2012

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Magnetic micro-structural uniformity of die-upset Nd-Fe-B magnets

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(Presented 3 November 2011; received 23 September 2011; accepted 8 December 2011; published online 12 March 2012)

Die-upset $Nd_{13.62}Fe_{75.70}Co_{4.45}B_{5.76}Ga_{0.47}$ magnets have been prepared with height reduction (h) in the range of 60 to 88%. The energy product as high as 50.4 MGOe was obtained in the sample with $h \sim 70\%$. The magnetic domains of the samples are revealed by using magnetic force microscopy (MFM). The average domain widths of the die-upset samples with surface normal parallel (//) and perpendicular ($\perp$) to the loading direction are in the range of $w^{//}$: 0.4-0.6 μm; $w^{\perp}$: 0.9-3.8 μm, respectively. These interaction domains are formed due to the strong inter-granular exchange interaction and magnetostatic interaction between grains. It was found that the ratio of $\phi_{rms}{}^{//}$ to $\phi_{rms}{}^{\perp}$ is a good indicator for the quality of the magnet, where the $\phi_{rms}{}^{//}$ and $\phi_{rms}{}^{\perp}$ are defined as the root-mean-square values of phase shift for the MFM images. The microstructures have been investigated by scanning electron microscopy (SEM), MFM and SEM results indicate the magnetic and crystalline microstructures are uniform for the sample with $h \sim 70\%$, giving rise to the highest magnetic performance among these samples. © *2012 American Institute of Physics*. [doi:10.1063/1.3679423]

I. INTRODUCTION

The knowledge of magnetic domain structures is not only of fundamental interests, but also of technological significances. In particular, it is very important for the understanding of the magnetic properties of magnets and for the development of high-performance magnets.[1] Die-upsetting is a thermomechanical process used to produce fully dense textured nano-scale magnets from magnetically isotropic melt-spun Nd-Fe-B ribbons.[2] The alignment of the magnetically easy axes of the grains in die-upset magnets (i.e., strong magnetic anisotropy) leads to the desired increase in remanence and magnetic energy products.[3–5] The size and properties of the magnetic domain structures, in relation to sample preparation procedures, play an important role for understanding the magnetic reversal process in nanocrystalline magnetic materials.

Magnetic force microscopy (MFM) with high lateral resolution (~10 nm), as a powerful tool to detect magnetic domains of magnetic materials,[6–8] has been used to investigate the interaction domains in die-upset Nd-Fe-B magnets.[9,10] The magnetic domains presented in the previous reports are not enough to illustrate the magnetic characteristics in high performance die-upset Nd-Fe-B magnets, especially on the uniformity of the die-upset Nd-Fe-B magnets that is one of important issues for the practical application of high-performance magnets.

In this paper, we present an investigation of the magnetic microstructures and the structure uniformity of the high-energy-product die-upset Nd-Fe-B magnets.

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II. EXPERIMENTAL

Die-upset Nd-Fe-B magnets were prepared from $Nd_{13.62}Fe_{75.70}Co_{4.45}B_{5.76}Ga_{0.47}$ melt-spun ribbon powders. The die-upsetting was in argon atmosphere at temperatures about 850 °C with different height-reduction (h) of 60%–88%. The size of the die-upset magnets is approximately Φ24 mm × 8 mm. Cylindrical specimens approximately 6 mm in diameter and 4 mm in height were cut from the center of die-upset magnets. And their magnetic properties were measured using hysteresis graph parallel to the loading (die-upset) direction at room temperature. Cubic samples for MFM studies (~4 mm^3) were also cut from the center region of the die-upset magnets to characterize the magnetic properties measured above. Magnetic domains of the samples were revealed using a Digital Instruments D3100 magnetic force microscopy using high coercivity Fe-Pt tips. All the MFM images were obtained at the same scanning height of 50 nm. We have prepared two types of magnetic surfaces for domain imaging studies. One is with surface normal parallel (//) to the die-upset direction; the other is with surface normal perpendicular ($\perp$) to the die-upset direction. The crystalline microstructures of the samples are studied using scanning electron microscopy (SEM).

III. RESULTS AND DISCUSSIONS

The magnetic parameters, such as remanence B_r, intrinsic coercivity H_{cj}, and maximum energy product $(BH)_{max}$, are listed in Table I. The optimal magnetic properties [$(BH)_{max}$ ~50.4 MGOe] are obtained when the h is 70%. The energy products of these samples are comparable to those previously reported in Nd-Fe-B magnets prepared using spark plasma

TABLE I. Magnetic properties (remanence B_r, coercivity H_{cj}, and maximum energy product $(BH)_m$) and parameters of MFM images (average domain width w, root-mean-square values of phase shift of the images ϕ_{rms}) of the die-upset Nd-Fe-B magnets with high reduction h (60, 70, and 88%).

Samples	h (%)	B_r (kGs)	H_{cj} (kOe)	$(BH)_m$ (MGOe)	$w^{//}$ (μm)	$w^{\perp}$ (μm)	$\phi_{rms}^{//}$ (°)	$\phi_{rms}^{\perp}$ (°)	$(\phi_{rms}^{//})/(\phi_{rms}^{\perp})$
A	60	13.54	14.27	43.3	0.40	0.90	4.2	3.5	1.2
B	70	14.16	12.85	50.4	0.53	1–3.5	8.2	1.7	4.8
C	88	13.93	11.32	46.8	0.40	1–3.8	3.4	1.2	2.8

sintering technique[11] and produced from amorphous Nd-Fe-Co-Ga-B materials.[12]

The obtained MFM images (10 μm × 10 μm) are shown in Fig. 1. As shown in the left column [Figs. 1(a), 1(c), and 1(e)], the magnetic domain structures of the parallel samples have maze-like pattern. One can conclude that many grains participate in the formation of an interaction domain in which the grains exhibit the similar orientation of magnetization.[13] Average domain width (w) of MFM images was measured by using the stereological method proposed by Bodenberger and Hubert[14] for these complex domains. Detailed explanation on this method can also be found in Ref. 1. The $w^{//}$ values of these samples are about 0.40 μm, 0.53 μm, and 0.40 μm for the domain patterns at Figs. 1(a), 1(c), and 1(e), respectively. The $w^{//}$ of the samples with $h \sim 70\%$ is the largest, indicating there exists the strongest interaction between the grains. The formation of these magnetic domain patterns can be interpreted by considering the energies that are associated with the surface free magnetic poles and the domain walls.[15] Its total energy is minimized when the reduction in magnetostatic energy due to having smaller domains that matches the energy spent in creating new domain walls. In these complex domain structures, the domain walls are likely laying along grain boundaries that is similar to that reported by Griffiths *et al.*[16] Moreover, there is no elongation in the shape of the interaction domains for these samples so that the two-dimensional Fourier transform (TDFT) of the corresponding MFM image, as shown in the insets in Figs. 1(a), 1(c), and 1(e), exhibits no directionality.

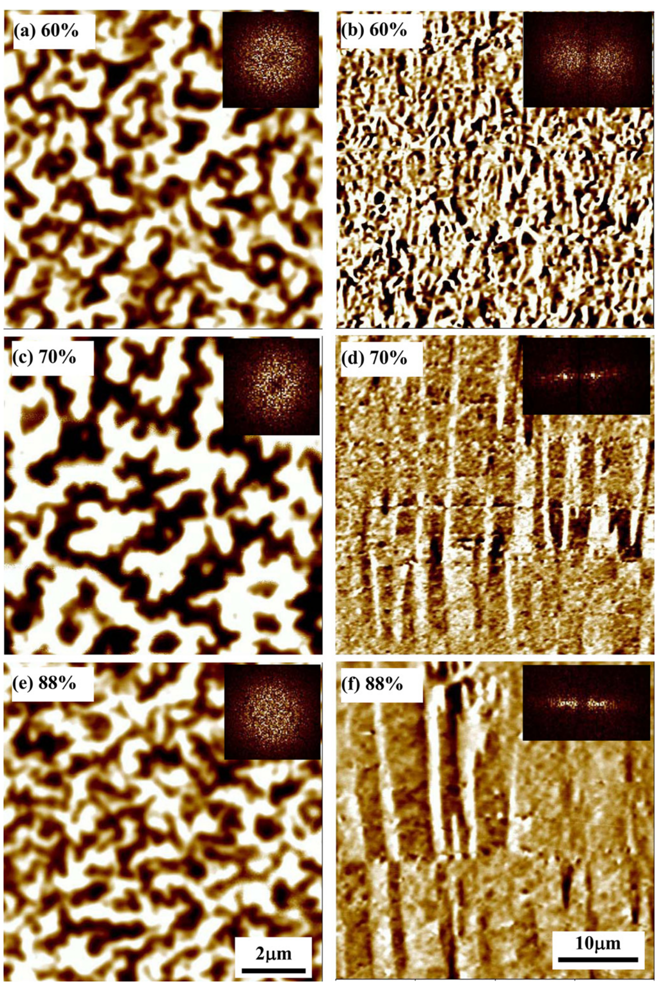

FIG. 1. (Color online) MFM images of the die-upset Nd-Fe-B parallel (left column) and perpendicular (right column) samples with different height reduction (60%–88%). (a) and (b): 60%, (c) and (d): 70%, (e) and (f): 88%. Insets are the corresponding two-dimensional Fourier transform patterns.

For a better understanding of magnetic domain structures, we have also studied the sample with surface normal perpendicular to the loading direction. The corresponding MFM images are shown in Figs. 1(b), 1(d), and 1(e) (right column) for h of 60–88%, respectively. A large scan size (40 μm × 40 μm) is needed to present the domain configurations fully. On the whole, their magnetic domain show plate-like patterns that are different from those maze-like patterns presented in the left column of Fig. 1. The average domain width $w^{\perp}$ is in the range of 0.9 μm–3.8 μm. From the corresponding TDFT patterns of the sample with $h = 60\%$ [inset of Fig. 1(b)], the Fourier transform pattern shows a slightly directionality that have a narrow plate-like pattern configurations with domain width (~0.9 μm). When increasing h, the directionality is more pronounced as shown in the TDFT patterns (see the insets of Figs. 1(d) and 1(f)). In general, the $w^{\perp}$ values are much larger than $w^{//}$. It is due to the surface demagnetization field for perpendicular samples are much smaller than that of parallel samples.

From the MFM images (Figs. 1(a)–1(f)), the root-mean-square values of phase shift of the MFM images, ϕ_{rms}, were calculated using *Roughness Analysis* assembled with MFM. The $\phi_{rms}^{//}$, $\phi_{rms}^{\perp}$, and $\phi_{rms}^{//}/\phi_{rms}^{\perp}$ of the parallel and perpendicular samples are listed in Table I. In general, the $\phi_{rms}^{\perp}$ is less than the $\phi_{rms}^{//}$ because stray field in the perpendicular sample surface is smaller than that of the parallel sample. The magnetic domains are revealed by MFM through detecting the interaction between magnetic tips and magnetic stray fields emerged from sample surface. The value of ϕ_{rms} is the standard deviation of the phase shift ϕ within the given scan area, and it can be calculated by

$$\phi_{rms} = \sqrt{\frac{\sum_i (\phi_i - \phi_{ave})^2}{N}},$$

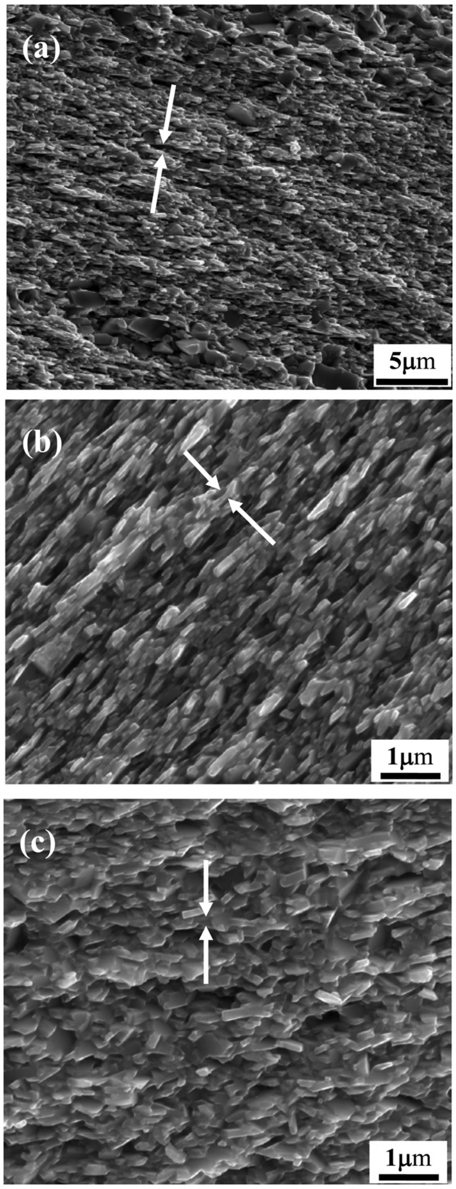

FIG. 2. SEM images of the die-upset Nd-Fe-B perpendicular samples with $h = 60–88\%$. The arrows show the die-upset direction.

where ϕ_{ave} is the average of the ϕ values within the given area, ϕ_i is the ith ϕ value, and N is the number of the pixels within the given area ($\sim 512 \times 512$). Generally, the evolution of $\phi_{rms}^{//}$ of the MFM images can be used to indicate the variation of the magnetic properties of the samples. However, we think that the ratio of ($\phi_{rms}^{//}$) to ($\phi_{rms}^{\perp}$) may be a better indicator for the quality of the die-upset Nd-Fe-B magnets. The higher the $\phi_{rms}^{//}/\phi_{rms}^{\perp}$ values, the better the magnetic performance of the samples. The highest $\phi_{rms}^{//}/\phi_{rms}^{\perp}$ value of $\sim$4.8 for the sample with h of 70% is in good agreement with the best magnetic properties [$(BH)_{max}$ of $\sim$50 MGOe].

In order to have a better understanding of the structure uniformity of the die-upset magnets, we have carried on SEM on perpendicular sample. As shown in Fig. 2, the size and shape of the grains of the samples could be recognized in the SEM images. The loading directions are marked in these images. It was found that the grain size distribution of the sample with $h = 60\%$ is non-uniform. The existence of some big grains at the left-bottom region of Fig. 2(a) is clear. However, for the sample with $h = 70\%$, the microstructure was composed of elongated nanocrystalline grains with their length perpendicular to the loading direction. The grains have even size distribution with their width of about 50–65 nm and their length of 300–500 nm. The uniform microstructure was expected to be responsible for the excellent magnetic properties of the magnets. With increasing h by 88%, it is clear that the grain distribution becomes non-uniform as compared with Fig. 2(b) even if the grain size in these two images is similar.

IV. CONCLUSIONS

The energy product of the die-upset Nd-Fe-B magnet with height reduction of 70% is as high as $\sim$50.4 MGOe. The ratio of root-mean-square values of phase shift of MFM images, $\phi_{rms}^{//}$ to $\phi_{rms}^{\perp}$, is found to be a good indicator for the quality of the die-upset Nd-Fe-B magnets. The higher the $\phi_{rms}^{//}/\phi_{rms}^{\perp}$ values, the better the magnetic performance of the samples. Uniformity on microstructure plays an important role on the achievement of high magnetic properties.

This work was in part supported by DOD (ARO) W911NF-08-1-01311(Development of advance cantilevers for magnetic domain imaging), W911NF-10-2-0099 (High-performance magnets), and NSF MRSEC DMR-0820521 (Magnetic nanostructures), the National Natural Science Foundation of China (50901021, 50931001), the National Basic Research Program of China (2010CB934601), and the National High-tech R&D Program of China (2011AA03A401).

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