Irreversible magnetization reversal in some Co-based alloy thin films

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Irreversible magnetization reversal occurs either by coherent or incoherent spin rotation or by wall displacement. In electrodeposited Co-W, Co-Fe, and Co-P 300–500-Å films, vibrating sample magnetometer hysteresis loop analyses indicate that magnetization reversal takes place by wall displacement. The formation and movement of domain walls has been put in evidence by Lorentz electron microscopy.

I. INTRODUCTION

The use of continuous metallic thin films as the recording medium in magnetic memory devices has been the subject of numerous papers and reviews. Beside pure element coatings, a great number of binary, ternary, and higher alloy thin films have been prepared by various plating techniques. Many of these films display metastable structures and possess a variety of highly interesting properties. Among the many electrodeposits Co-P and Co-Ni-P have been the most widely investigated. Indeed, the first commercial thin-film metallic magnetic disks were fabricated on the basis of these two alloys. 

For longitudinal recording, the minimal transition width (namely, the width of the region between oppositely magnetized adjacent bits) is given by

\[ l_{\text{min}} = m_t H_c, \]

where \( m_t \) is the in-plane remanent magnetization, \( t \) the layer thickness, and \( H_c \) the coercivity. Consequently, the recording density may be increased by the use of films having lower remanence, lower thickness, or higher coercivity. Electrodeposited Co-W and Co-P films fall within the category of high remanence and medium to high coercivity (of the order of a few hundreds Oe) systems.

The nature of the irreversible magnetization reversal which takes place within the recording medium under the influence of an external magnetic field is of practical significance. Magnetization reversal by wall displacement is 2–4 orders of magnitude slower than magnetization reversal by any mode of coherent or incoherent rotation (such as curling, buckling, or fanning). The nature of the irreversible reversal process can be determined by the so-called macroscopical approach [a vibrating sample magnetometer (VSM) hysteresis loop analysis] and by microscopical observation of the domain structure by Lorentz electron microscopy.

In the present work, three cobalt-based alloy thin-film electrodeposits, namely Co-W, Co-P, and Co-Fe, were studied. VSM analysis was applied to determine the nature of the irreversible magnetization reversal process. The existence and movement of domain walls were shown by Lorentz electron microscopy.

II. EXPERIMENT

A. VSM measurements

In ferromagnetic thin films, the nature of the irreversible magnetization reversal (its occurrence by wall displacement or by any mode of coherent or incoherent rotation) can be determined by a simple experiment. The experiment is aimed at measuring the critical field, \( H_{\text{crit}} \), which is the coercive field required to induce the process, as a function of the angle \( \alpha \) between the field direction and the plane of the film (Fig. 1).

Regardless of the exact details of the mechanism, or of the spin distribution within a wall, \( H_{\text{crit}} \) is proportional to \( 1/ \cos \alpha \) whenever the irreversible magnetization reversal occurs by wall displacement. This behavior is due to the fact that the pressure exerted by the external magnetic field on a domain wall which is pinned to some obstacles hindering its motion is proportional to \( M_s \cos \alpha \), where \( M_s \) is the magnetization within the domains. On the other hand, when coherent rotation is the dominant mechanism, \( H_{\text{crit}} \) is structure and texture dependent and varies with \( \alpha \) between a minimal value \( H_0 \) and twice this value. For uniaxial materials, like hexagonal cobalt, \( H_0 \) is given by \( K/M_s \), where \( K \) is the magneto-crystalline anisotropy constant and \( M_s \) the saturation magnetization. In multiaxial materials, like cubic cobalt, \( H_0 \) obeys a similar rule. It follows that a measurement of the angular dependence of \( H_{\text{crit}} \) provides evidence regarding the
actual mechanism operating within the films.

The measurements were carried out using a Foner-type VSM at fields of 0–10 kOe at Twente University, as described elsewhere. For the three alloy systems studied, samples with a high in-plane coercivity were chosen for the analysis. For each sample, ten hysteresis loops were measured, $\alpha$ ranging from 0° to 90° in 10° increments.

B. Lorentz electron microscopy

The Lorentz electron micrographs were obtained using the out-of-focus (Fresnel) method in a JEOL JEM-7A transmission electron microscope operated at 100 kV. The conical specimen holder of the top-entry goniometer stage of the microscope was modified so that the specimen was raised about 5 mm above its normal position in the objective lens. By varying the excitation of the objective lens, the in-plane component of the magnetic field could be changed from values having a negligible interference with the existing domain structure to values high enough to induce domain nucleation and wall displacement.

C. Sample preparation

Three cobalt-based alloy thin films, namely, Co-W, Co-Fe, and Co-P, were studied. The films (300–500 Å thick) were electrodeposited onto copper-coated microscope slides. The details of the deposition process are described elsewhere. Basic and acidic baths at various compositions, pH values, and temperatures were used. Prior to the vacuum deposition of the copper, a thin layer of Formvar (polyvinyl formal) was applied to the slides. This was done in order to avoid the peeling off of the film during electrodeposition and to enable the separation of the coating from the slides for the TEM study.

The specimens for the magnetic measurements were 1 × 1-cm$^2$ squares cut from the slides after deposition. The squares were then attached to the sample holder of the VSM by means of a two-sided adhesive tape.

The samples for the transmission electron microscope were prepared by first scratching 2 × 2-mm$^2$ squares on the coated glass and then dissolving the Formvar and the copper substrates as described elsewhere.

III. RESULTS

A. Magnetic measurements

A typical set of hysteresis loops for a high in-plane coercivity Co-W film, with $\alpha$ varying from 0° to 80° in 10° increments, is shown in Fig. 2. From the loops, $H_{\text{en}}(\alpha)$ was obtained. $H_{\text{en}}(\alpha)/H_{\text{en}}(0°)$ for Co-W, Co-Fe, and Co-P is plotted in Fig. 3. The solid curves stand for the function $1/\cos\alpha$. The agreement between the measured values and the $1/\cos\alpha$ function clearly indicates that in the three systems wall displacement is the dominant mechanism responsible for the irreversible magnetization reversal. The solid curve is the function $1/\cos\alpha$, indicating that in the absence of an external field $m$, is parallel to the film plane. This behavior is accounted for by the very large magnetostatic energy associated with free magnetic poles on the film surface.

Figure 5 is a plot of the relative hysteresis loss $W_H(\alpha)/W_H(0°)$ (where $W_H = \int H dM$) obtained from the hysteresis loops of Fig. 2, as a function of $\alpha$. For $\alpha = 0°–60°$ the measured values are very close to unity, which is the theoretical value obtained by assuming magnetization reversal by domain wall displacement. At angles above 60° the curve decreases steeply. A similar behavior was observed in the case of Ni sheets. This effect may be attributed to the increasing influence of the demagnetizing field, which becomes more pronounced at higher angles and at lower film thickness.

B. Lorentz electron microscopy

Figure 6 shows the nucleation and growth of magnetic domains in a cobalt film, induced by the magnetic field of the objective lens of the microscope (the black and white contrast of the domain walls is inherent to the method of observation). Within certain limits of the field, this process is reversible: a domain at A appeared when the field was increased and disappeared when the field was decreased [Figs. 6(a) and 6(b)]. Upon further increase of the field, these tongue-shaped domains widened at the expense of the rest of the film until they coalesced to form a sawtooth domain wall structure [Figs. 6(c) and 6(d)]. It is noteworthy that defects like inclusions [A in Figs. 6(a) and 6(b)] act as pinning points for the domain walls, thus hindering their motion. Had it been possible to increase the objective lens field sufficiently, full saturation would have been achieved, the domains favored by the field taking over the entire sample area.

IV. DISCUSSION

An electron microscopy study of deposits of the three systems (Co-W, Co-Fe, and Co-P) revealed that the depos-
FIG. 3. Normalized critical field $H_{\text{crit}}(\alpha)/H_{\text{crit}}(0^\circ)$ for Co-W, Co-Fe, and Co-P thin electrodeposits. The solid lines show the function $1/\cos \alpha$.

FIG. 4. In-plane orientation ratio $\text{OR}_1(\alpha) = m_x(\alpha)/m_x(0^\circ)$. The solid curve is the function $1/\cos \alpha$. In the isotropic case, $\text{OR}_{\text{iso}} = 1$ (dotted line).

FIG. 5. In-plane hysteresis loss $W_{\text{HIG}}(\alpha)/W_{\text{HIG}}(0^\circ)$. The theoretical value for domain wall displacement is 1 (dotted line).

FIG. 6. Nucleation of magnetic domains in Co-P electrodeposits is induced by the magnetic field of the objective lens. The magnetic domains appear when the field is increased (b) and disappear when the field is decreased (a). When the field is further increased, domains grow and coalesce [(c), (d)] to form a sawtooth wall pattern.
its 12,18-20 is associated with this range of grain sizes, 21 as well as to wall pinning effects.

The wall displacement mechanism for magnetization reversal seems inconsistent with the observation of the medium containing particles individually too small to allow the formation of walls. The apparent inconsistency may be reconciled by assuming that the magnetic coupling between adjacent grains bridges the discontinuity caused by the grain boundaries and enables the formation of walls which extend through several grains across their width.

Figure 6 illustrates the nucleation and growth of domains, and the occurrence of magnetization reversal by the wall displacement mechanism. This observation supports the conclusion based on the macroscopic measurements.

Evidently, the domain structure depends on the grain morphology, crystallography and texture. As described in detail previously, 13-15 by a proper choice of the deposition variables it is possible to control these structural parameters and obtain, for instance, deposits with a varying degree of texture perfection (the c axis of the hexagonal crystallites pointing in a direction normal to the film plane). Figure 7(a) shows the magnetic structure of a film having a strong perpendicular texture, and Fig. 7(b) of a film with a low degree of texture. In the former case the magnetization seems to be perpendicular to the film plane, while in the latter it is parallel to the film plane. Similar observations were made by Hughes 22 in Co-P and by Lodder and Wielinga 23 in sputtered Co-Cr films.

V. CONCLUSION

The wall displacement mechanism rather than coherent rotation is responsible for the irreversible magnetization reversal in electrodeposited Co-W, Co-Fe, and Co-P thin films. This was established by measurements of $H_{\text{rem}}(\alpha)$ and of the in-plane hysteric loss, as well as by Lorentz electron microscopy. Magnetic coupling between adjacent grains enables the formation of walls, which may be thicker than the size of the individual grains present within the films. The high coercivity observed in the films is associated with grain size and wall pinning effects.

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FIG. 7. Influence of film texture on the domain structure of a Co-P film: (a) strong perpendicular c-axis texture, (b) low degree of texture. The clear distance between adjacent grid bars is approximately 100 μm.