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Influence of stresses and magnetostriction on the soft magnetic behavior of metallic films

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Abstract

Nanocrystalline soft magnetic Fe–Zr–N films have been successfully deposited by DC magnetron reactive sputtering. For thick films (>200 nm), the compressive stress in the as-deposited films and the positive magnetostriction produce perpendicular anisotropy. The magnitude of this effect is smaller than the value obtained from the magnetic parameters indicating that in these films other sources of perpendicular anisotropy like columnar structure or texture play an important role. For thin films (<200 nm) with in-plane orientation of the magnetization, we have found a surprising difference between the magnitude of the induced uniaxial anisotropy in Fe–Zr–N and Fe–Co–Ta–N materials. This effect is not due to a difference in the magnitude of the magnetostriction or the strain. A possible explanation for the effect is a difference in the electronic configurations in the two materials.
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1. Introduction

A ferromagnetic film can be used as magnetic flux amplifier in ultra-high frequency applications if it has a simple magnetic domain structure, with the magnetization in all points of the film oriented into the same direction [1]. Often the magnetic domain structure in thin films is far from such a simple situation. Due to magnetoelastic anisotropy created by stresses, complicated magnetic domain structures can appear with a negative influence on the soft magnetic behavior of the film. Therefore, determination of the saturation magnetostriction and the characterization of the state of stress of the

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films are very important in the process of optimizing the performance of such films in devices.

For continuous thin films (thinner than about 200 nm) of soft magnetic alloys, the shape anisotropy dominates and the magnetization is practically always oriented in the plane of the film. Such a film can have an almost ideal domain structure if the magnetocrystalline anisotropy is negligible and an in-plane uniaxial magnetic anisotropy is present. This situation is realized in nanocrystalline films with induced anisotropy. When the films are thicker than 200 nm, a perpendicular anisotropy can appear leading to remarkable stripe domain structures [2].

The presence of interstitial atoms in the lattice produces strain and consequently stress. Therefore, the induced uniaxial anisotropy by interstitial atoms is another phenomenon that might be influenced by the magnetoelastic phenomenon.

The paper presents the determination of the magnitude of the magnetostriction and stress in sputtered Fe–Zr–N and Fe–Co–Ta–N films and the study of the influence of the magnetoelastic phenomenon on their magnetic properties.

2. Experimental

Fe–Zr–N and Fe–Co–Ta–N films with a thickness between 50 and 1000 nm were deposited by DC magnetron reactive sputtering. Different substrate temperatures were used in the range between −60 and 200 °C. The targets were pure (99.96%) Fe sheets covered with Zr wires or alloys of $Fe_{70}Co_{30}$ covered with Ta chips. For the depositions at room temperature (RT) of the substrate, a number of growth rates and Ar/N_2 ratios were used. When the deposition temperature was varied, the sputtering mixture contained 7 and 16 vol% N_2, respectively. An 800 Oe magnetic field was applied in the plane of the substrate during deposition. The composition and thickness of the films were determined by Rutherford backscattering (RBS) and elastic recoil detection (ERD). The phases, lattice spacing and nitrogen content were determined by standard X-ray diffraction (XRD) measurements. Nitrogen atoms, randomly dis-

solved in the octahedral interstices of the α-Fe phase, produce lattice expansion. This expansion is proportional to the N content [3]. Consequently, the N content can be estimated from the value of the lattice parameter of planes parallel to the surface of the sample. Hysteresis loops were recorded using a vibrating sample magnetometer (VSM). For samples with well-defined uniaxial anisotropy, the anisotropy field was considered to be equal to the saturation field in the hard axis direction. In the other cases, the anisotropy field was obtained using the distribution function of the anisotropy determined by the method described in Ref. [4]. The magnetostriction and stresses were determined using a curvature-measuring device.

3. Results and discussions

3.1. Magnetostriction and stress

The values of the saturation magnetostriction as a function of the N content for films with different compositions are presented in Fig. 1. The Zr-containing films have a magnetostriction which is positive for N concentrations higher than 5 at%. A positive magnetostriction means that the length of the specimen in the magnetized state is larger than that corresponding to the demagnetized state. At an N-content of 15 at%, the magnetostriction in
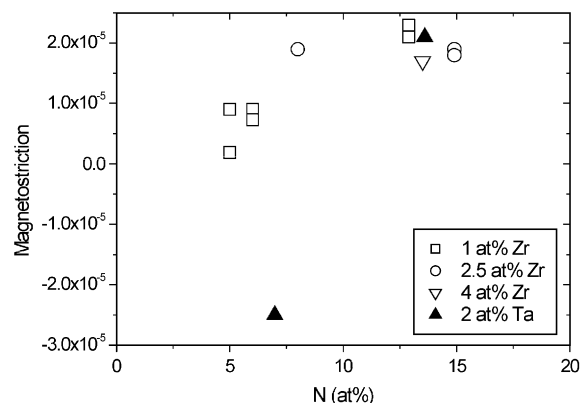

Fig. 1. Dependence of the saturation magnetostriction on the N content for $(Fe_{1−X}Zr_X)_{1−Y}N_Y$ and $((Fe–Co)_{1−X}Ta_X)_{1−Y}N_Y$ films. X is given in the legend. Y is given by the abscissa.

these films is of the order of 2×10^{-5} and varies little with the Zr content.

At 7 at% N, the Ta-containing films have a negative magnetostriction. The magnetostriction of these films becomes positive and practically equal to that of the Zr-containing films at N contents of the order of 15 at%.

Stress measurements show that all sputtered films are under a state of compressive biaxial stress with a magnitude in the 300–600 MPa range. There is no systematic correspondence between the magnitude of the stress and the deposition parameters.

3.2. Stripe domains and induced anisotropy

3.2.1. Perpendicular anisotropy

When the thickness of the sputtered films with a high N content surpasses 200 nm, degradation of the soft magnetic behavior occurs, leading to a complete disappearance of the soft magnetic properties. For such films, the typical shape of the hysteresis loop (Fig. 2) indicates the formation of so-called stripe domains. This particular magnetic domain structure, described in detail in Ref. [2], is typical for films with perpendicular anisotropy.

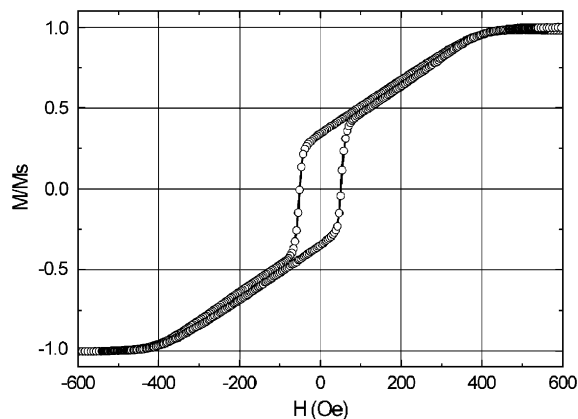Perpendicular magnetic anisotropy in sputtered films might be caused by microshape anisotropy,

Fig. 2. Hysteresis curve of a 400 nm thick Fe–Zr–N film showing perpendicular anisotropy. The coercive field is 60 Oe and the film saturates at 400 Oe.

magnetocrystalline anisotropy and/or magnetoelastic anisotropy [5]. The microshape anisotropy appears as a result of columnar structure of the film when the columns are separated by a nonmagnetic phase or voids. For nanocrystalline films with random oriented grains, usually the magnetocrystalline anisotropy is averaged out [6]. However, for films with a specific crystallographic texture, a net magnetocrystalline anisotropy may remain. Generally speaking, any of the three mechanisms may be dominant, depending on the particular process, and needs to be carefully identified. The aim of this section is to estimate the magnetoelastic contribution to the perpendicular magnetic anisotropy and to compare this contribution with the effective perpendicular anisotropy extracted from hysteresis loops like that presented in Fig. 2.

Via the magnetoelastic coupling, stresses produce magnetoelastic anisotropy in thin films [7]. The stress and the stress-induced magnetic anisotropy have the same direction. The sign of the induced anisotropy depends on the sign of the magnetostriction and on whether the stress is tensile or compressive. As an example, if a film is under uniaxial compressive stress and the magnetostriction is positive, the in-plane anisotropy is reduced. If this reduction is sufficiently large, magnetic anisotropy in a plane perpendicular to the direction of the stress is promoted. Following the same reasoning, one can easily predict that the effect of in-plane biaxial stresses and positive magnetostriction is a perpendicular anisotropy.

The magnetoelastic anisotropy energy constant K can be calculated using the formula:

$$K = -\tfrac{3}{2}\sigma\lambda,$$

where σ is usually taken to be the largest component of the biaxial stress and λ is the saturation magnetostriction. Using 2×10^{-5} for the saturation magnetostriction (Fig. 1), it can be estimated that in the films the magnitude of the stress-induced perpendicular anisotropy is in the range from 9 to 18 kJ/m^3.

The magnitude of the perpendicular anisotropy that governs the magnetic domain structure in the films can be estimated from hysteresis loops like the one presented in Fig. 2. The perpendicular

anisotropy constant ($K_\perp$) is given by the formula [8]:

$$K_\perp = \frac{M_S H_S}{2[1 - (t_C/t)^{2/3}]},$$

where H_S is the saturation field, M_S is the saturation magnetization, t_C is the critical film thickness and t is the film thickness. For the magnetic film presented in Fig. 2, one obtains with $H_S = 400\,\text{Oe}$ ($\approx 32\,\text{kA/m}$), $M_S = 17\,\text{kG}$ (1.7 T), $t_C = 200\,\text{nm}$ and $t = 400\,\text{nm}$ a value for $K_\perp = 73\,\text{kJ/m}^3$. This value is larger than the contribution given by the magnetoelastic effect, indicating that in this film the other two contributions to the perpendicular anisotropy are dominant.

3.2.2. In-plane anisotropy

For films with a thickness smaller than 200 nm, deposition at low temperature (below RT) leads to films with excellent soft magnetic properties. Both Fe–Zr–N and Fe–Co–Ta–N films obtained in this fashion are nanocrystalline and present a very low value for the coercive field ($<1\,\text{Oe}$). At the same time, these films have high values of the induced in-plane anisotropy (H_K), 26 Oe and, respectively, 48 Oe (Figs. 3 and 4).

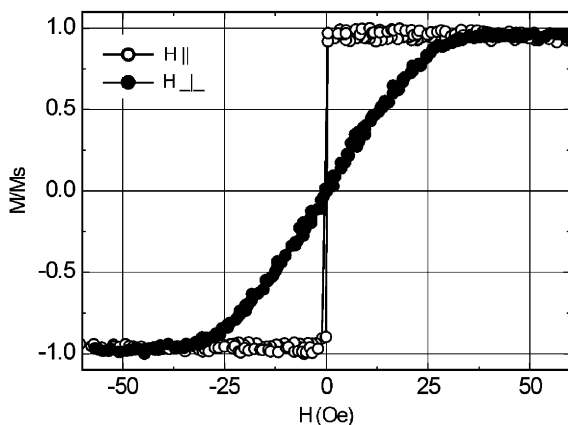The induced uniaxial anisotropy in Fe–Zr–N films originates from an anisotropic distribution of

Fig. 3. Easy axis and hard axis hysteresis loops for a $Fe_{81}Zr_2N_{17}$ film with the highest value of the induced anisotropy ($H_K = 26\,\text{Oe}$).
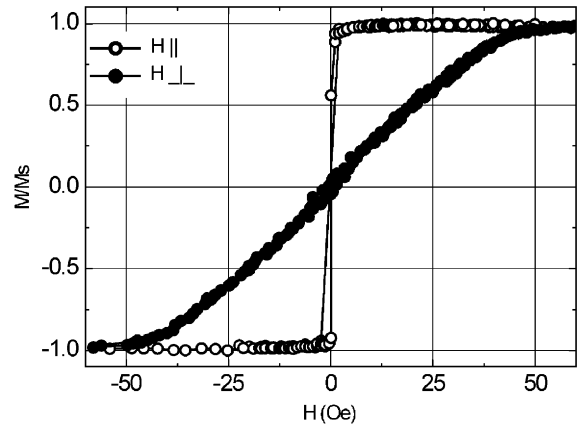
Fig. 4. Easy axis and hard axis hysteresis loops for a $Fe_{58.3}Co_{25}Ta_{1.7}N_{15}$ film with the highest value of the induced anisotropy ($H_K = 48\,\text{Oe}$).

interstitial N atoms in the octahedral interstices of the Fe lattice [1]. For the Fe–Co–Ta–N system, the same observation holds. Two arguments support this statement. First, if the film contains no N, Fe–Co–Ta films do not present detectable induced anisotropy. This is somewhat surprising, since in films containing two atomic species (e.g. Fe–Ni), deposition under the influence of a magnetic field can create induced uniaxial magnetic anisotropy [7]. Second, in the N-containing films, the induced anisotropy can be reoriented in the plane of the films by thermo-magnetic treatments at relatively low temperature of 200 °C. After such a treatment, the magnitude of the induced anisotropy is preserved. As illustrated in Ref. [1], this fact can only be explained by a reorientation of N atoms in the lattice. Other reconstructions of the lattice are not expected to occur at such low temperature.

The microscopic source of the induced anisotropy is the N-filled interstitial octahedron (Fig. 5). The films presented in Figs. 3 and 4 have practically the same values of the N content and the same saturation magnetization, but the anisotropy field is almost double in the Fe–Co–Ta–N film as compared to the Fe–Zr–N film. The same relationship was observed at 7 at%. In materials containing Fe, the anisotropy energy per N atom is practically independent of the N concentration and it is of the order of 40 meV/atom [9]. In order
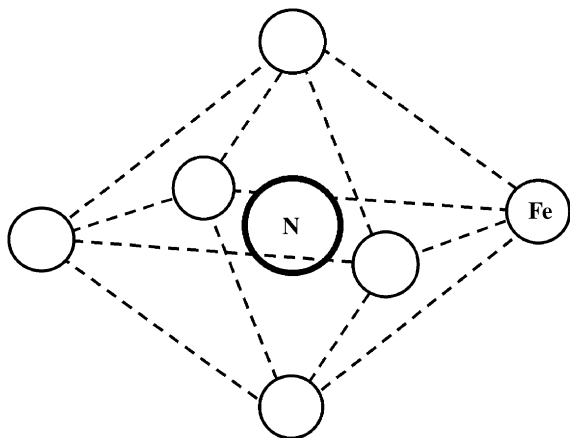
Fig. 5. Schematic representation of a N-filled octahedral interstice in Fe.

to explain the magnitude of the effect in the Fe–Co–Ta–N material, this quantity should be almost two times larger. The question is if this effect can be explained by differences in the magnitude of the magnetoelastic interaction.

The octahedral site is formed by six Fe atoms. Four atoms are situated in a $\{1\,0\,0\}$ plane at a distance $a/\sqrt{2}$ from the center of the octahedron and two Fe atoms at a distance $a/2$ on an $\langle 1\,0\,0 \rangle$ axis perpendicular to the $\{1\,0\,0\}$ plane, the octahedron axis (Fig. 5). These octahedral interstitial sites have a tetragonal symmetry and their shape and volume depend on the N content dissolved in the lattice [3]. As demonstrated in Ref. [9], in nanocrystalline materials the extra space required for accommodation of the N atoms is obtained by an overall lattice dilation. Consequently, the shape of the octahedron remains the same but the volume increases with increasing the N content. The associated changes in strain suggest that the anisotropy might be related with the phenomenon of magnetoelasticity.

From XRD measurements, we found that the dilation of the lattice in the two materials differs by less than 10% for the same N content. The strain produced by the presence of N in the lattice is almost the same in the two systems. From Fig. 1 one can see that the magnetostriction of the two samples is practically equal. Therefore, the mag-

netoelastic energy in the two alloys cannot be very different.

From the values of the composition of the Fe–Co–Ta–N material, it can be easily estimated that for each octahedral interstitial site one or two atoms forming the octahedron is Co while, in the Fe–Zr–N material the octahedrons are formed almost exclusively by Fe atoms. A difference in hybridization of the orbitals in the Fe–Co–N compound compared to the Fe–N compound may offer an alternative explanation for the difference of the in-plane anisotropy between the two materials.

4. Conclusions

Magnetostriction and stress are two important phenomena which influence the magnetic domain structure of sputtered films. For thick films ($>200\,\text{nm}$), the perpendicular component of the anisotropy is influenced by the magnetoelastic effect. The compressive stress in the as-deposited films and the positive magnetostriction produce perpendicular anisotropy. The magnitude of this effect is smaller than the value obtained from the magnetic parameters indicating that in these films other sources of perpendicular anisotropy like columnar structure or texture play an important role. For thin films ($<200\,\text{nm}$) with in-plane orientation of the magnetization, we have found a surprising difference between the magnitude of the induced niaxial anisotropy in Fe–Zr–N and Fe–Co–Ta–N materials. This effect is not due to a difference in the magnitude of the magnetostriction or the strain. A possible explanation for the effect is a difference in the electronic configurations in the two materials.

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