Interplay of uniform and random anisotropy in nanocrystalline soft magnetic alloys

Sybille Flohrer a,*, Rudolf Schäfer a, Christian Polak b, Giselher Herzer b

** a IFW Dresden, Institute for Metallic Materials, Helmholtzstrasse 20, D-01069 Dresden, Germany
b VACUUMSCHMELZE GmbH & Co. KG, Grüner Weg 37, D-63450 Hanau, Germany

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Abstract

The magnetization process in nanocrystalline Fe 73Cu1Nb3Si16B7 ribbons with different magnitudes of a magnetic field induced transverse anisotropy was investigated by Kerr-microscopy and magnetic hysteresis measurements at room temperature and elevated temperatures. Comparing samples with strong \( K_u \approx 30 \text{ J/m}^2 \) and weak \( K_u \approx 3 \text{ J/m}^2 \) induced anisotropy \( K_u \), we find significant differences in the magnetic microstructure, reflecting the interplay of the uniform, field induced and the random magneto-crystalline anisotropy. In particular for weak \( K_u \), we find irregular magnetization patches within the wide regular domains connected to the induced anisotropy. These magnetization patches are fluctuating on the scale of a few micrometers and are interpreted in terms of the random anisotropy.

Keywords: Magnetic domain; Nanocrystalline materials; Magnetic anisotropy; Kerr-microscopy; Magnetization process

1. Introduction

Nanocrystalline Fe–Cu–Nb–Si–B alloys reveal a homogeneous ultrafine grain structure of body-centered cubic FeSi with grain sizes \( D \) of typically 10–15 nm and random orientation, embedded in an amorphous minority matrix. 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 [1–4]. The composition investigated here, i.e., Fe 73Cu1Nb3Si16B7, moreover reveals near-zero saturation magnetostriction \( (|\lambda_s| < 0.2 \text{ ppm}) \) (cf. [4]) and meanwhile has successfully entered into application under the trademark VITROPERM® 800 [5].

The soft magnetic properties basically originate from the suppression of the random local anisotropy by exchange interaction [2,3] similar to the case of amorphous metals [6]. However, the reduction of the relatively short wave random magneto-crystalline anisotropy is only one side of the coin. Additional, comparatively uniform anisotropies have to be considered for understanding the soft magnetic properties in more detail (cf. [4]). Thus, a low or near-zero magnetostriction constant is needed to avoid magneto-elastic anisotropy due to internal mechanical stress. Uniaxial anisotropy induced by magnetic field annealing plays a tremendously important role in order to tailor the hysteresis loop according to the needs of the application.

Indeed, it appears that, as in amorphous metals, the average random anisotropy of optimized nanocrystalline alloys is negligibly small and that the soft magnetic properties are predominantly controlled by magneto-elastic and, in particular, by field induced anisotropies. The corresponding evidence is given by:

1. a vanishing...
or modified grain size dependence of coercivity for grain sizes below about 15–20 nm [7,8], (2) wide regular domain patterns [9,10] and (3) the shape of the hysteresis loop after magnetic field annealing, which almost perfectly matches the theoretical expectations for a uniform uniaxial anisotropy (cf. [4]).

Nonetheless there are also significant differences to amorphous metals, which bring about the more prominent role of the random anisotropy even in optimized nanocrystalline soft magnets. A well-known example is the degradation of the soft magnetic properties with increasing temperature, since the exchange coupling between the nanocrystallites is reduced when approaching the Curie temperature of the amorphous matrix [2,4,9].

The present paper intends to illuminate in more detail the interplay between random and small uniform anisotropy in highly permeable nanocrystalline alloys. For this purpose, we analyze the magnetization process of nanocrystalline Fe75Cu1Nb3Si16B7 (VITROPERM® 800F) with a small induced anisotropy of $K_u$. We investigated two toroidally wound cores of nanocrystalline Fe75Cu1Nb3Si16B7 with different magnitudes of field induced anisotropy and compare it to that of an amorphous Co-base alloy with similarly high permeability.

2. Experimental

We investigated two toroidally wound cores of nanocrystalline Fe75Cu1Nb3Si16B7 (VITROPERM® 800F) with transverse field induced anisotropies of $K_u \approx 3$ and $K_u \approx 30 \text{ J/m}^3$. The cores had an inner and outer diameter of 13 and 20 mm, respectively, and were wound from 20 μm thick and 20 mm wide ribbons of the amorphous precursor material. These samples were annealed for 0.5 h at 570 °C in order to achieve the nanocrystalline state. During the heat treatment, the cores were homogeneously magnetized by a saturating magnetic field applied either along or transverse to the ribbon axis. The strength of the transversely induced anisotropy was varied by appropriate choice of the annealing time and temperature in the transverse field (cf. [4]): one core was subjected to the transverse field during the entire heat treatment resulting in a relatively strong induced anisotropy of $K_u \approx 30 \text{ J/m}^3$, which corresponds to an average permeability of $\mu = J_s^2 / (2\mu_0 K_u) \approx 20 \times 10^3$. A second core was transverse field annealed only for 3 h at 420 °C after having been crystallized in a longitudinal magnetic field. This annealing condition results in a significantly smaller induced anisotropy of $K_u \approx 3 \text{ J/m}^3$, corresponding to an average permeability as high as $\mu \approx 200 \times 10^3$. For comparison, we also investigated a transverse field annealed core of an amorphous Co67Fe4Mo2Si16B11 alloy (VITROVAC® 6025F) with a small induced anisotropy of $K_u \approx 0.6 \text{ J/m}^3$ corresponding to a permeability of $\mu \approx 200 \times 10^3$. The saturation polarizations of the nanocrystalline and the amorphous alloy are $J_s = 1.22$ and $J_s = 0.55 \text{ T}$, respectively. Both alloys have a near-zero saturation magnetostriiction of $|\lambda_s| < 0.2 \text{ ppm}$ in the annealed state and are optimized commercial grades for applications which require high permeabilities.

The magnetization process of the samples was investigated by measurement of the hysteresis loops and by domain observations. The experiments were performed at room temperature and, for selected samples, also at elevated temperatures up to 550 °C. The hysteresis loops were determined by inductive measurements with a frequency of $f = 0.1 \text{ Hz}$ at room temperature and, for sensitivity reasons, with $f = 50 \text{ Hz}$ at elevated temperatures. For the hysteresis loops as well as for the domain observations, the cores were magnetized along their circumferential direction, i.e., transverse to the induced magnetic easy axis. The domain structure was observed by digital enhanced Kerr-microscopy [11] without further sample preparation such as polishing or coating. At room temperature the observations were performed directly on the outer surface of the magnetic core. Domain observations at elevated temperatures up to 550 °C were conducted in a heating stage under vacuum on small pieces of ribbons of about 7 mm in diameter.

3. Results and discussion

The investigated samples all show the typical features expected for a transverse uniaxial anisotropy: the hysteresis loops shown in Fig. 1 are characterized by a low remanence to saturation ratio and an essentially linear loop up to ferromagnetic saturation. The domain patterns in Fig. 2 reveal wide transverse slab domains and magnetization rotations as the prevailing magnetization process.

![Fig. 1. Quasi-static hysteresis loop of: (a) the amorphous core (induced transverse anisotropy $K_u \approx 0.6 \text{ J/m}^3$); (b) the nanocrystalline core with high induced anisotropy ($K_u \approx 30 \text{ J/m}^3$); and (c) the nanocrystalline core with low induced anisotropy ($K_u \approx 3 \text{ J/m}^3$).](image-url)
However, homogeneous magnetization rotation is only found for the amorphous core (Fig. 2(a)) and the nanocrystalline core with strong induced anisotropy $K_u$ (Fig. 2(b)). The magnetization process of the nanocrystalline core with weak $K_u$ (Fig. 2(c)) is dominated by inhomogeneous rotations, domain nucleation and domain splitting. The more complex domain structure of the nanocrystalline core with low $K_u$ is also manifested in a slightly enhanced remanence to saturation ratio (Fig. 1). Indeed, if the induced anisotropy of the nanocrystalline alloy is reduced further, below about 2–3 J/m$^3$, the remanence increases rapidly and the hysteresis loop becomes non-linear. In contrast, the transverse field annealed amorphous alloy still shows a low remanence and a linear hysteresis loop at even smaller induced anisotropy of a few tenths J/m$^3$ only.

Fig. 2. Magnetization process in hard axis fields of: (a) the amorphous core ($K_u \approx 0.6$ J/m$^3$); (b) the nanocrystalline core with high induced anisotropy ($K_u \approx 30$ J/m$^3$); and (c) the nanocrystalline core with low induced anisotropy ($K_u \approx 3$ J/m$^3$). Arrows indicate the magnetization direction, which is estimated from inductive magnetization measurements in dependence of the applied magnetic field. The magnetization is given in fractions of the saturation polarization $J_s$.

Significant contributions from the magneto-elastic anisotropy due to internal mechanical stress or the local

Fig. 3 shows additional characteristics which become visible at higher optical resolution if the easy axis is oriented perpendicular to the axis of the magneto-optical sensitivity (which emphasizes transition regions like domain walls). The most interesting findings here are the patchy magnetization fluctuations within the wide domains of both nanocrystalline cores. These short-range fluctuations occur on the μm scale and are most pronounced in the nanocrystalline sample with low induced anisotropy (Fig. 3(a)). In the nanocrystalline core with the stronger induced anisotropy, corresponding magnetization fluctuations can be only observed at higher optical resolution (Fig. 3(b) and (c)). In comparison, the domains of the amorphous core (Fig. 3(d)) appear magnetized homogeneously, even at high optical resolution.

Inhomogeneous magnetization patterns discussed above ultimately originate from inhomogeneous anisotropy contributions $\delta K$ which become relevant if they exceed the homogeneous field induced anisotropy $K_u$.

Fig. 3. The high-resolution domain image of the tape wound core with $K_u \approx 3$ J/m$^3$ (a) exhibits a patchy fluctuation of magnetization. The core with higher $K_u \approx 30$ J/m$^3$ (b) and (c) reveals smaller patches at higher optical resolution. The amorphous core with $K_u \approx 0.6$ J/m$^3$ (d) shows homogeneously magnetized domains. The images were taken in the demagnetized states.
shape anisotropy around surface defects can be largely ruled out here for the following reasons:

1. The saturation magnetostriction of the investigated samples is close to zero ($|\lambda_s| < 0.2$ ppm) and the typical stress domain patterns (cf. [10]) are totally absent.
2. Typical surface defects like the air pockets on the wheel side of the ribbons typically extend over a scale of 50–100 $\mu$m, which is too large in order to account for the patchy magnetization fluctuations on the $\mu$m scale (Fig. 3).

The circumstance that the amorphous alloy, despite its very low induced anisotropy, reveals no magnetization fluctuations rather suggests that the magnetization inhomogeneities of the nanocrystalline sample are related to the microstructure. Indeed, the experimental observations can be understood coherently if they are interpreted in terms of the residual average contribution of the random magneto-crystalline anisotropy. That is:

1. Theoretical estimates within the random anisotropy model typically yield $\langle K_i \rangle \approx 2 - 4 \text{ J/m}^3$ for the average random magneto-crystalline anisotropy in optimized nanocrystalline Fe-base alloys and a negligibly small value of $\langle K_i \rangle \ll 10^{-3} \text{ J/m}^3$ for the average random local anisotropy in amorphous transition metal alloys [6,8,12]. Thus, a significant impact of the random anisotropy is only expected for the nanocrystalline sample with low induced anisotropy ($K_u \approx 3 \text{ J/m}^3$) where we have $\langle K_i \rangle \approx K_u$. Correspondingly, we roughly have $K_u/\langle K_i \rangle \approx 5 - 10$ for the nanocrystalline sample with the stronger induced anisotropy and $K_u/\langle K_i \rangle \approx 10^3$ for the amorphous sample. Accordingly, the magnetization process in these samples is increasingly homogenized.
2. The magnetization patches shown in Fig. 3 are modulated on a scale of a few micrometers, which is of the order of the exchange length $L_{ex} \sim (A/(K))^1/2$ ($A$ is the exchange stiffness constant and $K$ the average anisotropy constant). This makes sense since the easy axis resulting after averaging over the grains within the volume of the exchange length is randomly fluctuating on this very scale. Furthermore, the size of the patches decreases as the induced anisotropy increases (cf. Figs. 3(a) and (c)). Similarly, the patchy modulation decreases as increasing $K_u/\langle K_i \rangle$ since the orientation of the total anisotropy axis is more and more determined by the uniform induced anisotropy.

Fig. 4 illustrates the interplay of a uniform uniaxial anisotropy $K_u$ and the average random anisotropy $\langle K_i \rangle$ as obtained by numerical simulations [8]. For small $K_u$ the easiest magnetic axis is dominated by the random anisotropy. Accordingly, the easiest axis reveals a large angular dispersion from one region of exchange coupled

Fig. 4. Average orientation $\vartheta$ of the easiest magnetic axis for a system of randomly oriented particles with average anisotropy constant $\langle K_i \rangle$ and a superimposed, uniform uniaxial anisotropy $K_u$ (solid symbols: randomly oriented cubic grains; open symbols: uniaxial grains, dashed line: limit for $K_u = 0$; taken from [8]).

Fig. 5 demonstrates how the domain pattern of the nanocrystalline sample with low induced anisotropy ($K_u \approx 3 \text{ J/m}^3$) changes reversibly from wide domains at room temperature to small irregular domains at $350 \text{ °C}$, i.e., above $T_c^{am}$. Simultaneously coercivity and permeability are degraded by almost two orders of

Fig. 5. Transition from wide domains at room temperature to a patchy domain pattern at $350 \text{ °C}$ of the tape wound core with $K_u \approx 3 \text{ J/m}^3$ in the demagnetized state.
magnitude and the remanence to saturation ratio increases to about $J_r/J_s > 0.8$. The behavior is similar to that reported previously for a nanocrystalline Fe$_{73.5}$Cu$_1$Nb$_3$Si$_{13.5}$B$_9$ alloy [4,9] and indicates the dominance of the random magneto-crystalline anisotropy as the temperature exceeds $T_{am}$.

In comparison, Fig. 6 shows that the nanocrystalline sample with strong induced anisotropy ($K_u = 30$ J/m$^3$) reveals wide domains oriented more or less regularly along the direction of the induced easy axis even far above $T_{am}$. Fig. 7 complements the hysteresis loops at the corresponding temperatures. The regular domain structure and the linear shape of the hysteresis loop clearly indicate that the field induced anisotropy of this sample still plays a significant role in the magnetization process even at elevated temperatures. At the same time, however, the increase in coercivity and remanence and the more complex behavior of the domain patterns above 300 °C reflect an increasing contribution from the random magneto-crystalline anisotropy. Thus, for example, demagnetizing along the hard axis results in more irregular wide domains than demagnetizing along the easy axis (Fig. 6).

4. Summary and conclusion

We have investigated the magnetization process in nanocrystalline Fe$_{73.5}$Cu$_1$Nb$_3$Si$_{13.5}$B$_9$ ribbons with different magnitudes (i.e., $K_u = 3$ and 30 J/m$^3$) of a magnetic field induced transverse anisotropy. The results are compared to an amorphous Co-base alloy with an even lower induced anisotropy of $K_u = 0.6$ J/m$^3$ and can be summarized as follows:

1. All the samples reveal a linear hysteresis loop and wide transverse slab domains characteristic for a transverse uniaxial anisotropy.
2. However, homogeneous magnetization rotation within a domain is only found for the amorphous sample and the nanocrystalline sample with large $K_u$. The magnetization process of the nanocrystalline core with weak induced anisotropy is dominated by inhomogeneous rotations, domain nucleation and domain splitting.
3. The inhomogeneous processes appear to have their origin in magnetization patches fluctuating on the scale of a few micrometer, i.e., in the order of the exchange length $L_{ex}$. These fluctuations are most pronounced in the nanocrystalline sample with low $K_u$ and are virtually invisible in the amorphous sample.

It is suggested that the patchy magnetization fluctuations reflect the angular dispersion of the easiest
magnetic axis from one region of exchange coupled grains to the other. The effect is most pronounced if the random anisotropy is dominating other anisotropy contributions. Correspondingly, the angular dispersion of the easiest axis decreases and the patchy magnetization fluctuations disappear, the more the uniform anisotropy is becoming dominant over the random anisotropy.

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References