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Microstructure of nanocrystalline FeZr(N)-films and their soft magnetic properties

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Abstract

The microstructure of nanocrystalline FeZr(N) films, deposited by DC sputtering in a $Ar+N_2$ atmosphere was studied in correlation with their soft magnetic properties. The micromagnetic properties of the films were investigated using the Fresnel mode of Lorentz microscopy.

Keywords: Soft magnetic films; Lorentz microscopy; Nanocrystalline material

It has been found that certain nitrogen-containing nanocrystalline films composed of iron with 1–10 at% of a transition element, like Ta, Ti, Cr, Zr [1–4] have good soft magnetic properties. For thin films (<200 nm), where eddy current losses are small, magnetic losses at high frequencies are mainly determined by the presence of ferromagnetic resonance (FMR) with a frequency $f_{FMR} \approx \gamma(4\pi M_s H_K)^{1/2}$. Here, γ is the gyromagnetic ratio (0.446 MHz/Oe), M_s the saturation magnetization and H_K the effective anisotropy field in the plane. To obtain material with f_{FMR} in the GHz range, the anisotropy H_K should not be too small. On the other hand, it also should not be too high, otherwise the permeability $\mu = M_s/H_K$ will be too small. A reasonable compromise is in the range $H_K = 10-20\,G$, which allows keeping $\mu \approx 1000$ up to $f_{FMR} \approx 1-2\,GHz$. The ferromagnetic response at high frequencies is also influenced by the width of the FMR, which is determined by various dissipating processes and non-homogeneities of magnetization. One of the most efficient tools to study such magnetic non-homogeneities is the Lorentz transmission electron microscopy (LTEM), which has been intensively used before to study micromagnetic structures of Permalloy and other Ni–Fe based films (see reference list in Ref. [5]). Here, we report on our study of the micromagnetic features in soft magnetic FeZrN-films, as observed by LTEM.

Fe–Zr–N films were prepared by DC magnetron reactive sputtering. Films with thicknesses between 50 and 1000 nm were deposited at several temperatures between room temperature and 200°C. The films were grown in a nanocrystalline structural state on different substrates including glass, silicon wafers with and without a polymer underlayer. Pure (99.96 at%) Fe sheets partially covered with Zr wires were used as targets. The N and Zr content were controlled by varying the sputtering power and/or the Ar/N_2 gas mixture. An 800 Oe magnetic field was applied in the plane of the samples during deposition.

The deposition conditions were chosen to obtain a composition $Fe_{99-x}Zr_1N_x$, where the concentration of nitrogen was in the range of $x \leqslant 25$ at%. Standard $\theta-2\theta$ XRD scans show that up to $x = 10$ at% as-sputtered films were BCC single-phase materials with a strong (1 1 0) fibrous texture. The grain size, estimated from the width of the (1 1 0) peak, decreased monotonically with N content from typically 100 nm in the case of N-free films to <10 nm for films containing 8 at% N. The uniaxial in-plane anisotropy H_k, induced by the magnetic field during the deposition, increased from 5 to 20 Oe when the N-content increased from 5 to 8 at%, while coercive field H_c was about 2–10 Oe in this composition range [6].

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XRD, as well as conventional TEM and selected area diffraction (SAD), reveal a 10–30 nm size of crystallites for most of the investigated sputter deposited films, see, for example, Fig. 1a. Cross-sectional TEM, Fig. 1b, reveals a columnar structure in the films tested.

In Figs. 2 and 3, some results of the LTEM investigation in the out-of-focus (Fresnel) mode [7] are presented. A ripple structure, corresponding to a local variation of the magnetization vector on a submicron scale is visible. The ripple patterns are perpendicular to the local magnetization vector, shown by arrows. A cross-tie wall is illustrated in Fig. 2b. The singularities in the domain boundary correspond to the Bloch lines, a narrow region where the magnetization rotates out of the plane of the film [5]. As illustrated by the figure, circular Bloch lines, where the magnetization is rotating around the Bloch line, are followed by cross-Bloch lines, where the magnetization is directed either in or out of the singularity.

Due to a residual magnetic field in the sample region, there will be an in-plane magnetic field component, which magnetizes the sample when the sample is tilted in the microscope. Tilting around the hard direction perpendicular to the cross-tie wall forces the circular and cross Bloch lines to approach each other and annihilate. Tilting around the easy direction causes bending of the domain walls and a number of closely spaced circular- and cross-Bloch lines in cross-tie walls appear as shown in Fig. 3a, where the sample is tilted by 2.8° in the hard direction. Finally, at a certain tilt angle, corresponding to a critical value of the magnetizing field, the domain walls are "washed out", leaving the sample as a single domain, Fig. 3b. Nevertheless, the magnetic ripple microstructure persists. Small holes, present in the film prepared for TEM, serve as pinning points for such

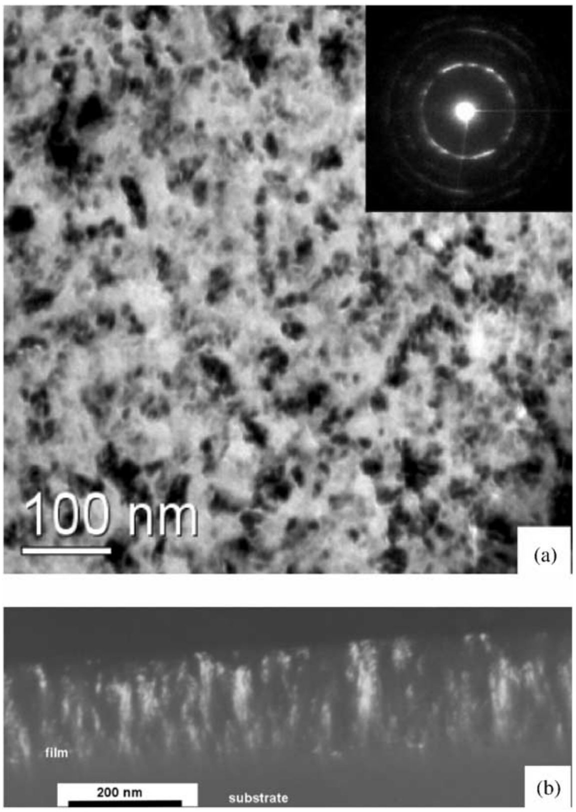

Fig. 1. In-plane (a) and cross-sectional TEM (b) images of a sputter-deposited FeZrN film.

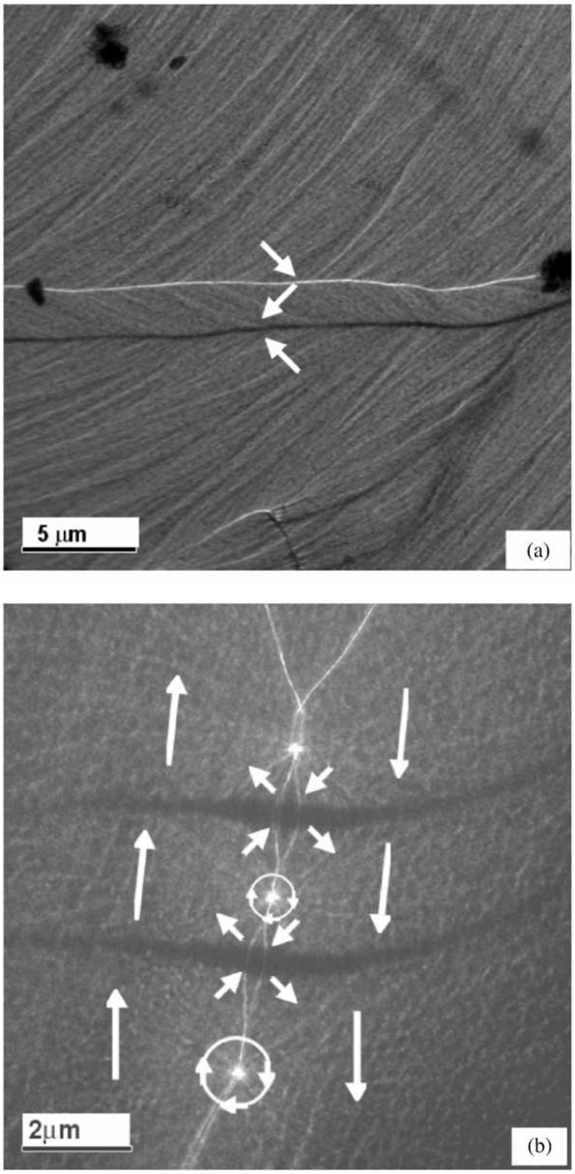

Fig. 2. Magnetic microstructure of sputtered films. (a) A 90°-domain structure with a ripple pattern. (b) Cross-tie wall with alternating circular and cross Bloch lines.

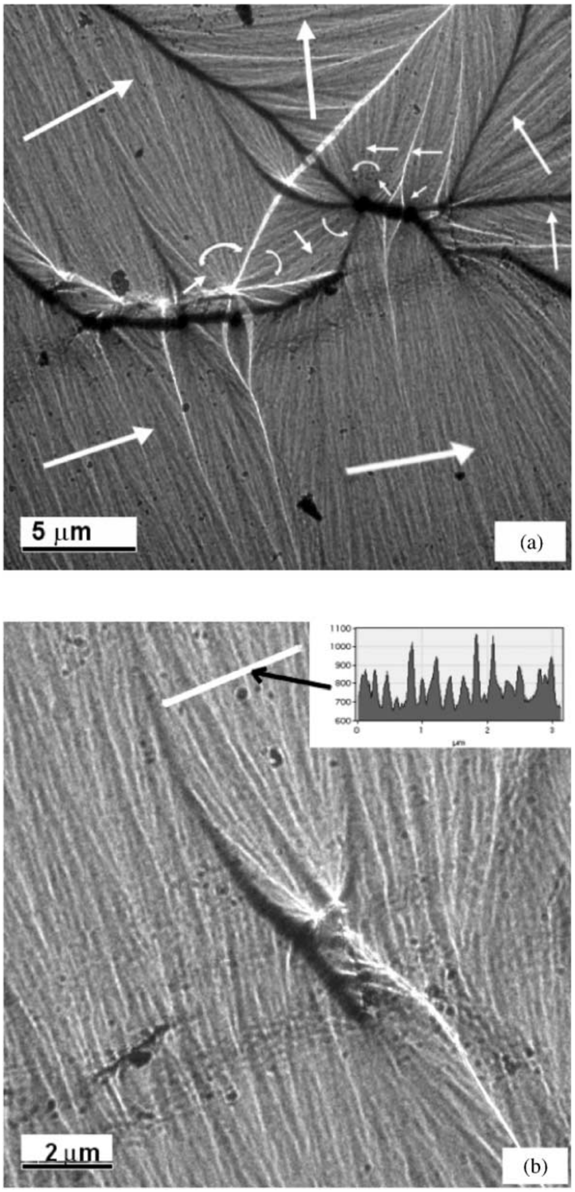

Fig. 3. (a) LTEM image for a tilt angle of 2.8°, with the tilt axis parallel to the easy axis. (b) At a certain critical tilt, the domain walls disappear but ripple structure remains. The ripples are pinned by defects (holes, cracks). The ripple contrast variation is shown in the inset.

ripples, which rotate around these holes during the magnetization, Fig. 3b.

Analysis of the image intensity variation outside the singularities, as shown in the inset of Fig. 3b, shows that the average wavelength of the magnetization ripples is about $\lambda = 200 \pm 20$ nm. The uncertainty in λ is, in fact, somewhat larger, because of the influence of the defocusing conditions, thus we take it as a rough estimate which serves the current purpose. This estimate is significantly smaller than the value $\approx 2\,\mu$m obtained by Hale and Fuller for Permalloy film [7]. The theory of Hoffmann [8] relates the wavelength with the exchange constant A and uniaxial anisotropy constant K_u: $\lambda_T = 2\pi(A/K_u)^{1/2}(H/H_k \pm 1)^{-1/2}$, where H is the external field in the plane of the film, H_k is the anisotropy field and + or − is to be taken for the case of the applied field parallel or perpendicular to the easy axis, respectively. Taking $(H/H_k \pm 1) = 2$ for the case illustrated by Fig. 2b, $A = 1.5 \times 10^{-6}$ erg/cm, $K_u = M_s H_k/2 \approx 3.75 \times 10^4$ erg/cm^3 (for $M_s \approx 1.5 \times 10^4$ Oe and $H_k \approx 5$ Oe) we obtain $\lambda_T \approx 280$ nm. The discrepancy with the experimental value 200 ± 20 nm could be assigned to the reason mentioned above and also to the uncertainties of the parameters used in the estimation of λ_T.

Applying the treatment of Wohlleben [9], we obtain for the average deviation of the local magnetization from the overall mean direction $\varphi \approx 0.59 \times 10^5 K/(Bt\lambda) \approx 0.6°$, where B is the magnetic induction (in T) in the film, t is the thickness of the film (in nm), λ is in nm. The magnitude of the contrast is determined as $K = 2\Delta I/I_0$, where I_0 is the average intensity of the image, ΔI is the rms variation of the intensity due to ripples. For the investigated film, the values $B = 1.5$ T, $t = 75$ nm and $\lambda = 200$ nm were applied. For the mean deviation angle Hoffmann suggested [8] $\varphi \approx DK/(4\pi^{1/2}(2t)^{1/4}(M_s^{1/2}(AK_u(H/H_k + 1))^{3/8})$. Using this theory with the parameters listed above and the grain size $D = 15$ nm, $K = 4.7 \times 10^5$ erg/cm^3, we obtain $\varphi \approx 1.5°$, which is 2.5 times larger than the experimental one 0.6°. The discrepancy could be assigned to the same reasons as for the wavelength.

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