 / h6 / h6 / h6 / h6 / h6 / h6 / h7 / h8 / h8 / h8 / h9 /

Key words: heat-assisted magnetic recording, media design, anisotropy constant ratio, thermal stability factor, thermal gradient

#### 1. Introduction

Heat-assisted magnetic recording (HAMR) has been proposed with the aim of solving the trilemma problem<sup>1)</sup> of magnetic recording. HAMR is a recording method in which the medium is heated to reduce the coercivity at the time of writing.

We have already reported the design method for HAMR<sup>2, 3)</sup>. We have introduced a new parameter, the anisotropy constant ratio  $K_{\mathrm{u}}/K_{\mathrm{bulk}}$  , which is the intrinsic ratio of film anisotropy constant  $K_{\mathrm{u}}$  to bulk  $K_{\mathrm{u}}$  <sup>2)</sup>. If the  $K_{\mathrm{u}}/K_{\mathrm{bulk}}$  values for two media are identical, the difficulties involved in preparing those media are the same, even if the  $K_{\mu}$  values are different. On the other hand, increasing  $K_{\mu}/K_{\text{bulk}}$  is a challenging task. Therefore, it is necessary to design a medium with a smaller  $K_{\mathrm{u}}/K_{\mathrm{bulk}}$  . The many relationships between design parameters and  $K_{\rm u}/K_{\rm bulk}$ have been revealed<sup>3)</sup>. We have reported the dependence of  $K_{\rm u}/K_{\rm bulk}$  on one parameter change. As a result, we find that increasing the writing temperature  $T_{\rm w}$  is only effective for reducing  $K_{\rm u}/K_{\rm bulk}$  when we compare  $K_{\rm u}/K_{\rm bulk}$  value calculated using standard parameter values. Although the combination of more than two parameters may be expected to reduce  $K_{\rm u}/K_{\rm bulk}$ , it is difficult to examine all parameter combinations. Our method is a model calculation, and it is easy to understand the physical meanings of the parameters.

In this study, we investigate the relationship between the design parameters and limiting factors of HAMR design with the aim of reducing  $K_{\rm u}/K_{\rm bulk}$ , and we provide the examination results of  $K_{\rm u}/K_{\rm bulk}$  for the combination of more than two parameters.

## 2. Media Design Method

From the following four conditions, we estimated

whether or not the media have the potential for HAMR. That is, information stability (1) during 10 years of archiving, (2) on the trailing side located 1 bit from the writing position during writing, (3) in adjacent tracks during rewriting, and (4) under the main pole during rewriting<sup>2, 3)</sup>.

(1) The first condition, namely the information stability during 10 years of archiving, is expressed as

$$\frac{K_{\rm u}(T_{\rm a})V_{\rm m}}{kT_{\rm a}} \ge \text{TSF}_{10},\tag{1}$$

where  $K_{\rm u}(T_{\rm a})V_{\rm m}/kT_{\rm a}$  is a medium thermal stability factor,  $T_{\rm a}$  is the ambient temperature (assumed to be 330 K), which is the maximum working temperature of the hard drive,  $V_{\rm m}$  is the grain volume for the mean grain size, k is the Boltzmann constant, and  ${\rm TSF_{10}}$  is the statistical thermal stability factor during 10 years of archiving.

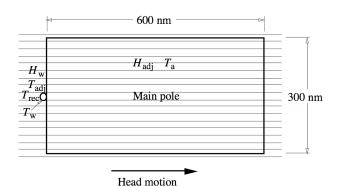


Fig. 1 Schematic illustration of writing-head configuration.

Figure 1 shows the writing-head configuration. It is assumed that the main-pole size of the head is 600 nm (down-track direction) × 300 nm (cross-track

direction), and the writing position is located on the trailing side of the main pole. The maximum temperature under the main pole is  $T_{\rm a}$ .

(2) The second condition, namely the information stability on the trailing side during writing, is expressed as

$$\frac{\Delta T}{\Delta x} = \frac{T_{\rm w} - T_{\rm rec}}{\Delta x} \le \frac{\partial T}{\partial x},$$
 (2)

where  $\Delta T/\Delta x$  is the medium thermal gradient for the down-track direction,  $T_{\rm w}$  is the writing temperature, and  $T_{\rm rec}$  is the maximum temperature at which the information on the trailing side can be held during writing as shown in Fig. 1.  $\Delta x$  is the distance from the position of  $T_{\rm w}$  to that of  $T_{\rm rec}$ , and  $\partial T/\partial x$  is the heat-transfer thermal gradient for the down-track direction.

(3) The third condition, namely the information stability in adjacent tracks during rewriting, is expressed as

$$\frac{\Delta T}{\Delta y} = \frac{T_{\rm w} - T_{\rm adj}}{\Delta y} \le \frac{\partial T}{\partial y},\tag{3}$$

where  $\Delta T/\Delta y$  is the medium thermal gradient for the cross-track direction, and  $T_{\rm adj}$  is the maximum temperature at which the information in adjacent tracks can be held during rewriting as shown in Fig. 1.  $\Delta y$  is the distance from the position of  $T_{\rm w}$  to that of  $T_{\rm adj}$ , and  $\partial T/\partial y$  is the heat-transfer thermal gradient for the cross-track direction.  $\Delta T/\Delta x$  and  $\Delta T/\Delta y$  are the minimum thermal gradients required by the medium for information stability.  $\partial T/\partial x$  and  $\partial T/\partial y$  are the thermal gradients calculated by a heat-transfer simulation<sup>3)</sup>.

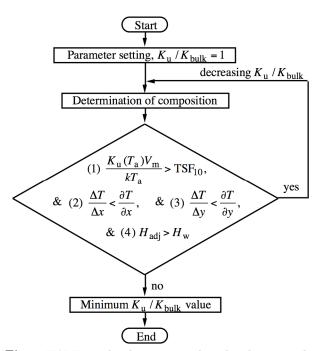
(4) The fourth condition, namely the information stability under the main pole during rewriting, is expressed as

$$H_{\rm adj} \ge H_{\rm w},$$
 (4)

where  $H_{\rm adj}$  is the maximum head field that can hold the information under the main pole, and  $H_{\rm w}$  is the writing field at the position of  $T_{\rm w}$  as shown in Fig. 1.

The details have been reported elsewhere<sup>3)</sup>.

The HAMR media were designed to obtain the minimum  $K_{\rm u}/K_{\rm bulk}$  value using the procedure shown in Fig. 2<sup>3)</sup>. First, the design parameters and  $K_{\rm u}/K_{\rm bulk}$  = 1 were set. Four HAMR conditions (1), (2), (3), and (4) were examined after determining the composition of the recording layer. If there are some margins for all four conditions,  $K_{\rm u}/K_{\rm bulk}$  can be reduced. When one of the four conditions reaches the limit, the minimum  $K_{\rm u}/K_{\rm bulk}$  value can be determined. That condition becomes a limiting factor.



**Fig. 2** HAMR media design procedure for obtaining the minimum  $K_{\rm u}/K_{\rm bulk}$  value<sup>3)</sup>.

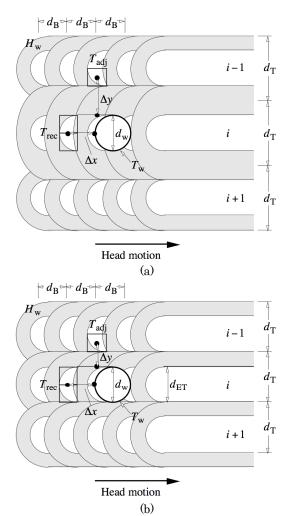
The medium was assumed to be granular. The standard parameter values of the media design are summarized in Table 1. The bit area S is fixed, and Sis the product  $d_{\rm B} \times d_{\rm T}$  of the bit pitch  $d_{\rm B}$  and the track pitch  $d_{\mathrm{T}}$ . The method for determining  $d_{\mathrm{T}}/d_{\mathrm{B}}$ was reported in a previous paper2). The arrangement of the grains is not considered. The effective track width  $d_{\rm FT}$  has been explained in a previous paper<sup>3)</sup>. The standard medium consists of four layers, that is, a recording layer RL (Fe-Pt base, thickness h = 8 nm), interlayer 1 IL1 (MgO base, 5 nm), interlayer 2 IL2 (Cr base, 10 nm), and a heat-sink layer HSL (Cu base, 30 nm).  $D_{\rm m}$  is the mean grain size, and  $V_{\rm m} = D_{\rm m}^{2} \times h$ . The inter-grain exchange coupling  $J^{(4)}$  is a newly introduced design parameter. A comparison of magnetic recording (MR) method was carried out of HAMR and HAMR combined with shingled magnetic recording (SHAMR). The light-spot diameter  $d_{\mathrm{L}}$  and the heat-spot diameter  $d_{\rm w}$  have also been defined in previous papers<sup>2, 3)</sup>.

Figure 3 (a) is a schematic illustration of the area near the writing position for HAMR. The writing field  $H_{\rm w}$  is applied to a wide area including the writing position. The circle denoted by  $T_{\rm w}$  is an isotherm of  $T_{\rm w}$ , and  $d_{\rm w}$  is the heat-spot diameter. The white regions indicate upward or downward magnetization, and the gray regions indicate the magnetization transition. The transition region spreads to adjacent tracks as a result of rewriting operations on the ith track.

On the other hand, the maximum rewriting number for SHAMR is considerably smaller than that for HAMR since the tracks are laid to overlap each other as shown in Fig. 3 (b).

Table 1 Standard parameter values of media design.

User areal density (Tbpsi)	4
Bit area $S(nm^2)$	140
Effective track width $d_{\rm ET}$ (nm)	10
Ambient temperature $T_a$ (K)	330
Writing temperature $T_{\rm w}$ (K)	500
Grain number per bit n (grain / bit)	4
RL thickness h (nm)	8
Standard deviation of grain size $\sigma_{\rm D}/D_{\rm m}$ (%)	10
Inter - grain exchange coupling $J$ (erg/cm <sup>2</sup> )	0
MR method	HAMR
Light-spot diameter $d_{\rm L}$ (nm)	9.0
Heat-spot diameter $d_{\rm w}$ (nm)	10
Linear velocity $v$ (m/s)	10
Thermal conductivity of IL1 $K$ (W /(cmK))	0.5



**Fig. 3** Schematic illustrations of writing position for (a) HAMR and (b) SHAMR.

## 3. Design Parameters and Limiting Factors

We have reported the dependence of  $K_{\rm u}/K_{\rm bulk}$  on one parameter change<sup>3)</sup>. As a result, increasing  $T_{\rm w}$  is only effective for reducing  $K_{\rm u}/K_{\rm bulk}$  when we compare

the  $K_{\rm u}/K_{\rm bulk}$  value calculated using standard parameter values. Increasing J from 0 to 0.19 erg/cm² has little effect. Although the combination of more than two parameters may be expected to reduce  $K_{\rm u}/K_{\rm bulk}$ , it is difficult to examine all parameter combinations. Therefore, we investigated the relationship between the parameters and the limiting factors. Conditions (2) and (3) can be combined as

$$\frac{\Delta T}{\Delta x} = \frac{\Delta T}{\Delta y} \le \frac{\partial T}{\partial x} = \frac{\partial T}{\partial y},\tag{5}$$

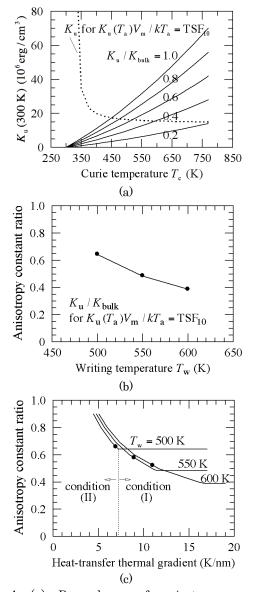
since  $\partial T/\partial x \approx \partial T/\partial y^2$ . Condition (4) has some margins for all the cases we examined. Therefore, the major limiting factors in the media design are condition (I) given by Eq. (1)  $(K_{\rm u}(T_{\rm a})V_{\rm m}/kT_{\rm a} \geq {\rm TSF_{10}})$  and condition (II) given by Eq. (5) (hereafter, shown as  $\Delta T/\Delta x = \Delta T/\Delta y \equiv \Delta T/\Delta x(y)$ ,  $\partial T/\partial x = \partial T/\partial y \equiv \partial T/\partial x(y)$ , and  $\Delta T/\Delta x(y) \leq \partial T/\partial x(y)$ ).

# 3.1 Condition (I) - increasing $K_{\rm u}(T_{\rm a})V_{\rm m}/kT_{\rm a}$

 $K_{\rm u}(T_{\rm a})V_{\rm m}/kT_{\rm a}$  must be increased or  ${\rm TSF_{10}}$  decreased to relax condition (I) given by Eq. (1)  $K_{\rm u}(T_{\rm a})V_{\rm m}/kT_{\rm a} \geq {\rm TSF_{10}}$ . First, we discuss the parameter for increasing  $K_{\rm u}(T_{\rm a})$ .

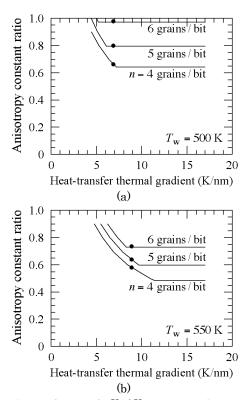
Figure 4 (a) shows the dependence of  $K_{\rm u}(300~{\rm K})$  on Curie temperature  $T_{\rm c}$ , in which the calculation parameter is  $K_{\rm u}/K_{\rm bulk}$ . The dotted line shows the  $K_{\rm u}(300~{\rm K})$  value that satisfies  $K_{\rm u}(T_{\rm a})V_{\rm m}/kT_{\rm a}={\rm TSF}_{10}$ . Although the  $K_{\rm u}(T_{\rm a})$  value that satisfies  $K_{\rm u}(T_{\rm a})V_{\rm m}/kT_{\rm a}={\rm TSF}_{10}$  is independent of  $T_{\rm c}$ , the  $K_{\rm u}(300~{\rm K})$  value that satisfies  $K_{\rm u}(T_{\rm a})V_{\rm m}/kT_{\rm a}={\rm TSF}_{10}$  increases as  $T_{\rm c}$  decreases, and it diverges at  $T_{\rm a}=330~{\rm K}$ . The intersections of the solid lines and the dotted line show the  $K_{\rm u}/K_{\rm bulk}$  dependence that satisfies  $K_{\rm u}(T_{\rm a})V_{\rm m}/kT_{\rm a}={\rm TSF}_{10}$  on  $T_{\rm c}$ . Therefore, the required  $K_{\rm u}/K_{\rm bulk}$  can be reduced by increasing  $T_{\rm c}$ , and then,  $K_{\rm u}/K_{\rm bulk}$  can be reduced by increasing  $T_{\rm w}$  as shown in Fig. 4 (b). In other words, if  $K_{\rm u}/K_{\rm bulk}$  is constant, increasing  $T_{\rm w}$  is effective for increasing  $K_{\rm u}(T_{\rm a})$ . If  $T_{\rm w}$  is low, the extra  $K_{\rm u}/K_{\rm bulk}$  is needed to realize HAMR.

is low, the extra  $K_{\rm u}$  / $K_{\rm bulk}$  is needed to realize HAMR. Furthermore,  $K_{\rm u}$  / $K_{\rm bulk}$  is a function of the heat-transfer thermal gradient  $\partial T/\partial x(y)$  as shown in Fig. 4 (c). As  $\partial T/\partial x(y)$  increases,  $K_{\mu}/K_{\text{bulk}}$  first becomes lower, and then becomes constant with respect to  $\partial T/\partial x(y)$ . In the range where  $K_u/K_{\text{bulk}}$  changes, the limiting factor is condition (II)  $\Delta T / \Delta x(y)$  $\leq \partial T / \partial x(y)$ . And it is condition (I)  $K_{u}(T_{a})V_{m}/kT_{a}$  $\geq$  TSF<sub>10</sub> in the range where  $K_{\rm u}/K_{\rm bulk}$  is constant. The constant  $K_{\rm u}/K_{\rm bulk}$  value in Fig. 4 (c) corresponds to that in Fig. 4 (b). Although increasing  $T_{\rm w}$  is effective for reducing  $K_u / K_{bulk}$  (Fig. 4 (b)), a higher  $\partial T / \partial x(y)$ is needed to obtain a lower  $K_{\rm u}/K_{\rm bulk}$  (Fig. 4 (c)). The closed circles show the  $K_{\mathrm{u}}$  / $K_{\mathrm{bulk}}$  values and their  $\partial T/\partial x(y)$  values calculated for the standard medium structure and the standard thermal constants2). Increasing  $T_{\rm w}$  is effective for reducing  $K_{\rm u}/K_{\rm bulk}$  in consideration of calculated  $\partial T / \partial x(y)$ .

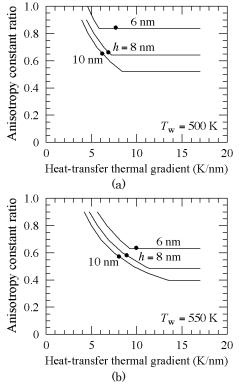


**Fig. 4** (a) Dependence of anisotropy constant  $K_{\rm u}(300~{\rm K})$  on Curie temperature  $T_{\rm c}$ , (b) dependence of anisotropy constant ratio  $K_{\rm u}/K_{\rm bulk}$  on writing temperature  $T_{\rm w}$ , and (c) dependence of  $K_{\rm u}/K_{\rm bulk}$  on heat-transfer thermal gradient  $\partial T/\partial x(y)$ .

Next, we discuss the parameter for increasing  $V_{\rm m}$ . One way of increasing  $V_{\rm m} = D_{\rm m}^2 \times h$  is to reduce the grain number per bit n. Although  $TSF_{10}$  becomes larger as n becomes smaller<sup>4</sup>,  $D_{\rm m} = \sqrt{S/n} - \Delta$ increases as n decreases where S is the bit area and  $\Delta = 1$  nm is the non-magnetic spacing between grains (e.g.,  $D_{\rm m}$  = 4.92 nm for n = 4). The solid line, in the range where  $K_{\mathrm{u}}/K_{\mathrm{bulk}}$  is constant in Fig. 5, becomes noticeably low as n decreases. In that range, the limiting factor is condition (I) as mentioned above. Therefore, reducing n relaxes condition (I) through increasing  $V_{\mathrm{m}}$ . Furthermore, the closed circles can also be reduced as n decreases. Therefore, reducing n is effective for reducing  $K_{\rm u}/K_{\rm bulk}$ . When we compare (a)  $T_{\rm w}$  = 500 K and (b) 550 K, the n dependence of the  $K_{\mu}/K_{\rm bulk}$  values of the closed circles is reduced by increasing  $T_{\rm w}$  since the limiting factor becomes condition (II).



**Fig. 5** Dependence of  $K_{\rm u}/K_{\rm bulk}$  on  $\partial T/\partial x(y)$  at (a)  $T_{\rm w}=500~{\rm K}$  and (b) 550 K (calculation parameter: grain number per bit n).



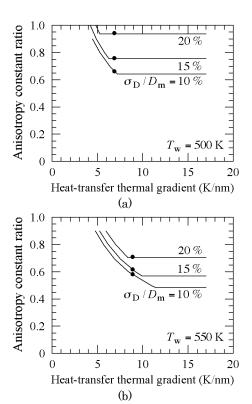
**Fig. 6** Dependence of  $K_{\rm u}/K_{\rm bulk}$  on  $\partial T/\partial x(y)$  at (a)  $T_{\rm w}=500~{\rm K}$  and (b) 550 K (calculation parameter: RL thickness h).

Another way of increasing  $V_{\rm m}=D_{\rm m}^{-2}\times h$  is to increase the RL thickness h. It can also be seen from the solid lines where  $K_{\rm u}/K_{\rm bulk}$  is constant in Fig. 6 that increasing h relaxes condition (I). However, when h increases from the standard value of 8 to 10 nm, the  $K_{\rm u}/K_{\rm bulk}$  values of the closed circles are almost the same for both (a)  $T_{\rm w}=500$  K and (b) 550 K since the limiting factor is condition (II), and  $\partial T/\partial x(y)$  is decreased by increasing h due to the adiabatic effect of RL<sup>2)</sup>. If  $\Delta T/\Delta x(y)$  can be decreased or  $\partial T/\partial x(y)$  can be increased in some way, increasing h will be effective for reducing  $K_{\rm u}/K_{\rm bulk}$ . The way of decreasing  $\Delta T/\Delta x(y)$  will be discussed in 3.3, and the combination of the parameters will be discussed in 3.5.

## 3.2 Condition (I) - decreasing TSF<sub>10</sub>

 $TSF_{10}$  must be decreased to relax condition (I) given by Eq. (1)  $K_{\rm u}(T_{\rm a})V_{\rm m}/kT_{\rm a} \ge TSF_{10}$ .

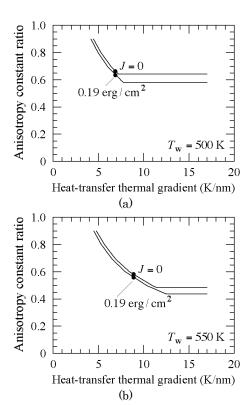
One way of decreasing  ${\rm TSF_{10}}$  is to reduce the standard deviation of the grain size  $\sigma_{\rm D}/D_{\rm m}$  <sup>4)</sup>. Decreasing  $\sigma_{\rm D}/D_{\rm m}$  relaxes condition (I) as seen from the solid lines in Fig. 7. Furthermore, the  $K_{\rm u}/K_{\rm bulk}$  values of the closed circles can be reduced by decreasing  $\sigma_{\rm D}/D_{\rm m}$  as shown in Fig. 7 (a). However, it seems that  $\sigma_{\rm D}/D_{\rm m}$  cannot be reduced from the standard value of 10 %



**Fig. 7** Dependence of  $K_{\rm u}/K_{\rm bulk}$  on  $\partial T/\partial x(y)$  at (a)  $T_{\rm w}=500$  K and (b) 550 K (calculation parameter: standard deviation of grain size  $\sigma_{\rm D}/D_{\rm m}$ ).

In Fig. 7 (b), it should be noted that the difference between the  $K_{\rm u}/K_{\rm bulk}$  values of the closed circles ( $\sigma_{\rm D}/D_{\rm m}$  = 10 % and 15 %) is very small. If  $T_{\rm w}$  can be

increased, a relatively large  $\sigma_{\rm D}/D_{\rm m}$  value may be allowable.



**Fig. 8** Dependence of  $K_{\rm u}/K_{\rm bulk}$  on  $\partial T/\partial x(y)$  at (a)  $T_{\rm w}=500~{\rm K}$  and (b) 550 K (calculation parameter: inter-grain exchange coupling J).

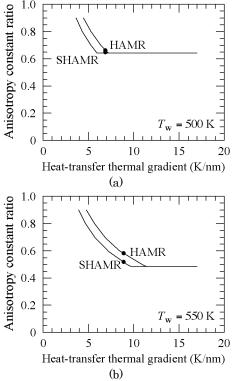
Although another way of decreasing  $\mathrm{TSF}_{10}$  is to introduce inter-grain exchange coupling  $J^{\,4)}$  as seen from the solid lines in Fig. 8.  $J=0.19~\mathrm{erg/cm^2}$  corresponds to an inter-grain exchange coupling  $e_{\mathrm{m}}$  of  $1.1^{\,4)}$  for  $D_{\mathrm{m}}$ . The inter-grain exchange coupling e makes the statistical thermal stability factor e times larger. It has little effect on reducing the  $K_{\mathrm{u}}/K_{\mathrm{bulk}}$  values of the closed circles since the limiting factor is condition (II). As seen from Fig. 8 (b), increasing  $T_{\mathrm{w}}$  has little effect for the same reason. If  $\Delta T/\Delta x(y)$  can be decreased or  $\partial T/\partial x(y)$  can be increased in some way, introducing J will also be effective for reducing  $K_{\mathrm{u}}/K_{\mathrm{bulk}}$ . The combination of the parameters will be discussed in 3.5.

# 3.3 Condition (II) - decreasing $\Delta T / \Delta x(y)$

 $\Delta T/\Delta x(y)$  must be decreased or  $\partial T/\partial x(y)$  increased to relax condition (II) given by Eq. (5)  $\Delta T/\Delta x(y) \leq \partial T/\partial x(y)$ .

Introducing SHAMR is effective for decreasing  $\Delta T/\Delta x(y)$  as seen from the solid lines where  $K_{\rm u}/K_{\rm bulk}$  changes in Fig. 9 (a).  $T_{\rm adj}$  in Eq. (3) for SHAMR is higher than that for HAMR since the maximum rewriting number for SHAMR is considerably smaller than that for HAMR³. However, the  $K_{\rm u}/K_{\rm bulk}$  values of the closed circles are almost the same since the limiting factor is condition (I). If we

can find a way to increase  $K_{\rm u}(T_{\rm a})V_{\rm m}/kT_{\rm a}$  or decrease TSF<sub>10</sub>, introducing SHAMR will be effective for reducing  $K_{\rm u}/K_{\rm bulk}$ . Figure 9 (b) shows the results for  $T_{\rm w}=550$  K. Since the limiting factor can be changed to condition (II), introducing SHAMR becomes effective.



**Fig. 9** Dependence of  $K_{\rm u}/K_{\rm bulk}$  on  $\partial T/\partial x(y)$  at (a)  $T_{\rm w}=500$  K and (b) 550 K (calculation parameter: HAMR and SHAMR).

# 3.4 Condition (II) - increasing $\partial T / \partial x(y)$

Although increasing  $T_{\rm w}$  is effective for increasing  $\partial T/\partial x(y)$  as mentioned above in **3.1**,  $\partial T/\partial x(y)$  can be changed very little by changing the parameters, namely the light-spot diameter  $d_{\rm L}$ , the heat-spot diameter  $d_{\rm w}$ , and the linear velocity v <sup>3</sup>. If the thermal conductivity K of IL1 decreases from 0.5 W/(cmK),  $\partial T/\partial x(y)$  becomes smaller as shown in a previous paper<sup>3</sup>. One possibility for increasing  $\partial T/\partial x(y)$  involves examining a media structure with a large  $\partial T/\partial x(y)$ .

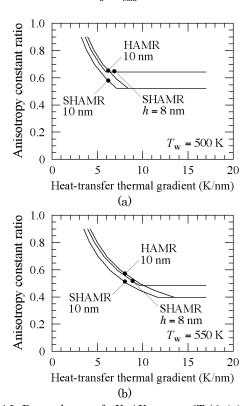
# 3.5 Combination of parameters

The combination of one parameter that relaxes condition (I) and another that relaxes condition (II) may be useful for reducing  $K_{\rm u}/K_{\rm bulk}$ .

When we replaced parameter h=8 nm with 10 nm,  $K_{\rm u}/K_{\rm bulk}$  changed from 0.66 to 0.65, and when only SHAMR was utilized,  $K_{\rm u}/K_{\rm bulk}$  became 0.64 at  $T_{\rm w}=500$  K. Alone they had scarcely any effect on reducing  $K_{\rm u}/K_{\rm bulk}$ . Next, we combined h=10 nm, which increases  $K_{\rm u}(T_{\rm a})V_{\rm m}/kT_{\rm a}$  of condition (I), and SHAMR, which decreases  $\Delta T/\Delta x(y)$  of condition (II). Figure 10 (a) shows the result of the combination, and

the resultant  $K_{\rm u}/K_{\rm bulk}$  is 0.58. The case where  $T_{\rm w}=550~{\rm K}$  is shown in Fig. 10 (b). When we compare HAMR and SHAMR, we find that SHAMR is more effective. This can be explained by Fig. 9 (b). On the other hand, when we compare a SHAMR of  $h=8~{\rm nm}$  and a SHAMR of  $h=10~{\rm nm}$ , the  $K_{\rm u}/K_{\rm bulk}$  values are almost the same since the limiting factor is condition (II) as seen in Fig. 6.

Therefore, the design parameters of HAMR, SHAMR, h, and  $T_{\rm w}$  are related in a complex manner in terms of reducing  $K_{\rm u}/K_{\rm bulk}$ .



**Fig. 10** Dependence of  $K_{\rm u}/K_{\rm bulk}$  on  $\partial T/\partial x(y)$  at (a)  $T_{\rm w}=500$  K and (b) 550 K (calculation parameter: HAMR, SHAMR, and h).

Next, we consider the relationship between HAMR, SHAMR, J, and  $T_{\scriptscriptstyle w}$ .

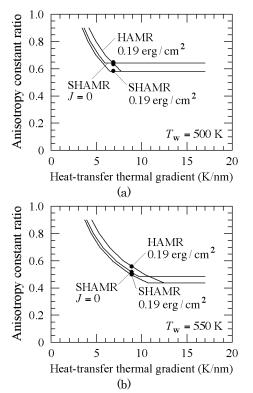
We combined J=0.19 erg/cm², which decreases  $\mathrm{TSF}_{10}$  of condition (I), and SHAMR, which decreases  $\Delta T/\Delta x(y)$  of condition (II). Figure 11 (a) shows the result of the combination, and the resultant  $K_{\mathrm{u}}/K_{\mathrm{bulk}}$  is 0.58. The case of  $T_{\mathrm{w}}=550$  K is shown in Fig. 11 (b). When we compare a SHAMR of J=0 and a SHAMR of J=0.19 erg/cm², the  $K_{\mathrm{u}}/K_{\mathrm{bulk}}$  values are almost the same since the limiting factor is condition (II).

The results are summarized in Fig. 12. The standard  $K_{\rm u}/K_{\rm bulk}$  value (calculated using the standard parameter values) is 0.66. The parameters  $T_{\rm w}$ , h, and J relax condition (I)  $K_{\rm u}(T_{\rm a})V_{\rm m}/kT_{\rm a} \ge {\rm TSF}_{10}$ , and the  $K_{\rm u}/K_{\rm bulk}$  values become 0.58, 0.65, and 0.63, respectively, by changing the parameters. And the parameters HAMR or SHAMR and  $T_{\rm w}$  relax condition (II)  $\Delta T/\Delta x(y) \le \partial T/\partial x(y)$ , and the  $K_{\rm u}/K_{\rm bulk}$ 

values become to be 0.64 and 0.58, respectively.

If h=10 nm and SHAMR or  $J=0.19~{\rm erg/cm^2}$  and SHAMR are combined,  $K_{\rm u}/K_{\rm bulk}=0.58$  can be achieved without increasing  $T_{\rm w}$ . A lower  $T_{\rm w}$  is better from the viewpoint of the heat resistance of the writing head and/or the surface lubricant.

If  $T_{\rm w}$  can be increased to 550 K, we can expect a  $K_{\rm u}/K_{\rm bulk}$  value of 0.51 by combination with SHAMR.



**Fig. 11** Dependence of  $K_{\rm u}/K_{\rm bulk}$  on  $\partial T/\partial x(y)$  at (a)  $T_{\rm w}=500$  K and (b) 550 K (calculation parameter: HAMR, SHAMR, and J).

Standard 
$$K_u / K_{bulk} = 0.66$$
(I)  $\frac{K_u (T_a) V_m}{k T_a} \ge \text{TSF}_{10} \text{ relaxing}$ 
 $T_w = 550 \text{ K} \leftarrow 500 \text{ K}$  0.58

 $h = 10 \text{ nm} \leftarrow 8 \text{ nm}$  0.65

 $J = 0.19 \text{ erg} / \text{cm}^2 \leftarrow 0$  0.63
(II)  $\frac{\Delta T}{\Delta x(y)} \le \frac{\partial T}{\partial x(y)} \text{ relaxing}$  0.58

SHAMR ← HAMR 0.64

 $T_w = 550 \text{ K} \leftarrow 500 \text{ K}$  0.58

Fig. 12 Media design guideline.

#### 4. Conclusions

The major limiting factors as regards obtaining the minimum anisotropy constant ratio  $K_{\rm u}/K_{\rm bulk}$  for media are (I) the information stability during 10 years of archiving and (II) the information stability on the trailing side during writing and in adjacent tracks during rewriting.

The parameters, namely the writing temperature  $T_{\rm w}$ , the recording layer thickness h, and the inter-grain exchange coupling J, relax condition (I). On the other hand, HAMR combined with shingled magnetic recording (SHAMR) and  $T_{\rm w}$  relax condition (II).

The combination of one parameter that relaxes condition (I) and another that relaxes condition (II) is useful for reducing  $K_{\rm u}/K_{\rm bulk}$ . If increasing h and SHAMR or increasing J and SHAMR are combined,  $K_{\rm u}/K_{\rm bulk}$  can be reduced successfully without increasing  $T_{\rm w}$ . If  $T_{\rm w}$  can be increased,  $K_{\rm u}/K_{\rm bulk}$  will be reduced by the combination with SHAMR.

It is important for the media design to distinguish whether the limiting factor is condition (I) or (II).

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# The hysteresis properties analysis of Fe-4 wt% Si prepared by high purity metallurgy

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An investigation on the improvement in hysteresis properties of high purity Fe-4 wt% Si prepared by cold crucible levitation melting was presented compared with a Fe-0.1 wt% C-4 wt% Si alloy formed by the identical process. The results showed that thin elongated carbon compounds were precipitated in both grains and grain boundaries of Fe-0.1 wt% C-4 wt% Si sample observed by means of Kerr effect microscopy and STEM, leading to an apparent degradation in coercivity, permeability and hysteresis loss. With regard to hysteresis loss in Fe-4 wt% Si, it increases linearly with the increase of maximum magnetic flux density below 1.4 T, whereas for Fe-0.1 wt% C-4 wt% Si, it behaves nonlinearly even at low maximum magnetic flux density and rises sharply with the increase of maximum magnetic flux density. Additionally, the precipitated carbon compounds in the grain boundaries change the morphology of the grain boundaries.

Key words: Fe-4 wt% Si, high purity, hysteresis loss, magnetic domain

#### 1. Introduction

The iron loss  $P_{\text{loss}}$  of electrical steel can be divided into three components: hysteresis loss  $P_{\text{h}}$ , classical eddy current loss  $P_{\text{e}}$ , and anomalous loss  $P_{\text{a}}$  according to the equation:

$$P_{\text{loss}} = P_{\text{h}} + P_{\text{e}} + P_{\text{a}} \tag{1}$$

Among the three components, hysteresis loss is the fundamental loss in electrical steel, especially at high magnetic flux density in the typical frequency range of transformers. Hysteresis loss is typically associated with obstacles to domain wall motion such as grain boundaries, precipitates 1) and crystallographic orientations 2). The improvement possibility in hysteresis loss of silicon iron alloy by reducing impurities has been recognized clearly <sup>3, 4)</sup>. Kaido et al. 1) gave a theoretical calculation about the effect of the precipitates on the hysteresis loss by reducing impurities further than commercial non-oriented steel 35H210, and their results turned out that the influence can be decreased substantially to 1/4. On the other hand, the eddy-current loss is closely related to the domain size and mobility of domain wall 5), which also can be significantly influenced by the existence of precipitates, and its mechanism has been reported by several authors <sup>6)-8)</sup>.

Most of the precipitates are originated from exterior impurities during a series of metallurgy processes and reduction of impurities is running up against potency in the conventional metallurgy processes. Therefore, in this current study, a method called cold crucible levitation melting (CCLM) was embraced to minimize the impurities in the ingots. The most important feature of CCLM is that the

molten alloy can be levitated from the cold crucible by magnetic force, so as to reduce the impurities apparently by isolating the contamination from the crucible during melting <sup>9)</sup>. Meanwhile, highly pure raw iron and silicon with a purity of 99.99 wt% and 99.999 wt% respectively were utilized to prepare the alloys.

In this paper, to clarify the potential of improvement in hysteresis properties by high purity, Fe-4 wt% Si alloy was firstly prepared by CCLM. Another Fe-4 wt% Si alloy with 0.1 wt% carbon added was also prepared as a comparison to reveal the effect of carbon on the magnetic properties and its interaction with domain structures. The permeability, coercivity and hysteresis loss of both alloys were studied. Additionally, the morphology of microstructure and the demagnetized domain structure were investigated by means of Kerr effect microscopy. The factors which influenced the hysteresis properties were discussed in detail by taking the domain structure and microstructure into consideration.

## 2. Materials and methods

Two kinds of Fe-4 wt% Si samples without and with addition of 0.1 wt% carbon were used for this study, identified as 4N and 3N. Two samples were both prepared from raw iron and silicon with a purity of 99.99 wt% and 99.999 wt% respectively in CCLM under high vacuum, except 3N with addition of 0.1 wt% carbon (99.999 wt%), which can be summarized as 4N (Fe-4 wt% Si) and 3N (Fe-0.1 wt% C-4 wt% Si). The detail of the identical CCLM furnace and its effect of impurities elimination, the homogeneity of element distribution can be found in the papers <sup>10, 11)</sup>. Both the

samples were maintained at melted condition and stirred for about 20 min at approximately 1600 °C, then followed by furnace cooling to room temperature (without subsequent annealing).

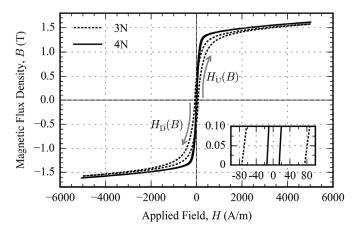
Hysteresis loops were measured at maximum magnetization from approximately 0.5 T to 1.5 T, and 5000 A/m applied field on both 5-mm-thick ring samples of 35 mm inner and 45 mm external diameter. These tests were performed under direct current (DC) using a magnetization measurement instrument, and various hysteresis properties were calculated based on the hysteresis loops.

The samples for domain observation were cut from the ring samples. The thickness of both samples was 2.5 mm and they were both finely grounded with an adequate finish polish with 0.02 µm colloidal silica, so as to reduce the agitation of surface stress. The demagnetized domain structures were observed using Kerr effect microscopy. The determination of crystallographic direction and microstructure were performed by Electron Back-Scattered Diffraction (EBSD) equipped in a SU-70 Scanning Electron Microscopy and analyzed by Orientation Image Microscopy (OIM) software. The morphology of precipitates in 3N sample were observed by Scanning Transmission Electron Microscopy (STEM) equipped in the FEI TECNAI-F20 TEM, and its samples were prepared by ion milling after a mechanical polish to about 30 μm.

#### 3. Results and Discussion

## 3.1. Hysteresis loop

The hysteresis loops in Fig. 1 show that 4N sample has better hysteresis properties with lower coercivity of 18 A/m compared with 76 A/m, and remanence of 0.32 T compared with 0.48 T of 3N. All of the compared data are summarized in Table 1. Additionally, 4N can be magnetized up to its



**Fig. 1** DC hysteresis loops measured in 3N and 4N ring samples at maximum applied field  $H_{\rm m}$  of 5000 A/m, associated with an inset for clarity of the coercivity and low magnetization part of hysteresis loops. Two arrows indicate the ascending branch  $H_{\rm U}(B)$  and descending branch  $H_{\rm D}(B)$  of hysteresis loops, respectively.

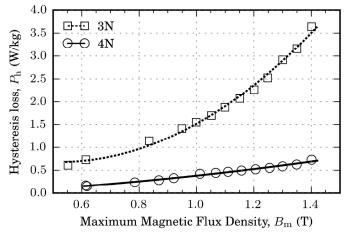
**Table 1** Comparison of maximum magnetic flux density, coercivity, remanence and permeability of two samples at  $H_m = 5000 \text{ A/m}$ 

Hysteresis properties	4N	3N
Maximum magnetic flux density (T)	1.60	1.58
Remanence (T)	0.32	0.48
Coercivity (A/m)	18	76
Maximum permeability (H/m)	0.010	0.004
Relative maximum permeability	8000	3200

maximum permeability with value of 0.010 H/m at lower applied field of 30 A/m with comparison to 0.004 H/m of 3N at 120 A/m, which can be concluded that 4N sample can be easily magnetized to higher magnetization at lower applied field compared to 3N.

The hysteresis losses  $P_h$  in Fig. 2 are calculated from a series of hysteresis loops measured at maximum magnetic flux density  $B_m$  from approximately 0.5 T to 1.45 T, and its relationship with each corresponding  $B_m$  are shown in Fig. 2. It can be found that  $P_h$  of 4N is significantly lower than that of 3N in the whole range of  $B_m$ , especially at higher Bm. Additionally, the increase of  $P_h$  in 4N has a linear connection with  $B_m$  within the range of 0.5 and 1.45 T applied in the experiment, whereas  $P_h$  of 3N increases gradually at the low applied field and fast along with the increase of  $B_m$ .

During magnetization, it is generally agreed that  $180^{\circ}$  domain wall displacement takes place at low applied field which are mainly controlled by the second phases and grain size  $^{12)}$ , and  $90^{\circ}$  domain wall displacement needs a fairly large magnetic field which are mainly controlled by the crystallographic texture  $^{12)}$ . The increment of hysteresis loss of high purity 4N sample only linearly relates with  $B_{\rm m}$ , which is totally independent with the sample parameters such as grain size, texture, regardless of whether on the condition of  $180^{\circ}$  or  $90^{\circ}$  domain wall displacement stage. However, the



**Fig. 2** Comparison of relationship of maximum flux density  $B_{\rm m}$  (less than 1.45 T) and hysteresis loss  $P_{\rm h}$  in 3N and 4N samples.

increment of hysteresis loss in 3N samples reveals its significant sensitivity on the structural parameters. During the 180° domain wall displacement stage at  $B_{\rm m}$  lower than 0.6 T, the increment of hysteresis loss is linear without relying much on the structural variation, but at the 90° domain wall displacement stage, the energy loss dissipated by the motion of domain wall increases aggressively.

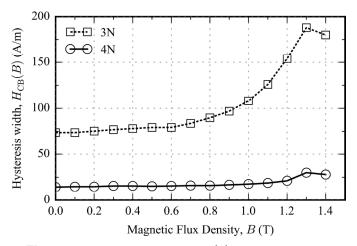
As known, hysteresis loss for ferromagnetic material is equivalent to the area of hysteresis loop, that is,  $P_{\rm h}=\oint HdB$ . The integration can also be treated as the summation of slices of width of hysteresis loop mathematically. Each individual width  $H_{\rm CB}(B)$  of hysteresis loop can be defined as the difference of the ascending branch  $H_{\rm U}(B)$  and the descending branch  $H_{\rm D}(B)$  (see Fig. 1) at a specific magnetization B according to Eq. 3  $^{1}$ :

$$H_{\rm CB}(B) = H_{\rm U}(B) - H_{\rm D}(B)$$
 (3)

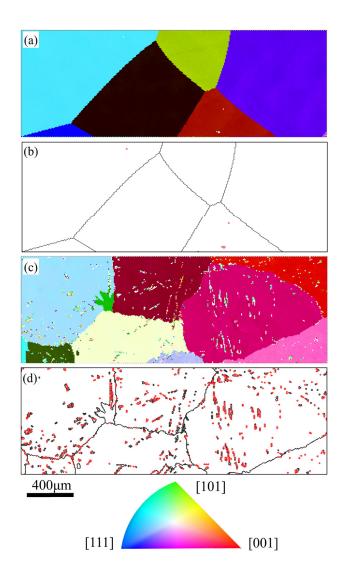
The variation of calculated  $H_{CB}(B)$  as a function of B in the hysteresis loops of the two samples are shown in Fig. 3. It can be seen that  $H_{CB}(B)$  maintains constantly till magnetization up to 1.2 T, then slightly increases and start to decrease afterwards in 4N sample, which is in accordance with the results of Fig. 2. As a comparison,  $H_{CB}(B)$  of 3N approximately maintains as a small constant below magnetization of 0.6 T, then starts to grow gradually and finally increases abruptly to a very large value and start to decrease afterwards. This difference of hysteresis loop width during magnetization between those two samples will be discussed in the following sections by combining with the morphology of precipitates, crystallographic texture, grain and domain structure.

# 3.2. Effect of microstructure and phase

The phase, grain morphology and orientations of part of the two rings samples are observed by EBSD and some of the results are shown in Fig. 4. By summarizing all the results, the structure of near equiaxed grains exist in both of the



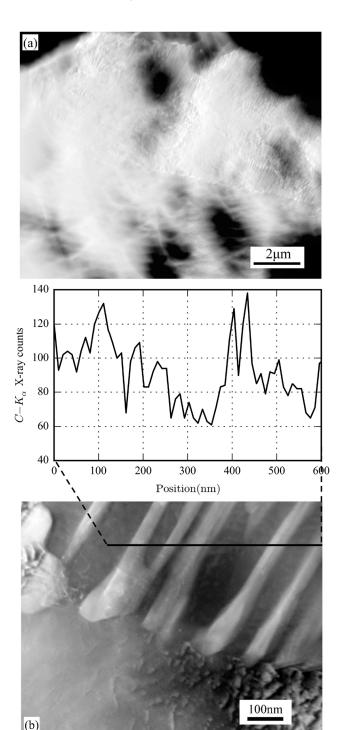
**Fig. 3** Relationship of width  $H_{CB}(B)$  of hysteresis loop at series of magnetic flux density B in 3N and 4N samples.



**Fig. 4** Orientation maps of (a) 4N and (c) 3N samples, grain boundary maps and phase maps of (b) 4N and (d) 3N samples observed by means of EBSD. In (b) and (d). The areas surrounded by black lines represent base phases, and the areas surrounded by red lines represent the second phases.

samples and their average sizes are similar, which are approximately 5 mm, and the orientations of grains are random without any preferred orientations found (Fig. 4 (a) and (c)). Therefore, the influence of grain size below 0.6 T (180° domain wall displacement stage), and the influence of crystallographic orientations on the difference of hysteresis loop width above 0.6 T (90° domain wall displacement stage) in Fig. 3 can be ignored. So the big differences on hysteresis properties between the two samples are roughly considered as a consequence of the existence of precipitates introduced in by carbon concentration. The morphology of the grains in 4N (Fig. 4 (b)) is irregular polyhedron by the equilibrium growth of adjacent grains, whereas near-polyhedron in 3N (Fig. 4 (d)) with the addition influence of the carbon precipitates. On the other hand, only A2 base phases exist in

4N (Fig. 4 (b)) but many of the second phases are precipitated in 3N indicated by the area surrounded by red lines in Fig. 4 (d). These phases are distributed in both grain and grain boundaries. Therefore, it can be stated here that the second



**Fig. 5** The precipitates observed in the 3N by STEM high-angle annular dark-field (HAADF). (a) Observation in a wider area indicates high density of thin elongated phase, and (b) one more specific observed area in (a). The middle figure shows the EDS mapping of carbon element along the line in (b).

phase is the main reason for the difference of hysteresis properties of the two samples, as well as the morphology difference of grain boundaries.

The specific morphology of the precipitates were investigated by STEM and its results are shown in Fig. 5. The overall view in Fig. 5 (a) indicates that the precipitates exist at a state of thin elongated columnar structures and aligned with each other in parallel. More specific observation in Fig. 5 (b) indicates that these precipitates grow in the grain as insertions, with a similar width of approximately 50 nm. The EDS results (center of Fig. 5) show that the carbon atoms are enriched in the precipitates, which can be considered as carbon compounds. These compounds can act as strong pinning obstacles against the structures and motion of domain wall, as demonstrated in the following section.

#### 3.2. Domain structure

The hysteresis loss is the energy dissipated in the domain wall motion, therefore, it is important to clarify the condition of domain structures in both of the samples. Fig. 6 gives the demagnetized surface domain structures in part of each sample, from which it can be found that small complex lancet domain patterns dominates in the observed areas, and size of both surface domains are in the same order of magnitude.

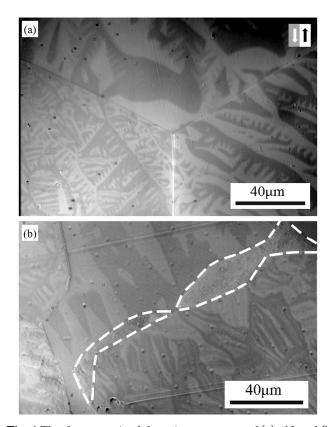


Fig. 6 The demagnetized domain structures of (a) 4N and (b) 3N samples observed by means of Kerr microscopy. The dark and bright areas indicate upward and downward magnetization components, respectively. The area surrounded by white dash lines in (b) indicate the carbon precipitates.

This proves that the precipitates dominate the difference in the hysteresis loss of the two samples, regardless of the influence of surfaces. The precipitates exist as non-magnet because no domain structures are observed in them (indicated by the arrows in Fig. 6). These non-magnetic precipitates weaken the interaction between magnetic components in the samples, resulting in the degradation of the magnetization process, which leads to the increase of hysteresis loss and reduction of permeability, which has been described previously.

It can be also found that domain structures in each individual grain of each sample differ. This kind of differences are mainly dominated by the deviation angle of the closest easy magnetization axis  $\{001\}$  from the sample surface, which has been reported in detail by Williams et al  $^{5)}$ .

#### 4. Conclusions

The hysteresis properties, specifically the relationship between hysteresis loss and maximum magnetic flux density, and magnetic domain of high purity Fe<sup>-</sup> 4 wt% Si (4N) alloy with comparison to Fe<sup>-</sup> 0.1 wt% - 4 wt% Si (3N) carbon created by CCLM were investigated.

- 1. High purity 4N alloy has super low coercivity, permeability and hysteresis loss than 3N, due to pinning of domain 180° and 90° wall displacement by the elongated thin carbon compounds which are distributed in both of grains and grain boundaries.
- 2. By increasing the purity of Fe-4 wt% Si, the relationship between hysteresis loss and maximum flux density are correlated linearly, by comparison to the nonlinear relationship in 3N.
- 3. The presence of carbon compounds precipitated in the grain boundaries changes the morphology of the grain boundaries. The compounds also retard the continuity

of the domain structures, thus finally resulting in degradation of the hysteresis properties.

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# Structure and Magnetic Properties of (001) Oriented CoPt-Ag and CoPd-Ag Alloy Films

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20-nm-thick (CoPt)<sub>100-x</sub>Ag<sub>x</sub> and (CoPd)<sub>100-x</sub>Ag<sub>x</sub> (001) films (x = 0–20) were grown on MgO(001) substrate at a temperature of  $300^{\circ}$ C by molecular beam epitaxy, and their crystal structure and magnetic properties were compared with those of FePt-Ag and FePd-Ag films [Y. Tokuoka *et al.*: *J. Appl. Phys.*, **115**, 17B716 (2014)] for the systematic study of the effect of Ag addition into transition metal – noble metal alloy films. For the CoPt-Ag, the addition of 5% Ag increased perpendicular magnetic anisotropy (PMA) and coercivity. However, for the CoPd-Ag, Ag addition enhanced neither the PMA nor the coercivity. Structural analysis of the CoPt-Ag and CoPd-Ag films revealed that Ag addition promoted L10 phase ordering for CoPt-Ag, while L10 ordering was not found in CoPd-Ag. Although there were some similarities in the effect of Ag addition between CoPt-Ag and FePt-Ag, such as increased PMA, coercivity, and L10 ordering, there were different trends in PMA and L10 ordering between CoPd-Ag and FePd-Ag, except that both films showed reduced coercivity with Ag addition. Ag segregation and L10 ordering are considered important factors to explain the variations of structure and magnetic properties with Ag addition.

Key words: perpendicular magnetic anisotropy, Ag segregation, CoPt, CoPd

## 1. Introduction

Tetoragonal L1<sub>0</sub> phase magnetic materials, e.g., FePt and CoPt, have been extensively studied as potential candidates for future high-density perpendicular recording media because extremely large perpendicular magnetic anisotropies (PMA). However, for the practical application of these materials, there still remain challenges such as fabrication of nano-sized grains, control of (001) orientation, and lowering the ordering temperature of L1<sub>0</sub> phase<sup>1), 2)</sup>. One of the effective ways to realize these requirements is the addition of the third element. Ag or Cu addition into FePt is known to be effective to reduce its L1<sub>0</sub> ordering temperature<sup>3)-6)</sup>. The addition of Ag into CoPt is also known to promote L1<sub>0</sub> ordering and (001) orientation, which is crucial to obtain the large PMA<sup>7), 8)</sup>. Moreover, it is reported that the ordering temperature of FePd is reduced by insertion of the Ag underlayer9).

We have also studied FePt-Ag and FePd-Ag films to investigate the effect of Ag addition into L10 phase magnetic materials 10)-12), and reported Ag addition to FePt with the amount of around 10 at. % is effective to promote L1<sub>0</sub> ordering and granular growth of FePt, resulting in the increase of the PMA and coercivity of the film normal direction. From the structural analysis magnetic properties measurements, it was concluded that the Ag is segregated from FePt, and this phase separation promotes L10 ordering and granular growth of FePt<sup>12)</sup>. Unlike the case of FePt-Ag, Ag addition into FePd does not contribute to the corecivity increase and granular growth, and the formation of FePd-Ag alloy was observed<sup>12)</sup>. This suggests that the phase separation is a key factor to control the microstructure of L10 phase magnetic materials and to

obtain suitable magnetic properties for the perpendicular recording media.

Recently, L. Zhang *et al.* reported that FePt-Ag:C films have large PMA and small grain size of around 6 nm<sup>13)</sup>. They reported a substantial increase of the PMA of FePt by the Ag addition, and the increase of the PMA is considered to be due to the phase separation of Ag and FePt by the heat treatment, in which the L1<sub>0</sub> ordering of FePt is promoted through the atom replacement by the Ag diffusion<sup>14)</sup>.

In this paper, CoPt-Ag and CoPd-Ag films have been studied in detail for the systematic understanding of the effect of Ag addition in 3d transition metal and noble metal alloy films. Although no ordered phase is reported in bulk CoPd<sup>15)</sup>, metastable L1<sub>0</sub> phase CoPd was reported in evaporated CoPd alloy films<sup>16)</sup>. The phase separation of Ag and CoPt or CoPd as well as the enhancement of L1<sub>0</sub> ordering by Ag addition are discussed. In addition to the structural analysis, the relationship between the structure and magnetic properties is studied in detail. Furthermore, the effect of Ag addition into CoPt and CoPd is compared with the results of previously reported FePt-Ag and FePd-Ag.

# 2. Experiment

In our previous study, 5 nm-thick (FePt)<sub>100-x</sub>Ag<sub>x</sub> and (FePd)<sub>100-x</sub>Ag<sub>x</sub> were grown directly on MgO(001) substrates<sup>12)</sup>. However, c-axis orientation of 5 nm-thick CoPt on MgO(001) was poor compared to the FePt-Ag, due to the lattice mismatch between CoPt and MgO. Thus in this study, 20 nm-thick (CoPt)<sub>100-x</sub>Ag<sub>x</sub> and (CoPd)<sub>100-x</sub>Ag<sub>x</sub> films (x = 0–20) were grown by molecular beam epitaxy (MBE) method. The CoPt-Ag was grown directly on MgO(001) substrates while CoPd-Ag was

grown on 10 nm thick Pd (001) buffer layer to reduce the lattice mismatching between CoPd and MgO(001) substrate. The Pd buffer layer was grown at room temperature, and CoPt and CoPd layers were grown at 300°C. Co, Pt (Pd), and Ag were co-evaporated from independently controlled e-beam sources, for which the deposition rate of each element was monitored by quartz thickness monitors. During the deposition, the MBE chamber was kept at a pressure of less than  $3\times10^{-7}$  Pa. Before the deposition, the MgO substrate was cleaned by 1 keV Ar<sup>+</sup> ion bombardment followed by the heat treatment at 1000°C in the MBE chamber.

The film structure was characterized by *in-situ* reflection high energy electron diffraction (RHEED) and ex-situ X-ray diffraction (XRD) using Cu  $K\alpha$  radiation source ( $\lambda = 0.15418$  nm). Hysteresis loops and magnetic anisotropies were measured by an alternating gradient field magnetometer and a torque magnetometer, respectively. The surface topography was checked by the atomic force microscopy (AFM).

## 3. Results and Discussions

## 3.1 Crystal structure and surface morphology

Figure 1 shows  $\omega$ –2 $\theta$  XRD profiles of (CoPt)<sub>100-x</sub>Ag<sub>x</sub> and (CoPd)<sub>100-x</sub>Ag<sub>x</sub> grown at 300°C. In the profiles of CoPt-Ag, 001 superlattice and 002 fundamental diffractions were clearly seen, which indicates the

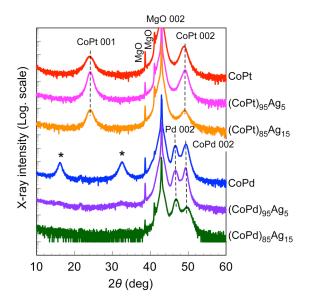


Fig. 1 ω-2θ XRD profiles of (CoPt)<sub>100-x</sub>Ag<sub>x</sub> and (CoPd)<sub>100-x</sub>Ag<sub>x</sub> alloy films (x = 0, 5, and 15) grown at 300°C. X-ray scattering vector is normal to the film surface. For the profile of CoPd, there exist reflections indicated as "\*" around 2θ = 16 deg and 32 deg. The lattice spacings estimated from these peaks are 0.55 nm and 0.28 nm, respectively, which coincide with 3- and 1.5-times of CoPd 002 spacing of 0.184 nm.

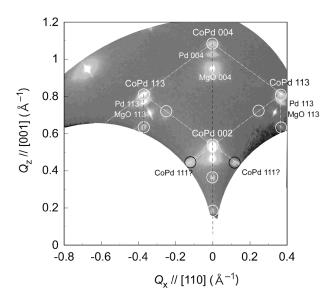
existence of (001) oriented L10 phase. Moreover, CoPt 001 peak intensity increased by adding Ag and no significant changes in the peak positions of CoPt 001 and 002 with the Ag addition were found. In the profiles of CoPd-Ag, Pd 002 and CoPd 002 peaks indicate the (001) oriented growth of Pd buffer and CoPd layers on the MgO(001) substrate. At x = 0, there exist reflections indicated as "\*" in the figure around  $2\theta = 16 \deg$  and 32deg. The lattice spacings estimated from these peaks are 0.55 nm and 0.28 nm, respectively, which coincide with 3- and 1.5-times of CoPd 002 spacing of 0.184 nm. This suggests the existence of the long range order structure along the CoPd [001] direction. With the increase of Ag content, this long range order structure of CoPd disappeared. The lattice constant estimated from the CoPd 002 was 0.366 nm, and the peak position of CoPd 002 did not change significantly with increasing Ag content.

The reciprocal space mapping of CoPd also showed the existence of the long range order structure along [001] direction. Figure 2 shows the reciprocal space mapping for a slice of (110) plane of CoPd grown on MgO (001) at 300°C. The  $Q_x$  and  $Q_z$  are along MgO[110] and [001] directions, respectively, and the radial component of the scattering vector was calculated as:

$$\sqrt{Q_x^2 + Q_z^2} = \frac{2\sin\theta}{\lambda} \,, \tag{1}$$

where  $\lambda = 1.5418 \text{ Å}$  and  $\theta$  is Bragg angle.

The diffraction spots at  $Q_z = 0.18 \text{ Å}^{-1}$  and  $0.36 \text{ Å}^{-1}$  correspond to the peaks indicated as "\*" in Fig. 1. Besides these spots, several diffraction spots, such as  $(Q_x, Q_z) = (-0.37 \text{ Å}^{-1}, 0.63 \text{ Å}^{-1})$  and  $(0.37 \text{ Å}^{-1}, 0.63 \text{ Å}^{-1})$  are seen, suggesting the existence of the long range order structure along CoPd [001] direction. More detail structural analysis is necessary to elucidate the

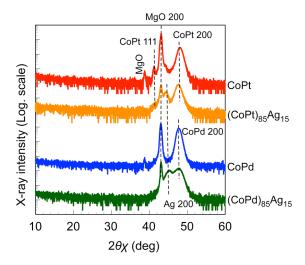


**Fig. 2** Reciprocal space mapping of (110) plane of CoPd film grown on MgO (001) at 300°C.

structure of CoPd alloy films, but at present we consider two possibilities: atomic layered structure along [001] direction with a period of 3 times of 002 spacing (0.184 nm) or periodic anti-phase domain boundary similar to that observed in  $CuAu^{17}$ ,  $Cu_3Pd^{18}$ , etc.

Figure 3 shows in-plane  $\phi$ -2 $\theta\chi$  XRD profiles of  $(\text{CoPt})_{100-x}\text{Ag}_x$  and  $(\text{CoPd})_{100-x}\text{Ag}_x$  with x = 0 and 15. The profiles were taken under the condition that the X-ray scattering vector is along MgO [100] direction. Besides the reflection of MgO 200, intense peak from CoPt 200 or CoPd 200 is seen in the profile at x = 0. No superlattice reflections of CoPt and CoPd were seen in the profiles. The addition of Ag did not significantly change the peak position of CoPt 200 and CoPd 200, and Ag 200 peak was seen in the profiles at x = 15. The lattice constants a estimated from the CoPt 200 and CoPd 200 peaks were 0.380 nm and 0.379 nm, respectively. From these structural analyses, we concluded that the Ag is segregated from CoPt or CoPd, and promotes L10 ordering of CoPt. L10-CoPt is well known as a stable phase near equiatomic composition of Co and Pt, while L10-CoPd is reported to be metastable<sup>16)</sup>. Although, the addition of the third element, e.g., C, was reported to stabilize the metastable L1<sub>0</sub>-MnAl phase in the Mn-Al system<sup>19)</sup>, the Ag addition to Co-Pd seems to have no significant effect to obtain metastable L10-CoPd phase.

Figure 4 shows the RHEED patterns of  $(\text{CoPt})_{100-x}\text{Ag}_x$  with Ag amount of x=0-15. The incidence of the electron beam was along [100] direction of MgO(001) substrate and the patterns were taken after the deposition of CoPt-Ag films. For x=0, streak patterns with a space corresponding to the in-plane lattice spacing of CoPt were observed. The "vertical" streak pattern means slightly broadened reciprocal



**Fig. 3** In-plane  $\phi$ -2θχ XRD profiles of  $(\text{CoPt})_{100-x}\text{Ag}_x$  and  $(\text{CoPd})_{100-x}\text{Ag}_x$  alloy films (x = 0 and 15) grown at 300°C. X-ray scattering vector is parallel to MgO [100] direction.

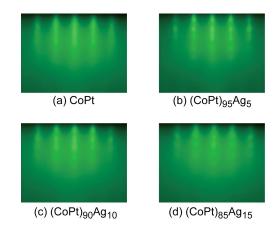


Fig. 4 RHEED patterns of (CoPt)<sub>100-x</sub>Ag<sub>x</sub> grown at 300°C with Ag amounts of (a) x = 0, (b) 5,
(c) 10, and (d) 15. The incidence of electron beam was along [100] direction of MgO(001) substrate. All patterns were taken after deposition of CoPt-Ag films.

lattice rods of CoPt, indicating rather flat surface of CoPt with atomic steps. With increasing the Ag content, the RHEED pattern changed from streaks to vertically aligned spots. This means the incident electrons passed inside the CoPt crystals, and thus indicates the granular growth of the CoPt-Ag. The AFM images of (CoPt)<sub>95</sub>Ag<sub>5</sub> and (CoPt)<sub>85</sub>Ag<sub>15</sub> are shown in Figs. 5 (a) and (b), respectively. With increasing the Ag content, the granular growth of the CoPt-Ag was confirmed, which coincides well with the RHEED observation. A similar granular growth by the addition of Ag was also confirmed in CoPd-Ag films. The granular structures of CoPt-Ag and CoPd-Ag are considered to be induced by the segregation of Ag from CoPt or CoPd, and we consider the Ag covers CoPt or CoPd grains, forming core/shell structure as observed in the FePt-Ag:C film<sup>14)</sup>. White protrusions in Fig. 5 (b) are quite similar to Ag re-precipitates at the top of the FePt-oxide film reported by Yang et al.<sup>20</sup>. In such case, the actual Ag content in

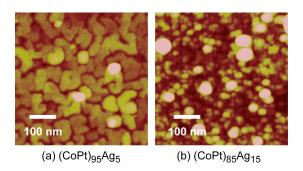
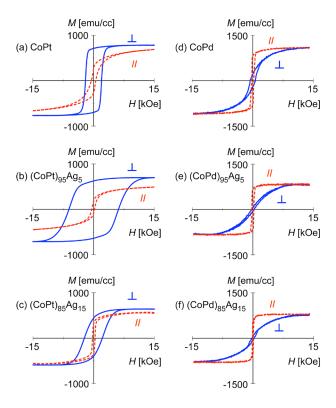


Fig. 5 AFM images of (CoPt)<sub>95</sub>Ag<sub>5</sub> and (CoPt)<sub>85</sub>Ag<sub>15</sub> grown at 300°C. Image area is 500 nm × 500 nm and vertical scale of image is 20 nm. Mean roughness R<sub>a</sub> of image is (a) 1.47 nm and (b) 2.26 nm.

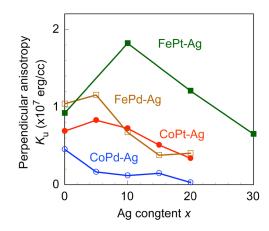
the CoPt-Ag may be smaller than the designed value. A similar granular growth by the addition of Ag was also confirmed in CoPd-Ag films.

#### 3.2 Magnetic properties

The structural variations of CoPt-Ag and CoPd-Ag strongly affect their magnetic properties. Figure 6 hysteresis loops of $(CoPt)_{100-x}Ag_x$ (CoPd)<sub>100-x</sub>Ag<sub>x</sub>, where the external field was applied parallel and perpendicular to the film plane. Figure 7 shows the Ag content dependence of the PMA, Ku, of CoPt-Ag and CoPd-Ag films, where the Ku was estimated from the torque curve applying a magnetic field of 15 kOe. For comparison, the Ku of the previously reported FePt-Ag and FePd-Ag are also shown in the same figure. For the CoPt-Ag, the increases of PMA and coercivity were found by the addition of Ag with an amount of 5 at. % as shown in Fig. 6 (b) and Fig. 7. However, for the CoPd-Ag, Ag addition reduced the PMA (see Fig. 7) and did not contribute to enhance the coercivity as shown in Fig. 6 (d)-(f). Interestingly, the CoPd film has some PMA and coercivity, which may be related to the long range order structure along CoPd [001] direction as discussed in Fig. 2. The variations of the magnetic properties of CoPt and CoPd with Ag content will be explained by the structural variation. For CoPt-Ag, Ag addition promotes L10 ordering, and Ag



**Fig. 6** Hysteresis loops of (a)–(c)  $(CoPt)_{100-x}Ag_x$  and (d)–(f)  $(CoPd)_{100-x}Ag_x$ , where the external field was applied parallel and perpendicular to the film plane. Ag contents are (a), (d) x = 0, (b), (e) x = 5, and (c), (f) x = 15.



**Fig. 7** Ag content dependence of perpendicular anisotropy  $K_{\rm u}$  of CoPt-Ag and CoPd-Ag films, where  $K_{\rm u}$  was estimated from torque curve applying a magnetic field of 15 kOe. For comparison, the  $K_{\rm u}$  of the previously reported FePt-Ag grown at 250°C and FePd-Ag grown at 400°C are also shown<sup>12)</sup>.

atoms are considered to segregate from CoPt, which will make the L1<sub>0</sub>-CoPt grains smaller as observed in Figs. 4 and 5. These structural variations will increase the PMA and coercivity of the CoPt-Ag films. On the other hand, Ag did not assist the formation of the L1<sub>0</sub> phase CoPd, which does not contribute to enhance the PMA and coercivity even though the granular growth was promoted by the Ag addition.

By comparing these results with those of the previously reported FePt-Ag and FePd-Ag<sup>12)</sup>, we found a similarity between FePt-Ag and CoPt-Ag. For both alloys, Ag addition enhanced their PMA and coercivity as shown in Fig. 7 and Ref. 12), owing that the Ag addition promoted the L10 ordering and granular growth. Also, there exists a similarity between FePd-Ag and CoPd-Ag; Ag addition did not contribute to enhance the coercivity in both alloys. However, there is a difference between FePd-Ag and CoPd-Ag. For FePd-Ag, the Ag tend to form an alloy with FePd, which lowers the coercivity even though its PMA slightly increases with the Ag addition of 5 at. % due to the increase of L10 ordering. For CoPd-Ag, the Ag atoms segregate from the CoPd, which makes the grains smaller, but the Ag does not contribute to promote L10 ordering.

### 4. Conclusion

The effect of Ag addition to CoPt and CoPd on their crystal structures and magnetic properties was investigated, and the results were compared with those of previously reported FePt-Ag and FePd-Ag for the systematic understanding of the effect of the Ag addition. It was confirmed that Ag atoms in the CoPt segregate from CoPt, which promotes L1<sub>0</sub> ordering and granular growth of CoPt. Such structural variations improved the magnetic properties of CoPt for a

perpendicular recording medium, e.g., increases of PMA and coercivity. These tendencies were similar to the results of FePt-Ag, in which Ag segregation effectively increases PMA and coercivity of FePt. On the other hand, Ag addition does not contribute to the increase of coercivity of CoPd, which is similar to the result of FePd-Ag. However, the microstructure of CoPd-Ag is different from that of FePd-Ag. For CoPd-Ag, Ag atoms segregate from CoPd, but do not assist the formation of the L1<sub>0</sub> CoPd phase. On the other hand, for FePd-Ag, Ag tends to form an alloy with FePd, which slightly promote the L10 ordering of FePd but does not contribute to the granular growth. From these results, it is considered that the reason why Ag is an effective material to obtain high ordered L10 phase and small sized grains is a low solubility of Ag to FePt and CoPt as well as a low surface energy of Ag, which will be effective to make core/shell structure as reported in Ref. 14).

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日本磁気学会 第 206 回研究会

# 発電用電子・磁気材料の現状と今後の展望

地球温暖化の抑制と温室効果ガス排出量削減するために化石資源に代わるエネルギー源の有効活用の必要性が高まっています。中でも、近年、環境からの微小なエネルギーを電力に変換する技術の研究が精力的に行われています。本研究会では発電用電子・磁気材料に着目し、振動発電、熱電変換、磁気冷凍について第一線でご活躍されている6名の研究者の方を講師としてお招きし、ご講演いただきます。発電用電子・磁気材料の現状と今後の展望について活発に議論していただけます。多くの方のご参加をお待ちしております。

日時: 2016年1月29日(金)13:00~16:45

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## プログラム

座長:東 大地(日立金属)

13:00 - 13:35 「エナジーハーベスティング技術の現状と将来」

○竹内敬治(NTT データ経営研究所)

13:35 - 14:10 「磁歪式振動発電を用いたバッテリーフリーIoT の実用化展開」

○上野敏幸(金沢大)

14:10 - 14:45 「積層同時焼成磁気回路を用いた MEMS マイクロエアタービンの開発」

○内木場文男(日大)

休憩(15分)

座長:関 剛斎(東北大)

15:00 - 15:35 「エレクトレット振動発電器のマーケット調査」

○諸口 登(小西安)

15:35 - 16:10 「異常ネルンスト効果を利用した環境発電素子のための磁性材料開発とその展望」

○桜庭裕弥(物材機構)

16:10 - 16:45 「磁気と熱のエネルギー変換機能磁性材料とその応用」

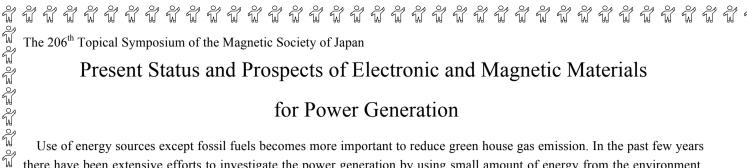
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OY. Sakuraba (NIMS)

OA. Saito (Toshiba)

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The 206<sup>th</sup> Topical Symposium of the Magnetic Society of Japan

# Present Status and Prospects of Electronic and Magnetic Materials

# for Power Generation

Use of energy sources except fossil fuels becomes more important to reduce green house gas emission. In the past few years there have been extensive efforts to investigate the power generation by using small amount of energy from the environment. In this symposium, six lecturers will give recent trends and prospects of electronic and magnetic materials for vibrational power generator, thermoelectric conversion, magnetic refrigeration etc. We welcome the participation of all of you.

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Information: The Magnetic Society of Japan / Tel: 03-5281-0106

and its prospect"

16:10 - 16:45

URL: http://www.magnetics.jp/archive/english/seminar/206.html

Organizers: Daichi Azuma (Hitachi Metals), Shigenobu Koyama (Daido Steel), Takeshi Seki (Tohoku Univ.)

#### Program

13:00 - 13:35	Chair: D .Azuma (Hitachi Metals) "Current status and prospects of energy harvesting technologies"  OK. Takeuchi (NTT DATA INSTITUTE OF MANAGEMENT CONSULTING)
13:35 - 14:10	"Practical applications of battery free IoT using magnetostrictive vibrational power generator"  OT. Ueno (Kanazawa Univ.)
14:10 - 14:45	"Development of MEMS micro air turbine generator with multilayer Co-fired magnetic circuit"  oF. Uchikoba (Nihon Univ.)
	Break (15 min.)
15:00 - 15:35	Chair: T. Seki (Tohoku. Univ.)  "Market research for electret vibration energy harvester"  ON. Moroguchi (Konishiyasu)
15:35 - 16:10	"Development of the magnetic materials for energy harvesting devices using anomalous Nernst effect

The presentations will be given in Japanese. / Audio and/or visual recording is prohibited.

"Magnetic materials for thermal and magnetic energy conversion and application to refrigeration"

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