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Magneetoelastic modelling in soft nanocrystalline alloys

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Abstract

Magneetoelastic effects in ultra soft nanocrystalline alloys are investigated theoretically and experimentally. From $H_c$ measurements, extraction of magnetoelastic contribution is carried out using a formalism obtained revisiting random anisotropy model (RAM) in the light of domain walls (DW) displacements, our approach based on theoretical investigations on the way of a reversal of a correlated volume (CV) located in the vicinity of a DW. Modelling of magnetoelastic effects shows that even in perfectly relaxed samples, a magneetoelastic contribution exists due to elastic frustration experienced by a CV during its magnetization reversal. Magnitude of this energy is large enough to drive coercivity of samples featuring grain diameter $D$ around 10 nm, which are of major interest for applications.

Keywords: Nanocrystalline alloys; Magnetoelastic effects; Random anisotropy

1. Introduction

Magneetoelastic effects in soft nanocrystalline alloys are of major importance. Experimental data show that it is necessary to consider, apart from classical random anisotropy contribution, a second source of coercivity usually attributed to magnetoelastic effects [1,2]. Until now, the involved mechanisms are unclear and the present work proposes to answer this question.

Magneetoelastic coercivity results due to internal frustration and can be investigated at different scales. At first, one can look to the frustration occurring at the frontier of the nanocrystal and surrounding amorphous, these media have different magnetostriction coefficients $\lambda_a \approx 22 \times 10^{-6}$ (amorphous), $\lambda_c \approx -5 \times 10^{-6}$ (FeSi nanocrystals) [3–5]. At the scale of the so-called correlated volume (CV), characterized by dimensions comparable to the ferromagnetic correlation length $L_0$, this effect is subjected to the same averaging process as the magnetocrystalline anisotropy and leads to a negligible contribution compared with the magnetocrystalline one [6]. This allows to consider the ribbon as isotropic and elastically homogeneous, with magnetostriction coefficient $\lambda_a$ and Young Modulus $E$ leading to a magnetoelastic energy density $W_{me} \approx E\lambda_a^2$. With $\lambda_a \approx$ several $10^{-6}$ and $E \approx 2 \times 10^{11}$ Pa, one obtains $W_{me} \approx$ several J/m$^3$, which, dealing with macroscopic properties, can play a role only if coherent at the scale of the CV.

Looking for a mechanism able to induce such a coherent stress, we consider the reversal of the magnetization in a CV. A mismatch arises from its immersion in a medium differently magnetized, maximum frustration occurring when the magnetizations are perpendicular to each other. The magnetoelastic effects will be those of a coherent uniaxial anisotropy, denoted in the following as $K_{me}$. A quantitative modelling of $K_{me}$ is therefore of great interest and proposed in Section 3.

Experimental data concern ribbons (atomic composition Fe$_{75.5}$Cu$_{17}$Nb$_{2.8}$Si$_{14.3}$B$_{6.2}$, thickness $\approx 21 \mu$m) manufactured by Imphy Alloys. The ribbons are annealed at different temperatures $490 < T < 570$ °C and durations $1 < t < 600$ min.

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Crystalline fraction \( f \) and grain diameter \( D \) are characterized by X-ray diffraction. The large database (30 samples) obtained feature \( 8 < D < 13 \) nm and \( 10 < f < 75\% \). Confrontation with the modelling is based on \( H_c \) measurements. The part due to random anisotropy being not negligible, the extraction of magnetoelastic contribution to \( H_c \) involves dedicated formalism as proposed in Ref. [3]. We propose, in part 2, an alternative formalism considering CV reversal in a macroscopic scheme governed by domain wall (DW) displacement. Extraction of \( K_{me} \) from experiments will be then carried out, allowing confrontation with modelling, marking the end of part 3.

2. CV reversal and DW displacement: Application to the extraction of experimental \( K_{me} \)

In the frame of random anisotropy model (RAM), the basic acting entity is the CV. Since the exchange stiffness \( A \) prevents changes in the direction of magnetization within very short distance, the magnetization mechanism is assumed to be of the uniform rotation. As a result, the anisotropy of an assembly of nanograins, settling \( CV \), is assumed to be of the uniform rotation. As a result, the effective anisotropy governs the reversal field \( H_r \approx 2 K_c/J_s \) (\( J_s \) = saturation polarization), associated to the CV and thus the resulting coercivity [3–5].

The preceding scheme considers the CV as an isolated particle. In fact, from a macroscopic point of view, the magnetization variation is due to DW displacements. In other words, an acting CV behaves in a co-operative way with the neighbours by the way of magnetostatic interactions. Our goal is thus to determine the quantitative influence of these interactions upon \( H_r \) and, at a larger scale, upon \( H_c \).

To do this, we will consider a peculiar CV, denoted \( CV_0 \), and neighbouring CVs, those acting as a global entity, that is, the DW. In this frame, \( CV_0 \) is considered as a pinning centre with \( H_{0o} \) as pinning field. Looking to the evolution of the interaction energy \( E(z_d) \) with \( z_d \), \( z_d \) denoting the relative position \( CV_0/DW \), allows to determine \( H_{0o} \). Finally and according to Ref. [7], \( < < H_0 > > \) obtained by averaging \( H_{0o} \) on the population of CVs coinciding with the DW, will be directly related to \( H_c \) as

\[
H_c = \langle \langle H_r \rangle > + \left( \langle \langle H_r \rangle > - \langle \langle H_r \rangle > \right)^{1/2}
\]

(1)

(Horizontal bars indicate an average process on the different absolute positions of the wall). Usually, \( E(z_d) \) exhibits even parity, leading, with defects uniformly distributed, to \( < < H_0 > > = 0 \) [7]. \( H_c \) is so determined by the second term of Eq. (1). As we shall see, the situation corresponding to our case is different.

The first step is to determine \( E(z_d) \). To do this, we picture the DW by a region of thickness \( L_0 \) (Fig. 1). Magnetizations of CV and surrounding media (DW or adjacent domains (AD)) are different, leading to a magnetostatic energy \( E_m \). For convenience, magnetization of different media are assumed uniform, the DW being transversally magnetized. We obtain the uniform densities of the magnetic charges deposited on faces \( a, c \) (part of \( CV_0 \) surrounded by DW), and \( b, d \) (part of \( CV_0 \) surrounded by AD), and opposite faces. \( E_m \) thus involves self-contributions \( E_{mi} \) corresponding to rectangles \( a_i - c_i - b_i - d_i \) or \( (L_0 - L_a) \times L_0 \) with charge density \( \sigma_i \), and interaction terms \( E_{mij} \) (calculated with centred charges instead of charge densities). \( E_{mi} \) is given as follows [9]:

\[
4\pi\mu_0 E_{mi} = L_0^2 \int F[p], \quad p_i = \frac{L_a}{L_0} \quad \text{or} \quad \frac{(L_0 - L_a)}{L_0},
\]

\[
F[p] = p^2 \ln \left[ \frac{1}{p + \sqrt{1 + p^2}} + p \ln \left( p + \sqrt{1 + p^2} \right) \right] + \left[ 1 + p^2 - (1 + p^2)^{-3/2} \right]/3.
\]

(2)

In the domain of interest (\( 0 < p_i < 1 \)), \( F \) accepts—and will be replaced by—a more easily analytically tractable approximation function \( G \) defined as

\[
G[p] = a p^2 - b p^3; \quad a = 2.58525, \quad b = 1.13266.
\]

(3)

With \( E_k = K \sin^2 \theta \), denoting the CV volumetric density of anisotropic energy, we write the total energy \( E_t = (E_m + E_k)_0 \), using dimensionless parameters \( k = K \pi \mu_0 / J_s^2 \) and \( u = z_d/L_0 \), as

\[
E_t = k \sin^2 \theta + (1 - \cos \theta - \sin \theta) u(1 - u) \left( 1 - 1/\sqrt{5} \right)
\]

\[
+ (1 - \cos \theta) \frac{(a - 1/2)(1 - u)}{2(b(1 - u)^2)}
\]

\[
+ (1 - \sin \theta) \frac{(a - 1/2)u^2 - bu^3}{2}, \quad 0 < u < 1.
\]

(4)
Symmetric formula is obtained for $1 < u < 2$. Minimizing $E_r$ with respect to $\theta$, one obtains equilibrium angle $\theta_e(z_0)$ and equilibrium energy $E_e(z_0) = E_r(\theta_e(z_0))$, plotted on Fig. 2.

The $k = 0$ curve shows that, dealing with magnetostatics $\theta_e(0) = 0$, $\theta_e(L_0) = \pi/2$ and $\theta_e(2L_0) = \pi$, $\theta_e$ is characterized by a continuous evolution. CVs featuring easy magnetization along Oy show the same behaviour with $\theta_e(L_0) = \pi/2$ again. Situation of CVs featuring $k > 0$ differs : with $K$ increasing, $\theta_e(z_0)$ lags more and more towards 0, and $E_e(z_0)$ drawing nearly asymptotic curve $E_r(0)$. If $k$ is large enough, $\theta_e(L_0)$ reaches a value $\theta_{bk} < \pi/2$. On increasing $z_0$ slightly, the magnetization would like to reach direction $\theta_e(L_0) = \pi - \theta_{bk}$, but cannot, because of the barrier featured by the costly direction $\pi/2$. The switching thus occurs latter (cf. point B on curve $k = 0.8$), breaking the symmetry of $E_e(z_0)$. The transition between the two behaviours occurs for a $k_1$ value $\approx 0.5$. The point is that CVs featuring $k > k_1$ give a non zero $\ll H_r \gg$ term in Eq. (1), and from this point of view, settle a relevant population looking to $H_c$.

Surprisingly, $K_1 = k_1J_s^2/\pi\mu_0$—and related quantities—are determined by magnetostatics instead of anisotropy and are of unrealistically high magnitude. This paradox vanishes, noticing that, dealing with the magnetostatic energy, the crude assumption of uniformly magnetized regions leads to a high charges concentration and an overestimated evaluation calling upon $\mu^*$ effect [10], which leads to a reduction factor between 1 and $1 + J_s^2/(2\mu_0K_e)$ depending on the magnetizations of CV and surrounding medium. $K_1$ is now controlled by the anisotropy and is much lower. Nevertheless, the reduction is obtained through the dilution of charges, which, to be effective, involves a reversing cell probably greater than the basic CV. This shows that looking to the basic properties (volume and shape) of CVs, we cannot be unaware of the magnetostatics, justifying future investigations.

At this step, we retain only a general teaching overview of our scheme, i.e., looking to $H_c$, the relevant population of CVs features $K > K_1 > 0$, leading to $K_e = \langle K \rangle$. Numerical value of $K_1$ being strongly dependent on details of reversal scheme, we take it null in the following. With a probability distribution $p(K)$ Gaussian, we thus obtain:

$$K_e = \int_0^\infty Ke^{-K^2/(2\sigma^2)}dK/\int_0^\infty e^{-K^2/(2\sigma^2)}dK = \sqrt{2\pi}/\pi$$

(5)

This result is comparable to the one obtained in the classical frame of RAM, our average value and usual fluctuation term both are controlled by $\sigma$.

Extending our analysis to the case of a superimposed longitudinal anisotropy $K_u > 0$ (which will correspond in our case to $K_{me}$), $p(K)$ is now centred on $K_u$. Increasing $K_u$, population of CVs featuring $K < 0$ progressively vanishes, leading to the asymptotic case $K_e = K_u$. Using dimensionless parameters $k_a [K_u/(f^2K_0^4A^{-3}D^6)^{1/3}$ and $k_e = K_e/(K_u k_a)$, it is finally obtained [9],

$$k_a = \left[ 1 + \sqrt{2/\pi} k_e^{3/2} \exp[-1.06 k_e^{-0.92}] \right] / k_e$$

(6)

where $k_a$ measures respective weight of random anisotropy and $K_u$ in the resulting $K_e$. $K_u$ calculated using Eq. (6) with typical numerical values for nanocrystalline alloys is plotted in Fig. 2. In our case, $K_{me}$ plays the role of $K_u$ and is extracted as follows.

At first, one notices that if $f$, $K_{0b}$, $A$, $D$ are known, couples ($K_u$, $K_{me}$) are $k_a$ dependent; $k_a$ is given, $k_a$ is deduced from Eq. (6), allowing to calculate $K_e/K_{me}$ (definition of $k_a$) and $K_{me}$ (definition of $k_a$). From $K_{me}$ and $k_a/K_{me}$, one obtains $K_e(k_a)$.

$K_e$ is determined from $H_c$ measurements provided by our database, assuming with [4] the law

$$H_c = 2axK_e/J_s; \quad x = 0.065$$

(7)

$K_e$ known, denoted $K_{0b}$, an iterative calculation of $K_e(k_e)$ can be carried out, varying $k_e$. A peculiar value $k_{0b}$ is obtained, satisfying $K_e(k_{0b}) = K_{0b}$. The value of $K_{me}$ corresponding to $K_{0b}$ is so $K_{me}(k_{0b})$.

Fig. 2. Evolution of equilibrium energy of CV with respect to DW position for the situation described on Fig. 1.

Fig. 3. Effective anisotropy $K_e$ calculated with Eq. (6) ($A = 10^{-11}$ J/m, $K_0 = 8 kJ/m^3$). Curve $k_a = 1$ separates regions where $K_e$ is governed by $K_u$ (left side) or $D$ (right side).
Fig. 4. Experimental values of $K_{mc}$ deduced from $H_c$ measurements using Eq. (6) and (7) and confrontation with model: (a) magnetizations along edges, no buffer zone; (b) magnetizations along edges with BZ; and (c) magnetizations along diagonals with BZ.

Values of $K_{mc}$ obtained this way from $H_c$ measurements are plotted on Fig. 4. One notices a sudden increase for $f>0.65$, corresponding to an increase of $H_c$ as already observed in Refs. [11,12]. Of course, new phenomena occur here, which have nothing to do with magnetoelastics. Noticing that corresponding samples feature annealing treatments (530 °C, $t>60$ min), (550 °C, $t>10$ min) or (570 °C, $t>2$ min), we explain the increase of coercivity calling upon firstly apparition of Cu nucleus, followed by the beginning of the formation of borides [13]. As a result, the relevant region allowing confrontation with the quantitative model of $K_{mc}$ is $f<0.65$.

3. Modeling of $K_{mc}$

Magnetoelastic energy due to reversing of CV is evaluated looking at the costly situation featured by CV and AD perpendicularly magnetized ($\theta = \pi/2$, AD magnetized along Oz). CV is supposed to switch when surrounded by the DW, and we will thus pay attention to the case $\varepsilon_d = L_0$. The situation involves three elastically identical media, i.e. AD (superscript d), DW (superscript w) and CV (superscript c) with magnetostriction coefficient $\lambda_a$ measured by SAMR method [14], plotted on Fig. 6 and fitted by Eq. (8),

$$\lambda_a(f) = 24.5 \times 10^{-6} - 31.5 \times 10^{-6}f$$

Young modulus $E$ is calculated in the frame of an effective-medium theory [8], with nanocrystals ($E_c = 210$ GPa) embedded in amorphous residual phase ($E_a = 160$ GPa [12]). One notices that it was not possible to reproduce evolution reported in Ref. [12], even introducing an interfacial medium elastically weak as proposed in Ref. [15] according to Ref. [16]. We finally introduce Poisson’s ratio $\nu \approx 0.33$ according to Ref. [17] and shear modulus $G = 0.5E/(1 + \nu)$.

DW and reversing CV ($\theta = \pi/2$) feature the same equilibrium strains $\lambda_{eq}: \lambda_a$, and $\lambda_{eq}^\perp = \lambda_{eq}^\perp = -\lambda_a/2$, which differ from those of AD, i.e. $\lambda_{eq}^\perp = \lambda_a$, $\lambda_{eq}^\perp = \lambda_{eq}^\perp = -\lambda_a/2$. As a result, media experiment compressive stresses $\sigma_\text{c}^\perp$ and shearing stresses $\tau_\text{s}^\perp$ related to strains $\lambda_{eq}^\perp$ and $\gamma_\text{s}^\perp$ by the way of generalized Hooke’s laws are written following [18] in Eq. (9) as:

$$\sigma_\text{c}^\perp - \nu(\sigma_\text{c}^\perp + \sigma_\text{c}^\perp) = E(\lambda_{eq}^\perp - \lambda_{eq}^\perp), \quad \tau_\text{s}^\perp = 2G\gamma_\text{s}^\perp$$

$$W^\perp = \frac{E}{2(1 + \nu)} \left[ \nu \left( \sum_j \Delta \lambda_j^\perp \right)^2 + \sum_j \left( \Delta \gamma_j^\perp \right)^2 + 2 \sum_j \gamma_j^\perp \right]$$

A basic simple hypothesis is that domains impose strains everywhere, i.e. inside DW and CV. Energy variations are localized in CV, allowing to identify $K_{mc}$ as $W^c$ obtained from Eq (10) and (11) and plotted in Fig. 5.

$$W^c(\pi/2) - W^c(0) = K_{mc} = p9E\lambda_a^2/[4(1 + \nu)]$$

where $p$ is a numerical prefactor equaling 1 in this first approach. Regarding experimental data, calculated $K_{mc}$ appears strongly overestimated. We thus improve our modelization as follows

In the first step, CV not considered yet, we introduce two identical buffer zones (BZ) (superscript b) at either side of DW, allowing a progressive matching of strain between DW and AD. BZ strains are calculated assuming linear variations from those of DW (now adjustable but assumed still uniform) to those of AD (equilibrium strains), the thickness $\alpha L_0$ of BZ being adjustable too. Magnetization of BZ, controlled by ferromagnetic exchange interactions,
A next step in improvement could consist in adding a second buffer zone (cf. Fig. 6) in charge of accommodating mismatch between CV and DW. According to Ref. [8], one can expect a new significant decrease in magnetostatic energy with a much better agreement between experiment and model.

### 4. Conclusion

The present work investigates the way a nanocrystalline soft material develops coherent stress at the scale of, a condition required to obtain a noticeable effect on coercivity from magnetoelastic origin. The reversal mechanism itself is a good candidate, leading us to study the CV reverse in connection with the macroscopic magnetization process featured by DW displacement. The role of magnetostatics is underlined, with questions still open on how to include it in a quantitative tractable approach; this problem shared by all the materials exhibiting vanishing anisotropies [19]. As a result, an evolution of the RAM is proposed and used to extract magnetoelastic contribution from $H_c$ measurements provided by a large database offered for investigations. Comparison with model is not perfect; correct evaluation involving numerous degrees of freedom to express the natural behaviour of any physical system is required to take advantage of all possibilities to minimize its energy.

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