Photoemission electron microscopy study of remanent magnetic domain states in ferromagnetic wedge films deposited on substrates with micrometer-sized square plateaus

L. J. Heyderman,^{a)} S. Czekaj, F. Nolting, and E. Müller
Paul Scherrer Institut, 5232 Villigen PSI, Switzerland

P. Fischer
LBNL/CXRO, 1 Cyclotron Road, Berkeley, California 94720

Ph. Gasser
EMPA Materials Science and Technology, Überlandstrasse 129, 8600 Dübendorf, Switzerland

L. López-Díaz
Departamento de Física Aplicada, Universidad de Salamanca, Plaza de la Merced, Salamanca 37008, Spain

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We present a detailed study of the remanent magnetic domain configurations in demagnetized polycrystalline ferromagnetic thin film wedges of cobalt and Permalloy deposited on prepatterned silicon substrates with micrometer-sized square plateaus, which have a height of 125 nm, using photoemission electron microscopy. We have observed the continuous evolution of the magnetic domain states in the square ferromagnetic elements on top of the plateaus as a function of film thickness. At high film thickness we observe the Landau state, which is the expected lowest energy state, but at lower thickness we see a variety of metastable states which are trapped as a result of local pinning. In a small thickness range below 10 nm, the square elements contain 360° walls and small domains which are likely to be a result of local effects such as magnetocrystalline anisotropy and edge roughness. We are able to simultaneously observe the development of the magnetic domains in the continuous polycrystalline film surrounding the plateaus and, rather than the expected large domains, we observe at intermediate film thickness a significant modification of the domain configuration to small domains. Here the roughness of the silicon substrate surrounding the plateaus, which is due to the reactive ion etching process used to prepare the prepatterned substrates, gives rise to local stray fields in the ferromagnetic film which play an important role in determining the resulting domain structure. © 2006 American Institute of Physics. [DOI: 10.1063/1.2174119]

I. INTRODUCTION

The study of magnetic spin structures in small magnetic elements has been a key area of scientific focus for more than a decade and has been facilitated by the development of lithography methods for patterning magnetic thin films.¹ Small magnetic elements are not only of scientific interest for the study of magnetic spin structures in laterally confined systems but are also of paramount importance for technological applications such as ultrahigh density information storage and sensor devices.^{2–6}

Square and rectangular elements are of particular interest as model systems because their basic fourfold symmetry results in simple domain states which have been simulated^{7–14} and experimentally observed using various magnetic imaging methods.^{15–20} The magnetic domain configurations present are predominantly determined by the stray field energy associated with the shape boundaries, and a number of key states have been identified which fall into two categories. The first category contains flux-closure states, including the Landau (or vortex) state [Fig. 1(a)] and the diamond state [Fig. 1(b)], which has a slightly higher energy than the

Landau state due to the additional exchange energy associated with additional domain walls and vortices. The second category contains states with a high remanence and incomplete flux closure. These states are found in smaller, thinner elements which have a reduced stray field energy and do not include vortices because of the high exchange energy associated with them. These states include the *C* state [Fig. 1(c)], the *S* state [Fig. 1(d)], and the flower state [Fig. 1(e)]. The final magnetic state observed for a given element depends on the size of the element, film thickness, and the magnetic material, i.e., its magnetization at saturation, exchange length, and the magnetic anisotropies present. In polycrystalline systems the effect of the magnetocrystalline anisotropy is dependent on the size and orientation of the individual crystallites and can be observed as magnetization ripple.

In the current work, we present a detailed study of the magnetic domain configurations in polycrystalline ferromagnetic thin film wedges of cobalt and Permalloy deposited on prepatterned silicon substrates with micrometer-sized square plateaus using photoemission electron microscopy (PEEM). This allows us to observe the continuous development of magnetic domain states in the micrometer-sized square ele-

^{a)}Electronic mail: laura.heyderman@psi.ch

FIG. 1. Schematic diagrams of the expected PEEM contrast for magnetic configurations in ferromagnetic square elements. The details of the spin structures which occur in real elements, and therefore the measured contrast, will be dependent on the square size, thickness, and material properties.

ments on top of the plateaus as a function of film thickness. In addition, we are able to simultaneously observe the development of the magnetic domains in the continuous film surrounding the plateaus.

II. EXPERIMENT

Prepatterned silicon substrates with square plateaus were produced by electron beam lithography. First, rows of micrometer-sized squares, with center-to-center distances of $8\ \mu\text{m}$ and edge lengths of 4 , 2 , and $1\ \mu\text{m}$, were exposed in a polymethylmethacrylate resist spin coated on a naturally oxidized silicon (001) substrate using a Leica LION LV1 electron beam writer.²¹ These patterns were transferred into a chromium metal hard mask by evaporation of the chromium onto the electron beam resist followed by lift off in acetone. The pattern was then transferred from the chromium hard mask into the silicon by reactive ion etching using a $\text{CHF}_3/\text{O}_2/\text{SF}_6$ plasma. The substrates were etched to give a plateau height of $125\ \text{nm}$ and, in a final step, the chromium hard mask was removed using a chlorine plasma etch. The top surface of the plateaus is smooth but the silicon surface in the valleys is relatively rough due to the plasma etching [Fig. 2(a)]. An estimation of the roughness from high resolution scanning electron microscope (SEM) images gives the height, width, and center-to-center distance of the small silicon hills of 15 , 15 , and $40\ \text{nm}$, respectively.

Two different ferromagnetic wedge films were investigated; Permalloy [$\text{Ni}(83\%) \text{Fe}(17\%)$], deposited by thermal evaporation (base pressure of 4×10^{-6} mbar), and cobalt, deposited by dc sputtering (deposition at 1.4×10^{-4} mbar). The wedge was created by moving a shutter across the substrate during deposition. The saturation at magnetization of the films was determined from hysteresis loops measured using a

FIG. 2. High resolution SEM images of (a) a $1\ \mu\text{m}$ square silicon plateau and then with a cobalt layer deposited onto the plateau with thicknesses of (b) $40\ \text{nm}$, (c) $110\ \text{nm}$, and (d) $240\ \text{nm}$. The sample is tilted at 45° .

FIG. 3. XMCD images of a cobalt wedge deposited on a prepatterned silicon substrate with rows of square plateaus with 4, 2, and 1 μm edge lengths. The square number (in black) and the cobalt thickness in nm (in white) are indicated.

vibrating sample magnetometer and found to be 880×10^3 A/m for Permalloy and 1300×10^3 A/m for cobalt, which are comparable to the literature values.²² The preferential growth of the magnetic films on the top surface of the silicon substrate rather than on the plateau edges and self-shadowing effects resulted in the isolation of the square magnetic elements sitting on top of the plateaus from the continuous film in the valleys, as observed with a high resolution SEM and by transmission electron microscopy. The gap between the upper and lower magnetic films is clearly observed up to a film thickness of $t=140$ nm [see Figs. 2(b) and 2(c)], and even up to 240 nm [Fig. 2(d)] there still appears to be some separation. The thickness of the ferromagnetic wedges was determined from high resolution SEM images of the magnetic film cross sections.

The magnetic imaging was carried out at the SIM beamline²³ at the Swiss Light Source, Paul Scherrer Institut, with an Elmitec PEEM.²⁴ Employing x-ray magnetic circular dichroism (XMCD), the magnetic domains in cobalt or Permalloy are imaged by tuning the x-ray energy to the Co or Ni L_3 edge, respectively. Dividing two images taken with the left and right circular polarized light leads to an XMCD image where the intensity is a measure of the angle between the circular x-ray polarization vector, or magnetization sensitivity direction, and the magnetic spins in the domains.²⁵ Ferromagnetic domains with magnetic spins parallel or antiparallel to the polarization vector appear black or white in the XMCD image, and oppositely magnetized domains with magnetic spins perpendicular to the polarization vector will both have the same gray contrast. For all images presented in this paper, the magnetization sensitivity direction is vertical. Before imaging, the magnetic films were demagnetized by rotating them about an in-plane axis parallel to the rows of squares in a dc magnetic field which was reduced from a value well above the saturation field to zero. This means that the applied demagnetizing field is essentially perpendicular to the rows of squares and parallel to the magnetization sensitivity direction. Our detailed observations of the magnetic domain configurations in the 4 and 2 μm elements, and the surrounding continuous films, are described in the next section.

III. RESULTS

A. Cobalt square elements

The XMCD images of magnetic domain states for the demagnetized cobalt wedge with thickness up to 300 nm are given in Fig. 3. Due to the difficulty of determining the exact thickness of the films below $t \approx 10$ nm with the SEM, we have assumed a zero film thickness at the first square with no magnetic contrast (position indicated with white square frames and labeled “square 0” in the figure) which is in agreement with the absorption spectra taken at this position. This results in an uncertainty in the nanometer range of the low thickness values. From magnetic simulations,^{9,11} it seems that the lowest energy state for the 4 and 2 μm square elements should be the Landau state, with a transition to single domain behavior at very low (around 1 nm) thickness. However, rather than the Landau state we have observed a

variety of metastable states not predicted by the simulations. For the 4 μm square elements, we have observed four key thickness regions.

- (i) For $t \approx 2$ nm (square 1), we observe a three-domain state with the domain walls oriented at 45° to the square boundaries [see Fig. 4(a)]. The orientation of the domain walls is similar to that of the continuous film surrounding the plateaus and may therefore be determined by the intrinsic uniaxial anisotropy of the cobalt film.
- (ii) For $t=4-8$ nm (squares 2–4), we observe very narrow domains, which seem to result from the piling up of 360° walls [Fig. 4(b)]. Such 360° walls are common in highly dispersed magnetic films which have been demagnetized in a similar manner.²⁶ The magnetization dispersion is linked to the variation in the orientation of the magnetocrystalline anisotropy axes from crystal grain to grain. Roughness of the square edges may also help to pin magnetic spins, facilitating the formation of the 360° walls.
- (iii) For $t=10-18$ nm (squares 5–18), several different magnetic states are observed which can be divided into two main categories: near-diamond states [Fig. 4(c)] and asymmetric Landau states [Fig. 4(e)]. We also observe one square element with an S state [Fig. 4(d)]. These various states are the results of trapping of magnetic states at local energy minima via pinning of magnetic spins, e.g., in domain walls or vortices, at material defects.
- (iv) At a $t=19$ nm (square 19), the elements finally fall into the centered Landau state [Fig. 4(f)] and remain in this state up to the maximum thickness of 300 nm (square 216). It is interesting here to see that the Landau state is maintained by the stray field energy associated with the square shape up to the highest film thickness, despite the fact that the film thickness is 2.5 times thicker than the original etched silicon substrate plateaus.

The study of the magnetic domain configurations in the 4 μm squares as a function of thickness has allowed us to observe how local effects, i.e., pinning and magnetocrystalline anisotropy, compete with the stray field energy associated with the square shape. This results in a series of metastable states existing up to a thickness where the stray field energy dominates and the centered Landau state prevails. For the 2 μm squares we see a similar competition (see Fig. 5), with the centered Landau state first occurring at a film thickness of $t=15$ nm (square 13) and remaining up to $t=300$ nm, although at a thickness of 25 nm (square 28), one last metastable diamond-like state occurs. While we see an indication of comparable behavior from the XMCD contrast in the 1 μm square elements, we have concentrated our study here on the 4 and 2 μm square elements which requires a field of view too large (i.e., resolution is too low) to determine the details of the magnetic states present in the smaller elements.

FIG. 4. XMCD images of $4\ \mu\text{m}$ cobalt squares showing (a) three-domain state, (b) piled up 360° walls, (c) near-diamond state, (d) S state, (e) asymmetric Landau state, and (f) centered Landau state.

FIG. 5. XMCD images of $2\ \mu\text{m}$ cobalt squares with the square number indicated. At low thicknesses a series of metastable states is observed which include a C state (square 1), a thickness range where the magnetocrystalline anisotropy dominates and small domains are present (squares 2–4), a state with white contrast at the borders and a central gray contrast (squares 6 and 11), an S state (square 9), and asymmetric Landau states (squares 7, 8, 10, and 12). The centered Landau state is attained at square 13 ($t=15\ \text{nm}$), with one last metastable diamond-like state observed in square 28 ($t=25\ \text{nm}$).

FIG. 6. XMCD images of a Permalloy wedge deposited on a prepatterned silicon substrate with rows of square plateaus with 4, 2, and 1 μm edge lengths. The square number (in black) and the cobalt thickness in nm (in white) are indicated.

FIG. 7. XMCD images of $4\ \mu\text{m}$ Permalloy squares showing (a) centered Landau state, (b) three-domain state, (c) small domains with small black and white edge domains reminiscent of the flower state, (d) S state, (e) zigzag state with schematic diagram, (f) C state, (g) asymmetric Landau state, and (h) centered Landau state.

B. Permalloy square elements

The XMCD images of magnetic domain states for the demagnetized Permalloy wedge with thickness up to 96 nm are given in Fig. 6. Again, we see different thickness regions for the $4\ \mu\text{m}$ squares.

- (i) At $t \approx 1\text{--}2\ \text{nm}$ (squares 1 and 2), we observe different domain configurations. In square 1 [Fig. 7(a)] we observe a Landau state, and in square 2 [Fig. 7(b)] we again observe a three-domain state with the domain walls oriented at 45° to the square edges.
- (ii) For $t \approx 3\ \text{nm}$ (squares 3 and 4), we observe small domains [Fig. 7(c)]. In this region it seems that the magnetocrystalline anisotropy associated with the individual crystal grains dominates, resulting in a fine domain structure.
- (iii) For $t \approx 4\text{--}5\ \text{nm}$ (squares 5 and 6), we observe an S state [Fig. 7(d)].
- (iv) For $t \approx 6\text{--}10\ \text{nm}$ (squares 7–12), we observe two magnetic configurations: either a zigzag configuration [Fig. 7(e)] or a C state [Fig. 7(f)].
- (v) For $t = 11\text{--}16\ \text{nm}$ (squares 13–19), we observe an asymmetric Landau state [Fig. 7(g)].
- (vi) For $t = 17\text{--}96\ \text{nm}$ (squares 20–112), the elements finally fall into the centered Landau state [Fig. 7(h)].

For the $2\ \mu\text{m}$ Permalloy squares we also observe a similar competition with thickness until the centered Landau state is attained at a thickness of 13 nm (square 15).

C. Continuous cobalt film

In a continuous thin polycrystalline film, we expect to see fairly large domains, which are elongated along the mag-

netization direction and are several micrometers across, and also the appearance of cross-tie walls over a certain thickness range.⁷ For the cobalt film (Fig. 3), we do indeed see cross-tie walls separating large domains with the film thickness in the range of 26–65 nm (squares 31–91), and large domains continue to be present up to the largest thickness of 300 nm. However, in a thickness range starting sharply just after square 1 ($t \approx 2\ \text{nm}$) and running up to about square 30 ($t \approx 26\ \text{nm}$), we observe small domains which are isotropic in character and increase in size as the cobalt film thickness increases. It seems that in this thickness range, the roughness of silicon substrate, which is produced during the reactive ion etching process used to create the silicon plateaus, plays an important role in determining the domain structure. The local stray fields associated with the resulting film roughness gives a dramatic change in the magnetic domain configuration, providing nucleation sites and pinning centers which favor the formation of small domains. The domain size increases with thickness as the effect of the surface roughness decreases, and at a thickness of 26 nm, the expected large domains appear as the surface roughness becomes insignificant compared with the total film thickness. It is surprising to see that the large domains reappear at lower film thicknesses (just before square 1 at a nominal thickness below $\approx 2\ \text{nm}$). However, if one considers that stray fields become negligible when a ferromagnetic film becomes ultrathin, a possible explanation for the reappearance of large domains is that the stray field energy associated with the film roughness becomes too small at the lower thicknesses to have an influence on the magnetic domain configuration.

The presence of the square plateaus in the silicon results in square holes or antidots in the continuous films. The effect

of the stray field energy associated with the antidots can be most clearly seen in Fig. 3, squares 88–91 and 150–153. Here typical antidot configurations, i.e., whisker domains emanating from the 2 and 1 μm squares and diagonal domains running between the squares in adjacent rows, can be seen.^{27,28} Between the 4 μm squares we also observe Landau patterns (between squares 34 and 35) and hexagon-shaped domain configurations, with (black and white contrast between squares 35 and 36) and without (black contrast between squares 37 and 38) the presence of a domain wall, all of which we have outlined with a dashed-white line and are the results of stray field energy associated with the square holes.

For very thick films with a direct exchange interaction (i.e., physical contact between the films), the neighboring XMCD contrast is expected to be the same. It is also conceivable that there are stray field interactions between the continuous film and the square elements when they are isolated from each other. For the larger film thicknesses, as we have seen, the stray field energy associated with the square shape dominates and results in a Landau state in the square elements. For the thickness range where we observe metastable states in the square elements (film thickness is below 20 nm and vertical separation between the magnetic layers is 100 nm or more), the stray field interactions between the two layers may play a more important role in determining the resulting domain structures.

It should also be noted that, at the lower half of the XMCD image in Fig. 3 between squares 10 and 17, a piling up of 360° walls is observed in the continuous film, indicated in several places by the white arrows, which again is an indication of a high anisotropy dispersion in the cobalt films.²⁶

D. Continuous Permalloy film

For the continuous Permalloy film there is also a large region containing small domains (Fig. 6), which starts at a thickness of $t \approx 6$ nm and continues beyond square 33 ($t \approx 28$ nm). We do not observe cross-tie walls in the Permalloy films which would normally occur in the region where we observe small domains.

IV. CONCLUSIONS

We have studied the magnetic domain configurations in ferromagnetic square elements with continuously increasing ferromagnetic film thickness using PEEM. At high film thicknesses, where the stray field energy associated with the shape boundaries dominates, we observe the centered Landau state. Although this is predicted to be the lowest energy state for practically all film thicknesses, we observed that for lower thicknesses the system is trapped in some high energy, more complex configurations. These metastable states include three-domain states, diamond states, zigzag domain states, *C* states, *S* states, and asymmetric Landau states, and their presence can be attributed to two causes: First, for low film thickness, material defects such as the surface and edge roughness result in strong local pinning sites and the demagnetizing field is not sufficient to overcome them. Indeed, the

lower the thickness the more effective the pinning site will be. Secondly, as the film thickness decreases it becomes more and more difficult to switch between equilibrium states because at lower thickness, the magnetic spins are forced to lie in plane resulting in higher energy barriers between metastable states. In very thin films the observed magnetic spin configuration is almost never the ground state because a few roughness-induced pinning sites suffice to trap the system into high energy equilibrium states, with the coercive field associated with these pinning sites creating a high energy barrier. The presence of local effects, such as magnetocrystalline anisotropy and shape edge roughness, is emphasized by the appearance of small domains or tightly packed 360° walls in a thickness region below 10 nm where these effects appear to dominate.

In the continuous films surrounding the square plateaus, we observe a thickness range with small, isotropic domains which are the results of the surface roughness of the substrate in the valleys due to the etch processes used to prepattern the silicon substrate. It seems that creation of small domains results from the stray fields associated with the substrate roughness which provide a dense network of nucleation and pinning centers.

This work highlights the importance of local magnetic effects in the determination of the final magnetic domain states in magnetic thin films and small elements. Material defects cannot realistically be eliminated and therefore an understanding of their role in the behavior of magnetic thin film elements is essential for the fabrication of future magnetic devices.

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