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1990 J. Phys.: Condens. Matter 2 1885

(http://iopscience.iop.org/0953-8984/2/7/018)

J. Phys.: Condens. Matter **2** (1990) 1885–1890. Printed in the UK

Fluctuations of the saturation magnetostriction constant in amorphous ferromagnets

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Received 29 June 1989, in final form 16 October 1989

Abstract. Fluctuations of the magnetostriction constant λ with correlation length larger than the correlation length of the magnetic moments, affect the anisotropy field introduced by applying external stress. The experimental anisotropy field contains two terms. The first one is the anisotropy field corresponding to the average λ. The second term is a series in increasing powers of the stress. The coefficient of the nth power encloses the nth moment of the λ distribution. When the experimental value of λ is determined by means of magnetoelastic effects, it is obtained from the stress derivate of the magnetoelastic anisotropy field. Since the existence of fluctuations leads to a series of powers of the stress, the experimental λ would also depend on the stress. This dependence is expected to be enhanced when the average λ vanishes. It is suggested that the stress dependence of λ observed in nearly zero magnetostriction, Co-rich alloys could be related to fluctuations in λ.

1. Introduction

A ferromagnetic sample would exhibit zero macroscopic magnetostriction when either the local magnetostriction coefficients are zero at each atomic site or the average of non-zero local coefficients, over the different orientations of the structural units, vanishes.

As a well-known example let us consider a polycrystalline sample composed of cubic crystallites distributed isotropically. The overall magnetostriction should be zero if $\lambda_{100} = \lambda_{111} = 0$ at each crystallite, but a zero overall magnetostriction is also expected when $\lambda_{100} \neq 0$ and $2\lambda_{100} = -9\lambda_{111}$, since for this relation the average λ is zero.

Amorphous ferromagnets are characterised by orientational fluctuations of the local easy axes. The correlation length of these fluctuations, l, is the correlation length of the amorphous structure, i.e. typically $l \sim 10$ Å. It is known that l is much lower than the correlation length of the magnetic moments, L, for 3d-transition-metal alloys. Exchange interactions average the short wavelength fluctuations of anisotropy and magnetostriction, thus giving rise to unique values of K and λ in volumes of the order of L^3. Our interest will be focused on the long-range fluctuations of λ, i.e. fluctuations with correlation length $\delta \sim L$. Note that long-range fluctuations are not an intrinsic characteristic of the amorphous state, rather they describe the coexistence of different amorphous phases, as well as long-range fluctuation of the short-range order.

The aim of this work is to point out that the experimental magnetostriction obtained by means of magnetoelastic effects, in a sample with long-range spatials fluctuations of λ around its average value $\langle\lambda\rangle$, is given by

$$\lambda^{\exp} = \langle\lambda\rangle + \sum_{n=2}^{\infty} a_n \langle(\lambda - \langle\lambda\rangle)^n\rangle\sigma^{n-1} \tag{1}$$

where the a_n, which depend on the type of experiment performed, are functions of the applied field and anisotropy and σ is the applied stress.

Now it is well established that $\lambda^{\exp}$ of nearly zero-magnetostriction amorphous alloys decreases linearly with the applied stress as

$$\lambda^{\exp} = \lambda_{\sigma=0} - a\sigma \tag{2}$$

where a is a positive value of about $10^{-10}\,\mathrm{MPa}^{-1}$.

This behaviour was observed by the use of different experimental methods based on magnetoelastic effects [1–3].

Different mechanisms have been proposed to explain the stress dependence of $\lambda^{\exp}$ [4–7]. Some reviews dealing with the overall behaviour of magnetostriction in Co-rich alloys are available in the literature [8–10].

In this work the long-wavelength fluctuations of λ are analysed as a new possible origin of the stress dependence described by equation (2).

2. Dependence of the magnetisation on the applied stress

In a magnetised sample the action of a tensile stress along the direction of the applied stress induces a change of magnetisation. The sign of such increment is the sign of the magnetostriction constant of the sample. In Co-rich alloys with magnetostriction small and positive, the increment of magnetisation per unit stress, $\Delta M/\Delta\sigma$, evolves from positive values, at low stress, toward negative values at higher stresses. This behaviour seems to indicate a stress dependence of the magnetostriction constant according to that described by equation (2). It will be shown that fluctuations in the magnetostriction around a small and positive average value $\langle\lambda\rangle$ give rise to a stress dependence of $\Delta M/\Delta\sigma$ consistent with that observed in the experiments.

Let us consider a metallic glass ribbon, subjected to a longitudinal magnetic field H high enough to overcome the 'knee' of the magnetisation curve. Under this condition the action of a tensile stress applied along the ribbon axis can be envisaged as an increment of H by an amount $3\lambda\sigma/\mu_0 M_s$.

The discussion will be restricted to those values of H and σ for which $H + H_k$ lies in the region corresponding to the approach to saturation.

The increment of magnetisation produced by an increment of stress, $\Delta\sigma$, when the ribbon is subjected to H and σ will then be given by

$$\Delta M = \chi\, 3\lambda\Delta\sigma/\mu_0 M_s \tag{3}$$

where χ is the positive differential susceptibility at $H + H_k$, M_s the saturation magnetisation and λ the saturation magnetostriction, supposed to be uniform everywhere along the sample. Since χ decreases with increasing H, equation (3) supplies direct information about the strength and sign of ΔM, according to the sign of λ. For positive λ, ΔM is always positive and its strength decreases as either H or σ increases. When λ is negative, ΔM is negative everywhere but its absolute value increases with σ, since H_k is negative. Therefore any change in the sign of ΔM has to be explained as originating from a change in the sign of λ.

Let us now consider the existence of spatial fluctuations of λ around the average value $\langle\lambda\rangle$. The average expected ΔM for long-range fluctuations should be

$$\langle\Delta M\rangle = (\chi\lambda)\, 3\Delta\sigma/\mu_0 M_s. \tag{4}$$

By putting $\delta\lambda = \lambda - \langle\lambda\rangle$, the second order expansion of χ yields

$$\langle\Delta M\rangle = (\chi\langle\lambda\rangle + 3\chi'\langle\delta\lambda^2\rangle\sigma/\mu_0 M_s)\, 3\Delta\sigma/\mu_0 M_s \tag{5}$$

where χ and χ' are the corresponding functions at $H + 3\langle\lambda\rangle\sigma/\mu_0 M_s$.

For the sake of clarity let us assume that χ behaves as $a/(H + H_k)^n$. Under this assumption equation (5) leads to

$$\langle\Delta M\rangle = \{[\mu_0 M_s H\langle\lambda\rangle + 3\sigma(\langle\lambda\rangle^2 - n\langle\delta\lambda^2\rangle)]/(\mu_0 M_s H + 3\langle\lambda\rangle\sigma)\} \times (3a\Delta\sigma/\mu_0 M_s|H + 3\langle\lambda\rangle\sigma/\mu_0 M_s|^n). \tag{6}$$

For a sample with uniform magnetostriction the same susceptibility behaviour would have led, through equation (3), to

$$\Delta M = \lambda 3a\Delta\sigma/\mu_0 M_s|H + 3\lambda\sigma/\mu_0 M_s|^n. \tag{7}$$

Comparison of equations (6) and (7) points out that the effective λ in a sample with fluctuating λ is given by

$$\lambda^{\exp} = [\mu_0 M_s H\langle\lambda\rangle + 3\sigma(\langle\lambda\rangle^2 - n\langle\delta\lambda^2\rangle)]/(\mu_0 M_s H + 3\langle\lambda\rangle\sigma). \tag{8}$$

Let us suppose $\langle\lambda\rangle$ to be small and positive. In a sample with uniform $\lambda = \langle\lambda\rangle$, ΔM should always be positive, according to equation (7). However in a sample with fluctuations of λ in which $\langle\delta\lambda^2\rangle > \langle\lambda\rangle^2$ there should be a change of sign of ΔM, from positive to negative, at $\sigma = \sigma_c$ given by:

$$\sigma_c = \mu_0 M_s H\langle\lambda\rangle/3[n\langle\delta\lambda^2\rangle - \langle\lambda\rangle^2]. \tag{9}$$

Equation (8) points out that λ fluctuation, with $\langle\delta\lambda^2\rangle \gg \langle\lambda\rangle^2$, gives rise to a stress dependence of the experimental magnetostriction. At zero stress the experimental λ is the average value $\langle\lambda\rangle$, but as σ increases the experimental λ value depends on σ as

$$\lambda^{\exp} = \langle\lambda\rangle - 3n\langle\delta\lambda^2\rangle\sigma/\mu_0 M_s H. \tag{10}$$

However, as was expected from elementary statistical theory, equation (6) converges to equation (7) when $\langle\lambda\rangle^2 \gg \langle\delta\lambda^2\rangle$.

3. Magnetoelastic anisotropy field

The stress dependence of magnetisation analysed above does not supply a direct value of λ. This is due to the fact that the expression relating ΔM and λ encloses the differential susceptibility (see equations (3), (5), (6) or (8)). Most of the experimental magnetoelastic methods used to determine λ are based on the measurement of the stress dependence of the anisotropy. Particularly, the transverse susceptibility technique has been customarily applied in different versions for measuring λ. In this method a magnetic field, H_z, high enough to achieve technical saturation is applied along the z axis. Then a tensile stress, σ, with variable strength is also applied along the same direction. An AC field $H_y < H_z$, induces rotations of the magnetisation from the z axis towards the y axis. The component of the magnetisation, M_y, along the y axis is given by (2)

$$M_y = M_s H_y/(H_z + H_k) \tag{11}$$

where $H_k = 3\lambda\sigma/\mu_0 M_s$.

The stress derivate of the inverse susceptibility, M_y/H_y is, according to equation (11)

$$\mathrm{d}(1/\chi)/\mathrm{d}\sigma = (1/M_s)\mathrm{d}H_k/\mathrm{d}\sigma = 3\lambda/\mu_0 M_s^2. \tag{12}$$

When λ fluctuates spatially around $\langle\lambda\rangle$, with a long wavelength, H_k does likewise and the average M_y, measured by experiment, should be written as

$$\langle M_y\rangle = M_s H_y\langle 1/(H_z + H_{\langle k\rangle} + \delta H_k)\rangle \tag{13}$$

where

$$H_k = 3\lambda\sigma/\mu_0 M_s = 3(\langle\lambda\rangle + \delta\lambda)\sigma/\mu_0 M_s = H_{\langle k\rangle} + \delta H_k.$$

Expansion to second order in equation (13) and performing average procedure yields

$$1/\chi = [(H_z + H_{\langle k\rangle})/M_s](1 - \langle\delta H_k^2\rangle/(H_z + H_{\langle k\rangle})^2 + \ldots). \tag{14}$$

By comparing equations (14) and (11) it turns out that as a consequence of the fluctuations the effective anisotropy field becomes

$$H_k^{\exp} = H_{\langle k\rangle} - \langle\delta H_k^2\rangle/(H_z + H_{\langle k\rangle}) + \ldots. \tag{15}$$

In nearly zero-magnetostriction alloys $H_{\langle k\rangle} = 3\langle\lambda\rangle\sigma/\mu_0 M_s$ is much lower than H_z, therefore equation (15) can be written as

$$H_k^{\exp} = H_{\langle k\rangle} - \langle\delta H_k^2\rangle/H_z. \tag{16}$$

The experimental magnetostriction results in

$$\lambda^{\exp} = \tfrac{1}{3}\mu_0 M_s\,\mathrm{d}H_k^{\exp}/\mathrm{d}\sigma = \langle\lambda\rangle - 6\langle\delta\lambda^2\rangle\sigma/\mu_0 M_s H_z. \tag{17}$$

4. Discussion

It has been shown that fluctuations in λ give rise to a stress dependence of the experimental magnetostriction consistent with that observed and reported in Co-rich amorphous alloys. According to equation (16) the rate of the stress dependence (which is negative) takes the absolute value $6\langle\delta\lambda^2\rangle\sigma/\mu_0 M_s H_z$. By considering $\mu_0 M_s = 0.9$ T and $H_z = 8 \times 10^4$ A m^{-1} its order of magnitude is $10^{-4}\langle\delta\lambda^2\rangle$ Pa^{-1} or $10^2\langle\delta\lambda^2\rangle$ MPa^{-1}.

Therefore $\langle\delta\lambda^2\rangle$ must take a value of about 10^{-12} to account for the experimental rate of 10^{-10} MPa^{-1}.

There are two experimental observations which tend to support the model developed here as being the true source of the behaviour of λ. The first one is the negligible dependence on the temperature of the decreasing rate of λ with σ, as shown by measurements performed at liquid nitrogen temperature [11]. The second important aspect is the influence of the bias field strength on the rate of the stress dependence, as indicated in equation (16). Hernando and co-workers [11] have recently shown that the higher H_z, the higher the stress required to change the sign of λ from negative to positive. Moreover, the variation of λ on the applied field was found experimentally to be about 10^{-10} m A^{-1} for fields of about 5×10^3 A m^{-1} and stresses of about 500 MPa, in very good agreement with the value deduced from equation (16) using the same parameters (1.33×10^{-10} A m^{-1}).

There are also two aspects which must be explained in order for the idea proposed here to be considered as being reliable. The first question deals with the existence of long-range fluctuations in λ, which are not an intrinsic characteristic of the amorphous structure. The long-range character of fluctuations is required to make valid the averaging procedure performed to obtain equations (4) and (13). The second aspect is connected with the high value of the distribution broadness $\langle \delta\lambda^2 \rangle \sim 10^{-12}$.

It is worth noting that the existence of two amorphous phases in Co-rich amorphous alloys has recently been evidenced by different experimental techniques [12–15]. These phases are characterised by slightly different short-range order, local symmetry and therefore local magnetic anisotropy. The coexistence of two phases with λ of about 10^{-6} and different sign to each other would justify both the high $\langle \delta\lambda^2 \rangle$ value as well as the long-range character of the fluctuations.

The coexistence of different amorphous phases is proposed as a possible source of the surprising behaviour of magnetostriction in nearly zero-magnetostriction Co-rich amorphous alloys.

Measurements of the law of approach to saturation in non-magnetostrictive alloys seem to indicate that the magnetoelastic coupling between magnetisation and local defects corresponds to local λ of about 10^{-6} [15]. This result reinforces the idea that zero macroscopic magnetostriction is reached by averaging the spatial fluctuation of local magnetostriction.

In a previous work, some of the authors [16] have thoroughly analysed the influence of the distribution of the residual stress σ_r on the experimental λ value obtained by using magnetoelastic effects. Although such influence has been shown to be noticeable, it cannot account for a change of sign of λ^{exp}. To demonstrate this, let us consider λ uniform but H_k fluctuating as a consequence of fluctuations in σ_r. In this case H_k can be written as

$$H_k = H_{\langle k \rangle} + \delta H_k \tag{18}$$

with

$$H_{\langle k \rangle} = 3\lambda(\sigma + \langle \sigma_r \rangle)/\mu_0 M_s = 3\lambda\sigma/\mu_0 M_s.$$

Thus

$$\langle \sigma_r \rangle = 0$$

and

$$\delta H_k = \frac{3\lambda\delta\sigma_r}{\mu_0 M_s}.$$

The macroscopic observations of λ^{exp} can also be predicted in this case through the averaging procedure shown in equation (13). Notice, however, that $\langle \delta H_k^2 \rangle$ does not increase with σ^2. Therefore it can be easily seen that λ^{exp} obtained from equation (16) does not depend on σ.

We conclude that fluctuations in λ account for the general aspect of the stress dependence of λ^{exp} observed in low-magnetostriction metallic glasses.

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