Random Anisotropy Model for Nanocrystalline Soft Magnetic Alloys with Grain-Size Distribution

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A simple model considering grain-size distribution is proposed based on the random anisotropy model. When the maximum grain size (D_m) is less than the exchange correlation length and induced anisotropies are sufficiently small, the effective magnetic anisotropy constant ($\langle K_1 \rangle$) is given by using a distribution function ($f(D)$) for the grain size (D) as $\langle K_1 \rangle \approx K_1^4 \{ \int_0^{D_m} D^3 f(D) dD \}^2 / (\varphi^6 A_c^3)$, where K_1 is the magnetocrystalline anisotropy constant, φ is a parameter which reflects both the symmetry of $\langle K_1 \rangle$ and the total spin rotation angle over the exchange-correlated coupling chain and A_c is the exchange stiffness constant. The log-normal distribution function reproduces well the observed grain-size distribution and yields $\langle K_1 \rangle \approx K_1^4 \langle D \rangle^6 \exp(6\sigma_D^2) / (\varphi^6 A_c^3)$, where $\langle D \rangle$ is the mean grain size and σ_D is the geometric standard deviation for the distribution. This result satisfies the well-known $\langle D \rangle^6$ law. However, $\langle K_1 \rangle$ increases with increasing σ_D even if $\langle D \rangle$ is constant. Our model has been extended by taking into account the effect of the coherent induced anisotropies on the exchange correlation length. The coercivity ($H_c \propto \langle K_1 \rangle / J_s$, where J_s is the saturation magnetization) of the nanocrystalline Fe-Nb-B(-P-Cu) alloys with different grain-size distribution have been calculated. Our model explains well the dependence of H_c on the grain-size distribution. These results suggest that one should pay attention on not only the mean grain size but also on the grain-size distribution since the inhomogeneity of the grain size increases H_c .

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1. Introduction

Nanocrystalline soft magnetic alloys have been studied extensively over decade.¹⁻³⁾ The origin of magnetic softness in the nanocrystalline alloys is due to the important condition that the structural correlation length (grain size) is shorter than the exchange correlation length (L_{ex}) over which spins are coupled via the exchange interaction. Alben *et al.* have described the effective anisotropy energy of the amorphous materials based on a statistical argument, the so-called random anisotropy model (RAM).⁴⁾ The structural correlation length for the amorphous materials corresponds to atomic distance, and the effective anisotropy energy density in the RAM is given by the square root of the mean-square fluctuations of the anisotropy energy in the exchange coupled volume ($\sim L_{ex}^3$). Herzer has shown that the RAM explains the effective anisotropy energy even in nanocrystalline systems and predicted that the coercivity (H_c) varies as the sixth power of the grain size (D) in the range of $D < L_{ex}$.^{5,6)}

Since Herzer's first application of the RAM to nanocrystalline Fe-Si-B-Nb-Cu alloy,⁵⁾ this model has been employed widely to explain the origin of the magnetic softness in various nanocrystalline systems.⁷⁻⁹⁾ However, the original RAM only deals with single-phase systems. Most of the nanocrystalline soft magnetic alloys contain a considerable amount of an amorphous phase.^{3,10)} Herzer,¹¹⁾ Hernandez *et al.*,¹²⁾ Suzuki and Cadogan¹³⁾ have proposed extended RAMs for the two-phase nanocrystalline alloys. The influence of induced anisotropies (e.g., magneto-elastic or annealing-induced anisotropies) should be also considered. In actual nanocrystalline alloys, the contributions of the induced anisotropies to magnetization process always complicate the treatment of the intrinsic physical problems. Suzuki *et al.* reported that the magnetization process of the

nanocrystalline alloys with sufficiently small D is mainly dominant for the induced anisotropy and D^6 law for H_c is not established.¹⁴⁾ If the induced anisotropy is long-range and uniaxial, then H_c is proportional to D^3 , not D^6 .

Another important fact is grain-size distribution. In the analysis based on the RAM, the magnetic softness has been discussed using the mean grain size. However, the grain size has a distribution in actual nanocrystalline alloys. In the two samples in which the mean grain size is the same, the magnetic softness may be different if the feature of the distribution of the grain size is different. Recently, we reported that the Fe₈₅Nb₆B₉ alloy has as-quenched structure composed of an amorphous phase and α -Fe grains with 20–45 nm in size, and has crystallized nanostructure including the relatively coarse grains.^{15,16)} The effect of the structural inhomogeneities on the soft magnetic properties is very interesting. In this paper, the effect of the grain-size distribution on the magnetic softness of nanocrystalline soft magnetic alloys is discussed based on the RAM.

2. Random Anisotropy Model with Grain-Size Distribution

2.1 Basic random anisotropy model

In Herzer's random anisotropy model (RAM),^{5,6)} the reduction of the effects of the intrinsic magnetocrystalline anisotropy constant (K_1) is based on the following random-walk consideration:

$$\langle K_1 \rangle = \frac{K_1}{\sqrt{N}}, \quad (1)$$

where $\langle K_1 \rangle$ is the random magnetocrystalline anisotropy (i.e., the fluctuating part of the magnetocrystalline anisotropy) which governs the magnetization process in the sample. N is

the number of grains in a magnetically coupled volume ($V_{\text{ex}} = L_{\text{ex}}^3$) which, in bulk-form systems, is

$$N = \frac{V_{\text{ex}}}{V} = \left(\frac{L_{\text{ex}}}{D} \right)^3, \quad (2)$$

where $V = D^3$ is the grain volume. The exchange correlation length (L_{ex}) is determined by the competition between the anisotropy and exchange energy terms and is defined as

$$L_{\text{ex}} = \varphi \sqrt{\frac{A_c}{\langle K \rangle}}. \quad (3)$$

Here, A_c is the exchange stiffness constant of crystalline phase and φ ($= 1$ in the Herzer's model^{5,6}) and $(4/3)^{1/2}$ in the original RAM proposed by Alben *et al.*⁴) is a parameter which reflects both the symmetry of the effective anisotropy constant ($\langle K \rangle$) and the total spin rotation angle over the exchange-correlated coupling chain.¹³ Provide that the random magnetocrystalline anisotropy is the system and the effective anisotropy constant is approximated by $\langle K \rangle$ (*i.e.*, $\langle K \rangle \approx \langle K_1 \rangle$), the effective length of L_{ex} can then be determined self-consistently using eqs. (1)–(3), yielding

$$L_{\text{ex}} \approx \varphi^4 \frac{A_c^2}{K_1^2 D^3} = L_0 \left(\frac{L_0}{D} \right)^3, \quad (4)$$

which leads to the well-known relation

$$\langle K_1 \rangle \approx \frac{1}{\varphi^6} \frac{K_1^4 D^6}{A_c^3} = K_1 \left(\frac{D}{L_0} \right)^6, \quad (5)$$

where

$$L_0 = \varphi \sqrt{\frac{A_c}{K_1}} \quad (6)$$

is the intrinsic exchange correlation length.

2.2 Grain-size distribution

In general, D has a distribution in actual nanocrystalline alloys. The original idea was proposed by Herzer.¹¹ For simplicity, let us consider that the maximum grain size (D_m) does not exceed L_{ex} . The random magnetocrystalline anisotropy and the exchange correlation length for a system with more than two grain sizes are given by

$$\langle K_1 \rangle = \left(\sum_i \frac{v_i}{\varphi^3} \frac{D_i^3 K_i^2}{A_c^{3/2}} \right)^2, \quad (7)$$

$$L_{\text{ex}} = \left(\sum_i \frac{v_i}{\varphi^4} \frac{D_i^3 K_i^2}{A_c^2} \right)^{-1}, \quad (8)$$

where the index i accounts for each representative grain with identical magneto-crystalline anisotropy constant (K_i) and grain size (D_i), and v_i is the volume fraction of the grain i . If all the grains have a same magneto-crystalline anisotropy constant (K_1), then L_{ex} and $\langle K \rangle$ are given by using a distribution function of the grain volume ($f(V)$), or a distribution function that of the of the grain size ($f(D)$), as

$$L_{\text{ex}} = L_0^4 \left\{ \int_0^{V_m} V f(V) dV \right\}^{-1} = L_0^4 \left\{ \int_0^{D_m} D^3 f(D) dD \right\}^{-1}, \quad (9)$$

Fig. 1 Log-normal grain-size distribution function with different geometric standard deviation (σ_D) as a function of normalized grain size ($D/\langle D \rangle$, $\langle D \rangle$ is the mean grain size).

$$\langle K_1 \rangle = \frac{K_1}{L_0^6} \left\{ \int_0^{V_m} V f(V) dV \right\}^2 = \frac{K_1}{L_0^6} \left\{ \int_0^{D_m} D^3 f(D) dD \right\}^2, \quad (10)$$

where $V_m = D_m^3$ is the maximum grain volume. For simplicity, we assume further a log-normal distribution function (Fig. 1) as the grain-size or grain-volume distribution:¹⁷

$$f(D_r) = \frac{1}{\sqrt{2\pi\sigma_D D_r}} \exp\left(-\frac{\ln^2 D_r}{2\sigma_D^2}\right), \quad (11)$$

$$f(V_r) = \frac{1}{\sqrt{2\pi\sigma_V V_r}} \exp\left(-\frac{\ln^2 V_r}{2\sigma_V^2}\right), \quad (12)$$

where $D_r = D/D_0$ and $V_r = V/V_0$ are the reduced grain size and the volume, respectively, D_0 and V_0 are the medians, and σ_D and σ_V ($= 3\sigma_D$) are the geometric standard deviations for distributions. If $f(D_r)$ is negligible small at $D_r > D_m$, then D_m can be regarded as infinity and we obtain

$$\begin{aligned} L_{\text{ex}} &= L_0 \left(\frac{L_0}{D_0} \right)^3 \exp\left(-\frac{9}{2} \sigma_D^2\right) \\ &= L_0 \left(\frac{L_0}{\langle D \rangle} \right)^3 \exp(-3\sigma_D^2), \end{aligned} \quad (13)$$

$$\langle K_1 \rangle = K_1 \left(\frac{D_0}{L_0} \right)^6 \exp(9\sigma_D^2) = K_1 \left(\frac{\langle D \rangle}{L_0} \right)^6 \exp(6\sigma_D^2), \quad (14)$$

where

$$\langle D \rangle = \int_0^\infty D f(D) dD = D_0 \exp\left(\frac{\sigma_D^2}{2}\right) \quad (15)$$

is the mean grain size. The eqs. (13) and (14) indicate that L_{ex} decreases and $\langle K_1 \rangle$ increases with increasing σ_D , *i.e.*, soft magnetic properties of nanocrystalline alloys deteriorates with increasing σ_D , even if $\langle D \rangle$ is constant. It should be noted that this result is essentially established in other distribution functions.

2.3 Induced anisotropies

Naturally, the effective anisotropy in the nanocrystalline alloys may have contributions from induced anisotropies such as magneto-elastic anisotropy other than the random magnetocrystalline anisotropy and hence the effective anisotropy constant in actual materials is more correctly¹⁴⁾

$$\begin{aligned}\langle K \rangle &= \sqrt{K_u^2 + \langle K_1 \rangle^2} = \sqrt{K_u^2 + \frac{K_1^2}{N}} \\ &= \sqrt{K_u^2 + K_1^2 \left(\frac{D}{L_{\text{ex}}} \right)^3},\end{aligned}\quad (16)$$

where K_u is the induced uniaxial anisotropy energy density. Most of the nanocrystalline Fe-M-B alloys with good soft magnetic properties exhibit remanence ratio (J_r/J_s) of around 0.5. This means that the magnetization process of the alloys is mostly governed by the induced anisotropies, *i.e.*, $K_u > \langle K_1 \rangle$. Let us consider that L_{ex} is determined by K_u instead of $\langle K_1 \rangle$, *i.e.*,

$$L_{\text{ex}}^{\text{ua}} \approx \varphi \sqrt{\frac{A_c}{K_u}} = L_0 \sqrt{\frac{K_1}{K_u}}, \quad (17)$$

where $L_{\text{ex}}^{\text{ua}}$ is the exchange correlation length as the induced uniaxial anisotropy is dominant. In a limiting condition of $K_u^2 \gg \langle K_1 \rangle^2$ enables us to arrive at

$$\begin{aligned}\langle K \rangle &\approx K_u + \frac{\sqrt{K_u K_1}}{2L_0^3} \int_0^{D_m} D^3 f(D) dD \\ &= K_u + \frac{\sqrt{K_u K_1}}{2} \left(\frac{\langle D \rangle}{L_0} \right)^3 \exp(3\sigma_D^2).\end{aligned}\quad (18)$$

2.4 Two-phase random anisotropy model

In above calculations, the existence of the intergranular amorphous phase is not considered. However, in actual nanocrystalline alloys, the intergranular amorphous phase exists. The anisotropy energy density and the exchange stiffness constant (A_a) of the amorphous phase would be different from those of the crystalline phase. In order to consider the existence of the intergranular amorphous phase on the exchange correlation length, we adopted the model proposed by Herzer assuming that $A_a \approx A_c$.¹¹⁾ This assumption will be established when the volume fraction of the crystalline phase is sufficiently large.^{12,13)} We assumed that the grain-size distribution is given by the log-normal function, and have obtained the following results from eqs. (7)–(10) and (18) for the grains embedded in a matrix with $K_i = 0$:

$$\begin{aligned}L_{\text{ex}}^* &\approx \varphi^4 \frac{A_c^2}{v_c K_1^2} \left\{ \int_0^{D_m} D^3 f(D) dD \right\}^{-1} \\ &= \frac{L_0}{v_c} \left(\frac{L_0}{\langle D \rangle} \right)^3 \exp(-3\sigma_D^2),\end{aligned}\quad (19)$$

$$\begin{aligned}\langle K_1 \rangle^* &\approx \frac{v_c^2 K_1^4}{\varphi^6 A^3} \left\{ \int_0^{D_m} D^3 f(D) dD \right\}^2 \\ &= v_c^2 K_1 \left(\frac{\langle D \rangle}{L_0} \right)^6 \exp(6\sigma_D^2),\end{aligned}\quad (20)$$

$$\begin{aligned}\langle K \rangle^* &\approx K_u + v_c \frac{\sqrt{K_u K_1}}{2L_0^3} \int_0^{D_m} D^3 f(D) dD \\ &= K_u + v_c \frac{\sqrt{K_u K_1}}{2} \left(\frac{\langle D \rangle}{L_0} \right)^3 \exp(3\sigma_D^2),\end{aligned}\quad (21)$$

where v_c (≤ 1) is the volume fraction of the crystalline phase, L_{ex}^* and $\langle K_1 \rangle^*$ are the exchange correlation length and effective anisotropy constant as the induced anisotropy is negligible small, respectively, and $\langle K \rangle^*$ are the exchange correlation length and the effective anisotropy constant for $K_u^2 \gg \langle K_1 \rangle^2$. Under the condition of $v_c = 1$, eqs. (19)–(21) equal to eqs. (13), (14) and (18), respectively.

2.5 Coercivity

It is well known that coercivity (H_c) of materials generally depends on both saturation magnetization (J_s) and $\langle K \rangle$ and can be expressed as

$$H_c = p_c \frac{\langle K \rangle}{J_s}, \quad (22)$$

where p_c is a constant. If $K_u^2 \ll \langle K_1 \rangle^2$, then $\langle K \rangle$ in eq. (22) is given by $\langle K_1 \rangle^*$ of eq. (20) (or $\langle K_1 \rangle$ of eq. (14)). For $K_u^2 \gg \langle K_1 \rangle^2$, $\langle K \rangle$ in eq. (22) is given by the second part of $\langle K \rangle^*$ of eq. (21) (or $\langle K \rangle$ of eq. (18)) if K_u is coherent in space or if its spatial fluctuations are negligible to $\langle K_1 \rangle^*$ or $\langle K_1 \rangle$.¹⁴⁾

Figure 2 shows the normalized coercivity ($H_c(\sigma_D)/H_c(0)$) as a function of σ_D . The upper line shows H_c for K_u is negligible small. The lower line indicates H_c when coherent K_u is dominant. It should be noted that the same result for dependence of the reduced coercivity on σ_D is obtained for the one-phase RAM and for the two-phase one. The coercivity varies as $\exp(3n\sigma_D^2)$, where $n = 1$ for $K_u^2 \gg \langle K_1 \rangle^2$ and $n = 2$ for $K_u^2 \ll \langle K_1 \rangle^2$. At $\sigma_D = 0.5$,

Fig. 2 Normalized coercivity as a function of geometric standard deviation of grain-size distribution.

$H_c(\sigma_D)/H_c(0)$ reaches about 4.5 for $K_u^2 \ll (K_1)^2$ and about 2.1 for $K_u^2 \gg (K_1)^2$.

3. Experimental Procedure

$\text{Fe}_{84}\text{Nb}_7\text{B}_9$, $\text{Fe}_{85}\text{Nb}_6\text{B}_9$ and $\text{Fe}_{84.9}\text{Nb}_6\text{B}_8\text{P}_1\text{Cu}_{0.1}$ alloy ingots were prepared by arc melting a mixture of pure Fe, Nb and Cu metals, pre-alloyed Fe-P ingot and pure B crystal in an Ar atmosphere. A single-roller melt-spinning method in air or an Ar atmosphere was used to produce rapidly solidified ribbons with about 1 mm in width and 19–21 μm in thickness. The as-quenched ribbons were wound into a toroidal shape with 5 mm in inner diameter and 6 mm in outer diameter to be used as samples. Annealing treatment of the samples was carried out by treating the samples at 923 K for 300 s in a vacuum and the heating rate was 3 K/s.

The as-quenched and annealed structures were examined by X-ray diffractometry (XRD) with Co $K\alpha$ incident radiation and transmission electron microscopy (TEM). The samples for TEM observation were prepared by ion-milling from both sides of the ribbons. The saturation magnetization (J_s) was measured by a vibrating sample magnetometer (VSM) under a maximum magnetic field of 0.8 MA/m. The permeability (μ_e) was measured by a vector impedance analyzer at 1 kHz under a field of 0.4 A/m. The coercivity (H_c) and the remanence (J_r) were measured by a DC B - H loop tracer under a maximum magnetic field of 0.8 kA/m.

4. Experimental Results

Table 1 shows the mean grain size ($\bar{D}$), J_s , J_r/J_s , μ_e and H_c after the crystallization of the alloys. The mean grain size of the three alloys is almost the same. However, the soft magnetic properties in a crystallized state of the alloys are very different. The $\text{Fe}_{84}\text{Nb}_7\text{B}_9$ and $\text{Fe}_{84.9}\text{Nb}_6\text{B}_8\text{P}_1\text{Cu}_{0.1}$ exhibit high μ_e (36000 and 41000, respectively) and low H_c (5.6 A/m and 4.7 A/m, respectively). On the other hand, the soft magnetic properties of the $\text{Fe}_{85}\text{Nb}_6\text{B}_9$ alloy ($\mu_e = 14,000$ and $H_c = 10.5$ A/m) are inferior to those of the other alloys.

Figure 3 shows TEM images selected area electron diffraction (SAED) patterns of the as-quenched and crystallized alloys. The mean grain size and the distribution for the crystallized alloys evaluated by counting the α -Fe grains in the TEM images are shown in Fig. 4. The $\text{Fe}_{84}\text{Nb}_7\text{B}_9$ alloy has a single amorphous structure in an as-quenched state as shown in Fig. 3(a) and exhibits the narrow grain-size distribution in a crystallized state as shown in Figs. 3(b) and 4(a). On the contrary, the $\text{Fe}_{85}\text{Nb}_6\text{B}_9$ alloy has a mixed

as-quenched structure composed of an amorphous phase and α -Fe grains with 10–25 nm in size as shown in Fig. 3(b).^{15,16} These grains grow to a size of about 40 nm during the annealing treatment, and remain in the nanostructure after the crystallization as shown in Figs. 3(d) and 4(b). On the other hand, the simultaneous replacement of B by 1 at% P and Fe by 0.1 at% Cu for the $\text{Fe}_{85}\text{Nb}_6\text{B}_9$ alloy decreases the α -Fe grain size to nano-scale in an as-quenched state as shown in Figs. 3(e) and 3(f).^{15,16} Consequently, as shown in Figs. 3(g) and 4(c), the crystallized $\text{Fe}_{84.9}\text{Nb}_6\text{B}_8\text{P}_1\text{Cu}_{0.1}$ alloy exhibits the narrow grain-size distribution comparable to that of the $\text{Fe}_{84}\text{Nb}_7\text{B}_9$ alloy.

The volume fraction of the α -Fe phase (v_c) was estimated from the ratio of the integral intensity of the crystalline contribution to the total intensity of the XRD peak.¹⁸ In order to separate the contribution of the amorphous and α -Fe phase, a fitting procedure of the diffraction peaks was preformed. Both the first amorphous halo and the (110) diffraction peak of the α -Fe phase were fitted by means of the pseudo-Voigt function:

$$pV(x) = I_0\{n_L L(x) + (1 - n_L)G(x)\}, \quad (23)$$

where $L(x)$ and $G(x)$ are Lorentzian and Gaussian functions, respectively, I_0 represents the peak intensity and n_L is the Lorentzian contribution to the diffraction peak. The variable x is defined as

$$x = \frac{2\theta - 2\theta_0}{w}, \quad (24)$$

where 2θ is the angular diffraction variable, $2\theta_0$ is the peak's location and w is the peak's half width at half maximum. Figure 5 shows the results of the fitting procedure and v_c of the alloys. The amorphous halo turns out to be fitted by a nearly pure Lorentzian function (*i.e.*, $n_L \approx 1$) in all the cases, whereas the crystalline peaks show a Gaussian contribution with $(1 - n_L) \approx 0.3$. Obtained v_c values are about 0.7, which is consistent with the earlier observed value for nanocrystalline $\text{Fe}_{83}\text{Nb}_7\text{B}_9\text{Cu}_1$ alloy by Mössbauer analysis.¹⁹

5. Discussion

In this section we analyze the coercivity of the nanocrystalline Fe-Nb-B(-P-Cu) alloys with the different grain-size distribution on the basis of the two-phase RAM described in section 2.4. As shown in Table 1, J_r/J_s of the alloys is about 0.5. This indicates that the magnetization process of the alloys is mostly governed by the induced anisotropies as described in section 2.3. Therefore, we chose the two-phase RAM with the induced anisotropies, *i.e.*, eqs.

Table 1 Mean grain size ($\bar{D}$), saturation magnetization (J_s), remanence ratio (J_r/J_s), permeability (μ_e) and coercivity (H_c) in a crystallized state for the Fe-Nb-B(-P-Cu) alloys. As-quenched structure of the alloys is also shown.

	As-quenched	Crystallized				
	Structure	$\bar{D}^*/\text{nm}$	J_s/T	J_r/J_s^{-1}	μ_e^{**}	$H_c/\text{A}\cdot\text{m}^{-1}$
$\text{Fe}_{84}\text{Nb}_7\text{B}_9$	fully amorphous	8.9	1.52	0.48	36,000	5.6
$\text{Fe}_{85}\text{Nb}_6\text{B}_9$	amorphous + α -Fe	10.5	1.62	0.44	14,000	10.5
$\text{Fe}_{84.9}\text{Nb}_6\text{B}_8\text{P}_1\text{Cu}_{0.1}$	amorphous + nano α -Fe	8.6	1.61	0.48	41,000	4.7

* Evaluated from TEM images.

** 1 kHz, 0.4 A/m.

Fig. 3 (a)–(e), (g) TEM images and selected-area electron diffraction (SAED) patterns, (f) high-resolution TEM image and nano-beam ED pattern of as-quenched and crystallized Fe-Nb-B(-P-Cu) alloys.

(17) and (21). Table 2 shows the parameters used in the analysis. Here, L_0 ($= 37$ nm) is determined by the upper limit of the grain size in which H_c of the nanocrystalline Fe-M-B alloys is proportional to $D^{6/3}$, K_1 ($= 47$ kJ/m³) is the value for pure α -Fe and p_c ($= 0.64$) is adopted the theoretical value for cubic particles oriented at random.²⁰⁾

The solid curves in Fig. 6 show the fitting results of the grain-size distribution by using the log-normal distribution function (eq. (11)). The log-normal distribution function reproduces well the observed grain-size distribution. The $\text{Fe}_{84}\text{Nb}_7\text{B}_9$ and $\text{Fe}_{84.9}\text{Nb}_6\text{B}_8\text{P}_1\text{Cu}_{0.1}$ alloys exhibit the almost

same $\langle D \rangle$ ($= 9.1$ and 8.7 nm, respectively) and σ_D ($= 0.26$ for both the alloys). The fitting result for the $\text{Fe}_{85}\text{Nb}_6\text{B}_9$ alloy by using the grains less than 35 nm in size yields $\langle D \rangle = 10.0$ nm and $\sigma_D = 0.23$, which are close to those of the other alloys. In order to reproduce the bimodal grain-size distribution of the $\text{Fe}_{85}\text{Nb}_6\text{B}_9$ alloy more correctly, we consider the bimodal distribution function expressed by superimposing the two log-normal distribution functions with the different medians (D_0 and $d_b D_0$, $d_b > 1$) and the geometric standard deviations (σ_D and σ_b) as follows:

Fig. 4 Grain-size distribution and mean grain size ($\bar{D}$) evaluated by counting the α -Fe grains in transmission electron micrographs for crystallized Fe-Nb-B(-P-Cu) alloys.

Fig. 5 X-ray diffraction profiles of crystallized Fe-Nb-B(-P-Cu) alloys. The lines indicate fitting pseudo-Voigt functions of nanocrystalline and amorphous phases. For graphic reasons, only one experimental points every five is actually shown. The best-fit lines were obtained by considering all data points. The volume fractions of nanocrystalline phase (v_c) are also shown.

$$f_b(D_r) = \frac{1}{\sqrt{2\pi}\{r_b(d_b^3 - 1) + 1\}D_r} \left[\frac{1 - r_b}{\sigma_D} \exp\left(-\frac{\ln^2 D_r}{2\sigma_D^2}\right) + \frac{r_b d_b^3}{\sigma_b} \exp\left\{-\frac{\ln^2(D_r/d_b)}{2\sigma_b^2}\right\} \right], \quad (25)$$

where r_b is the ratio of the distribution function for the large gains to that of the small grains and

$$\int_0^\infty D^3 f_b(D) dD = \frac{D_0^3}{r_b(d_b^3 - 1) + 1} \left\{ (1 - r_b) \exp\left(\frac{9}{2} \sigma_D^2\right) + r_b d_b^6 \exp\left(\frac{9}{2} \sigma_b^2\right) \right\}. \quad (26)$$

The volume fraction of the large grains (v_l) is given by

$$\frac{v_l}{v_c} = r_b d_b^6 \exp\left(\frac{9}{2} \sigma_b^2\right) \left\{ (1 - r_b) \exp\left(\frac{9}{2} \sigma_D^2\right) + r_b d_b^6 \exp\left(\frac{9}{2} \sigma_b^2\right) \right\}^{-1}. \quad (27)$$

Figure 7 shows the normalized coercivity ($H_c(v_l/v_c)/H_c(0)$) as a function of the volume fraction of the large grains (v_l/v_c). With increasing D , then H_c is rapidly increases as D^3 or D^6 . However, H_c gradually increases first with increasing v_l/v_c , and rapidly increases around $v_l/v_c \approx 0.8$ as shown in

Fig. 7. Therefore, it can be said that the influence of the coarse grains on H_c is relatively small if v_l/v_c is small. The fitting result by using eq. (25) yields $d_b = 4.3$, $\sigma_b = 0.050$ and $r_b = 1.6 \times 10^{-4}$. Here, r_b was determined so that the calculated v_l/v_c agrees with the experimental one ($= 0.44$)

Table 2 Summary of materials parameters (L_0 : intrinsic exchange correlation length, K_1 : magnetocrystalline anisotropy constant for crystalline phase, p_c : prefactor for coercivity) used in this analysis.

L_0/nm	37
$K_1/\text{kJ}\cdot\text{m}^{-3}$	47
p_c	0.64

Fig. 6 Grain-size distribution of crystallized Fe-Nb-B-(P-Cu) alloys. The histograms were obtained from TEM images. The lines indicate fitting (a), (b) and (d) unimodal or (c) bimodal log-normal distribution functions. The inset in (c) is an enlarged view within 30–50 nm grain size. The fitting parameters are also shown.

obtained from the histogram shown in Fig. 4(b).

The induced anisotropy constant is determined by the experimental value of H_c ($= 5.6 \text{ A/m}$) of the $\text{Fe}_{84}\text{Nb}_7\text{B}_9$ alloy. The obtained K_u is 104 J/m^3 and it is assumed that the other alloys have the same value. These values give $L_{\text{ex}}^{\text{ua}}$ (eq. (17)) of about $0.8 \mu\text{m}$. The integrals in eqs. (21) and (26) converge around $D_r \approx 5$ ($D \approx 45 \text{ nm}$), which is sufficiently

Fig. 7 Normalized coercivity as a function of volume fraction of large grains.

smaller than $L_{\text{ex}}^{\text{ua}}$. Therefore, following two assumptions are adequate: one is to regard the upper limit of the integral range as infinity, and the other is that the maximum grain size is smaller than $L_{\text{ex}}^{\text{ua}}$.

Tables 3 and 4 summarize the obtained parameters. The effective anisotropy constant ($\langle K_1 \rangle^*$) given by eq. (20) are also shown in Table 4 in order to confirm whether it has satisfied $K_u^2 \gg \langle K_1 \rangle^{*2}$. The calculated coercivity ($H_c^{\text{calc.}} = 4.9 \text{ A/m}$) with the unimodal log-normal distribution function for the $\text{Fe}_{84.9}\text{Nb}_6\text{B}_8\text{P}_1\text{Cu}_{0.1}$ alloy is in good agreement with the experimental value ($H_c^{\text{mes.}} = 4.7 \text{ A/m}$). On the other hand, $H_c^{\text{calc.}} (= 7.0 \text{ A/m})$ for the $\text{Fe}_{85}\text{Nb}_6\text{B}_9$ alloy is 2/3 times as large as $H_c^{\text{mes.}} (= 10.5 \text{ A/m})$. This large difference originates in disregarding the existence of the coarse grains with about 40 nm in size. The calculated coercivity ($= 12.3 \text{ A/m}$) is consistent with the experimental one by considering the existence of the coarse grains. Furthermore,

Table 3 Summary of obtained volume fraction (v_c) of crystalline phase and parameters for log-normal grain-size distribution function.

	$\text{Fe}_{84}\text{Nb}_7\text{B}_9$	$\text{Fe}_{85}\text{Nb}_6\text{B}_9$	$\text{Fe}_{84.9}\text{Nb}_6\text{B}_8\text{P}_1\text{Cu}_{0.1}$
v_c	0.66	0.70	0.71
$\langle D \rangle / \text{nm}$	9.1	10.0	8.7
σ_D	0.26	0.23	0.26
d_b	—	4.3	—
σ_b	—	0.050	—
r_b	—	1.6×10^{-4}	—

Table 4 Summary of calculated magnetic anisotropy energy densities, effective exchange correlation length and coercivity.

$f(D)$	Fe ₈₄ Nb ₇ B ₉	Fe ₈₅ Nb ₆ B ₉		Fe _{84.9} Nb ₆ B ₈ P ₁ Cu _{0.1}	
	unimodal LN*	unimodal LN*	bimodal LN* ($r_b = 1.6 \times 10^{-4}$)	bimodal LN* ($r_b = 1.0 \times 10^{-4}$)	unimodal LN*
$K_u/\text{J}\cdot\text{m}^{-3}$	104	104	104	104	104
$\langle K_1 \rangle^*/\text{J}\cdot\text{m}^{-3}$	7	12	37	27	6
$\langle K \rangle^*/\text{J}\cdot\text{m}^{-3}$	117	122	135	130	116
$L_{\text{ex}}^{\text{ua}}/\mu\text{m}$	0.79	0.79	0.79	0.79	0.79
$H_c^{\text{calc.}}/\text{A}\cdot\text{m}^{-1}$	5.6	7.0	12.3	10.5	4.9

*LN: Log-normal distribution function.

$H_c^{\text{calc.}} = 10.5 \text{ A/m}$ is obtained by using $r_b = 1.0 \times 10^{-4}$. This gives the rather small v_l/v_c of 0.34. This value seems to be in the range of the experimental error because it is difficult to accurately obtain the distribution of the coarse grains with extremely small number.

A possible source of the induced anisotropy is magneto-elastic anisotropy:

$$K_u^{\text{el}} = -\frac{3}{2} \lambda \sigma_r, \quad (28)$$

where λ is the saturation magnetostriction constant and σ_r is the residual stress. The total magnetostriction of nanocrystalline alloys is given as¹¹⁾

$$\lambda_t = v_c \lambda_c + (1 - v_c) \lambda_a, \quad (29)$$

where λ_c and λ_a are the saturation magnetostriction constant of the nanocrystalline and amorphous phases, respectively. By using $\lambda_t = 0.1 \times 10^{-6}$ and $\lambda_c = -4 \times 10^{-6}$, then $\lambda_a = 8 \times 10^{-6}$ is obtained for the Fe₈₄Nb₇B₉ alloy.³⁾ In order to explain the obtained K_u value ($= 104 \text{ J/m}^3$), $|\sigma_r| \approx 17$ and 9 MPa for the nanocrystalline and amorphous phases are required, respectively. Polak *et al.* estimated σ_r and its wavelength of the nanocrystallized Fe_{73.5}Cu₁Nb₃Si_{13.5}B₉ alloy from the pinning field and the external stress dependence of H_c to be 20–60 MPa and to be 200–600 μm , respectively.²¹⁾ They also reported that the residual stress of the as-quenched alloy is 60–200 MPa and its wavelength is much larger than that of the nanocrystallized alloys. These results imply that it is impossible to perfectly remove the quenched-in stresses by the crystallization. The range of σ_r values estimated by Polak *et al.* are of the same order of magnitude as our estimated values. Furthermore, the wavelength of σ_r estimated by Polak *et al.* is sufficiently longer than L_{ex} whose typical value is about 2 μm .¹¹⁾ This means that the magneto-elastic anisotropy is not averaged out. It is possible that the residual stresses with the sufficiently long wavelength exist in the nanocrystallized Fe-Nb-B(-P-Cu) alloys. Therefore, the residual stress seems to be one of the origins of the coherent induced anisotropies.

6. Conclusions

The effect of grain-size distribution on coercivity (H_c) of nanocrystalline soft magnetic alloys has been investigated based on the random anisotropy model (RAM). The obtained results are summarized as follows.

(1) A simple model considering the grain-size distribution is proposed based on RAM. This model gives the similar

results of the original RAM. However, H_c increases with increasing width of the grain-size distribution, *i.e.*, soft magnetic properties of nanocrystalline alloys deteriorates with increasing the width of the grain-size distribution, even if the mean grain size is constant.

(2) The effect of the coarse grains in nanostructure on soft magnetic properties has been discussed. The analytical results show that the existence of the coarse grains causes the increase in H_c . However, that the influence is relatively small if the volume fraction of the coarse grains is small.

(3) Our model explains well the dependence of H_c for the nanocrystalline Fe-Nb-B(-P-Cu) alloys on the grain-size distribution.

(4) The magnetization process of the nanocrystalline Fe-Nb-B(-P-Cu) alloys is mostly governed by the induced anisotropies. A possible source of the induced anisotropy is magneto-elastic anisotropy due to the residual stress introduced during the quenching process.

These results suggest that one should pay attention on not only the mean grain size but also on the grain-size distribution since the inhomogeneity of the grain size increases H_c .

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