Magnetic properties of nanocrystalline FeCuNbSiB with huge creep induced anisotropy

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Abstract. Ribbons of originally amorphous Fe$_{73.5}$Cu$_1$Nb$_3$Si$_{15.5}$B$_7$ have been annealed under tensile stress for about 4 seconds at temperatures between 450°C and 750°C. Under such short time conditions the optimum nanocrystalline state is achieved for annealing temperatures between 600°C and 720°C. This is about 100°C higher than in more conventional heat treatments with annealing times in the order of an hour. The stress annealed ribbons reveal an almost perfectly linear hysteresis loop and a transverse domain pattern with zigzag domain walls characteristic for a magnetic easy plane perpendicular to the stress axis. The induced magnetic anisotropy increases proportional to the annealing stress and is comparable to that obtained after more prolonged annealing. We succeeded to induce anisotropy constants as high as $K_u \approx 12$ kJ/m$^3$ which even exceed the local magneto-crystalline anisotropy ($K_1 \approx 8$ kJ/m$^3$) of the crystalline Fe-Si phase. Nonetheless, the coercivity is still as small as $H_c \approx 8$ A/m. This is to be compared to $H_c \approx 0.5$ A/m for field annealed samples where $K_u (\approx 20$ J/m$^3$) is smaller by more than two orders of magnitude. The behaviour of $H_c$ can be understood within the framework of the random anisotropy model.

1. Introduction

Nanocrystalline Fe-Cu-Nb-Si-B alloys reveal a homogeneous ultrafine grain structure of bcc-FeSi with grain sizes of typically 10–15 nm and random orientation, embedded in an amorphous minority matrix [1], [2]. This particular microstructure is obtained by devitrification from the amorphous state and enables excellent soft magnetic properties comparable to those of permalloys and Co-based amorphous alloys. Optimised alloy compositions like Fe$_{73.5}$Cu$_1$Nb$_3$Si$_{15.5}$B$_7$ moreover reveal near-zero saturation magnetostriction in the nanocrystalline state which makes them particularly attractive for application [2].

Similar to other soft magnetic alloys, the hysteresis loop can be tailored by uniaxial magnetic anisotropies induced by annealing in a magnetic field [3], [4] or under tensile stress [5], [6]. For Si-contents larger than about 10 at% stress annealing of nanocrystalline Fe-Cu-Nb-Si-B yields a creep induced anisotropy characterised by an easy magnetic plane perpendicular to the stress axis. This macroscopic anisotropy originates from the local magneto-elastic anisotropy of the FeSi crystallites associated with their elongation induced by stress annealing [6]–[8].

The induced magnetic anisotropy energy is proportional to the stress applied during annealing and can amount to be several thousands of J/m$^3$ which is two orders of magnitude higher than the typical
magnitude of field induced anisotropies. Such strong and well-controlled anisotropies are of interest, for example, for magnetic energy storage cores which require low a permeability of typically a few hundreds being constant over a wide magnetic field range [9], [10].

The practical use of stress annealing for application ultimately requires a continuous reel-to-reel annealing technique with possibly short annealing times [10]. The objective of the present work is to investigate in more detail the magnetic properties of stress annealed Fe$_{73.5}$Cu$_1$Nb$_3$Si$_{15.5}$B$_7$ after such short time annealing treatments lasting a few seconds only. A particular emphasis hereby was to increase the creep induced anisotropy as far as possible.

2. Experimental

Amorphous ribbons of Fe$_{73.5}$Cu$_1$Nb$_3$Si$_{15.5}$B$_7$ were prepared by rapidly quenching from the melt in about 20 µm thickness. The nanocrystalline state was achieved by annealing under a tensile stress $\sigma_a$ along the ribbon axis. For this purpose the ribbon was continuously transported through a furnace with an approximately 10 cm long homogeneous temperature zone kept under nitrogen atmosphere. The ribbon was guided in close contact to an annealing fixture made of stainless steel in order to ensure a good heat transfer. The annealing temperature $T_a$ was measured by a thermocouple attached to the fixture in the centre of the oven. The annealing speed was 1.6 m/min which corresponds to an annealing time of 4 seconds at $T_a$ with heating and cooling rates in the range of 100-200 K/s. The set-up allows inline recording of the ac (60 Hz) hysteresis loop after annealing. The investigated ribbons were 1.6 mm and 6 mm wide. The narrower width was chosen to achieve highest tensile stress.

The saturation magnetostriction $\lambda_s$ was measured by the small angle magnetisation rotation (SAMR) method [11], [12]. The magnetic anisotropy was determined from the dc hysteresis loop recorded on straight ribbons. The samples were 0.5 m long which ensured negligibly small demagnetisation effects. Coercivity was determined either from the hysteresis loops or, in particular for high anisotropies, with a Förster Koerzimat 1.096 [13]. The domain structure was determined by magneto-optical Kerr microscopy using digital image processing to enhance the contrast [14]. The images shown are difference images between the demagnetised and the magnetically saturated state.

3. Results and Discussion

Figure 1 shows the saturation magnetostriction $\lambda_s$, the anisotropy field $H_a$ and the coercivity $H_c$ as a function of the annealing temperature $T_a$ after annealing for 4 seconds under a tensile stress of 45 MPa. The decrease of $\lambda_s$ and the simultaneous increase of $H_a$, both starting at about 550°C, indicate the onset of the first stage of crystallisation which is largely completed at annealing temperatures of about

![Figure 1](image_url)
600°C. As a result we obtain the typical nanocrystalline microstructure consisting of bcc FeSi grains embedded in an amorphous matrix [1], [2]. X-ray diffraction revealed an average grain size of about 12 nm. This structure remains stable up to about 720°C where the precipitation of Fe-B compounds leads to a severe degradation of coercivity and to an increase of the saturation magnetostriction. The optimum nanocrystalline state is thus achieved at annealing temperatures which are about 100°C higher than in more conventional heat treatments with annealing times in the order of an hour [1], [2]. Due to the very short annealing time the onset of crystallisation even occurs at higher temperatures than in typical DSC (differential scanning calorimetry) scans where the corresponding onset temperatures are \( T_{x1} = 508°C \) and \( T_{x2} = 694°C \) for a heating rate of 10 K/min.

Figure 2 shows the typical hysteresis loops obtained in almost the whole range of annealing temperatures \( T_a \). Only above \( T_a \approx 720°C \) the coercivity and the remanence start to increase (cf figure 1) indicating that the hysteresis loop is more and more controlled by the high magneto-crystalline anisotropy of Fe-B precipitates (e.g. \( K_1 \approx -400 \, \text{kJ/m}^3 \) for Fe12B [15]). At \( T_a \approx 745°C \), for example, where \( H_c \) approaches its maximum value, the hysteresis loop reveals a high remanence to saturation ratio of \( J_r/J_s \approx 80\% \) (\( \sigma_a = 20 \, \text{MPa} \)). The remanence slightly decreases to about 75\% when the annealing stress \( \sigma_a \) increases to about 200 MPa indicating the formation of a still transversely induced anisotropy.

Figure 3 depicts the induced anisotropy in the nanocrystalline state as a function of the annealing stress.

**Figure 2.** Typical hysteresis loops obtained after stress annealing. The arrow indicates the definition of the anisotropy field \( H_a \).

**Figure 3.** Induced anisotropy \( K_u \) of nanocrystalline Fe73.5Cu1Nb3Si15.5B7 as a function of the annealing stress.

**Figure 4.** Domain patterns of 1.6 mm wide ribbons annealed for 4 s at 600°C with a tensile stress of (a) 30 MPa, (b) 160 MPa, (c) 430 MPa and (d) 750 MPa.

**Figure 5.** Domain width \( w \) as a function of the magnetic anisotropy \( K_u \), induced by stress annealing 1.6 mm and 6 mm wide ribbons for 4 s at \( T_a \).
stress. The anisotropy constant $K_u$ was evaluated by $K_u = H_a J_s/2$ where $J_s$ denotes the saturation polarisation. Although the annealing time is only a few seconds, the magnitude of the induced anisotropy is comparable to previous results obtained for more prolonged annealing times in the order of an hour [5]-[8]. Indeed, we achieved induced anisotropies up to $K_u = 12 \text{ kJ/m}^3$ resulting in a relative permeability of $\mu = 50$ constant up to a magnetic field of about 20 kA/m (cf figures 2 and 3). Such record high values of $K_u$ even exceed the local magneto-crystalline anisotropy constant of the FeSi crystallites ($K_1 = 8 \text{ kJ/m}^3$ [2]) indicated by the horizontal, dash-dotted line in figure 3. The achievable maximum value of $K_u$ was limited by the circumstance that the ribbons broke when we tried to apply annealing stresses larger than about 800 MPa. The plastic elongation of the ribbons annealed under the maximum load close to 800 MPa was already as large as 50% relative to the original length.

Figures 4 and 5 supplement the characteristic features of the domain structure. The transverse domains separated by zigzag walls indicate a well developed magnetic easy plane perpendicular to the direction of the annealing stress. The domain width $w$ can be well described theoretically by the Landau-Lifshitz domain model [14], i.e.

$$w = \sqrt{8bA/K_u} \quad (1)$$

where $b$ denotes the ribbon width and $A (= 6\times10^{-12} \text{ J/m})$ is the exchange stiffness. In comparison field annealed samples reveal straight domain walls and significantly wider domains ($w = 100 – 200 \mu\text{m}$) due to their comparatively small anisotropy [16].

Figure 6, finally, shows the coercivity $H_c$ as a function of the induced anisotropy $K_u$. For comparison the figure also contains the data obtained after magnetic field annealing. The latter yields considerably smaller induced anisotropies of $K_u = 5 – 25 \text{ J/m}^3$ and correspondingly small coercivities of $H_c = 0.3 – 0.6 \text{ A/m}$ [2]-[4]. Yet even for the huge creep induced anisotropies exceeding the local magneto-crystalline anisotropy of the crystallites, $H_c$ remains well below 10 A/m.

The coercivity mechanism is schematically sketched in figure 7. Thus, the sample reveals a small remanent magnetisation $J_r$ which basically is determined by anisotropy fluctuations $\delta K$ [16], [17]. At $H_c$ the average magnetisation is slightly rotated out of the magnetic easy axis thus compensating the remanent magnetisation. Coercivity and remanence, hence, are related by $\mu_0\chi H_c = J_r$, where $\mu_0$ is the vacuum permeability and $\chi$ is the susceptibility associated with $K_u$. Assuming $J_r/J_s \propto \delta K/K_u$ [17], this results in

$$H_c = p_c \delta K/J_s \quad (2)$$

where $p_c$ denotes a dimensionless pre-factor related to the proportionality factor between $J_r/J_s$ and...
The anisotropy fluctuations $\delta K$ include a significant contribution arising from the randomly oriented magneto-crystalline anisotropy of the nanocrystals [16], [17]. The magnitude of these fluctuations can been evaluated within the framework of the random anisotropy model for mixed uniform and random anisotropy which yields for $K_u > \delta K$ [17]

$$\delta K = \frac{1}{2} x \sqrt{\beta K_1 K_u (D/L_0)^3}.$$  

Here $x$ is the crystalline volume fraction, $\beta$ a factor taking into account the cubic symmetry of the crystals, $D$ is the grain size and $L_0 = \varphi_0 (A/K_1)^{1/2}$ the basic exchange length related to the local magneto-crystalline anisotropy $K_1$. For a theoretical estimate we assumed the following material parameters: $K_1 = 8$ kJ/m$^3$, $\beta = 0.4$, $L_0 = 40$ nm, $x = 0.75$ [2], [17] and $D = 12$ nm as determined by X-ray diffraction. The experimental data can then be reasonably matched assuming $p_c = 0.16$. The numerical result is represented by the full line shown in figure 6.

It should be finally noted that the determination of $H_c$ is a non-trivial experimental task, in particular for very high anisotropies. Owing to the extreme loop shape where both $J_r/J_s$ and $H_c/H_a$ are less than 0.1%, the accuracy needed to reliably resolve $H_c$ is at the limit of the experimental resolution. The most reproducible results could still be obtained by using a commercial fluxgate magnetometer (Förster Koerzimat CS 1.096 [13]). In this case we used 60 mm long samples where 8 ribbons were stapled on each other in order to increase the signal to noise ratio. Yet the experimental error for $H_c$ is still relatively large ranging in the order of about ± 10 – 20%.

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