The magnetization and demagnetization processes in the 11 types of Dy-free Nd–Fe–B sintered magnets based on the NEOMAX-50 type magnets were investigated using the step method that is our original method of measurements and analyses of minor loops with step by step increased maximum applied fields. With the decrease in average grain sizes (DAVE), the single-domain grains (SDG) volume fraction increased and inversely that of the saturable multi-domain grains (SMDG) decreased, the changing behavior was monotonous. The dependence of coercivity on the DAVE, however, showed clear change around DAVE=3.0 μm, from the increase with decreasing DAVE to the opposite relationship less than DAVE <3.0 μm. We consider that the changing in the relationships should come from the ratios of the domain width (Wd) to the DAVE, and conclude that the well cutting of magnetic interactions at the grain boundaries should realize the coercivity of $\mu_0 H_C \approx 2$ T in fine grain size Dy-free samples.

**Key words:** Nd–Fe–B sintered magnets, coercivity, magnetization and demagnetization processes, domain structure, domain wall motion, grain sizes in sintered magnets.

### 1. Introduction

The author analyzed the magnetization and the demagnetization processes in Sm$_2$Fe$_{17}$N$_3$ magnetic particles by the categorization of the particles into the single-domain particles (SDP), the multi-domain particles (MDP) and the saturable multi-domain particles (SMDP) 1), 2). The SMDP is the MDP in thermally demagnetized state, and can be magnetized into the SDP by the application of a certain magnetic field, and after the magnetization they behave mainly as the SDP and a certain fraction of the SMDP return to multi-domain state in the 2nd and 3rd quadrants (2, 3Q) in hysteresis loops. The latter portion of SMDP is also can be magnetized in single-domain state to the inverse direction by the application of a certain magnetic field to the inverse direction.

The magnetic properties of the magnets basically composed of the isolated magnetic particles such as bonded magnets should be made up of the properties of individual particles, i.e. the magnetization and coercivity of such a bonded magnet can be treated as the summation of these properties in individual particles. The SMDP, which behave as the SDP and/or returning to multi-domain state in the 2, 3Q, have the coercivities as the particles themselves and their volume percentages in the bonded magnets are comparatively high as 50-70 % 1), 2). The categorization, therefore, has an important meaning for the discussion of coercivity mechanism in the bonded magnets.

The authors, however, studied in this study the coercivity in the sintered Nd–Fe–B magnets 3), 4), 5) and found that the magnetization and demagnetization in the magnets occur in the group of crystal grains, that is to say the region composed of crystal grains in the sintered magnet behaves collectively as a magnetic unit. It was also revealed that the domain wall motion (DWM) in the thermally demagnetized state in the 1st quadrant (1Q), and in the regions where domain walls spread over the crystal grains in the remanent states in the 2, 3Q was affected by the magnetic interactions among the crystal grains 5).

The purpose of this study, therefore, is to apply the categorization of the magnetic particles to the sintered Nd–Fe–B magnets, and to analyze the collective magnetic behavior in the regions composed of a certain number of crystal grains as the single-domain grains (SDG), the multi-domain grains (MDG), and also the saturable multi-domain grains (SMDG). It is very interesting to confirm the usefulness of the categorization in the sintered magnets in which the magnetic interactions among crystal grains are very strong compared to these in among particles in the bonded magnets.

In this paper, magnetizations (polarization) $(\mu_0 M)$ and magnetic field $(\mu_0 H)$ are both presented as only $M$ and $H$, respectively, using T (Tesla) unit.

### 2. Experiments and Results

#### 2.1 Samples and their magnetic properties

The samples in this study have basically the same chemical composition, and the same processing details with the NEOMAX-50 type commercial magnets of the former NEOMAX Co. Ltd., Japan. The A1 sample was finally annealed at the optimum temperature of about 773 K, but the A2 sample is finally annealed at about 1073 K. The B series samples were prepared using the fine starting powder of about 2 μm in diameter. The
sintering temperatures were increased from the B1 to the B3 samples step by step as shown in Table 1. The average grain sizes ($D_{\text{AVE}}$) of sintered samples are also increased from 3.1 $\mu$m of the B1 sample to 8.4 $\mu$m of the B3 sample. The C series of samples were prepared using a finer starting powder of about 1.6 $\mu$m in diameter. With the differences of sintering temperatures and the final annealing temperatures, the $D_{\text{AVE}}$ and the magnetic properties are different as shown in Table 1.

The chemical composition as rare-earth contents of the samples were measured using SEM-EDS, and A1 and A2 contain about 32 wt.% of Nd, but the B- and C-series samples were almost 27-28 wt.% of Nd, and in some C-series samples, Pr of 2-4 wt.% were detected at some surface positions in the samples.

The densities of the samples ($\rho$) are almost full ones ($\rho = 7.53-7.64$) in all samples in this study, but the mechanical hardness of the samples that should correspond to the mechanical bond strength between crystal grains are varied with the sintering temperature, i.e. the C series samples are comparatively softer than the other samples of A and B series.

The magnetic properties of the samples were measured using a normal vibrating sample magnetometer (TOEI Industry Co. Ltd., VSM-5-15 type), and the analysis of the magnetization and demagnetization mechanism was done by the step method that will be explained precisely in the following section. For the elimination of surface effects on the magnetic properties, the sample size was prepared as about 5 mm×5 mm×5.5 mm for all samples.

Table 1 shows the preparation conditions, i.e. $T_s$ : sintering temperature, $T_a$ : final annealing temperature, and the average grain sizes ($D_{\text{AVE}}$), and the magnetic properties, i.e. $H_C$ : coercivity and $M_S$ : saturated magnetization of the samples in this study.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$T_s$ (K)</th>
<th>$T_a$ (K)</th>
<th>$D_{\text{AVE}}$ ($\mu$m)</th>
<th>$H_C$ (T)</th>
<th>$M_S$ (T)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A1</td>
<td>773</td>
<td>1.20</td>
<td>1.38</td>
<td></td>
<td></td>
</tr>
<tr>
<td>A2</td>
<td>1073</td>
<td>0.80</td>
<td>1.45</td>
<td></td>
<td></td>
</tr>
<tr>
<td>B1</td>
<td>1303</td>
<td>3.1</td>
<td>1.62</td>
<td>1.41</td>
<td></td>
</tr>
<tr>
<td>B2</td>
<td>1333</td>
<td>5.1</td>
<td>1.35</td>
<td>1.41</td>
<td></td>
</tr>
<tr>
<td>B3</td>
<td>1373</td>
<td>8.4</td>
<td>1.11</td>
<td>1.40</td>
<td></td>
</tr>
<tr>
<td>C1a</td>
<td>1198</td>
<td>7.7</td>
<td>1.01</td>
<td>1.36</td>
<td></td>
</tr>
<tr>
<td>C1b</td>
<td>1198</td>
<td>1.8</td>
<td>1.37</td>
<td>1.37</td>
<td></td>
</tr>
<tr>
<td>C2a</td>
<td>1218</td>
<td>1.8</td>
<td>1.06</td>
<td>1.40</td>
<td></td>
</tr>
<tr>
<td>C2b</td>
<td>1218</td>
<td>1.8</td>
<td>1.37</td>
<td>1.39</td>
<td></td>
</tr>
<tr>
<td>C3a</td>
<td>1253</td>
<td>2.9</td>
<td>1.48</td>
<td>1.42</td>
<td></td>
</tr>
<tr>
<td>C3b</td>
<td>1253</td>
<td>2.9</td>
<td>1.27</td>
<td>1.41</td>
<td></td>
</tr>
</tbody>
</table>

Figure 1 shows the relation between the coercivity and the average grain sizes in the samples. As shown with the dashed line, the coercivities increase with the decreasing $D_{\text{AVE}}$ down to about 3 $\mu$m, but inversely decrease in the cases of finer $D_{\text{AVE}}$ such as the C series samples. An exceptional case is seen in the sample A2 that was finally annealed at 1073 K which is higher than the optimum annealing temperature of about 773 K. It should be also remarked that the coercivities of C1b and C2b samples finally annealed at 1073 K are clearly larger than those of the C1a and C2a samples treated at 773 K that has been considered to be the optimum final annealing temperature for normal industrial processes.

2.2 Domain structures in the samples

The domain structures and the average grain size of the samples were observed and determined using the magneto-optical Kerr effect microscopy (MOKE) and the magneto-force microscopy (MFM). For the observations, the samples were finally polished using a diamond paste, and domain structures of the samples were investigated in a previous study.

For the determination of the average grain sizes, the diameters of about 200 grains in each sintered magnet were measured directly from the observed microstructures. As shown in Fig. 2 (L) of the c-plane MOKE image of the thermally demagnetized A1 sample ($D_{\text{AVE}} = 5.6 \mu$m), the grain size was almost decided from the maze patterns of domain structure in each grain. The domain structures on this image were seemed to have the connections across crystal grains, i.e. the domain structures are not limited in each grain.

On the polished parallel plane to crystal c-axis as
shown in Fig. 2 (R), it is very difficult to observe clear domain structure images using the MOKE and the MFM, especially, in the highly orientated sintered magnets such as the samples in this study. However, it is possible in some points to observe the domain stripe-like structures spread over the group of crystal grains. In almost cases, the grain boundary (GB) is the edge of a domain, i.e. The domain walls exist at the grain boundaries. About the A and B series samples, we tried to observe the stripe structures on the planes using the MOKE, and the average domain widths (Wd) are about 1 µm and 1.5 µm in the B1 and B3 samples, respectively. The measured Wd will be shown and used in the discussion part of this paper.

Figure 3 is a schematic representation of domain structure in the samples in this study. Each magnetic domain spread over the group of crystal grains that means the domain walls exist through the grain boundaries between grains. The initial magnetization from the thermally demagnetized state of the magnets, therefore, starts by the domain wall motion (DWM) through the grains.

Figure 4 shows the MFM images of domain structures in the A2 and B1 samples as the typical examples of the samples in this study (all observed in the remanent state). Fig.4 (A) and (B) are the images of partially magnetized A2 and B1 samples, respectively, with the application of +1.2 T to the thermally demagnetized state. The monotonous bright grains in the images are the magnetized grains into single-domain state, and the black and white maze patterns in grains and/or spread over grains are domain structures in the samples. In Fig. 4 (A) and (B), it is obvious that the domain structures (DS) spread over the crystal grains more widely in the A2 samples than the B1 sample. The latter case, the DS seems to be only spread over some grains, but in the former case, that exist over a number of grains.

In Fig. 4 (C) and (D), the DS, which was once almost disappeared with the +5 T pulse-field application, returned with the application of -1.2 T in the 2, 3Q in the hysteresis loops can be observed. The DS in the A2 sample (C) seems to exist in each grain, but that in the B1 sample (D) is obviously spread over crystal grains where $D_{AVE}$ is 3.1 µm as shown in Table 1.

The interesting relationship between the coercivity and the DWM was discussed in the previous paper, and also will be precisely investigated in this paper.

2.3 The step method (Fig.5) for the categorization of crystal grains in the sintered magnets

In the initial state, the samples are thermally demagnetized, that means the domain structure spreads over the whole sample through the crystal grains. With the application of a certain magnetic field in a minor loop, the domain wall motion (DWM) results in the appearance of magnetization of the sample.

Fig. 2 The MOKE images of the c-plane of the thermally demagnetized A1 sample (L), and the polished plane parallel to c-axis of the same A1 sample (R).

Fig. 3 Schematic representation of the domain structure in a thermally demagnetized sintered sample.

Fig. 4 (A) and (B): MFM images of partially magnetized A2 and B1 samples with application of +1.2 T in the initial magnetization process, and (C) and (D): MFM images of partially demagnetized A2 and B1 samples after application of +5 T pulse-field and of -1.2 T in the demagnetization process. (All images are on the polished c-plane of the samples in remanent state)
When the applied field is more than 0.5 T, it is expected that some grains and/or groups of grains can be magnetized into single-domain state because of the demagnetizing fields of the grains and/or of regions are estimated to be \( H_{\text{demag}} = N M_s = 0.5 \) T in the case of Nd-Fe-B magnets, where \( N \approx 1/3 \) is the demagnetizing factor of the sphere grain and/or region and \( M_s \approx 1.5 \) T is the saturated magnetization.

When the magnetizations under the maximum applied field \( (M(H_{\text{appl}})) \) and the remanences \( (M_r(H_{\text{appl}})) \) in minor loops are obtained, the differences between them (\( \Delta M \)) correspond to the magnetization variation by the domain wall motion (DWM) in the minor loops \( (M_{\text{DWM}}) \). For examples, the magnetization differences between \( M(+0.9 \) T) and \( M_r(+0.9 \) T) in the 1st quadrant, and \( M(-1.2 \) T) and \( M_r(-1.2 \) T) in the 2nd quadrant in Fig. 5 correspond to the magnetizations caused by the DWM (the contribution of rotation of magnetic moments in the samples is negligibly small in the minor loops). The magnetization and demagnetization behaviors in the samples can be detected with the measurements of minor loops with step by step increasing of maximum applied field. The relation can be presented as the following equation.

\[
M_{\text{DWM}} = \Delta M = M(H_{\text{appl}}) - M_r(H_{\text{appl}}) \tag{1}
\]

Here, \( M_{\text{DWM}} \) is the total domain wall motion (DWM) in minor loops. Fig. 6 shows schematically the method to obtain the \( M_{\text{DWM}} \) based on the real measured results about the sample B1.

On the other hand, a certain small ratio of crystal grains is the MDG through the whole magnetic measurements in the step method. The ratios are ordinarily 3-4 vol.% in the highly orientated sintered magnets such as the samples in this study. The magnetization of MDG \( (M_{\text{MDG}}(H_{\text{appl}})) \) in a minor loop comes from the DWM in them, and can be determined as the small decrease in magnetization from the saturated magnetizations to the remanences after the magnetization under 5 T, for example. The magnetizations coming from the DWM except these from the MDG, \( (M_{\text{DWM}}) \), therefore, can be represented as the following.

\[
M_{\text{DWM}} = M_{\text{DWM}} - M_{\text{MDG}}(H_{\text{appl}}) \tag{2}
\]

The obtained \( M_{\text{DWM}}, M_{\text{DWM}} \) and \( M_{\text{MDG}}(H_{\text{appl}}) \) in the case of sample B1 are indicated in Fig. 7. The curve obtained as the \( M_{\text{DWM}} \) decreases in more than about 0.5 T applied fields. It can be understood that the total DWM measured as the magnetization decreases with increasing the maximum applied field in the minor loops in \( H_{\text{appl}} > 0.5 \) T of the estimated demagnetizing field of the sample.
The decrease of the $M_{DWM}$ should correspond to the magnetization of grains and/or regions magnetized into single-domain state, and they never return to multi-domain state in the 1Q in the measurements. This result of magnetic measurements well corresponds to the results of domain structure observations showed in Fig. 4 (A) and (B). The magnetizations calculated as the decrease of magnetizations from the maximum one in the obtained DWM as $M_{DWM}$ (cross-hatched magnetizations in Fig. 8) can be treated as the remanences coming from the magnetized SMDG.

In other words, The DWM as $M_{DWM}$ decreased with increasing the applied fields ($H_{appl}>0.5$ T) means the disappearance of movable domain walls in them, i.e. the multi-domain regions were magnetized into single-domain state. As shown in Fig.9, now we know the amounts of the magnetized SMDG as the $M_{SMDG}$ = ($\bigtriangleup M_{DWM}$ = $M_{DWM}$($H_{appl}$) - $M_{DWM}$($H_{appl}$)) see cross-hatched magnetizations in Fig.8 and $M_{SMDG}$ in Fig. 9. In general, the remanences ($M(H_{appl})$) in the minor loops correspond to the total magnetizations coming from the magnetized grains in the samples. The differences between $M(H_{appl})$ and $M_{SMDG}$, i.e. $\Delta M = M(H_{appl}) - M_{SMDG}$, therefore, should correspond to the magnetizations from another portions magnetized into the single-domain state in the applied field direction.

If there is the SDG in the samples, they change the direction of magnetic moments under a certain applied field that should also make the increase of remanences. The $\Delta M$ above is, therefore, can be treated as the magnetization coming from the magnetically reversed SDG as shown in Fig. 9. Therefore, we can rewrite the $\Delta M$ as the $M_{SDG}$ in the figure.

As a conclusion of this discussion, we can separate the three components of the magnetization in the studied samples that is the $M_{SDG}$, the $M_{SMDG}$, and the $M_{MDG}$.

The separated magnetization curves of three categorized grains in the B1 sample are shown in Fig. 9 ($M_{MDG}$ and $M_{SMDG}$) and in Fig. 8 ($M_{MDG}$).

### 2.4 Volume fractions of categorized three types of grains in the magnetization process in the samples

Using the step method explained in the former section, the volume fractions of three categories of crystal grains were determined for all samples in this study as shown in Table 2.

<table>
<thead>
<tr>
<th>Sample</th>
<th>SDG</th>
<th>MDG</th>
<th>SMDG</th>
</tr>
</thead>
<tbody>
<tr>
<td>A1</td>
<td>22.2</td>
<td>3.3</td>
<td>74.5</td>
</tr>
<tr>
<td>A2</td>
<td>31.0</td>
<td>3.7</td>
<td>65.3</td>
</tr>
<tr>
<td>B1</td>
<td>34.5</td>
<td>2.8</td>
<td>62.8</td>
</tr>
<tr>
<td>B2</td>
<td>19.6</td>
<td>3.2</td>
<td>77.2</td>
</tr>
<tr>
<td>B3</td>
<td>12.4</td>
<td>4.9</td>
<td>82.7</td>
</tr>
<tr>
<td>C1a</td>
<td>47.1</td>
<td>2.0</td>
<td>50.9</td>
</tr>
<tr>
<td>C1b</td>
<td>43.8</td>
<td>1.8</td>
<td>54.5</td>
</tr>
<tr>
<td>C2a</td>
<td>47.1</td>
<td>1.7</td>
<td>51.2</td>
</tr>
<tr>
<td>C2b</td>
<td>44.8</td>
<td>1.5</td>
<td>53.7</td>
</tr>
<tr>
<td>C3a</td>
<td>36.0</td>
<td>1.2</td>
<td>62.8</td>
</tr>
<tr>
<td>C3b</td>
<td>36.2</td>
<td>1.5</td>
<td>62.3</td>
</tr>
</tbody>
</table>

With the decrease of $D_{AVE}$, the fractions of SDG increase and inversely that of SMDG decrease, both monotonously. It is in contrast with the coercivity dependence on the average grain size in which the slope showed clear kink at around 3 μm of $D_{AVE}$ as shown in Fig. 9.
Concerning to the discussion here, the care must be taken in that the domain structure is formed through a certain numbers of crystal grains, not limited in a grain.

2.5 The DWM in the demagnetization process in the 2nd and 3rd quadrants

Based on the measurements explained in the former section, after the magnetization under 5 T pulse-field, for an example, more than 95 vol.% of grains in all samples can be magnetized into single-domain state. The coercivity of the samples concerns to the demagnetization process from this magnetized state in the 2, 3Q in hysteresis loops.

The magnetization changes corresponding to domain wall motion (DWM) in the samples can be separated also in the demagnetization process as explained in the following. In the step method measurements shown in Fig. 5, the magnetizations under the applied field ($M(H_{appl})$) and the remanences ($M_r(H_{appl})$) can be obtained in each minor loop measurement. The differences between them, i.e. $\Delta M = M(H_{appl}) - M_r(H_{appl})$, are physically composed by the DWM and the rotation of magnetic moments in the samples. Since the latter contribution, however, is very small in a minor loops in this study ($H_{appl}<1.6$ T), it is negligible as the magnetization change. The magnetization change, therefore, can be treated as the DWM in the sample.

Figure 11 shows the magnetization changes corresponding to the DWM ($\Delta M$ in the A2 sample of coercivity ($H_c$) = 0.8 T (up) and in the B1 sample of $H_c$ = 1.6 T (down). In the 2, 3Q in hysteresis curves, the partially demagnetized samples showed the domain structures as shown in Fig. 4(C) and (D). If there were domain walls in the samples, the DWM ($\Delta M$) could be measured as Fig. 11 (up).

The real measured results in Fig. 11, however, revealed that the magnitude of DWM obviously different in the samples, even the all samples showed the reformation of domain structures, the examples were shown as in Fig. 4(C) and (D). Since the $M_{SDG}$ are only 2-4 % of the saturated magnetizations ($M_S$) in all samples in this study, the DWM ($\Delta M$) should appear in the SMDG that showed the DWM in the thermally demagnetized state and the volume fractions are 50-80 % in the samples. Physically, the DWM generates in the regions in which the multi-domain state reformed in the demagnetization process.

This reformed domain wall motion (RDWM), therefore, can be characterized as the ratios of magnetizations to the SMDG ($M_{SMDG}$) in the thermally demagnetized state. We can decide the ratios as RDWM = $\Delta M_{RDWM}$ / $M_{SMDG}$ (in the 1Q). Fig. 12 shows the relationship between the RDWM and the coercivities in the samples in this study.
3. Discussion

3.1 The reason of existence of the SDG in the initial magnetization process

The feature of sintered magnets compared with the bonded magnets is the strong interactions among crystal grains. The interactions are mechanical, magnetic and chemical ones, but here we will focus our attention on the magnetic interaction.

As shown in Figs. 2 and 3, a domain spread over a number of crystal grains in the studied magnets. It is not easy to image the existence of the SDG in the magnets. Actually, we cannot find the SDG in the thermally demagnetized state of the A1 sample in Fig. 2. All crystal grains have domain structures that are also recognized from that the $D_{\text{AVE}}$ is clearly larger (about 2-8 $\mu$m) than domain width of about 1$\mu$m in the samples.

We can explain the reason why the SDG can be detected in the samples as shown in Fig. 13 by the following interpretation. The domain structures spread over the crystal grains regardless of the existence of grain surfaces, i.e. boundaries among them. The domain wall energy and/or the statistic magnetic interaction between the grains should be affected through the grain boundaries, nevertheless the observed domains spread over the grains.

As shown in Fig. 13, since the domain widths of about 1$\mu$m in the thermally demagnetized state are narrower than the $D_{\text{AVE}}$, almost all grains are in multi-domain state. With the application of magnetic field, the widths of advantageous domains become wider according to the relation of $H_a = N M$, here $H_a$ is the applied field, $N$ is the demagnetizing factor of grains and/or of collectively behaving regions, and $M$ is the magnetization of the sample, naturally $M = M(\rightarrow) - M(\leftarrow)$.

When the domain widths become wider than the grain sizes, it is possible that the grains are magnetized into single-domain state. If the field were sufficiently large to magnetize the grains into the SDG, then the grains would behave as the SDG hereafter. Since the threshold condition of the phenomenon above is the domain width becomes wider than the grain size, if the collectively behaving regions are sufficiently large as 50-100 $\mu$m, the applied magnetic field need not to be larger than the demagnetizing field of about 0.5 T in the case of the Nd-Fe-B magnets, ex. the domain widths easily reach 2-3 $\mu$m. And also another important point is the demagnetizing factor, $N$, need not to be 1/3 when the group of crystal grains forming the magnetically collective region, and it can be nearly zero for a needle-like shape of the region. The magnetization of the SDG, therefore, should start just after the application of magnetic field, that obviously agrees to the magnetization curve of the SDG shown in Fig. 9 ($M(\text{SDG})$ curve).

3.2 The relationship between the RDWM and the coercivity in the sample

The magnetic reversal starts from the formation of reversed nucleus at a certain position in a grain, and the reversed portion spreads to a whole grain and also to surrounding grains. The probability of nucleation and

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Fig. 12 The relationship between the RDWM ratio and coercivities in the samples. The extrapolated coercivity of $\mu_0H_c = 2$ T for the C-series samples.

Fig. 13 A schematic representation of the reason of appearance of the SDG in the initial magnetization process.

the difficulty of propagation of reversed region to surrounding grains should deeply correlate to the nature of grain boundary (GB). The cutting of magnetic interactions at the GB should be favorable for the appearance of large coercivity.

The RDWM in this study is the ratio of maximum magnetization by the DWM in the 2, 3Q in the remanent state to the total magnetization of SMDG in the 1Q, i.e. RDWM = ΔM_max (DWM in 2, 3Q) / M(SMDG in 1Q). It should arise from, therefore, the DWM in reformed multi-domain structure in the reversed regions. In the step method, the magnetic reversal in the 2, 3Q was interrupted in a certain intermediate state, and it appeared that the number and volume of reversed regions should increase with increasing the maximum applied fields in the minor loops.

In Fig. 12, the linear relationships between the coercivities and the RDWM are clearly shown. The 2 lines for the A and B series samples and for the C series samples are obviously independent, but both lines show the increase of RDWM deteriorates the coercivity. The large RDWM will be caused by the large total volume of reversed regions having the domain structure in the remanent state and also by large mobility of domain walls in the regions. Therefore, it can be concluded inversely from the data in Fig.12 that the samples, in which the total volume of reversed regions and the mobility of DW in the region are small, show the large coercivities. The preferable GB for coercivity means large pinning effects on DWM, and well magnetic cutting between the grains. Again, the well cutting of magnetic interactions at the GB, therefore, should be better for the appearance of large coercivity in the samples.

The next important result to be discussed is the origin of clear different slopes in the 2 lines for the A and B series samples and for the C series samples. The difference in slope implies that the interaction between GB and DW is clearly different in two groups of samples, otherwise the slope should only show parallel shift. The obvious difference in the 2 groups of samples is the DAVE as shown in Table 1. The A, B series samples have more than 3 μm, DAVE >3 μm, but the C series samples less than 3 μm, DAVE <3 μm.

The domain width (W0) increases with increasing the particle size in the case of isolated spherical samples and with increasing the thickness in the case of wide plate-like samples 8). It is possible to calculate the relationship between the W0 and the diameter of spherical samples (R), also and the thickness (L) of plate-like samples. The domain width in the Nd-Fe-B magnets starting from about 0.3 μm of the single-domain particles reaches to about 3 μm when the size and the thickness become to about 100 μm in the remanent state.

If we compared the DAVE, regarding as the W0 above, with the R and L, the DAVE (W0) of C-series samples were almost correspond to R, L = 30-70 μm of the particle and/or region sizes, and in the A and B series samples, only that of the B1 sample of the size of R, L = 100 μm could be realized but other samples should be too large. Based on the consideration here, Fig.14 (A) represents the relationship between the measured W0 as shown in Figs. 2, 3 using MOKE and the DAVE of the samples in this study, and Fig. 14 (B) shows the numbers of domain in an average grain sizes (DAVE) of the samples in this study. The calculation, based on the equations in ref. (8), reveals that the C-series samples and the B1 sample averagely contain 2-3 domains in a grain (N=2-3), and that the other samples, however, more than 4 domains exist averagely in a grain (N>4).

If we use 2-dimensional simple model as shown in Fig. 15, it is obvious that the (1/n) of total domain length exist in the GB when the n domains (N) exist in a grain. Therefore, it implies that a certain threshold length proportional to the (1/n) values exist concerning to the DWM difficulty in the samples, and the A, B series samples and C-series samples should be the larger or smaller than the threshold value, respectively. The domain walls existing possibility in the GB (DW(pinn)), where the DW pinning should appear.
Fig. 15 Schematic representations of 2-dimensional model of the domain walls (DW) in grain boundaries. (the \( n \) domains in a grain (\( N_D \)) mean the \((1/n)\) length of a DW exist at the grain boundary). (DW(pinn)) means the ratio of DW length existing at the GB)

The measured RDWM in the A and B series samples should mainly be the DWM in grains, but that in the C series samples corresponds to the DWM going across the GB. Therefore, the DWM in the A, B samples are naturally larger than these in the C series samples. Inversely, the small increase in the DWM in the C series samples means the DW can easily goes through the GB, i.e. the magnetic reversal easily propagates into surrounding grains, that should be harmful for the coercivity. The discussion here is on a model of appearance of RDWM in this study, but it is real results that the 2 lines are completely independent as shown in Fig. 12.

At the end of this discussion, it should be added that the total GB volume become double when the average grain size is a half (= \((1/2)D_{\text{AVE}}\)). The rare-earth contents of C-series samples are almost the same as these of B-series samples, that is, the thickness of GB should be less than a half of B-series samples in the C-series samples. The domain wall pinning and the stopping of reversal propagation to surrounding grains in the C-series samples should be very weak compared with these effects in the A and B-series samples.

4. Conclusion

The categorization of crystal grains into the SDG, the MDG and the SMDG successfully applied to the sintered Nd-Fe-B magnets. With the decrease in grain sizes (\( D_{\text{AVE}} \)), the SDG fraction increased, but inversely that of the SMDG decreased monotonously. The coercivities, however, showed the increase with decreasing grain sizes in \( D_{\text{AVE}}>3 \) \( \mu \)m, but oppositely decreased in \( D_{\text{AVE}}<3 \) \( \mu \)m. This phenomenon can be explained by the ratios of the \( D_{\text{AVE}} \) to the domain width (\( W_d \)) in crystal grains and/or the magnetically collective regions that correlate to the possibility of domain walls existing in grains or in grain boundaries (GB). The well cutting of magnetic interactions at the GB is expected to result in the coercivity of \( H_C = 2 \) T in fine grain size (C series) Nd-Fe-B sintered magnets in this study as shown in Fig. 12, as the extrapolated arrow.

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