Magnetic coupling in boron-rich NeFeB nanocomposites

B. Lu,[a)] M. Q. Huang, Q. Chen,[b)] B. M. Ma,[b)] and D. E. Laughlin
Department of Materials Science and Engineering, Carnegie Mellon University, Pittsburgh, Pennsylvania 15213

In this article, we report types of magnetic domain patterns caused by either dipolar coupling or exchange coupling inside a heat-treated boron-rich NdFeB melt-spun ribbon. The average grain size of the sample was about 38 nm. Single particle domains were commonly observed in Fresnel images all through the specimen, suggesting little exchange coupling among the grains in these regions. Snake-shaped long interactive domains consisting of chains of grains were found in many Foucault images at the regions where exchange coupling is weak. These domains are caused by dipolar coupling along the chains. Wider domains consisting of many grains were also observed by Fresnel images, although they were much less common than the single particle domains. The grains inside a wider domain are believed to be exchange coupled with each other. The magnetic properties of such boron-rich NdFeB composites are influenced more by the dipolar coupling than by exchange coupling between the nanocrystalline grains.
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I. INTRODUCTION

The magnetic properties of hard magnetic nanocomposites are strongly influenced by magnetic coupling between the nanocrystalline grains.[1] It has been found[2] that $Nd_2Fe_{14}B$ crystalline grains changed from multidomain grains to single-domain grains as the grain size decreases from 3000 to 30 nm. Since the critical single-domain particle size of $Nd_2Fe_{14}B$ is about 300 nm, the intergranular magnetic coupling becomes more pronounced when the grain size decreases below this value. At this stage, the remanence is enhanced through the exchange coupling effect.[3] However, the coercivity will decrease[4] due to both exchange coupling and dipolar coupling among the grains. Therefore there is a limit to the enhancement of the energy product that can be achieved by the refinement of grains.

A further improvement of remanence can be accomplished by adding soft ferromagnetic phases with larger M_s but a smaller grain size, which is comparable to the domain wall width of the hard magnetic phase.[5] Due to the exchange coupling between the soft and the hard phases, the entire exchange coupling effect inside the material will be enhanced. Also, due to the high M_s of the soft magnetic phase, the magnetostatic coupling between the nanocrystalline grains will be strengthened. Therefore it is important to obtain a clear picture of the magnetic domain structure inside this kind of nanocomposite.

In this article, a heat-treated boron-rich NdFeB melt-spun sample was studied by Lorentz transmission electron microscopy (LTEM). The remanence of the nanocomposite was confirmed to be enhanced by soft magnetic phases, such as, α-Fe and Fe_3B in a previous study.[6] The patterns of both exchange coupling and dipolar coupling among the grains were investigated by Fresnel imaging and Foucault imaging technique of LTEM. The aim of this research is to obtain a better understanding of the relationship between the magnetic couplings and the magnetic properties in this kind of NdFeB nanocomposite.

II. EXPERIMENT

Samples of NdFeB nanocomposites with nominal composition $(Nd_{0.95}La_{0.05})_{9.5}Fe_{bal}Co_5Nb_2B_{10.5}$ were prepared by the melt-spinning process followed by heat treatment for 10 min at different temperatures. It has been found by conventional transmission electron microscopy (CTEM)[6] that the crystalline grains grew from the as-spun amorphous state through a partially crystallized state and finally to a fully crystallized state. The 700 °C heat-treated sample has the optimum magnetic properties and the narrowest grain size dispersion. Hence it was chosen to be investigated by LTEM.

Since the 20 μm thick ribbon was brittle, it was ion milled directly without mechanical polishing in a Gatan 600 ion miller equipped with a liquid N_2 stage. This was followed by ion polishing for less than 5 min at room temperature with Gatan 691 precision ion polishing system (PIPS).

CTEM and LTEM observations were performed using a Phillip-420 transmission electron microscope (TEM) and a JEM-4000 TEM. The latter TEM was operated at 400 kV with its object lens current shut off. In this way the specimen was located in a magnetic field-free space suitable for the study of magnetic domain structures. Bright field images were obtained to display the grain size, while Fresnel and Foucault images from the same region were taken to provide information on the magnetic domain geometry and the direction of magnetization within individual domains.

III. RESULTS AND DISCUSSION

The grain size of the sample varies from 3 to 72 nm with an average of 38 nm. This feature is uniform through the specimen. Figure 1 shows a bright field image of the sample. Some small precipitates with a size less than 5 nm can be

[a)]Electronic mail: binlu@andrew.cmu.edu
[b)]Also with: Rhodia Inc., Rare Earths and Gallium, CN 7500, Cranbury, NJ 08512.

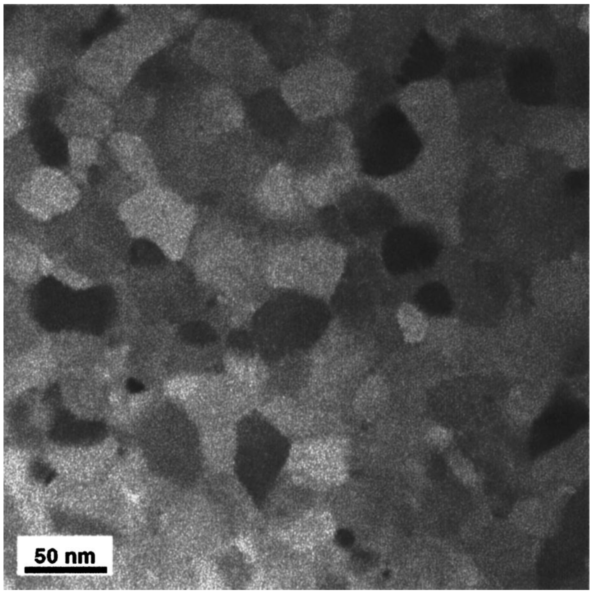

FIG. 1. Bright field image of the NdFeB nanocomposite.

seen at the grain boundaries. These are believed to be α-Fe and Fe_3B grains. This was confirmed[6] by microdiffraction of samples that were annealed at high temperature where these precipitates grew larger.

Figure 2(a) is a Fresnel image displaying a typical region where the magnetic domains are of the same size as the grains. The magnetic contrast arises only at grain boundaries, which indicate there is weak or no correlation between the magnetization of neighboring grains. Therefore, in a thermally demagnetized state, the direction of magnetization inside each grain is close to its own easy axis. Figure 2(b), which is also a Fresnel image, presents the domain structure in another area, which is less common in the sample. Here the domain size is much larger than the grain size. It can be seen that there is zero contrast within the domain, indicating the magnetization directions of the grains inside a domain are very close to each other. The multiparticle domains suggest there is strong exchange coupling between the magnetization of the neighboring grains inside each of them. In this case, the magnetization direction of each grain no longer remains along its own easy axis. The domain boundaries occur only at the grain boundaries where the intergranular exchange couplings are weak.

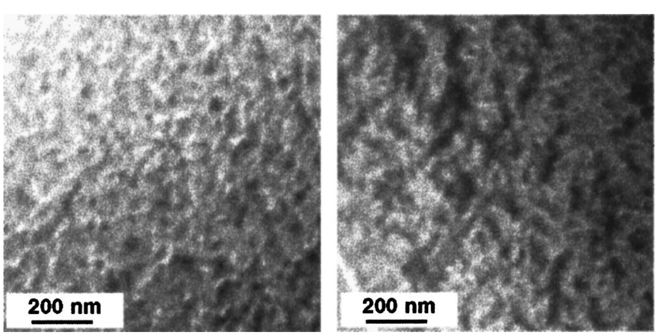

FIG. 2. Fresnel images showing: (a) single particle domains, and (b) multiparticle domains.

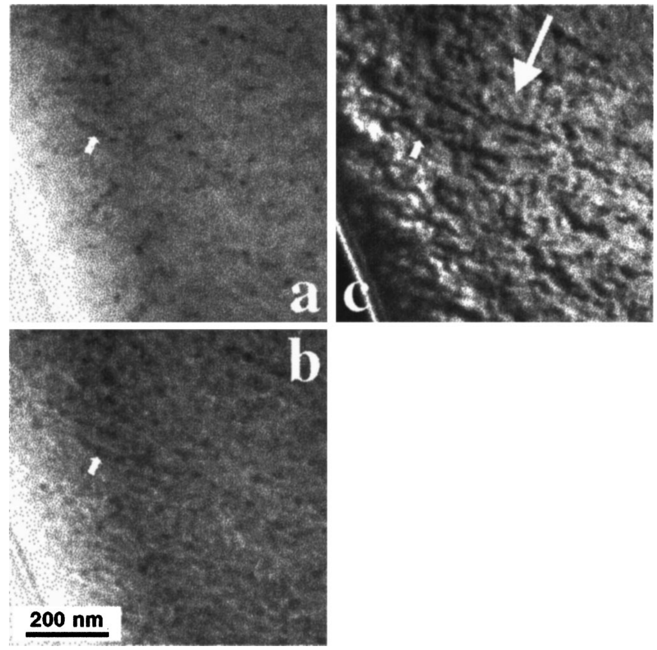

FIG. 3. Region of dipolar coupled grains: (a) bright filed images, (b) Fresnel image, (c) Foucault image.

Figure 3 shows a bright field image and corresponding Fresnel image and Foucault image. The domain size shown in the Fresnel [Fig. 3(b)] is nearly the same as the grain size in the bright field image [Fig. 3(a)], indicating there is little intergranular exchange coupling in this area. However, in the Foucault image [Fig. 3(c)] there are some stripes of about 200 nm length and 40 nm width. The long white arrow points the displacement direction of the aperture. The reason why there are no stripes in the Fresnel image is that the magnetization directions of the grains in the stripes in the Foucault image are not exactly parallel to each other. Therefore magnetic contrast arises at the boundaries of the neighboring grains. However, because the magnetization of the grains has components of the same sign in the direction perpendicular to the displacement of the aperture, they present the same contrast in the Foucault image.

According to the above analysis, it is interesting to note the stripe pointed out by an arrow in Fig. 3(c). This stripe has its corresponding bright field image and Fresnel image in Figs. 3(a) and 3(b) (shown by an arrow), respectively. Figure 3(a) shows that there is some diffraction contrast between the grains inside the strip. In Fig. 3(b) some magnetic contrast is added up to the diffraction contrasts at the grain boundaries due to the difference in magnetization directions of the grains. However, there is no contrast inside the stripe in Fig. 3(c). This feature of arrangement of directions in a stripe strongly suggests a dipolar coupling between the grains. The grains seem connected one by one by their magnetization in a zigzag way. This gives rise to the above observations. In contrast to this case, the exchange coupling causes the grains to be more parallel to each other side by side. Therefore the domains revealed by both Fresnel and Foucault images should be wider than those stripes.

Figure 4 comprises a bright field image and the corresponding Fresnel image and pair of Foucault images with

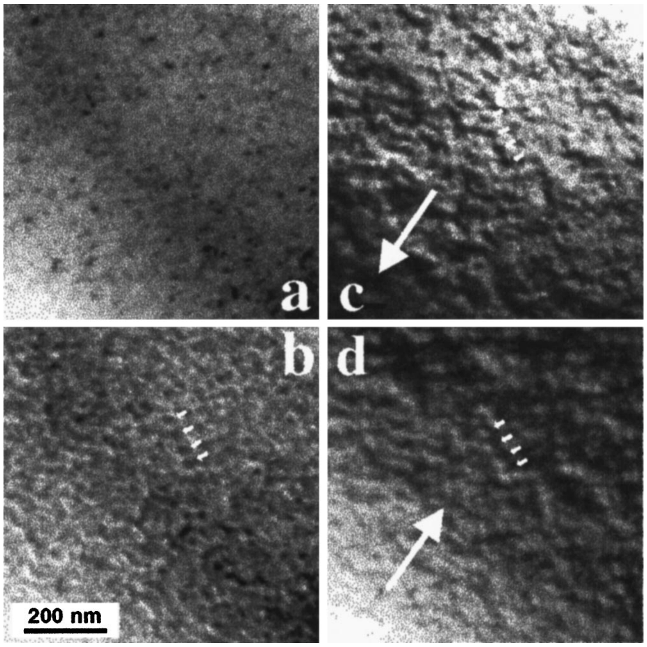

FIG. 4. Region of dipolar coupled grains showing snake-shaped long interactive chains: (a) bright filed images, (b) Fresnel image, (c) and (d) pair of Foucault images with opposite displacement of the aperture.

opposite aperture displacement (long white arrows). It can be concluded from the bright field image [Fig. 4(a)] and the Fresnel image [Fig. 4(b)] that little exchange coupling is observed in this region. On the other hand, the Foucault images [Figs. 4(c) and 4(d)] show some snake-shaped chains. The longest chain is shown by arrows in the center of Figs. 4(c) and 4(d). The wavy feature of the line also suggests dipolar coupling of the neighboring grains along the line. The magnetization directions in these grains are not parallel to each other. They are, however, roughly aligned in a zigzag way. Consequently, they have same contrast in the Foucault images, but they are observed as single particle domains in the Fresnel image. The four grains pointed out by arrows in the Fresnel image are separated by magnetic contrast at the grain boundaries due to the difference of the magnetization directions. The lower three grains can be seen in the Foucault images joined in forming the wavy interactive domain caused by dipolar coupling, while the top is out of the position.

It is very common to find weak intergranular exchange coupling areas inside the specimen. In these areas, dipolar coupling between the grains can be easily observed. This effect is probably due to the soft magnetic phases at the grain boundaries of the $Nd_2Fe_{14}B$ grains. On one hand, grains of soft phases cannot enhance the exchange coupling between the grains of hard phases, although they themselves are exchange coupled with the hard phases. On the other hand, due to their high M_s, the soft phase can enhance the dipolar coupling between the hard phases. Figure 5 is a typical Foucault image of the specimen showing more snake-shaped interactive domains caused by dipolar coupling.

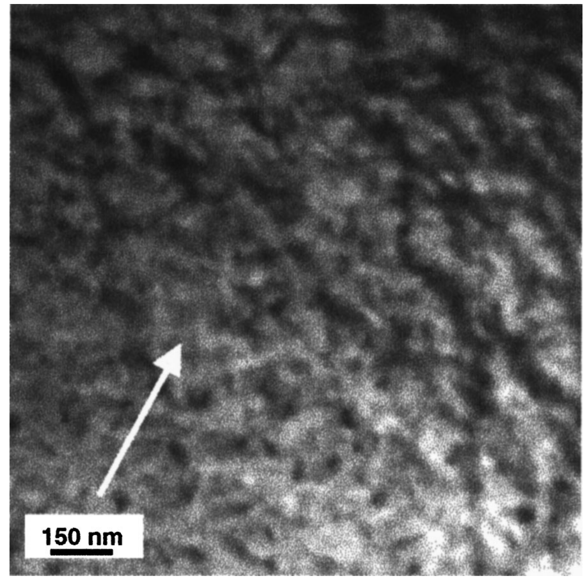

FIG. 5. Dipolar coupled wavy chains in a typical area of weak intergranular exchange coupling.

IV. CONCLUSION

We have shown the pattern of magnetic coupling inside a heat-treated boron-rich NdFeB melt-spun ribbon, with its nominal composition as $(Nd_{0.95}La_{0.05})_{9.5}Fe_{bal}Co_5Nb_2B_{10.5}$. Dipolar couplings between the grains are very commonly observed all through the specimen, where exchange coupling between the grains is very weak. Most of the interactive domains have a wavy feature. There are some parts of the sample where strong intergranular exchange coupling can be observed. But it is much less common than the dipolar coupling. The enhancement of dipolar coupling is probably due to the soft magnetic phases at the grain boundaries. It can be concluded that the magnetic properties of the studied NdFeB nanocomposite with a soft magnetic phase are influenced much more by the dipolar coupling than exchange coupling among the grains.

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