Relation between domain wall motion and coercivity in Nd-Fe-B sintered magnets prepared in various conditions

To cite this article: K Kobayashi et al 2009 IOP Conf. Ser.: Mater. Sci. Eng. 1 012035

View the article online for updates and enhancements.
Relation between domain wall motion and coercivity in Nd-Fe-B sintered magnets prepared in various conditions

K. Kobayashi¹, Y. Ochiai¹, K. Hayakawa¹, and M. Sagawa²

¹Department of Materials and Life Science, Shizuoka Institute of Science and Technology, Toyosawa 2200-2, Fukuroi, Shizuoka 437-8555, Japan
²Intermetallics Co., Ltd., 309 Kyodai Venture Plaza, I-36 Ohara-Goryoh, Nishikyo-ku, Kyoto 615-8245, Japan

E-mail: Koba@ms.sist.ac.jp

Abstract. For the study of the relationship between the domain wall motion (DWM) and the coercivity in the samples of Dy-free Nd-Fe-B sintered magnets, the DC susceptibility (χ_Dc) in the initial magnetization from the thermally demagnetized state and the DWM in the partially demagnetized state after the full magnetization in 5T-pulse field were measured. In the high coercivity (μ_Hc) samples of μ_Hc=1.35T and 1.62T, the stronger pinning in initial magnetization was detected as the χ_Dc compared to other samples of the smaller coercivities such as μ_Hc=0.80T. The former samples also showed little DWM in the partially demagnetized state even the domain structures were clearly detected by magneto-force microscopy (MFM). In the latter samples, however, the obvious DWM was measured as the differences (ΔM) between the magnetizations under applied field (M_a) and the remanences (M_r) in demagnetization process (ΔM = M_r - M_a). The pinning in DWM in initial magnetization process and the DWM in the partially demagnetized state are both originated from the interaction between the domain walls and grain boundary phases. The enhancement in coercivity, therefore, should come from the same interactions in the sintered samples.

1. Introduction

The ordinary theories of the coercivity of permanent magnets are based on the magnetic reversal mechanism of single-domain particles as the Stoner-Wohlfarth model (1). The authors have been studied the coercivity of Nd-Fe-B sintered magnets by the domain structure observations using MOKE and MFM (2,3). It was revealed in these studies that the magnetic reversal in the sintered magnets was not only developed by the summation of magnetizations in magnetic reversed, nucleated particles and/or crystal grains, but also the formation of regions composed by the groups of crystal grains where the domain structure would be reformed in zero applied field (3,4).

The domain motion, therefore, is very important even in the magnets categorized into the nucleation type in coercivity appearance mechanism. In this study, the domain wall motion (DWM) was studied by the DC susceptibility measurements and the measurements of differences between the magnetization under the maximum applied fields and the remanences in each minor loop that correspond to the DWM. The former method was applied to the initial susceptibility measurements

¹ To whom any correspondence should be addressed.
from the thermally demagnetized state, and the latter method was conducted in the demagnetization process from once fully magnetized state in the 2nd and 3rd quadrants in the hysteresis loops.

The pinning forces for the DWM in the initial susceptibilities, and the magnetizations correspond to the DWM show the clear relationship to the coercivities of the samples. The comparatively large coercivity appeared in the sample showed the strong pinning in the initial DWM and small DWM in the demagnetization process.

2. Experimental

2.1 Samples

The samples in this study are the type of NEOMAX-50 by former NEOMAX Co., Ltd.. The chemical compositions of the samples are almost same with ordinary Dy-free Nd-Fe-B magnets with 0.1-0.3 wt.% of Cu and Al additives from starting materials. The average grain sizes and the coercivities after magnetization in 5T-pulse field are indicated in Table 1. The A1 and A2 samples are the commercial Nd-Fe-B sintered magnets of NEOMAX-50, and in the B1, B2 and B3 samples, the starting powder of average particle size of about 2.1 μm was newly prepared, and sintered at about 1300K, 1330K and 1373K, respectively. The final annealing temperatures are 753-793K in the samples, except the A2 sample at 1073K. The saturated magnetizations of the samples are 1.38T (A1) – 1.45T (A2).

<table>
<thead>
<tr>
<th>Sample</th>
<th>$D_{ave}$ (μm)</th>
<th>$\Phi_0Hc$ (T)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A1</td>
<td>5.6</td>
<td>1.20</td>
</tr>
<tr>
<td>A2</td>
<td>5.0</td>
<td>0.80</td>
</tr>
<tr>
<td>B1</td>
<td>3.1</td>
<td>1.62</td>
</tr>
<tr>
<td>B2</td>
<td>5.1</td>
<td>1.35</td>
</tr>
<tr>
<td>B3</td>
<td>8.4</td>
<td>1.11</td>
</tr>
</tbody>
</table>

*Only this sample was annealed at about 1073K. (Others were annealed at about 773K)*

2.2 Observation of domain structure

The observation of domain structures was carried out using the magneto-optical Kerr effect microscopy (MOKE) and the magneto-force microscopy (MFM). The observations in this study were done in zero applied magnetic-field both in the thermally demagnetized initial state and after the application of magnetic fields to inverse direction for the initial magnetization for the 2nd and 3rd quadrant in hysteresis loops.

2.3 Magnetic measurements

The magnetic measurements were done using the vibrating sample magneto-meter (VSM). In the thermally demagnetized state, the initial susceptibilities ($\Phi_{int}$) measurements were carried out in adjusting the time constant and the waiting time conditions for eliminating the effects from the apparatus for the measurements. The linearity of susceptibility to applied field was kept for our apparatus when the time constant of 0.01sec and the waiting time of 1sec in this study.

For the 2nd and 3rd quadrant in hysteresis loops, i.e. for studying the coercivities of the samples, the magnetizations under the maximum applied field ($M_{appl}$) and the remanences ($M_r$) in the minor loops which measured for step by step increased maximum applied fields (each step was 0.1T in this study) to the inverse direction for the initially magnetized direction. The domain wall motion (DWM) in the loops can be detected as the differences between the maximum magnetization and the remanence in each minor loop ($DWM = M_r - M_{appl}$).
3. Results and Discussion

3.1 Domain structure observations

Fig.1 shows the typical examples of the domain structures (DS) observed on the polished c-plane using MOKE in thermally demagnetized state. The MFM images of the thermally demagnetized state do not give clear domain structure images compared with the MOKE images caused by the interaction between the magnetic fluxes from domains and that from the canti-lever material (Pt-Co) (5). Fig.1 (A) is the DS of sample A2 which is the lowest coercivity sample ($H_c = 0.8T$) in this study, and having the average grain size ($D_{ave}$) of 8.4 µm which can be confirmed in the figure. Fig.1 (B) that of the sample B1 of the highest coercivity ($H_c = 1.62T$) in this study, and is 3.1 µm which can also be ascertained in the figure.

Fig.2 is the MFM images on the polished c-plane of the partially demagnetized (applied -1.2T field for the initial magnetization direction) A2 (A) and B1 (B) samples. In the state, the MFM domain structure images are clear as the figure. In the A2 sample, the DS return mainly into the crystal grains. In the B2 sample, however, the DS spread over groups of grains. The widths of black domains in the images are narrower than the white domains that can be explained by the influence from the surrounding still magnetized grains.

It was ascertained, therefore, the existence of movable domain walls both in thermally demagnetized states as shown in Fig.1, and also in partially demagnetized states as in Fig.2. Since both observations were done in zero applied field, the DWM in the samples is expected to be detected.

(A)                                                        (B)

Figure 1 Images of the magneto-optical Kerr effect microscopy (MOKE) in thermally demagnetized state of the sample A2 (A) and of the sample B1 (B). (c-plane observation)

(A )                                                        ( B )

Figure 2 Images of magneto-force microscopy (MFM) after applying 5T pulse field and also –1.2T to the inverse direction for the initial magnetization of the sample A2 (A) and of the sample B1 (B). (c-plane observation)
### 3.2 Susceptibility measurements in the 1st quadrant

Fig.3 shows the initial magnetization curves from 0 to 0.1T applied field, the increasing rates of magnetic field were almost 0.2-0.6T/sec and the average rate was about 0.49T/sec. The conditions of measurements were optimized by the pre-observations about the sample samples as mentioned above. The samples B1 and B2, however, showed the obvious delay of the magnetization response for increasing magnetic field up to the initial 0.02-0.04T applied field.

The susceptibilities in these measurements (< 0.1T) correspond to the movement of domain walls that are observed as shown in Figs.1 and 2. The motive force of domain walls, \( F \), from the thermally demagnetized state can be treated as the effective applied field, \( H_{\text{appl}} \), worked to the magnetization of each domain, \( J_s \). If we consider the tilting angle, \( \theta \), between the applied field and the average magnetic moments in domains that are parallel to the crystal c-axis in Nd-Fe-B hard magnetic materials, that can be exhibited as the following.

\[
F = -2J_sH_{\text{appl}} \cos \theta
\]  

(1)

The differential equation of the domain wall motion can be represented by the well-known following style, where \( m \) is the apparent mass of domain wall, \( v \) is velocity of domain wall motion (DWM), and \( a \) is the coefficient of friction in the DWM. We assume that the average domain wall energy in the sample’s grain and/or in grain boundary is almost same wherever the domain wall exists, that is the term corresponding to the spring phenomenon of the domain wall at certain position can be negligible in this differential equation.

\[
m \frac{dv}{dt} = F - av
\]  

(2)

If the motive force, \( F \), and apparent mass of domain wall, \( m \), can be regarded as the constant values, which can be guaranteed by the increasing rate of applied field was almost constant, 0.49T/sec, in the measurements, and the moving domain walls have the almost same structure in the initial magnetization process now under consideration, the solution about the \( v \) in this equation is as the following.

\[
v = v_f (1 - \exp(-at/m))
\]  

(3)

In this equation, \( v_f \) is the final constant velocity of DWM that can be determined from the slopes in Fig.3, and \( t \) is time in second. Therefore, if we plotted the \( \text{ln}(1 - (v/v_f)) \) term versus time \( t \), we could determine the coefficient \( (a/m) \). The Y-intercepts of the obtained curves correspond to the initial pinning force in the DWM in the samples.

**Figure 3** The initial susceptibilities from the thermally demagnetized state in the samples in this study. (The conditions of the measurements were adjusted by the preliminary experiments as: the time constant 0.01sec and the waiting time as 1sec for keeping the linearity of the susceptibility to the applied field.)
and that the coefficients of friction in DWM ($\alpha/ m$) are not so largely different in the samples in the initial motion of domain walls up to 0.5 sec. Since the Y-intercepts in Fig.4 come from the constant terms after the integration, and the physical meaning of them is the initial velocity of domain walls just after applying the magnetic field, it is possible to understand that the stronger pinning in DWM results in the smaller Y-intercept in Fig.4. Then, it is possible to read that the former result above corresponds that the comparatively large coercivity samples show the strong pinning in the initial motion of domain walls. The latter result indicates that the domain wall mass, $m$, has the almost linear relation to the values of friction coefficient, $\alpha$, especially in the initial motion after applying magnetic field.

![Figure 4](image4.png)

**Figure 4** The plots of $\ln(1-(v/v_i))$ vs. time (sec) for the initial magnetization curves.

Fig.5 shows the relation between the coercivities and pinning strengths in DWM in the samples. It seems that the borderline between the strong and weak pinning exists at the coercivities change from 1.2T(A1) to 1.35T(B2). This phenomenon does not come from only the average grain size, i.e. $D_{ave.}=5.6 \mu m$ and $D_{ave.}=5.1 \mu m$ (B2), but also the state of grain boundary phases which should be different in the B-series samples from the commercial magnets of the A-series samples.

![Figure 5](image5.png)

**Figure 5** Analyses of domain wall pinning based on the equation (2), the y-intercepts correspond to the pinning force in each curves in figure 4.
3.3 Domain wall motion in the 2nd and 3rd quadrants

Fig. 6 shows the remanences and the magnetizations under the maximum applied field in each minor loop, and the differences between them that correspond to the DWM, hereinafter we treat the magnetization coming from domain wall motion as the DWM. The domain structure returned to both in the samples A2 and B1 in the 2nd and 3rd quadrants in hysteresis loops as shown in Fig. 2 that means domain walls existed in the samples. The DWM, however, was observed only in the sample A2 and was not clearly detected in the sample B1 as in Fig. 6. In case of the latter sample B1, we applied higher magnetic field up to ~7 T at the High field Laboratory of Tohoku university, but no DWM was detected.

As shown in Fig. 2, the demagnetization in the sintered samples starts from the returning into multi-domain state of groups of crystal grains. The DWM, therefore, is the fundamental index for considering the demagnetization process. The details are discussed in references (3) and (4).

Normally, the DWM in the samples should mainly be influenced by two factors; (1) the pinning force to the domain walls themselves which comes from the surface grain boundary phases and the size, the composition and/or microstructure of the crystal grain itself, and (2) the influence from the surrounding grains such as the affecting magnetic fluxes and as the local and sample’s demagnetizing fields. The DWM in the 1st quadrant, measured and discussed before in section 3.2, is mainly influenced by the pinning force to domain walls coming from grain boundary regions and domain wall masses in the grains themselves. The DWM studied here, however, is mainly related to the magnetic connections among crystal grains and also to the magnetic fluxes from surrounding magnetized grains to the reformed domain walls and/or domains themselves.

Figure 6 The demagnetization curves and the remanences obtained from the minor loop measurements in which the maximum applied fields were step by step increased, and the different between them that corresponds to the domain wall motion in each minor loop. (A): the sample A2, (B): the sample B1 in which no domain wall motion (DWM) was observed.

Fig. 7 shows the relationship between the coercivities and the relative DWM in reformed domain structures in 2nd and 3rd quadrants in the samples (RSMDG). The percentages (RSMDG) were calculated using the initial DWM in the saturable multi-domain grains (SMDG) that was discussed in the previous papers of our group (6), (7), and the maximum magnetization values such as Fig. 6 (A) (DWM in the figure). The SMDG behave as multi-domain grains (MDG) in the thermally demagnetized state, and magnetized into single-domain grains (SDG) after applying sufficiently large field such as 5T pulse-field. It keeps the single domain state in the remanent state. In the 2nd and 3rd quadrants, however, a portion of them behaves as the SDG and never returns to the MDG, but another portion of them returns to multi-domain state as shown in Fig. 2 and in Fig. 6 (A). The DWM under discussion corresponds to that in the latter case SMDG. Especially, the domain structure in the sample B1 in the demagnetization process spread over many grains. The DWM in the sample is clearly
affected by the grain boundaries, because of the domain walls penetrate into them. The motion of domain walls will be a collective phenomenon in the group of crystal grains that will be discussed in another paper (3), (4).

As the conclusion of the data represented in Fig.7, the comparatively large coercivities appeared in the samples of the limited DWM in the reform multi-domain state of SMDG (RSMDG) that can be understood as the connection in domains are strong and the motion of domain walls are very difficult even in the reformed multi-domain regions.

Figure 7 Measured coercivities and the fractions of the domain wall motion in the SMDG in which a portion of magnetized grains return to the multi-domain state in the 2nd and 3rd quadrants in the hysteresis loops (6) (RSMDG). The domain structures return to all samples in this study, but the DWM could not be observed in the B1 sample as in this figure.

3.4 Relation between the coercivity and the domain wall motion (DWM) in the initial magnetization and in the demagnetization processes.

The DWM in the initial magnetization process shows the clear pinning behaviour in the B1 and B2 samples of comparatively large coercivities as shown in Fig.5. Since the basic chemical compositions and the conditions of heat treatments are similar in the samples of this study, the domain wall energy inside of grains seems to be similar. The initial motion of domain wall, therefore, should be mainly influenced by the state of domain walls at the grain boundary (GB) composed by the non-magnetic phases, where the stability of domain wall will be increased (1). The area and/or volume of GB phases for unit area of domain wall in the sample B1 of \( D_{ave}=3.1 \) \( \mu \)m should be nearly tenfold for these of the B3 sample of \( D_{ave}=8.4 \) \( \mu \)m. In the commercial magnets of A1 and A2 samples, since the distribution of \( D_{ave} \) are comparatively large as 3-10 \( \mu \)m for the \( D_{ave}=5.0-5.4 \) \( \mu \)m of them, the area of GB phases being passed through by the domain walls should be smaller than the samples B1 and B2 in which the \( D_{ave} (=3.1 \text{ to } 5.1 \mu m) \) distribution is obviously narrower.

On the other hand, the difficulty of DWM in the reformed multi-domain grains as shown in Fig.7 should be originated by the magnetic interaction in group of crystal grains. The domain walls should be stabilized by the lowering of energy of domain wall passing through the GB phases and also the lowered wall energy itself at the surface of reformed multi-domain regions. It is obvious that the domain walls exist as shown in Fig.2 in both the samples A2 and B1, but only the domain walls in the A2 sample are movable and these in the B1 samples spread over the crystal grains did not show the mobility by the application of magnetic field as shown in Fig.6 (B).

Fig.8 shows that the relationship between the pinning force in the initial magnetization in the 1st quadrant and the fractions of detectable DWM in the reformed multi-domain regions in the demagnetization process in the 2nd and 3rd quadrants (RSMDG). The figure indicates the clear
inclination that stronger pinning behaviour in the initial magnetization process corresponds to the smaller fraction of movable domain walls in the multi-domain region in the demagnetization process. The domain structure spread over wide region in the thermally demagnetized initial state, and the domain structures are also spread over many grains in the partially demagnetized state even the multi-domain regions are limited in a certain volume. The DWM is responsible for the appearance of coercivity in the magnets.

Figure 8 The relationship between the pinning force for the initial DC susceptibilities from the thermally demagnetized state and the fractions of DWM as magnetizations in the reformed multi-domain regions (RSMDG) in the demagnetization process.

4. Conclusion
The measurements of the DC susceptibilities in the 1st quadrant and the domain wall motions (DWM) in the 2nd and 3rd quadrants in the NEOMAX-50 type of Nd-Fe-B sintered magnets revealed that the samples having the comparatively large coercivities such as 1.35-1.62T without the Dy addition showed the strong pinning in initial DWM and the difficulty of DWM in the reformed multi-domain regions. Both phenomena should be originated from the interactions between the domain walls and the grain boundary phases that result in the difficulty of DWM in the sintered magnets.

Acknowledgement
The research was partially supported by the Ministry of Education, Culture, Sports, Science and Technology, Scientific Research of Priority Areas “Panoscopic Assembling and High Ordered Functions for Rare Earth Materials”. The authors also deeply thank for the helpful discussion and the measurements for this paper by Mr.T. Kohno and by Mr.T. Matsushita.

References