Domain structure of hard magnetic NdAlFeCo bulk metallic glass

WEI Bingchen[1], WANG Weihua[1,2], PAN Mingxiang[2], HAN Baoshan[3], ZHANG Zhenrong[3] & HU Wenrui[1]

1. National Microgravity Laboratory, Institute of Mechanics, Beijing 100080, China;
2. Institute of Physics and Center for Condensed Matter Physics, Chinese Academy of Sciences, Beijing 100080, China;
3. State Key Laboratory of Magnetism, Institute of Physics and Center for Condensed Matter Physics, Chinese Academy of Sciences, Beijing 100080, China

Correspondence should be addressed to Wei Bingchen (e-mail: weibc @imech.ac.cn)

Abstract **Magnetic domain structure of hard magnetic $Nd_{60}Al_{10}Fe_{20}Co_{10}$ bulk metallic glass (BMG) has been studied by using magnetic force microscopy. In the magnetic force images it is shown that the exchange interaction type magnetic domains with a period of about 360 nm do exist in the BMG, which is believed to be associated with the appearance of hard-magnetic properties in this system. As the scale of the magnetic domain is much larger than the size of the short-range ordered atomic clusters existing in the BMG, it is believed that the large areas of magnetic contrast are actually a collection of a group of clusters aligned in parallel by strong exchange coupling interaction. After fully crystallization, the BMG exhibits paramagnetism. No obvious magnetic contrast is observed in the magnetic force images of fully crystallized samples, except for a small quantity of ferromagnetic crystalline phase with low coercivity and an average size of 900 nm.**

Keywords: metallic glass, hard magnetism, magnetic force microscopy, exchange coupling interaction.

Nd-Fe-Al based bulk metallic glasses (BMG) with a diameter up to 12 cm have been recently prepared[1,2]. This BMG system has evoked an extensive attention due to its hard magnetic properties at room temperature[3—7]. The high coercivity of these amorphous alloys is striking because no structural anisotropy should exist in the disordered packing solid. It is supposed that the BMG is an ensemble of short-scale ordered magnetic atomic clusters in uniform distribution. These clusters consist of Nd and transition elements (TM) and possess large random anisotropy. It is the magnetic exchange coupling among them that causes the high coercivity of this magnetic system[2]. Most of the consequent results supported this model[5,6]. However, no direct experimental evidence for the existence of exchange coupling interaction in this system has been presented so far. Therefore, it is of a special interest to study the magnetic domain structure of the BMG. In this note, the magnetic domain structure of $Nd_{60}Al_{10}$-$Fe_{20}Co_{10}$ was investigated by magnetic force microscopy (MFM).

1 Materials and methods

Ingots with compositions of $Nd_{60}Al_{10}Fe_{20}Co_{10}$ were prepared by arc melting from elemental Nd, Fe, Al and Co with a purity of 99.9% in a titanium-gettered argon atmosphere. Cylindrical specimens of around 3 mm in diameter and 50 mm in length were prepared from the ingots by die casting them into a copper mold under argon atmosphere. The structure of the as-cast cylinder was characterized by X-ray diffraction (XRD). Magnetic measurements were performed using vibrating sample magnetometer (VSM) with a maximum applied field of 1592 kA • m^{-1}. The study of domain structure was carried out by using Digital Instruments NanoScope Ⅲa D-3000 MFM. It allows the topographic and magnetic force images to be collected separately and simultaneously in the same area of the sample by using Tapping/Lift modes. The magnetic tips used were micro-fabricated Si cantilevers with pyramidal tip coated with magnetic Co-Cr thin film of a 40- nm thickness and a coercivity of about 32 kA • m^{-1}. In our experiment, the tip used was magnetized upward prior to imaging. Its f_0 = 80.6 kHz, and the lift-height during scanning was 30 nm. The details of principles and techniques for MFM study can be seen in refs. [8, 9]. The samples for MFM study were cut from the cylinder, ground and polished, followed by vacuum annealing to remove the stress built in the surface layer.

2 Results and discussion

The as-cast cylinder exhibits an XRD spectrum typical for amorphous phase without obvious crystalline reflection peaks. Fig. 1(a) shows the dependence of hysteresis loops on isothermal annealing temperature for the $Nd_{60}Al_{10}Fe_{20}Co_{10}$ BMG, and the annealing duration is 1.8 ks. It is shown that the as-cast $Nd_{60}Al_{10}Fe_{20}Co_{10}$ BMG exhibits hard magnetism with H_{CM} of 326 kA • m^{-1}, σ_s of 10.8 A • m^2 • kg^{-1}, and σ_r of 7.2 A • m^2 • kg^{-1}. These values are in consistence with the previous results of Nd-Fe-Al based BMGs[1—3], indicating that the BMG is permanent magnetic material with relative high coercivity and rather low magnetization compared with the nano-composite rare- earth permanent magnets. The coercivity of the BMG remains almost unchanged in the annealing temperature ranging from room temperature up to 740 K, and disappears after full crystallization of the BMG above 760 K. The presence of only one magnetic phase in the as-cast cylinders is confirmed by the curve of magnetization vs. temperature (fig. 1(b)), in which only one magnetic transition appears at Curie temperature of about 470 K before crystallization. The above results have demonstrated that the hard magnetic properties of the Nd-Fe-Al based BMGs are presented by the amorphous phase instead of the crystallized one, which has also been confirmed by Ding[3], Wei[4], Fan[5] and Xing et al.[6]. MFM was employed to image the magnetic domain structure of amorphous phase in this BMG.

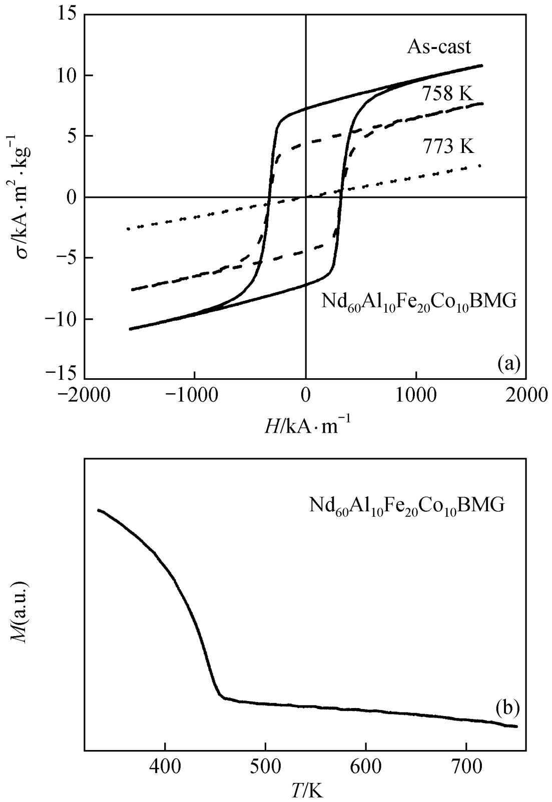

Fig. 1. Dependence of hysteresis loop on isothermal annealing temperature for $Nd_{60}Al_{10}Fe_{20}Co_{10}$ BMG. (a) Annealing for 1.8 ks; (b) *M-T* curve.

A typical 10 μm × 10 μm magnetic force image for the as-cast sample is shown in fig. 2(a). Obvious magnetic contrast was observed in the magnetic force images. It can be seen that the image is characterized by darker areas adjacent with brighter areas in sub-micron scale and in random distribution. Higher magnification images from the as-cast sample did not provide additional detail. In fact, the dark area indicates that the magnetization direction in this area is nearly parallel to the upward tip magnetization, and the bright area indicates the opposite. In order to verify the reliability of these images the samples were re-scanned under the same experimental conditions and the practically identical images as its first scanning ones were obtained. The average period (T) of the domain pattern and the average contrast between dark and bright areas were measured by means of the section analysis. A large number of sections were analyzed in order to get statistical results. The section analysis of $\Delta\phi$ vs. length of the A-A section is shown in fig. 2(b). It can be seen that the average roughness $(R_a)_{\Delta\phi}$ and root mean square $(RMS)_{\Delta\phi}$ describing the contrast of the image are 0.94° and 0.72°, respectively, and T = 360 nm.

After annealing at 773 K for 1.8 ks, the sample crystallizes completely, and exhibits paramagnetism as shown in fig. 1(a). Fig. 3(a) shows its magnetic force image with

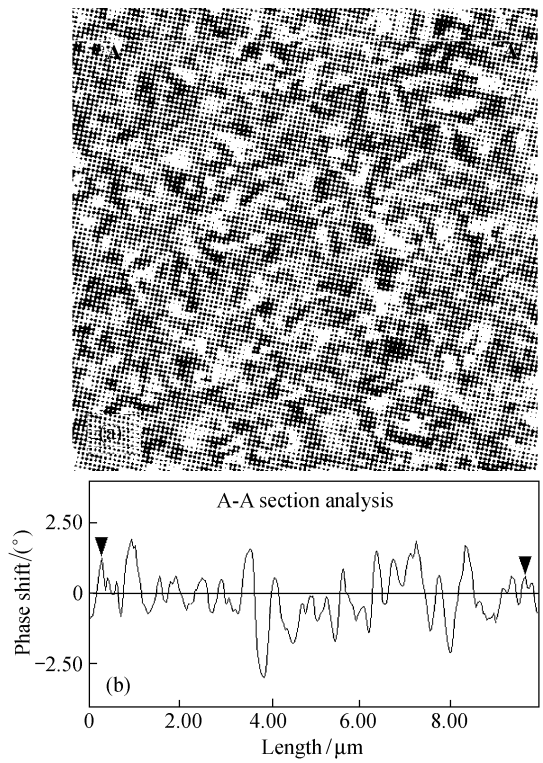

Fig. 2. Magnetic force image with a scan size 10 μm × 10 μm for the as-cast $Nd_{60}Al_{10}Fe_{20}Co_{10}$ BMG (a) and the result of A-A section analysis (b).

a scan size of 20 μm × 20 μm. It is revealed that the magnetic force image of the fully crystallized sample is much different with that of the as-cast one. No obvious widely-distributed magnetic contrast is observed in this figure, except for a small quantity of dark block-like magnetic domains. The magnetic contrast indicates that the magnetic domains are of ferromagnetic crystalline phase. A statistical analysis on the images shows that these ferromagnetic phases possess an average size of 900 nm, and volume fraction of 12%. Fig. 3(b) is the section analysis results of B-B section shown in fig. 3(a). It is shown that the average $\Delta\phi$ for the ferromagnetic phases is –2°, indicating that these phases are magnetized upward. Because no bright magnetic contrast is observed around these ferromagnetic phases, it is suggested that these phases possess a low coercivity, and they are magnetized by the magnetic tip as the tip scans the sample.

The magnetic contrast of the as-cast sample is similar to the so-called "exchange coupling domains" presented in the nano-composite RE (rare-earth)-TM (transition metal) permanent materials, in which strong exchange coupling interaction exists between hard and soft magnetic grains[10]. However, no ferromagnetic crystalline grains exist in the BMG. According to the cluster model of the Nd-Fe melt-spinning magnetic system, the exchange coupling interaction among magnetic clusters with ran-

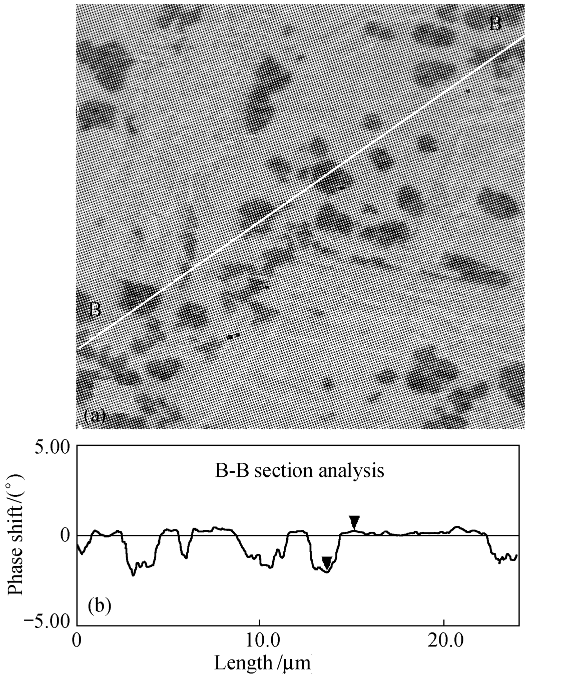

Fig. 3. Magnetic force image with a scan size 20 μm × 20 μm for the $Nd_{60}Al_{10}Fe_{20}Co_{10}$ BMG annealed at 773 K for 1.8 ks (a), and the result of B-B section analysis (b).

dom anisotropy could cause the high coercivity of the-magnetic system[11]. Inoue confirmed the presence of these short range ordered clusters in the Nd-Fe-Al system by high-resolution transmission electron microscopy and radial distribution function studies. Fan and Xing et al. have estimated the size of the clusters to be nearly 2 nm[5,6]. In the present study, the as-cast BMG exhibits a typical amorphous pattern without obvious crystalline reflection peaks. This gives an upper limit of a few nanometers for the cluster size[5]. However, the above section analysis of the magnetic force images of the $Nd_{60}Al_{10}Fe_{20}Co_{10}$ BMG has shown that their magnetic domains are in sub-micron scale, which is significantly larger than the size of the ordered atomic clusters composed of the Nd-Fe and Nd-Co atoms. We infer that this would be an experimental evidence of the existence of strong exchange coupling. It is by the exchange coupling that a great deal of short-scale ordered atomic clusters are aligned to form the large-scale domains. In other words, the large areas of magnetic contrast are actually a collection of a group of clusters with similar magnetic orientation aligned by exchange coupling. Furthermore, the presence of strong exchange coupling is also confirmed by the significant remanence enhancement of our BMG. Its M_r/M_s value is 0.67, which is obviously greater than 0.5. This value predicted randomly oriented and non-interacting particles. Considering that the dimension of the ordered magnetic clusters of present hard magnetic material has an upper limit of a few nanometers, the number of clusters in one group could be estimated as 10^3—10^4.

Hexagonal δ phase and hexagonal Nd phase are main crystallized products of $Nd_{60}Al_{10}Fe_{20}Co_{10}$ BMG annealed at 773 K for 1.8 ks. They exhibit paramagnetism at room temperature, thereby no widely distributed magnetic contrast is revealed in the magnetic force image as shown in fig. 3(a). However, a small quantity of dark magnetic domain exists in the magnetic force image of the fully crystallized sample, which indicates the presence of ferromagnetic crystalline phases in the matrix. The chemical ingredient and structure of these phases are still unknown. These phases do not obviously affect the systemic magnetic properties due to its low volume fraction and large interval.

3 Conclusions

The magnetic domain structures of hard magnetic $Nd_{60}Al_{10}Fe_{20}Co_{10}$ BMG were studied by magnetic force microscopy, and they exhibit similar features to those of nano-composite RE-transition metal permanent materials. The magnetic domains have a length scale of about 360 nm. The large area of magnetic contrast is composed of thousands of short-range ordered atomic clusters aligned by strong exchange coupling interaction. The presence of exchange coupling interaction in the BMG is confirmed by magnetic force microscopy studies. After the full crystallization, no widely distributed magnetic contrast is revealed in the magnetic force images, but a small quantity of magnetic domains still exists.

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