HTML ABSTRACT + LINKS

Influence of the dipolar interactions in the magnetization reversal asymmetry of hard–soft magnetic ribbons

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Partial crystallization of the metallic glass $Co_{66}Si_{16}B_{12}Fe_4Mo_2$ was performed by annealing at temperatures between 500 and 540 °C for 10–20 min, resulting in crystallite volume fractions of $(0.7–5)\times10^{-3}$ and sizes of 50–100 nm. This two-phase alloy presents a remarkable feature: a hysteresis loop shift that can be tailored by simply premagnetizing the sample in the adequate magnetic field. Shifts as large as five times the coercive field have been obtained which make them interesting for application as magnetic cores in dc pulse transformers. The asymetrical magnetic reversal is explained in terms of the magnetic dipolar field interaction and the observed hysteresis loops have been satisfactorily simulated by a modification of Stoner-Wohlfarth's model of coherent rotations. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1831552]

I. INTRODUCTION

It is well known that core/shell structures with a ferromagnetic core surrounded by a surface layer of an antiferromagnetic oxide exhibit exchange anisotropy when the samples are submitted to field cooling.[1,2] As a consequence of this exchange bias anisotropy, the hysteresis loop is displaced along its field axis and the coercive field is increased. Moreover, the phenomenon of the loop displacement has been observed in systems that have not been submitted to the mentioned thermomagnetic treatment but to a conventional annealing without field cooling.[3,4] The most outstanding difference of this loop shift in contrast with the bias one is that it can be tailored by simply premagnetizing the sample in the adequate magnetic field and without any further annealing treatment. In fact, the shift can be canceled by the convenient demagnetizing process of the sample in a decreasing ac field. Although the origin of this phenomenon is now commonly attributed to the precipitation of magnetically hard particles in a softer matrix, its detailed mechanism is still an interesting subject for further research.[5–7]

Apart from the fundamental interest of this research, its technological aspect is also of great importance: devices like dc pulse transformers in which the linear region of the hysteresis loop of the soft magnetic core (between zero field and saturation) can be enhanced by a loop displacement H_{shift}, as long as H_{shift} is greater than the coercive field H_c.

In this article shifted hysteresis loops corresponding to annealed samples of the soft magnetic alloy $Co_{66}Si_{16}B_{12}Fe_4Mo_2$ are studied. Our efforts have been directed to obtaining large ratios H_{shift}/H_c as well as to clarify the influence of the dipolar interactions on the magnetization reversal asymmetry.

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II. SAMPLES AND EXPERIMENTAL TECHNIQUES

For the experiment the alloy $Co_{66}Si_{16}B_{12}Fe_4Mo_2$ was chosen as amorphous precursor (ribbons provided by Vacuum Schmelze and Goodfellow, 25 and 40 μm thick and 20 mm wide) because of its nearly zero magnetostriction and nearly zero magnetic anisotropy after an adequate mechanical polishing.[8] Previous works have reported about the effects of thermal treatments on this alloy in the range of 250–480 °C and times between 10 and 90 min.[9,10] These treatments were directed to obtain the nanocrystallization of the samples, which results in an enhancement of their soft magnetic properties. In the present work the samples were annealed at higher temperatures, in the range of 500–540 °C, still below the crystallization temperature ($T_{crys}=558$ °C) and during short times of 10–20 min. Differential scanning calorimetry studies revealed that these treatments give rise to partially crystallized samples.[11] The samples were annealed in a quartz-tube preheated furnace in argon atmosphere. The temperature was continuously monitored by a thermocouple.

Previous to the annealing, some samples were polished with a 0.3 μm alumina suspension in order to reduce the surface roughness, as the magnetic anisotropy in this alloy has been proved to come mainly from its shape anisotropy.[8] Certain samples were cut disk shaped[12] with diameters of 15 mm which allows comparison of the hysteresis loop measurements along different in-plane directions of the disk without the effect of different demagnetizing factors. The magnetic hysteresis loops were obtained by means of a computer controlled inductive measurement system.

X-ray diffraction (XRD) spectra were obtained with a SIEMENS D-50 working with Bragg–Brentano configuration and using the Cu $K_{\alpha1}$ radiation ($\lambda=0.1541$ nm). Due to the absence of sharp x-ray diffraction peaks, transmission electron microscopy (TEM) observations were preferred to determine the size and shape of the nanometer-sized crystallites. Ion milling was used to prepare the thin foils for these

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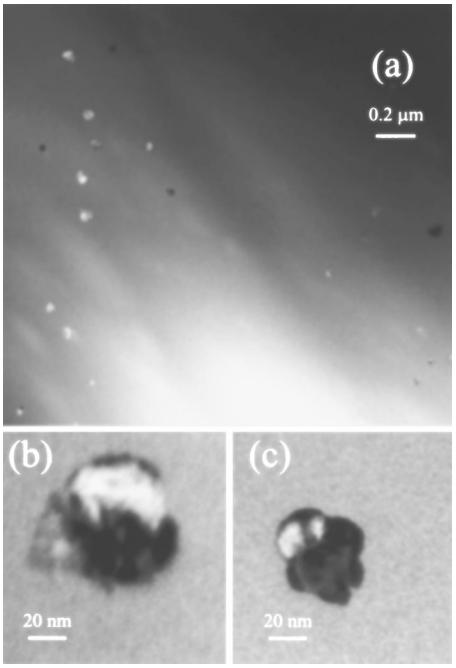

FIG. 1. (a) TEM image of the particles embedded in the amorphous matrix for a sample annealed 15 min at 520 °C, (b) detail of a spherical particle of 70 nm of the same sample and (c) spherical particle of 60 nm of a sample annelaed at 510 °C for 15 min.

studies. Surface density of particles was determined from the visible area of the micrographs. TEM images and selected area electron diffraction (SAED) patterns were obtained with an Hitachi MT800 electron microscope using and acceleration voltage of 200 kV.

III. RESULTS AND DISCUSSION

Figure 1(a) is a TEM picture of a sample that was annealed at 520 °C for 15 min. It shows some particles embedded in the amorphous matrix. As the white spots have all the same level of electron transmission, we think they can be attributed to holes left by particles that were removed from the matrix in the thinning process. Figure 1(b) is a TEM image of a spherical particle with 70 nm diameter of the same sample and Fig. 1(c) shows a particle of 60 nm from a sample that was annealed at 510 °C for 15 min. From these TEM observations it was determined that annealing at temperatures in the range of 500–540 °C with duration of 10–20 min resulted in volume densities of $(6–20)\times 10^9$ mm^{-3} of precipitated spherical grains with average sizes of 50–100 nm. No clear relation could be established between density and size of the particles and annealing temperature or time probably because, at the initial stages of the crystallization, the formation of clusters depends more on the number and distribution of the internal defects or impurities of the amorphous precursor that act as favorable nucleation sites.

A typical SAED pattern observed for these samples is shown in Fig. 2. The diffraction pattern shows at first glance the appearance of the diffuse rings characteristic of the amorphous state, consisting of three rings whose intensities decrease with increasing diameter (0.21 ± 0.01, 0.12 ± 0.01, and 0.08 ± 0.01 nm). This amorphous appearance can be explained by the low volume fraction of the grains that is about 0.1%, as deduced from the TEM micrographs like those in Fig. 1(a). A more detailed observation of the difractograms revealed reflections (not appreciable in Fig. 2) that could be indexed with the help of the free software ProcessDiffraction[13] as lines corresponding to fcc Co, hcp Co, Co_2B and Co_3B that would give rise to XRD peaks in the range 40–46 and 70–90 °C for 2θ, which would explain the breadth of the observed peaks. These are the most probable crystallites in this alloy, as found in similar compositions by other authors.[14,15]

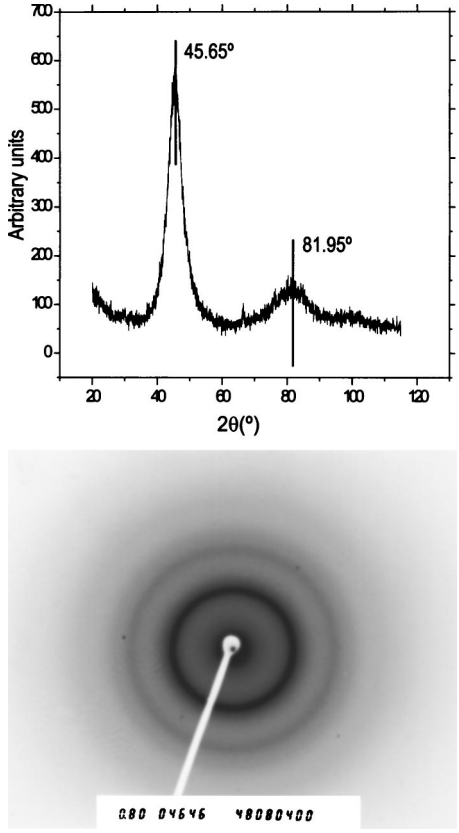

FIG. 2. X-ray and selected area diffraction patterns of the sample annealed 15 min at 520 °C.

Figure 3 shows a typical displaced hysteresis loop found

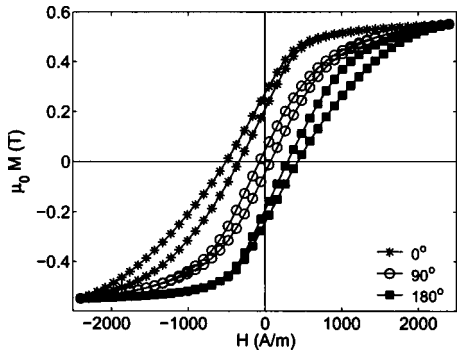

FIG. 3. Hysteresis loops of a disk-shaped sample annealed 20 min at 510 °C that was submitted to a field of 400 kA/m previous to the measurements. Asterisks, circles, and squares correspond, respectively, to measurements in which the initial applied field was parallel, perpendicular, and antiparallel to the premagnetizing field.

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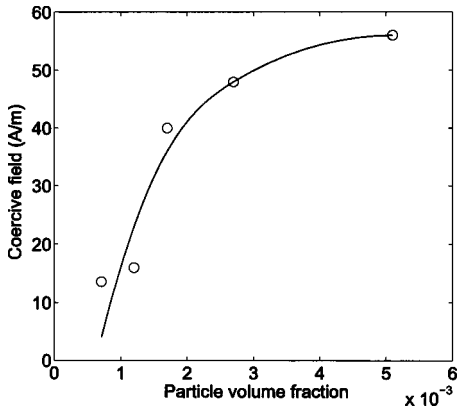

FIG. 4. Coercive field H_c vs particle volume fraction.

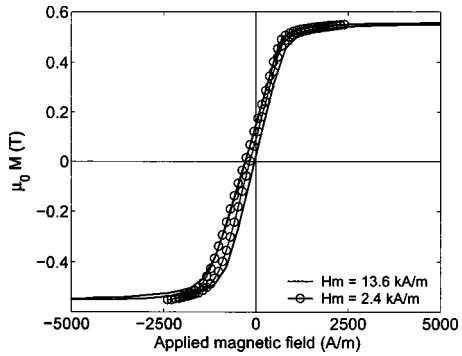

FIG. 5. Hysteresis loops of a sample annealed 20 min at 520 °C and premagnetized at 120 kA/m traced with maximum fields of 2.4 and 13.6 kA/m. In both loops $H_1=-300$ A/m while H_2 increased from -160 to -30 A/m with increasing maximum field.

in these samples. It corresponds to a 40 μm thick disk-shaped sample that was annealed at 510 °C during 20 min. The sample was premagnetized with a field of 400 kA/m before measuring. The hysteresis loops were obtained in three different orientations corresponding to the starting applied field forming 0°, 90°, and 180° with the premagnetizing field direction. The unidirectional character of the magnetization reversal asymmetry can be clearly seen, as it is opposite when the sample is rotated 180° while at 90° we obtained a loop that is symmetrical about zero field. Apart from the evident displacement along the field axis, different slopes of the left and right branches of the loop can be appreciated.

Figure 4 shows the dependence of H_c on the particle volume fraction f. This relationship suggests an influence of the local dipolar field in the microdomain structure of the neighboring amorphous material. For these H_c measurements the hysteresis loops were traced at a maximum field of 2400 A/m.

The asymmetrical magnetization reversal could be explained in terms of the local unidirectional stray field created in the soft amorphous matrix by the much harder particles. The magnetic moments of the particles have been aligned by the premagnetizing field H_p. Assuming spherical geometry for the hard particles, their mean dipolar field can be approximated by $H_d=1/3fM_{s2}$, M_{s2} standing for the saturation magnetization of the particles.[16] The internal field can be then written as $H_{\text{int}}=H-NM-H_d$, H being the external field and N and M the demagnetizing shape factor of the sample and the magnetization of the amorphous material in the direction of measurement.

Due to the big magnetic anisotropy of the particles, their magnetic moments remain unreversed when the hysteresis loop is traced with maximum fields of about $H_m=14$ kA/m, sufficient to reach saturation of the soft regions of the sample. The so-obtained hysteresis loops seem to be saturated as the volume fraction of the particles is so low that their contribution to the total magnetization of the sample is insignificant, but they are in fact minor loops.

As H_d is unchanged while H varies to trace the loop, all the loop is shifted by the quantity H_d and so the magnetization is zero at fields $H_1=-H_c-H_d$ and $H_2=H_c-H_d$, H_c being the coercivity of the amorphous matrix. In this case the measured displacement is $H_{\text{shift}}=H_d$ and the coercive field may be obtained as $(-H_1+H_2)/2=H_c$.

When the maximum field H_m is increased so that the magnetic moments of the particles can be reversed, the dipolar field is still H_d in the descending branch of the loop and so H_1 remains the same, but in the ascending branch the dipolar field is now $-H_d$ and so the curve cuts the abscissa axis at $H_2=H_c+H_d$. This results in a null shift of the saturated loop and a maximum coercive field given by $(H_1+H_2)/2=H_c+H_d$.

Our measurements of hysteresis loops confirmed this effect of H_m, as shown in Fig. 5 where we can see the loops of a sample annealed at 520 °C during 20 min and premagnetized at 120 kA/m traced with maximum fields of 2.4 and 13.6 kA/m. It can be appreciated that H_1 remains unchanged at a value of -300 A/m while H_2 increases from -160 to 30 A/m with increasing H_m.

The evolution of the coercive field H_c and the loop displacement H_{shift} with the maximum applied field H_m that was used to record the loops is shown in Fig. 6, where it can be seen that the change rate of H_c and H_{shift} are equal but opposite in sign for both magnitudes. These curves are typical for all the samples.

The effect of premagnetizing at different fields was an increment of the loop displacement, while H_c remained approximately constant. As shown in Fig. 7 this increment

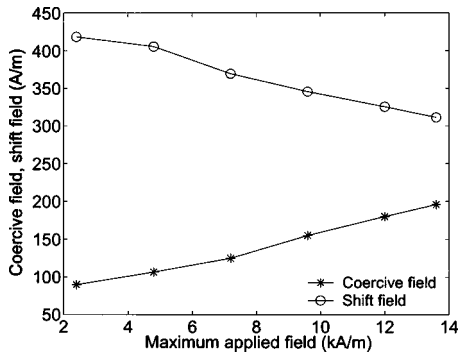

FIG. 6. Effect of the maximum field H_m applied to trace the hysteresis loop on the magnitude of the shift and the coercive fields for a sample that was annealed at 520 °C for 20 min and premagnetized in a field of 400 kA/m.

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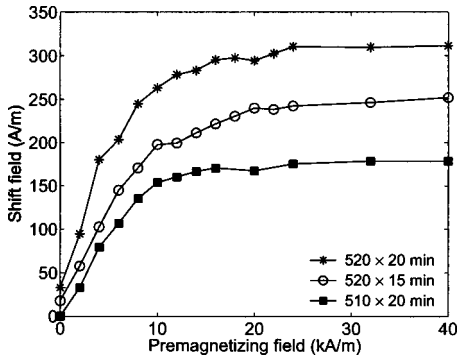

FIG. 7. Dependence of the shift field H_{shift} on the premagnetizing field for the sample annealed at 520 °C for 20 min, measured from loops traced with a maximum field of 2.4 kA/m.

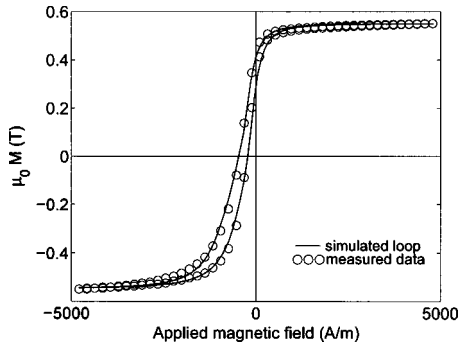

FIG. 9. Representative hysteresis loop of an annealed sample of $Co_{66}Si_{16}B_{12}Fe_4Mo_2$. The experimental data have been shown as circles and the solid line is the simulated loop.

ceased at about $H_p = 240$ kA/m in all the samples, the phenomenon reaching a saturation. This saturation value can be explained assuming that the particles are spherical and its own hysteresis loop will be saturated at a field that can be approximated by the addition of its uniaxial magnetic anisotropy field $K_1/2\mu_0 M_{s2}$ and its shape demagnetizing saturation field $(1/3)M_{s2}$. Assuming $K_1 = 10^4$ J/m^{-3} and $M_{s2} = 1$ T, as typical for Co compounds, we get $H_{\text{sat}} \approx 300$ kA/m.

As already mentioned, if the whole soft matrix were affected by the dipolar field, the loop would be shifted by $H_{\text{shift}} = (1/3) f M_{s2}$. Considering Co particles with $\mu_0 M_{s2} = 1$ T and the characteristic fractions of $(0.7–5) \times 10^{-3}$ found in these samples, we obtain $H_{\text{shift}} = 180–1400$ A/m, the same order of magnitude but systematically larger than those measured ($H_{\text{shift}} = 25–480$ A/m with $H_m = 2400$ A/m and $H_p = 400$ kA/m). This difference can be attributed to two reasons: (a) the easy directions of magnetization of the particles have a random distribution and so, even in the case of maximum premagnetization, all the magnetic moments may point in the same direction but are not perfectly alligned and (b) the dipolar field effect decreases rapidly with the distance to the particle and so not all the regions of the amorphous matrix are equally affected. Both reasons would give rise to inhomogeneity in the spatial distribution of the dipolar fields. In Fig. 8 a reasonable dependence of H_{shift} on the particle volume fraction can be observed.

In order to simulate the effect of the dipolar field in the

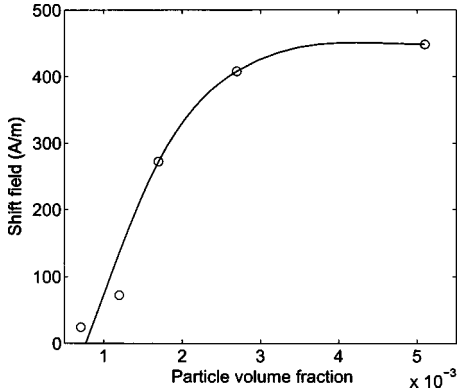

FIG. 8. Evolution of the shift field H_{shift} with the particle volume fraction for the annealed samples. The corresponding hysteresis loops were traced with a maximum field of 2.4 kA/m.

hysteresis loop, we have modified Nowak's model[17] of magnetic hysteresis. This author's work is itself a modification of Stoner-Wohlfarth's model of coherent rotations in which he includes an additional constant internal field that has two components parallel (L) and perpendicular (T) to the external field. The source of this additional field is connected to domain and ripple structure. While, following Nowak, L and T are considered positive when the direction of magnetization rotates clockwise and negative for counterclockwise rotations, H_d, representing the particle dipolar field contribution, remains unreversed. In fact, in the region of the loop near the applied field $H = -H_m$, the dipolar field may not be reversed but rotated a certain angle $\theta < 90°$, producing a transverse component $H_{d\perp} = H_d \sin\theta$.

In the case of magnetically isotropic samples, the condition of minimum total energy per unit volume can be analytically solved as

$$M = \pm M_s \cos \operatorname{atan} \frac{H_\perp}{H + H_\parallel}, \tag{1}$$

where $H_\parallel = H_d + L$ for the descending branch and $H_\parallel = H_d - L$ for the ascending one. Sign "−" and $H\perp = T + H_{d\perp}$ stand for $H < -H_\parallel$. This results in a hysteresis loop with coercive field $H_c = L$ and shifted by H_d. The loop is also more rounded for negative than for positive fields due to the term $H_{d\perp}$. We have then calculated the magnetization corresponding to each value of the applied field H as the media of the magnetizations M_i of N regions with volume fractions f_i affected by different values of the dipolar field H_d: $M = \Sigma_{i=1}^{N} f_i M_i$.

An example of the as-obtained loops is shown in Fig. 9. We tried here to simulate the hysteresis loop of a sample of $Co_{66}Si_{16}B_{12}Fe_4Mo_2$ that was annealed at 520 °C during 15 min. The sample is a ribbon 8 cm long that was mechanically polished before the annealing. The loop displacement calculated as $1/3 f M_{s2}$ would give, for this sample, 800 A/m and the measured one is 370 A/m, probably due to the fact that the entire bulk is not affected by the same dipolar field. For the simulation, four regions with volume fractions $f = 0.7$, 0.1, 0.1, 0.1, $L = 55$, 200, 340, 480 A/m and $H_d = 240$, 430, 610, 800 A/m, respectively, were considered. Values of $T = 320$ A/m and $H_{d\perp} = 240$ A/m were used. The resulting calculated loop has $H_c = 100$ A/m and $H_{\text{shift}} = 360$ A/m. In Fig.

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9 the theoretical magnetization corresponds to the solid line and fits the measured hysteresis loop, shown here as circles, very well.

IV. CONCLUSIONS

We have observed hysteresis loop shifts as large as five times the coercive field in samples of $Co_{66}Si_{16}B_{12}Fe_4Mo_2$ that were annealed for 10–20 min at temperatures slightly below the crystallization temperature. The loop displacement and the enhancement of coercivity produced by the annealing are related to the density and size of the precipitated crystallites and to the magnitude of the premagnetizing field and the maximum field used to trace the loop, in such a way that suggests that the dipolar field created by the hard particles is responsible for this phenomenon. Based on this hypothesis, we have developed a model based on Stoner-Wohlfarth's model of coherent rotations that satisfactorily simulates the shape of these loops.

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