Journal of Magnetism and Magnetic Materials 322 (2010) 2797–2800

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Domain imaging in FINEMET ribbons

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ARTICLE INFO

Article history:
Received 31 March 2010
Received in revised form
7 April 2010
Available online 22 April 2010

Keywords:
Magnetic domain
Bitter
Soft magnetic material
FINEMET
Magnetostriction

ABSTRACT

The magnetization behaviour of a ferromagnetic material depends on its domain structure, which in turn is largely determined by magnetic anisotropies. In this work, domain patterns were observed by a quite forgotten but still the simplest and the cheapest technique: the Bitter method. A systematic study of the evolution of the domain structure in FINEMET ribbons after thermal annealing is presented, correlating the results with the crystalline structure, magnetostriction and coercivity measurements.

1. Introduction

There has been a great scientific and technological interest on the development of magnetic materials for more than a century.

In 1988, Yoshizawa et al. [1] opened a whole new area for research and application purposes by developing the nanocrystalline alloy, the so-called FINEMET ($Fe_{73.5}Si_{13.5}$ $B_9Nb_3Cu_1$), which due to its special two phase structures, i.e., crystals 10–20 nm size embedded in an amorphous matrix exhibits extremely soft magnetic properties (low coercivity $H_C \leq 1$ A/m and high permeability ($\mu_{r(1\ kHz)} \geq 10^4$) together with high saturation magnetization. Such properties make them suitable for a wide variety of technological applications such as transformer cores, inductive devices, magnetic shielding, sensors, etc. [2].

The extrinsic magnetization behaviour of ferromagnetic materials depends strongly on their domain structure, which in turn is largely determined by magnetic anisotropies. However, the knowledge of such structure was not exhaustively considered on the development of soft magnetic materials. Two other magnitudes that play an important role on the domain structure are magnetostriction (which relates stresses with domains orientation) and coercivity (which indicates the mobility of domain walls).

Therefore, it was of great practical interest to the authors to characterize and understand FINEMET's domain structure together with the crystalline structure, magnetostriction and coercivity, not only in the as-quenched but also in the annealed state at 540 and 650 °C.

Nowadays, new imaging methods such as MFM (magnetic force microscopy) [3] and MOKE (magneto-optical Kerr effect) [4,5] became very popular. However, on the one hand, these techniques are neither suitable for studies of rough surfaces nor to sharply disclose maze structures present in these ribbons. On the other hand, latest advances in digital imaging techniques made the old and almost forgotten Bitter method much more practical and powerful.

The evolution of FINEMET's domain patterns with heat treatments at different temperatures was observed with this method and compared with magnetostriction and coercivity variation.

2. Principles of Bitter method

This is the oldest domain imaging technique which owes its name to Francis Bitter, who reported it in 1931 [6]. Although he was not able to deeply understand the observed patterns, some conclusions could be stated: domains were static, they could be rather wide and had usually a periodic appearance.

Although it was discovered almost eight decades ago, it is still the simplest method. The magnetic surface is covered by a very thin and uniform layer of ferrofluid, i.e. a colloidal suspension of Fe_3O_4 particles, which can be home-made or commercial (e.g. ferrofluids from Ferrofluidics Corp., Nashua, New Hampshire, and lignosite FML from Georgia Pacific Corp., Tacoma, Washington).

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0304-8853/$ - see front matter
doi:10.1016/j.jmmm.2010.04.030

The particles collect and agglomerate in regions where stray fields are present, typically on the domain walls. Therefore, with this technique it is not possible to directly observe domains as with MOKE, but only domain walls. It is often possible to indirectly determine the domains, but not the magnitude of the magnetization or its direction. Then, the decorated domain walls can be observed with a direct or inverse optical microscope. New advances on digital imaging techniques turn this traditional technique into a more practical and powerful tool. A weak field ($\sim$50 mT) may be perpendicularly applied in order to enhance the image contrast, but it should be done carefully so that it will not change the sample magnetostructure. In fact, the applied fields (with magnets or electromagnets) should be used to clearly distinguish magnetic domain patterns from the topography of the sample.

Since with the Bitter technique it is only necessary to wait for the colloid to stabilize for few minutes, the acquisition time is rather fast compared with MFM, which can take half an hour per image. However, it is not as fast to observe dynamic domain behaviour as with MOKE. The resolution depends on the quality of the colloid and the resolution of the microscope (typically 500 nm), and it is better than MOKE (1000 nm) but not as good as MFM (100 nm) [7]. A disadvantage is that it is a destructive technique: after the domain observation, a dirty layer will remain on the sample, which is difficult to remove. This might be a problem for some samples as for instance, thin layers fabricated by sputtering. Still, it remains the simplest and the cheapest technique for domain imaging.

3. Experimental

A FINEMET ($Fe_{73.5}Si_{13.5}B_9Nb_3Cu_1$) master alloy was prepared in an induction furnace. Ribbons 10 mm wide and 20 μm thick were obtained from these ingots by planar flow casting technique in air. The chemical composition was checked by inductively coupled plasma spectroscopy.

The samples were studied in their as-quenched form, as well as in an annealed state at 540 and 650 °C (1 h). The annealings were performed in vacuum in an electric resistance furnace.

X-ray diffraction (XRD) was performed using a HZG 4 with graphite monochromator Cu Kα_1 radiation.

A special device for direct measurement of magnetostriction (λ_s), designed and constructed at the Institute of Physics SAS [8], was used.

Coercivity values were obtained using a quasistatic fluxmetric method by applying a longitudinal magnetic field to the sample.

Domain observations with the Bitter technique were carried out with the set-up shown in Fig. 1 with an inverted microscope in the absence of applied magnetic field. A cover glass was used for two purposes: to spread the very small ferrofluid drop on the sample surface and to avoid the rapid drying of the colloid. The samples did not require any surface preparation.

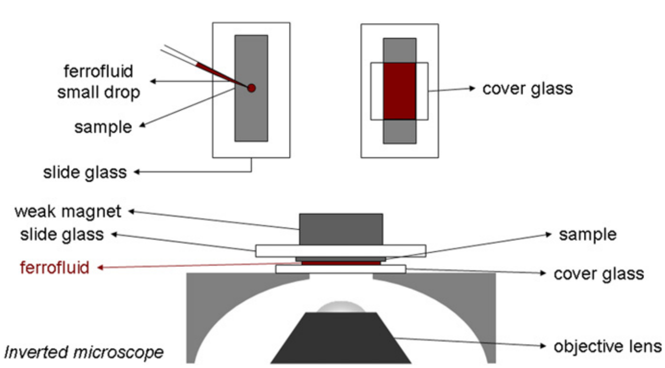

Fig. 1. Experimental set-up used for the Bitter technique.

4. Results and discussion

4.1. As-quenched ribbons

The amorphous structure of the rapidly quenched ribbons was checked on both sides by XRD (Fig. 2). Because of the random atomic arrangement, local magnetocrystalline anisotropies average out over large atomic distances, as explained by the random anisotropy model (RAM) [9]. Nevertheless, there is still an exchange coupling between the local moments that causes a long-range magnetic order. Thus, the amorphous material has good soft magnetic properties resulting in a very low coercivity value: $H_C = (3 \pm 2)$ A/m. Despite the lack of crystalline anisotropy of these materials, well defined domain patterns with locally fluctuating easy axes are observed as a result of residual anisotropies produced by internal stresses. The fabrication process is responsible for these stresses since there are differences in the quenching speed between the air and the wheel side, and at the same time, air bubbles are trapped between the ribbon and the wheel during the casting. The magnetization is coupled to the stress by the magnetostriction constant (λ_s). A value of λ_s equal to $(19 \pm 3) \times 10^{-6}$ was measured. The disordered magnetic microstructure of the as-quenched FINEMET is shown in Fig. 3 with the corresponding scale and ribbon axis (which are rather important for domain pattern analysis but not always shown [10]).

Two different kinds of patterns were observed: (i) wide curved in-plane domains with 180° walls, i.e. Bloch walls, product of dominating tensile stress for the positive magnetostriction and (ii) narrow fingerprint domains. These fingerprints are also called "stress patterns" (even though both (i) and (ii) are due to stresses in the material), "stripe" or "labyrinth". They are closure domains of underlying perpendicular domains, caused by planar compressive stress that induces an easy axis perpendicular to the surface. At increasing perpendicular anisotropy, i.e. stress level, the energy of the hard-axis closure domains rises and the "branching" mechanism appears in several "generations" (zoom in Fig. 3(b)). This occurs by a continuous modulation of the magnetization in a three-dimensional way [11]. Zig-zag walls are usually

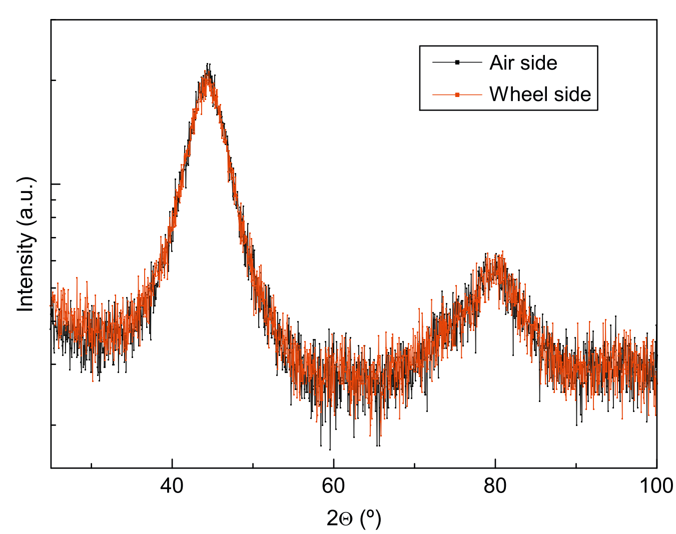

Fig. 2. X-ray diffraction pattern of as-quenched ribbon corresponding to the air and the wheel side.

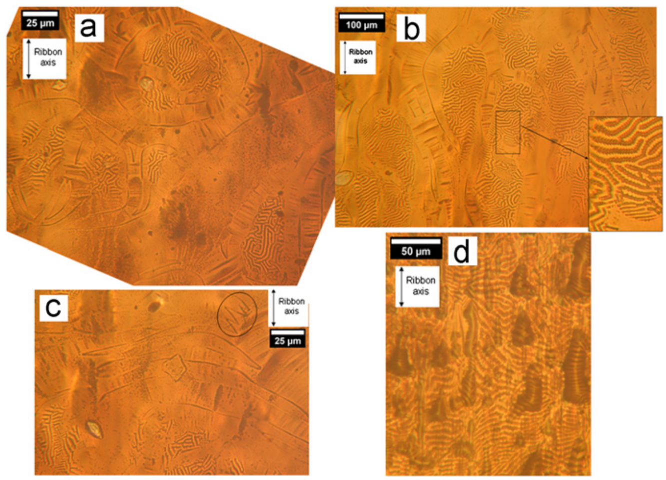

Fig. 3. (a)–(c) Bitter patterns of the bright side (air side) of as-quenched FINEMET showing wide in-plane domains and "stress patterns". Zoom in (b) branching mechanism in which anisotropy increases upwards. Circle in (c) shows typical zig-zag walls. (d) Bitter patterns of the rough side (wheel side) of as-quenched FINEMET with only "stress patterns" orientated nearly perpendicular to the longitudinal direction.

separating antiparallel domains, since this configuration minimizes the magnetostatic energy (Fig. 3(c)).

Island-like labyrinth domain patterns were seen surrounded by wide in-plane domains. Looking at the topography of the sample, it could be observed that the "stress patterns" were generally occupying "valley" regions, in agreement with the idea of compression stresses. Those stresses would be compensated by the wide in-plane domains next to them. However, it is not sure that these wide domains have a completely in-plane magnetization, since they present some diffuse lines oriented in radial direction towards the islands. Such ferrofluid accumulation may probably be indicating a certain perpendicular magnetization component.

The already shown images (Fig. 3) correspond to the bright side of the ribbon (air side), which is more representative of the effective anisotropy than the rough side (wheel side), which shows a more superficial anisotropy. The latter is more difficult to observe because of its topography and that is why some researchers were not even able to observe it. The image of the rough side of the as-quenched ribbon (Fig. 3(d)) showed a more uniform pattern: only "stress patterns" of practically uniform width were seen. Most of the stripe boundaries were nearly perpendicular to the longitudinal direction of the sample, i.e. direction of the remnant internal stresses. It has been estimated [12] that the stress level of the rough side is about four times greater than that of the bright side. The greater and more uniform stress in the rough side is due to its contact with the metallic wheel during the casting, causing a more rapid and uniform freezing.

4.2. Annealed ribbons at 540 °C

The controlled annealing (540 °C, 1 h) induced the precipitation of nanometric Fe–Si grains (~12 nm) (Fig. 4). As a result, a nanocrystalline structure of ferromagnetic nanocrystals embedded in a ferromagnetic amorphous matrix with magnetic correlation length larger than mean grain size is obtained. The random anisotropy model extended by Herzer [13] explained that when grain dimensions are kept below the ferromagnetic exchange length, the anisotropic of the homogenously distributed nanocrystals continues to average out [14]. In addition, this

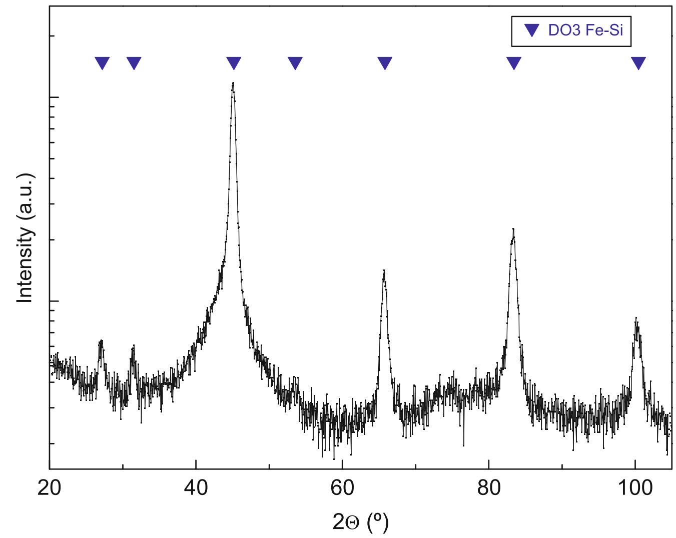

Fig. 4. X-ray diffraction pattern of sample annealed at 540 °C, 1 h.

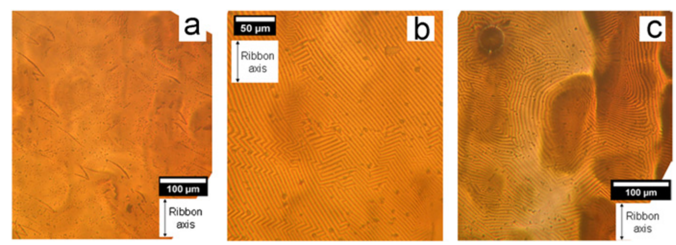

Fig. 5. Bitter patterns of the bright side (air side) of sample annealed at 540 °C, 1 h showing (a) in-plane domains, (b) regular stripe domains and (c) ordered stripe domains following the topography of the ribbon.

composite structure leads to the compensation of λ_s between the amorphous (positive contribution) and the crystalline (negative contribution) phases, decreasing the magnetoelastic energy. In fact, the measured λ_s was $(2.4 \pm 0.8) \times 10^{-6}$. Thus, the material is magnetically even softer than in the as-quenched state, as revealed by the H_C value: (0.6 ± 0.5) A/m.

In the heat treated sample, the complex residual stress distribution of the as-quenched ribbons apparently relaxed to a much more ordered and balanced stressed state with slight variations in orientation (Fig. 5). Not only ordered in-plane domains (Fig. 5(a)), but even "stress patterns" were occasionally detected (Fig. 5(b and c)) since λ_s was not exactly zero. In some regions of the sample, the pattern appeared twisted because of either damages of the ribbon or different topographies (Fig. 5(c)).The heat treatment apparently failed to achieve complete stress relaxation and a prevailing out-of-plane anisotropy can be inferred from the obtained domain patterns.

Our observations are in agreement with Tejedor and Hernando [12], who concluded that the domain structure of the as-cast sample is quite different from that displayed by the nanocrystalline ribbons. On the contrary, Hubert stated that since the crystallites of this material are so small, the samples do not differ from regular metallic glasses, and therefore, the magnetic microstructure of the nanocrystalline sample cannot be well distinguished at optical resolution from that of the amorphous one [11].

4.3. Annealed ribbons at 650 °C

In samples annealed at 650 °C (1 h), the precipitation of boride compounds and Nb_5Si_3 (Fig. 6) [15] caused the loss of the

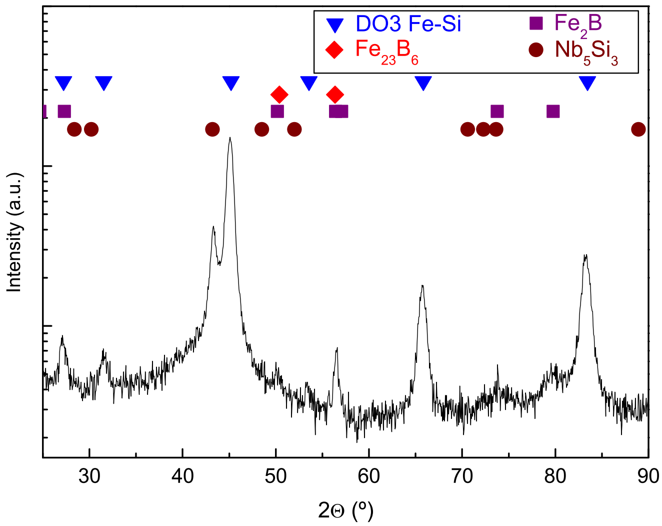

Fig. 6. X-ray diffraction pattern of sample annealed at 650 °C, 1 h.

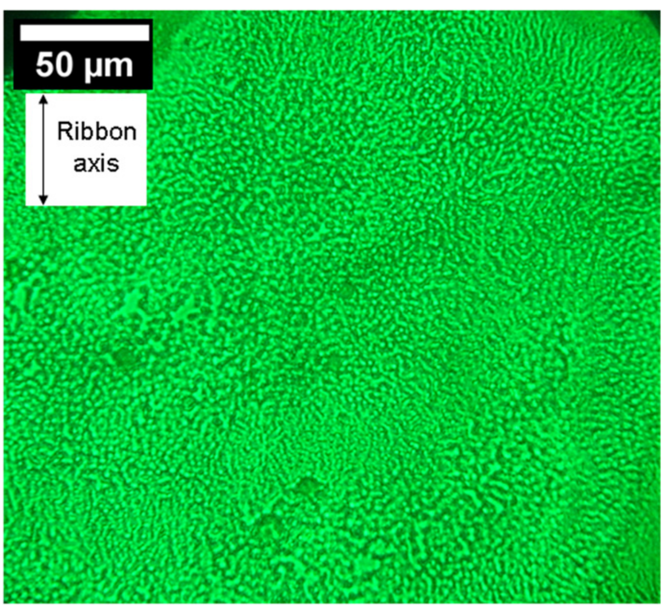

Fig. 7. Bitter patterns of the bright side (air side) of sample annealed at 650 °C, 1 h. A green filter was placed in the light path to improve the image contrast. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

material's soft magnetic properties and therefore its' interest from the technological point of view. Coercivity was found to be (120±80) A/m, and was determined more by the nucleation of many domains than by the motion of the walls. On the contrary, λ_s remained with a low value of $(3.9 \pm 0.9) \times 10^{-6}$, since iron-borides have positive magnetostriction [16], which compensates the negative magnetostriction of the Fe–Si grains.

The changes in the structure modified the magnetic microstructure resulting in more complex domain patterns (Fig. 7). The magnetization process in the samples annealed at 650 °C (1 h) was characterized by a cellular network configuration, i.e. a large number of small and immobile irregular domains. The cellular domain pattern is attributed to a balanced biaxial compression, which in turn leaded to a perpendicular anisotropy. Such stressed state was induced by the crystallization rather than being a residue from the casting process, as it was observed to relax at lower temperatures. The precipitation of borides induced compressive stresses to the ribbon, leading to such anisotropy distribution and the observed domain patterns. Wall contrast is barely visible in the samples, which means that the domain walls are very narrow because of either a high effective anisotropy or a reduced exchange interaction between the crystallites.

5. Conclusions

The evolution of the domain microstructure in FINEMET ribbons subjected to different heat treatments was studied by means of the traditional but still powerful, simple and cheap Bitter technique together with magnetostriction and coercivity measurements. A heterogeneous magnetization – including out-of-the-plane direction – was found on as-quenched samples due to stresses induced during the casting. When annealing at 540 °C most of the stresses were relieved, the magnetization turned towards the plane of the ribbon, the domain structure was ordered and the magnetostriction decreased as well as the coercivity. After annealing at 650 °C during 1 h, the material became magnetically hard, as the great increase of the coercivity revealed. This could also be observed in the domain patterns, which looked disordered in a cellular network configuration and moved only when a strong magnetic field was applied.

Acknowledgements

We thank Galina Kurlyandskaya and Juan Manuel Conde Garrido for the fruitful discussion of the results.

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