Critical phenomenon in soft magnetic properties of giant magnetostrictive materials: Computer simulation

Hirotooshi Fukunaga^{a)} and Takayuki Yamaguchi

Graduate School of Science and Technology, Nagasaki University, Nagasaki 852, Japan

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Dependence of magnetostrictive and magnetic properties on grain size was calculated numerically for an isotropic giant magnetostrictive material. The model magnet assumed is composed of 8000 cubic $(\text{Tb}_{0.3}\text{Dy}_{0.7})\text{Fe}_2$ grains. The direction of magnetization and the magnetostriction of each grain was determined for a given applied field under the consideration of the anisotropy, exchange, magnetoelastic, elastic, and Zeeman energies. Subsequently, the hysteresis loop and the magnetostriction versus applied field curve was obtained by varying the applied field. With decreasing the grain size D , the coercivity H_c and $d\lambda/dH$, where λ is the magnetostriction, decreased abruptly at $D=25$ nm. This improvement in soft magnetic properties exceeded the expected improvement from Herzer's calculation, in which H_c was proportional to D^6 . In order to study the significant improvement in detail, we calculated magnetic and magnetostrictive properties with varying magnetostriction and anisotropy constants. Resultantly, it was clarified that the critical phenomenon occurs when the total of elastic and magnetoelastic energies exceed the magnetic anisotropy energy in magnitude. © 2006 American Institute of Physics. [DOI: 10.1063/1.2176311]

Magnetostrictive materials require both giant saturation magnetostriction and soft magnetic properties. Although the cubic Laves phase TbFe_2 has the largest magnetostriction at room temperature to date,¹ it is magnetically hard because of its large crystalline magnetic anisotropy constant $K_{c1} = -6.3$ MJ/m³. Clark *et al.* have succeeded in decreasing K_{c1} without a large reduction in saturation magnetostriction by the replacement of a part of Tb with Dy, and Terfenol-D [$(\text{Tb}_{0.3}\text{Dy}_{0.7})\text{Fe}_2$] is widely known as one of the best magnetostrictive materials.¹

Recently, it was reported that a refinement of the grain size improves soft magnetic properties of Fe–Si–Nb–Cu–B alloys significantly,^{2,3} and a similar effect has been confirmed experimentally for magnetostrictive alloys.^{4–7} However, the details of the relationship between grain size and soft magnetic properties are left unknown because of the difficulties in controlling the size and alignment of $(\text{Tb}_{0.3}\text{Dy}_{0.7})\text{Fe}_2$ grains.

In this contribution, we numerically studied the dependence of magnetostrictive and magnetic properties on grain size for isotropic polycrystalline $(\text{Tb}_{0.3}\text{Dy}_{0.7})\text{Fe}_2$ alloy by computer simulation, and found that soft magnetic properties are improved abruptly when the grain size is smaller than a critical value. This critical phenomenon was discussed in terms of energies stored in the material.

The model magnet assumed is composed of 8000 cubic grains in the single domain state as shown in Fig. 1. Each grain has the cubic magnetic anisotropy, and its crystal orientation was distributed randomly. Thus, the model is an isotropic polycrystalline magnet. The magnetization of a grain is coupled to those of six neighboring grains by the exchange interaction. The periodic boundary condition was used in the three directions.

The direction of magnetization and the magnetostriction of each grain as determined for a given applied field by minimizing the total energy stored in the model magnet. The total energy W_{total} is given by the sum of the anisotropy, Zeeman, exchange, elastic, and magnetoelastic energies. The magnetostatic energy was neglected in this study, because its effect would be small compared to the other ones.

The anisotropy energy W_a is written as

$$W_a = V \sum_i K_{c1} (\alpha'_{1i}{}^2 \alpha'_{2i}{}^2 + \alpha'_{2i}{}^2 \alpha'_{3i}{}^2 + \alpha'_{3i}{}^2 \alpha'_{1i}{}^2), \quad (1)$$

where V and K_{c1} are the volume of the grain and the crystalline magnetic anisotropy constant, respectively. Furthermore, $\alpha'_i (\alpha'_{1i}, \alpha'_{2i}, \alpha'_{3i})$ is the direction cosine vector of magnetization vector projected to the crystal axes of the i th grain. The exchange energy W_{ex} can be expressed by

$$W_{\text{ex}} = -V \sum_i \frac{A}{D^2} \left(\sum_j^6 \alpha_i \cdot \alpha_j \right), \quad (2)$$

where A is the exchange stiffness constant, D is the grain size, and $\alpha_i (\alpha_{1i}, \alpha_{2i}, \alpha_{3i})$ is the direction cosine vector laying in the direction of the magnetization vector.

FIG. 1. Model magnet assumed. The magnet is composed of 8000 cubic $(\text{Tb}_{0.3}\text{Dy}_{0.7})\text{Fe}_2$ grains.

^{a)}Electronic mail: fukunaga@net.nagasaki-u.ac.jp

TABLE I. Material parameters used in calculation.

Material constants	(Tb _{0.3} Dy _{0.7})Fe ₂	Alloy 1	Alloy 2-5	Alloy 6
K_{c1} ($\times 10^5$ J/m ³)	5.48	5.48	0.0 to 10.96	5.48
I_s (T)	1.0	1.0	1.0	...
$\lambda_{100}, \lambda_{111}$ (ppm)	90, 1640	0, 0	variable	1000, 1000
c_{11}, c_{12}, c_{44} ($\times 10^{10}$ N/m)	14.1, 6.48, 4.87	...	14.1, 6.48, 4.87	9.74, 0.0, 4.87
A ($\times 10^{-11}$ J/m)	2.5	2.5	...	...
D (nm)	variable	variable	...	...

In this study, all the grains were assumed to have the same magnetostriction value for a given applied field. Under this assumption, the magnetostriction is expressed by the tensor $\mathbf{e}(e_{xx}, e_{yy}, e_{zz}, e_{xy}, e_{yz}, e_{zx})$. The elastic energy W_{elastic} and the magnetoelastic energy W_{magnet} are written as

$$W_{\text{elastic}} = V \sum_i \left[\frac{1}{2} c_{11} (e'_{xxi}{}^2 + e'_{yyi}{}^2 + e'_{zz i}{}^2) + \frac{1}{2} c_{44} (e'_{xyi}{}^2 + e'_{yzi}{}^2 + e'_{zxi}{}^2) + c_{12} (e'_{yyi} e'_{zz i} + e'_{zz i} e'_{xx i} + e'_{xx i} e'_{yy i}) \right], \quad (3)$$

$$W_{\text{magnet}} = V \sum_i \left\{ B_1 \left[e'_{xxi} \left(\alpha'_{1i}{}^2 - \frac{1}{3} \right) + e'_{yyi} \left(\alpha'_{2i}{}^2 - \frac{1}{3} \right) + e'_{zz i} \left(\alpha'_{3i}{}^2 - \frac{1}{3} \right) \right] + B_2 (e'_{xyi} \alpha'_{1i} \alpha'_{2i} + e'_{yzi} \alpha'_{2i} \alpha'_{3i} + e'_{zxi} \alpha'_{3i} \alpha'_{1i}) \right\}, \quad (4)$$

where c_{11} , c_{12} , and c_{44} are the elastic constants, and $\mathbf{e}'_i$ is the magnetostriction tensor projected to the crystal axes of the i th grain. Furthermore, B_1 and B_2 are the magnetoelastic coupling constants determined from the magnetostrictive constants, λ_{100} and λ_{111} , and the elastic constants.¹ The total of W_{elastic} and W_{magnet} is designated as the magnetostrictive energy $W_{\text{striction}}$.

From the above equations, it is clarified that total energy W_{total} is a function of α_i and $\mathbf{e}$ for given values of D and applied field. Therefore, we get the simultaneous equations for α_i and $\mathbf{e}$ by minimization of W_{total} . In our calculation, α_i of each grain were determined by using Gilbert's equation.⁸

The assumed material parameters are listed in Table I. Alloys 1-6 are made-up materials to make the discussion more clear.

Figure 2 shows typical λ - H loops for (Tb_{0.3}Dy_{0.7})Fe₂, where λ and H are the magnetostriction and applied field, respectively. It is clearly seen that soft magnetic properties

FIG. 2. Magnetostriction vs applied field for a (Tb_{0.3}Dy_{0.7})Fe₂ magnet composed by 100 nm (broken line) and 20 nm (solid line) grains.

depend on the grain size D . Therefore, we calculated hysteresis and λ - H loops with varying D , and the coercivity H_c and $(d\lambda/dH)_{\text{max}}$ are shown in Fig. 3 as a function of D , together with H_c of alloy 1 which is nonmagnetostrictive, where $(d\lambda/dH)_{\text{max}}$ is the maximum value of $d\lambda/dH$ in a λ - H loop, and λ_s is the saturation magnetostrictive constant for polycrystalline material defined as $\lambda_s = (2\lambda_{100} + 3\lambda_{111})/5$. As shown in the figure, H_c and $(d\lambda/dH)_{\text{max}}$ vary abruptly at $D = 25$ nm for (Tb_{0.3}Dy_{0.7})Fe₂, and soft magnetic properties of (Tb_{0.3}Dy_{0.7})Fe₂ are improved significantly for $D \leq 25$ nm. On the other hand, H_c of alloy 1 varies proportionally to D^6 between $D=10$ and 25 nm, which agrees with Herzer's predication neglecting magnetostriction.² Consequently, the critical phenomenon indicated for (Tb_{0.3}Dy_{0.7})Fe₂ is expected to originate from magnetostriction.

In order to study the relationship between the magnetostrictive and magnetic anisotropy energies, we calculated H_c with varying the magnetostriction and crystalline magnetic anisotropy constants, and the results are shown in Fig. 4 for alloys 2-5 and listed in Table I. For each alloy shown in Fig. 4, λ_s were varied under the condition of the constant $\lambda_{100}/\lambda_{111}$ value which is 90/1640. In addition, we neglected the exchange energy, which corresponds to the assumption of a large enough grain size. As shown in Fig. 4, H_c increases proportional to λ_s^2 and decreases abruptly at a critical λ_s value. After the abrupt decrease in H_c , the variation of H_c agreed with that of alloy 2 which does not have magnetic anisotropy. The chain line in Fig. 4 is the boundary between two regions: regions I and II. In region II, the effect of the magnetic anisotropy on H_c is suppressed by magnetostriction, while H_c is affected by a K_{c1} value in region I.

We compared the magnetic anisotropy energy W_a with the magnetostrictive energy $W_{\text{striction}}$, and found that $W_{\text{striction}}$ in the magnetic saturation state is larger than the magnetic

FIG. 3. Grain size dependence of coercivity and $d\lambda/dH$. The open circles indicate the results for alloy 1.

FIG. 4. Dependence of coercivity on saturation magnetostriction λ_s for alloys 2–5. K_{cl} values of these alloys are 0, 2.74, 5.48, and 10.96×10^5 J/m³, respectively. The saturation magnetostriction is reduced by the value for $(\text{Tb}_{0.3}\text{Dy}_{0.7})\text{Fe}_2$ which is 1020 ppm.

anisotropy energy in region II. This result resembles the variation of H_c of alloy 1 as shown in Fig. 3, for which soft magnetic properties are improved when the exchange energy is dominant to the anisotropy energy.

The variation of soft magnetic properties for alloy 1, as shown in Fig. 3, can be attributed to the exchange interaction that affects the directions of the neighbor magnetization and aligns them parallel to each other. Considering this effect, the results shown in Figs. 3 and 4 can be explained by assuming a similar effect to the exchange interaction for magnetostriction. Under this assumption, the critical decrease in H_c can be explained as follows. In the case of alloy 1, the exchange interaction has a tendency of aligning the direction of each grain and reduces the “effective” anisotropy energy with decreasing the grain size. In the case of $(\text{Tb}_{0.3}\text{Dy}_{0.7})\text{Fe}_2$, on the other hand, the magnetostriction boosts the effect of the exchange interaction. Consequently, the magnetostrictive energy becomes dominant to the anisotropy energy at $D = 25$ nm, and soft magnetic properties are improved significantly.

To confirm the above effect, we calculated $W_{\text{striction}}$ stored in two neighbor grains as shown in Fig. 5. The magnetization of grain 1 was fixed to the $[100]$ direction, and the direction of the magnetization in grain 2, α_2 , was varied, and the stored energy was calculated. The result is shown as a function of α_2 in Fig. 5. In the calculation, we assumed an isotropic magnetostrictive alloy and its material parameters are listed in Table I under alloy 6. As seen in Fig. 5, the minima of $W_{\text{striction}}$ lie down in the $[100]$ and $[-100]$ directions, while the maxima lie perpendicularly to them. This result suggests that the magnetostrictive energy affects the directions of magnetization and aligns them to the same or opposite direction and that it can boost the effect of the exchange energy.

To confirm the above-mentioned effect of magnetostriction for alloys with anisotropic magnetostriction, we calculated the average of $|\alpha_i \cdot \alpha_j|$ for all the grains of two alloys which are categorized in alloy 4 and have the same K_{cl} value and different λ_s values ($\lambda_s = 510$ and 1530 ppm). The calculated average of $|\alpha_i \cdot \alpha_j|$ for $\lambda_s = 1530$ ppm was larger than

FIG. 5. Calculated distribution of magnetostrictive energy $W_{\text{striction}}$. The energy is shown as a function of the direction of magnetization of grain 2. The inset shows the calculation model.

that for $\lambda_s = 510$ ppm at any given applied field, which is consistent with the results shown in Fig. 5 and suggests that anisotropic magnetostriction has an effect, that is, aligning magnetization into the same direction.

We have calculated the dependence of magnetostrictive and magnetic properties on grain size and found a unique phenomenon for isotropic materials with giant magnetostriction. Coercivity H_c and $d\lambda/dH$ varied abruptly at a critical grain size, and soft magnetic properties were achieved for a grain size smaller than the critical value. This critical phenomenon occurred when the magnetostrictive energy was larger than magnetic anisotropy energy and was explained by the effect that magnetostriction has a tendency of aligning magnetization into the same direction and boosting the effect of exchange interaction.

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