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Induced ferro-ferromagnetic exchange bias in nanocrystalline systems

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ABSTRACT

An unusual magnetic hysteresis consisting of horizontally shifted and distorted loops appears in some Co-based nanocrystalline systems in which soft and hard ferromagnetic phases coexist. The bias field can be tuned at room temperature by premagnetising treatments. Several works attributed the origin of this effect to the dipolar interaction, while little attention has been paid to the exchange interaction contribution due to its short-range nature. In this paper the relative importance of the dipolar and exchange interactions is investigated by means of micromagnetic simulations. It is demonstrated that the exchange coupling, though a nearest-neighbour interaction, has far-reaching repercussions in the magnetic configuration, and substantially prevails over the magnetostatic interaction as the cause of the asymmetrical magnetisation reversal. The straightforward conclusion is that we are dealing with a ferro-ferromagnetic exchange bias effect.

1. Introduction

The magnetic properties of nanocrystalline materials are mainly dictated by their grain size and intergrain distance. Densely compacted agglomerations of nanoparticles with sizes smaller than the exchange correlation length produce extremely soft magnetic behaviour [1,2], while larger sizes (typically above 20 nm) give rise to magnetic hardening with respect to the amorphous counterpart [3]. An intermediate kind of nanocrystalline systems may be achieved from an amorphous precursor by an adequate devitrification resulting in a low concentration of non-agglomerated nanoparticles embedded in the residual amorphous matrix. Depending on the nature of the crystalline phase and the magnetic character of the matrix, the material can be a combination of ferromagnetic hard and soft phases presenting biased hysteresis loops (HL) [4–6].

Biased HL are typical of systems in which ferro/antiferromagnetic interfaces are exchange coupled [7–10] and which are either field cooled or submitted to large pulsed fields [11]. Other studies can be found in the literature in which the exchange bias (EB) is produced by coupling of ferri/antiferro, ferri/ferri and antiferro/antiferro interfaces (see [12–14] and references there in).

Nonetheless, antiferro or ferrimagnetic phases are absent in the material studied in this work, in spite of which a clear biasing is observed without need of field cooling. This anomalous hysteresis has often been attributed to the dipolar interaction between the magnetically hard crystallites precipitated during the annealing and the still soft residual matrix [4,5,15], neglecting the exchange coupling between the two phases. Other hard/soft ferro/ferro multiphase composite systems with biased HL have been reported in which the effect has been attributed to magnetostatic interactions [16–20].

Contrariwise, this paper concerns the hard/soft ferro/ferro exchange interaction and demonstrates that, except for samples with very specific geometries, it is the main responsible for the shift of the HL and other features of the magnetisation reversal in this kind of nanocrystallised material. A simple model is presented in which the dipolar and exchange interactions between both hard and soft phases are taken into account giving rise to twisted magnetisation configurations as in exchange-spring multilayers [21–23], the large differences in the magnetisation behaviour coming from the extensiveness of the soft phase.

From the experimental point of view, the work focuses on partially devitrified samples of $Co_{66}Fe_4Mo_2Si_{16}B_{12}$ as representative of a group of Co-based nanocrystallised materials presenting biased HL with a common phenomenology [6,24,25]. HL shifts as large as five times the coercive field have been previously reported by the authors [15,26] in this system which, together with the persistence of the effect [27], points in the right direction for technical application in devices in which the hysteresis loops of their soft magnetic cores must be displaced, like switching power supplies, pulse transformers, magnetic sensors or giant magnetoimpedance-based sensors. But besides its potential technological relevance, this material constitutes a very interesting system to go further in the understanding of the influence of internal magnetic interactions on the hysteretic features.

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2. Experimental procedure and results

The experiments have been carried out on partially devitrified samples obtained by isothermal annealing of amorphous ribbons of $Co_{66}Fe_4Mo_2Si_{16}B_{12}$. In previous works [15,26] it was already reported that annealing this alloy at temperatures slightly below the crystallisation temperature ($T_{cr} = 558$ °C) give rise to samples with very low volume fractions (between 2×10^{-4} and 5×10^{-3}) of nanocrystals. X-Ray Diffraction and Selected Area Electron Diffraction results revealed that Co_3B and hcp Co, both ferromagnetic, are the most probable crystalline phases. Transmission Electron Microscopy (TEM) micrographs allowed the observation of particles with sizes between 15 and 80 nm. Fig. 1 shows two TEM images taken from a sample annealed at 530 °C for 12 min in which the typical morphology of this kind of hard-soft system can be appreciated: some agglomerates of only 1–6 nanoparticles completely surrounded by amorphous material.

The magnetic hysteresis curves were obtained at room temperature in an inductive computer-controlled hysteresismeter working at 3 mHz [28]. The loops of the as-quenched samples are typical of this soft alloy with very small coercive field ($H_c = 2.4$ A/m) and a saturation field that is mainly dependent on the shape demagnetising field. These are conventional odd-symmetrical M–H loops, in the sense that $M(H)$ in the descending branch is equal to $-M(-H)$ in the ascending one. After annealing, the hysteretic behaviour changes dramatically in several aspects: the coercive field increases remarkably and the loop becomes asymmetrical, appearing horizontally shifted and distorted. Moreover, the loop displacement and asymmetry can be modified at room temperature by subjecting the sample to a relatively strong premagnetising field H_p (20–200 kA/m) before measuring. It was previously stated that (i) the shift and distortion of the resulting loop are strongly dependent on the component of H_p along the measurement direction, (ii) the effect is saturated at $H_p \simeq 200$ kA/m, and (iii) opposite premagnetising fields give rise to opposite displacements and distortions. The dependence of the shift with the premagnetising field value is shown in Fig. 2 characterised by the bias field H_b defined as the applied field value at which the central point of the loop is placed.

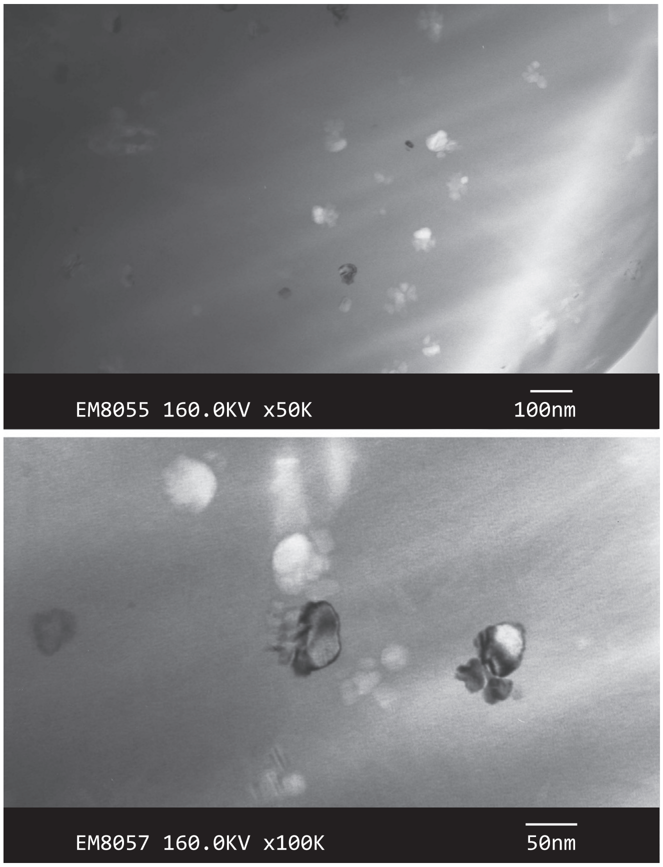

Fig. 1. TEM micrographs of the particles embedded in the amorphous residual matrix taken on a sample which was devitrified at 530 °C for 12 min.

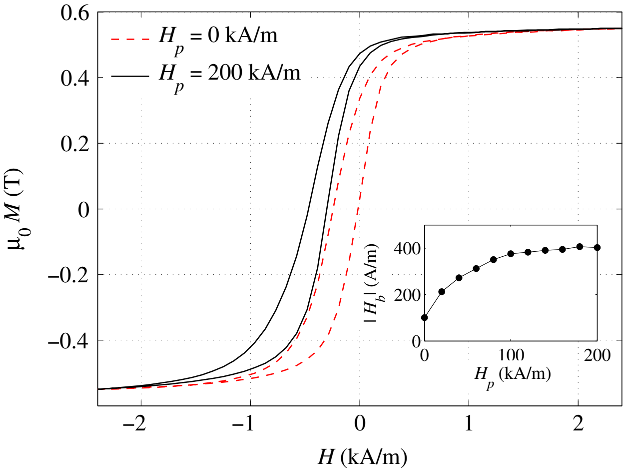

Fig. 2. Loops of a sample annealed at 530 °C for 12 min and measured before and after applying a premagnetising field H_p=200 kA/m. Inset: Bias field as a function of the premagnetizating field.

3. Model and discussion

Based on the fact that the magnetic system consists of a very low concentration of ferromagnetic hard crystallites embedded in a ferromagnetic soft amorphous matrix, the asymmetry of the magnetisation reversal can be explained in terms of the effect that the hard particles produce in their surroundings provided that (i) their magnetisation will remain practically unchanged while the external field sweeps from positive to negative values to trace the loop of the soft phase, and (ii) the ferromagnetic exchange coupling at the grain boundaries is strong enough to pin the magnetic moments of the amorphous matrix. Applying a premagnetising field produces then an orientation of the magnetisations of the crystalline grains whose effect is equivalent to an unidirectional anisotropy. Larger premagnetising fields induce larger biasing (see inset in Fig. 2); the fact that there is a little biasing even with no premagnetisation indicates that there is a certain degree of order in the orientation of the magnetisations of the precipitated grains just after annealing. It should be then remarked that, although they may seem saturated, all the biased HL are in fact minor loops.

The strong exchange interaction between a crystallite and the surrounding material will push the magnetic moments of the latter at the grain boundary to be parallel to the magnetisation of the former. As a consequence, both the magnetic poles due to the crystallite magnetisation M_1 and to the matrix magnetisation M_2 coexist on the grain boundary. In order to evaluate this effect, the particles will be approximated by uniformly magnetised spheres of radius R with an effective magnetisation of $M_1 - M_2$.

As a first approximation to the problem, we will analyse solely the effect of the magnetostatic interaction. With this purpose we have implemented numerically a simple model in which the magnetic moments of the amorphous material align with the magnetic field resulting from the superposition of the externally