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Magnetic force microscope study on anisotropic NdFeB permanent magnets①

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Abstract: NdFeB permanent magnets prepared by powder metallurgy were investigated using magnetic force microscopy (MFM). The excellent MFM images of sample along the surfaces parallel and perpendicular to the alignment axis were collected respectively. The results show the necessity of annealing procedure in the preparation of the samples to remove the polishing surface stress and to illustrate the real magnetic domain structure, so that the much information about both the magnetic structure and the topographic microstructure is obtained. The hard MFM tip is verified to be effective for this material especially for the sample with the examined surface parallel to alignment axis. By analyzing these well-captured magnetic force images, magnetic domains and alignment degree as well as the topographic information such as grain size and the nonmagnetic phases at the grain boundaries were demonstrated.

Key words: NdFeB permanent magnets; magnetic force microscopy; magnetic domain
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1 INTRODUCTION

Magnetic force microscopy (MFM) has been applied to study the micro-magnetic structure of magnets for more than one decade. Although the development of MFM technique enables it to investigate the magnetic structure of various magnets, especially for those moderate coercivity systems such as magnetic sensor materials and information storage media[1], the observation on the high performance permanent magnet NdFeB is still a hard nut to crack. However, MFM observation on NdFeB is really worthy of research efforts because of the very strong dependence of the magnetic properties of NdFeB on microstructure and such a powerful technique as MFM to correlate microstructure and magnetic properties. In fact, many valuable researches in this field have been carried out in recent years. Most of them focused on the NdFeB samples in form of melt spun ribbons or sputtered films[2-5]. For those amorphous or nanocrystalline NdFeB, the interaction domain pattern (a group of grains with their spins closely aligned by exchange coupling) is commonly found. How about the bulk samples prepared by powder metallurgy method? In order to avoid the strong stray field of sintered NdFeB, many methods were tried including modification of MFM instrument by certain attachments[6], utilization of the imaging processing[7], improvement of the test conditions[8] and so on. Systematic and illuminative results of MFM study on bulk sintered NdFeB were reported by Folks et al[9]. In addition to the valuable demonstration of MFM images, they examined the tip-sample interaction on the bulk NdFeB with the batch-fabricated tips of different coercivity and gave the result that the low coercivity tip coated with Fe/SiO_2 could provide the most readily understood contrast because of its "self-focusing" effect (or named "soft tip" effect, magnetic contrast generated between neighboring domains or at domain boundary walls depending on the disturbed degree of the tip). In present work, the well-captured MFM images of the anisotropic sintered NdFeB are successfully collected with the hard tip, which is almost an impossible thing according to Folks. As for our experimental result, the well-prepared sample after annealing and the MFM tip with the proper coercivity are two keys to obtain the optimum MFM images in which the magnetic domain pattern, domain size, alignment degree and grain size as well as the nonmagnetic phases at the grain boundaries can be clearly observed for their readily understood contrasts.

2 EXPERIMENTAL

$Nd_{15}Fe_{77}B_8$ powders prepared by jet milling with an average particle size of 3-4 μm were used in the

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experiment. Following compaction and magnetic alignment under 1T field and 200 MPa isostatic pressing, the green samples were sintered at 1 080 ℃ in 10^{-3} Pa vacuum for 1.5 h, then an aging procedure was performed at 600 ℃ for 1.5 h. Magnetic properties of the magnets were measured using magnetometer NIM2000HF system with the results of B_r= 1.2 T, $_bH_c$= 637 kA/m, and energy product $(BH)_{max}$= 288 kJ/m^3. Two MFM samples marked as No. 1 and No. 2 were prepared by cutting NdFeB bulk with the examined surface parallel and perpendicular to the orientation direction respectively (i.e. the perpendicular sample and the parallel sample). After the careful grinding and polishing, a 7h annealing procedure in vacuum at the temperature a little higher than Curie point was performed to remove the surface stress that is formed in the polishing procedure. So the samples are in the thermal demagnetized state. Nanoscope III a-D3000 MFM microscope (DI company) was used to obtain magnetic force images with two kinds of magnetic tips: the standard tip (MESP tip) and the high coercivity tip (MESP-HC tip) supplied by DI Company. Both tips were magnetized upward prior to imaging.

3 RESULTS

Fig. 1 shows the 40 μm × 40 μm scan size MFM images of sample No. 1 (the perpendicular sample). The main magnetic domain pattern consists of corrugation and spike domain. Those areas without any domain contrast are nonmagnetic phases such as the paramagnetic Nd-rich phase situated at the grain boundaries and the triple intersection points of the grains. Resulted from the exchange interaction between the magnetic $Nd_2Fe_{14}B$ phase and the nonmagnetic phases, some stripe domains appear around the nonmagnetic phases. By the comparison of Fig. 1(a) with Fig. 1(b), it is found that the similar corrugation and spike domain pattern are both presented before and after annealing procedure, but the grains separated by the nonmagnetic phases at the grain boundaries can be distinguished more clearly in Fig. 1 (b) than that in Fig. 1(a). Meanwhile, the fewer stripe domains at grain boundaries are observed in Fig. 1(b), which illustrates that the stress built in the surface layer during polishing procedure really can change the magnetic domain structure to some extent.

Fig. 2(a), collected with MESP standard tip, represents a greatly blurred image of sample No. 2 (the parallel sample) with serious streaks in the scan direction. Many repeats even with a new MESP tip yield the same result. Only when MESP - HC tip with the high coercivity is used, the

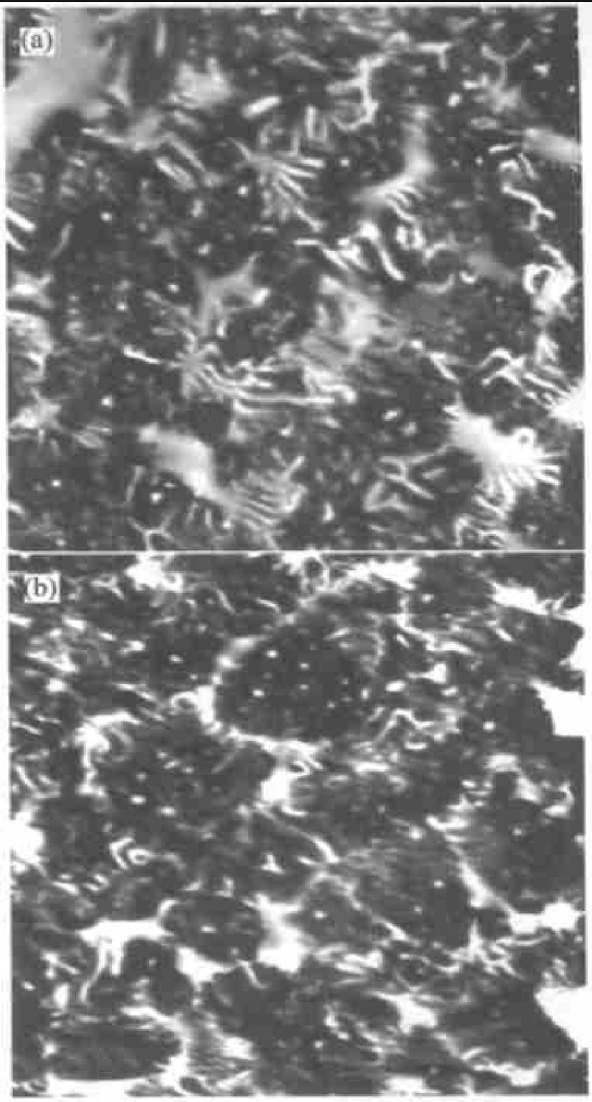

Fig. 1 MFM images of sample No. 1 with scan size of 40 μm × 40 μm and tip lift height of 100 nm
(a) —Before annealing, standard tip;
(b) —After annealing, standard tip

clear images of sample No. 2 before and after annealing procedure are collected as shown in Fig. 2(b) and Fig. 2(c). The plate domains aligning along the orientation direction appear in both images, but more details such as the clear grain boundaries contrast are shown in Fig. 2(c) of the sample after annealing procedure.

4 DISCUSSION

4.1 Effects of surface stress on MFM images

Considering the small magnetostrictive constant in comparison with anisotropy of $Nd_2Fe_{14}B$, Folks deemed that it was not worthwhile concerning the stress induced by polishing procedure because it did not tend to greatly alter the micro-magnetic distribution at the surface[9]. However, the effects of surface stress on the magnetic domain structure are observably indicated in our experiment. Comparing Fig. 1(a) with Fig. 1(b), and

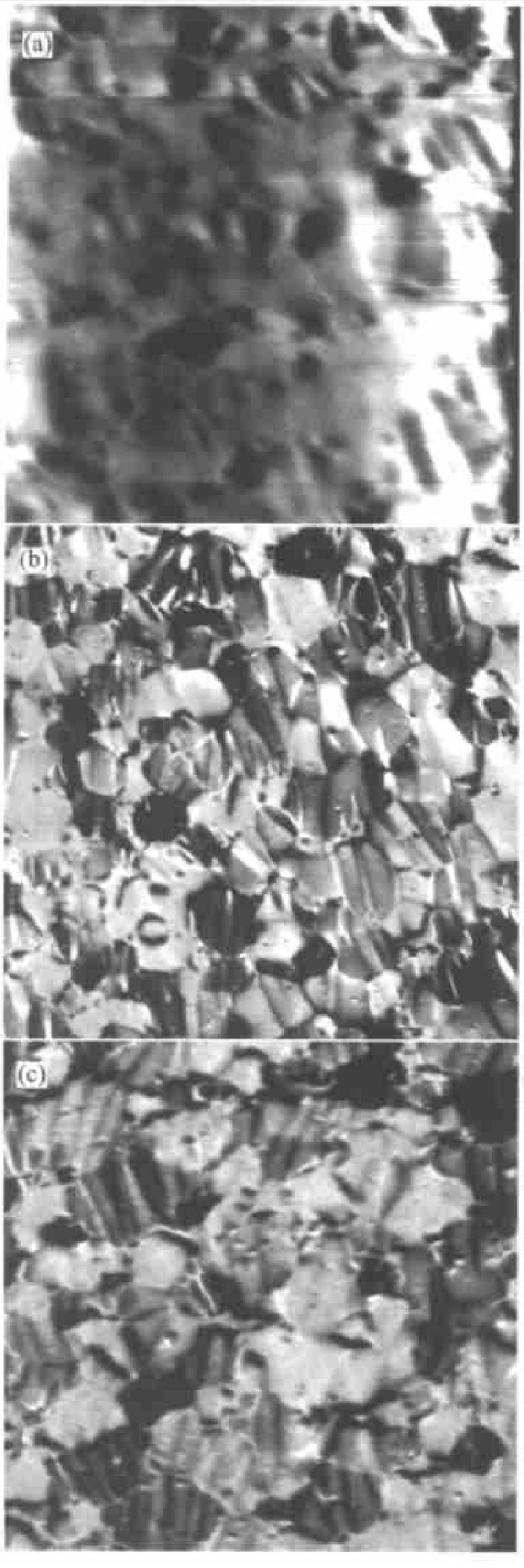

Fig. 2 MFM images of sample No. 2 with scan size of 40 μm × 40 μm and tip lift height of 100 nm
(a) —Before annealing, standard tip;
(b) —Before annealing, high coercivity tip;
(c) —After annealing, high coercivity tip

Fig. 2(b) with Fig. 2(c), it is true that there is not too many differences in the main magnetic domain pattern, for example the same corrugation and spike domain pattern in the perpendicular sample and the same plate domain pattern in the parallel sample are observed. But the stress resulted from polishing confuses the nature of domain structure to some extent. After the annealing procedure, the partial stripe domains of sample No. 1 caused by surface stress are removed and the clear grain boundaries appear in Fig. 1 (b). As the same thing for sample No. 2, the clearly distinguished grains with the plate domains of close orientation direction are shown in Fig. 2(c). This excellent image is of great help for the statistic of grain size and magnetic domain size. Therefore, the annealing procedure is considered to be necessary to obtain the more details and the real micro-magnetic structures in MFM imaging.

4.2 Effects of magnetic tip choice on MFM images

For the strong stray field of the sintered NdFeB, MFM imaging often suffers from the puzzling contrast phenomena for the uncontrolled disturbance on the magnetic MFM tip. Usually, there are two methods to solve this problem: 1) Use the hard tip that will not be remagnetized by sample's stray fields, i.e. the anisotropy field of tip is higher than the stray field of sample, which is termed "hard-tip response"; 2) Use the soft tip that can be easily remagnetized by sample's stray fields, i.e. the active magnetization volume of tip can be totally reversed over the domain boundary walls, which is termed "soft-tip response". The soft tip was verified successfully in Folks' work to give the readily understood MFM images of sintered NdFeB magnet[9]. Here two hard tips of MESP (standard) and MESP-HC (high coercivity tip) are also proved to be effective on this kind of material. Different to the results of Folks who found the serious perturbation of the high coercivity tip with $H_c = 1.114 \times 10^5$ A/m, the clear and readily understood MFM images are collected on sample No. 1 with MESP tip (Fig. 1). But such case does not occur on sample No. 2. MESP tip is greatly remagnetized by the stray field of sample No. 2 as shown in Fig. 2(a), and only MESP-HC tip with the high coercivity can bring through the ideal MFM images on this parallel sample. It seems incompatible with the fact that the perpendicular sample would generate the highest stray fields and gradients. Considering the magnetic flux direction, which is along the alignment axis of tip on the surface of perpendicular sample whereas always parallels to that of tip on the surface of parallel sample, the result is not strange. From this point, the perpendicular sample of high-energy permanent magnets is not the worst case for MFM imaging. It is the NdFeB sample with the easy axis lying in the examined surface that will give the greater disturbance to the magnetic tip. In this case, the real magnetic domain structure can be illustrated only using MESP-HC tip with the enough high coercivity to avoid the remagnetization in plane direction. Incidentally, the deteriorated tip can be repaired to some extent by re-orientation in a strong magnetic field.

4.3 Interpretation of MFM images of sintered Nd-FeB

Presenting as little white dots with diameters of 460 - 900 nm in Fig. 1(b), spike domains are formed on the surface of sample No. 1. To this perpendicular sample, direction of magnetic flux is up and down, resulting in the magnetic charge on the surface. In fact, these adjacent spike domains are the small cone-shaped reverse domains caused by surface magnetic charges to reduce the magnetostatic energy[10]. As for the corrugation and stripe domain pattern, it is due to the small angle of some grains' easy axis off the checked surface.

According to the energy calculation in the micro-magnetism, the adjacent domain on the surface of parallel sample keeps the same pattern as that of the main domain[11]. Therefore, the plate domain of the parallel sample shown in Fig. 2(c) can be taken as the real domain pattern inside the bulk NdFeB magnets.

It was pointed out that the first important thing to obtain the good MFM imaging of sintered NdFeB was to flat the examined surface "with peak-to-valley roughness of the order of tens of nanometers over scales of several micrometers"[9]. However, such careful polish often makes it difficult to discern the grain structure via AFM mode. Fortunately, with our well-collected images after annealing procedure, some topographic information such as grain size, grain boundaries with the nonmagnetic phases can be intuitively observed. For instance, the branched domain along grain boundary and the triple junction phase are shown clearly in Fig. 3. Moreover, the average statistics (for each sample at least three magnetic force images are measured in different areas) of sample's grain size is measured as about 6.0 - 7.5 μm.

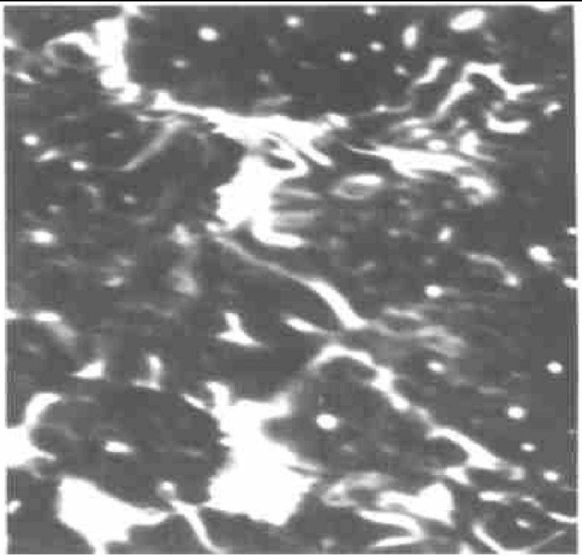

Fig. 3 Re-scanning image from Fig. 1(b) with scale of 20 μm × 20 μm and tip lift height of 100 nm

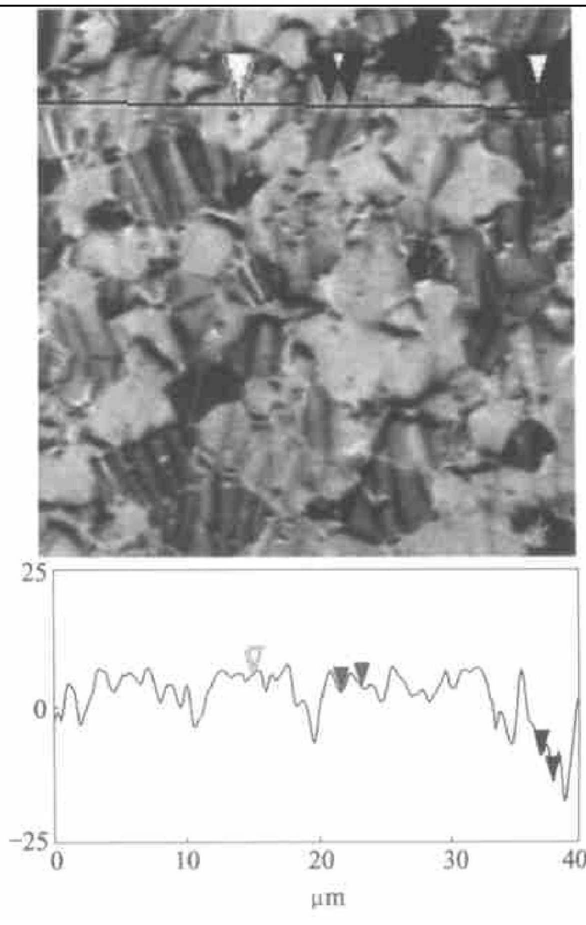

Fig. 4 Line section from MFM image shown in Fig. 2(c)

The domain size can be easily measured in MFM image of perpendicular sample. As Fig. 4 shows, a line scan taken from Fig. 2(c) illustrates the magnetic domain scale of 0.47 - 1.56 μm. Furthermore, the easily distinguished grains with plate domains of the close orientation direction could also be used to demonstrate the sample's alignment degree, which was investigated in Ref. [12]. With the function defined as Eqn. (1), the alignment degree f can be evaluated statistically from magnetic force images:

$$f = \left[\frac{\sum_n \theta_n^2 a_n b_n}{\sum_n a_n b_n} \right]^{\frac{1}{2}} \tag{1}$$

where θ is the angle between the direction of alignment axis and the plate domain orientation, a and b are the length and width of the grains, and n is the number of the grains.

For a completely isotropic magnet, f should be

45°, and for a perfectly oriented magnet, f should be 0°. Since our result calculated by this method is 17.2°, the magnetic orientation procedure should be improved.

5 CONCLUSIONS

1) As the first key to acquire the good MFM images of anitropic NdFeB magnet, well-prepared sample means not only the enough roughness but also the necessary annealing route following polishing to get rid of the surface stress. After this procedure the more details of magnetic structure and topographic information such as grain size and grain boundaries can be obtained.

2) Satisfied MFM images of anisotrophic sintered NdFeB magnets can be collected with the hard tips. For the perpendicular sample, MESP tip is enough whereas the parallel sample must be examined using MESP-HC tip with the high coercivity.

3) Well-captured MFM images provide both magnetic and topographic information of NdFeB magnet. The grain size and the nonmagnetic phases at grain boundaries can be observed in MFM images of the perpendicular sample, and the domain pattern, domain size as well as the alignment degree can be obtained in MFM images of the parallel sample.

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