Origin of the magnetization reversal of an Fe thin film on Si(111)

M. Cougo dos Santos, J. Geshev, J. E. Schmidt, S. R. Teixeira, and L. G. Pereira

Instituto de Física–UFRGS, Caixa Postal 15051, 91501-970, Porto Alegre, RS, Brazil

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The magnetic behavior of a 60-Å Fe film grown on Si(111) is reported and discussed. Scanning tunneling microcopy images showed Fe stripes along one of the $\langle 110 \rangle$ directions in the (111) plane, which favors easy magnetization axis parallel to the stripes. Inverted in-plane hysteresis loops and loops with unusual local peaks were measured and interpreted. A phenomenological model was proposed to interpret the magnetization data and an excellent concordance between the experimental and the calculated curves was obtained. The peculiarities in the magnetization curves are directly related to the very small misorientation of the sample's surface from the (111) plane, and to competing anisotropies. The model calculations showed that the magnetization leaves the sample's plane for small fields, forming an obtuse angle with the field for some configurations, thus resulting in negative remanence, which was confirmed by the nonzero values of the normal to the plane magnetization component from the magneto-optic Kerr effect polarimetry measurements.

I. INTRODUCTION

The recent scientific and technological advances in low-dimensional systems have revealed an interesting face of the properties of matter. Magnetic thin films, in particular, have shown phenomenology which cannot be observed in bulk materials. Certain growth conditions of the films and their crystallographic axes orientation relative to the substrate crystal axes, as well as the substrate morphological characteristics, can lead to a surprising magnetic behavior, one of the reasons being the complicated multicomponent magnetic anisotropy present. The question remains open whether the concepts elaborated for single-domain particles can be applied to thim films or whether specific behavior is to be expected.

Uniaxial anisotropy have been observed in iron[1–3] and cobalt thin films[4–7] grown on stepped substrates. The step-induced magnetic anisotropy is believed to originate from missing bonds at the step edges and strains within the film.[7] Deposition of iron with certain angle of incidence of the beam with the substrate creates shadows in the deposition and favors easy axis perpendicular to the beam.[8,9] Recently, strong uniaxial anisotropy has been found in stripes cut from Fe layers patterned by the "atomic saw" method.[10,11]

The Si(111) is favorable for growing epitaxial Fe(111) thin films.[12–15] During the cutting process of the Si wafer, small misorientation of the substrate's surface relative to its (111) plane creates atomic steps on the substrate surface, which is known to favor long-range iron stripes formation.[16] The present paper reports on the magnetic properties of a 60-Å Fe film grown on Si(111).

II. EXPERIMENT

The sample was prepared by electron gun evaporation in ultrahigh vacuum (10^{-9} Torr) at room temperature, at a deposition rate less than 1 Å/s. In order to prevent the oxidization of the film, it was covered by a 25-Å-thick Cr layer. The structural characterization was performed via conventional x-ray diffraction (XRD) performed on a Philips X'Pert MRD machine employing Cu $K\alpha$ radiation, as well as by scanning tunneling microcopy (STM) using a Digital Instruments Nanoscope IIIa. The STM images give a clear striped structure shown in Fig. 1. It can be interpreted as a consequence of the Si surface cut away within a few degrees of the (111) plane, leading to the iron stripes growth. The stripes are estimated to be approximately 4000 Å wide and 80 Å high, and are parallel to the [110] direction, as indicated in the figure. The XRD analysis confirmed that the wafer surface was tilted by 0.5° from the (111) plane. Ferromagnetic resonance data obtained recently for the same sample[15] reveal a deviation from the sixfold symmetry due to the combined effect of the induced uniaxial in-plane anisotropy and the small miscut of the substrate surface from the (111) plane.

III. RESULTS AND DISCUSSION

The magnetization data were acquired using two distinct methods, alternating gradient force (AGM) magnetometry, and magneto-optic Kerr effect (MOKE) polarimetry, the latter allowing components of the magnetization along two axes to be measured.[17] The magnetic field H is applied in the sample's plane and, unless otherwise stated, the magnetization is measured parallel to the field direction. The magnetization curves obtained by the two methods are practically the same, and all data presented here are from the MOKE measurements. Representative experimental hysteresis loops for several values of the angle ϕ, between the in-plane easy axis and the field direction, are shown in Fig. 2. The curves change from almost rectangular for fields along the [110] axis, to a curve with zero remanence for the perpendicular direction, but with a certain hysteresis for field values of about 10 Oe still existing. For higher ϕ, up to approximately 95°, the hysteresis loops become inverted, as it is shown in the figure for $\phi = 95°$. The two branches cross, resulting in negative remanence and coercivity. MOKE measurements gave nonzero normal to the plane magnetization component in the low-field range, which will be discussed latter.
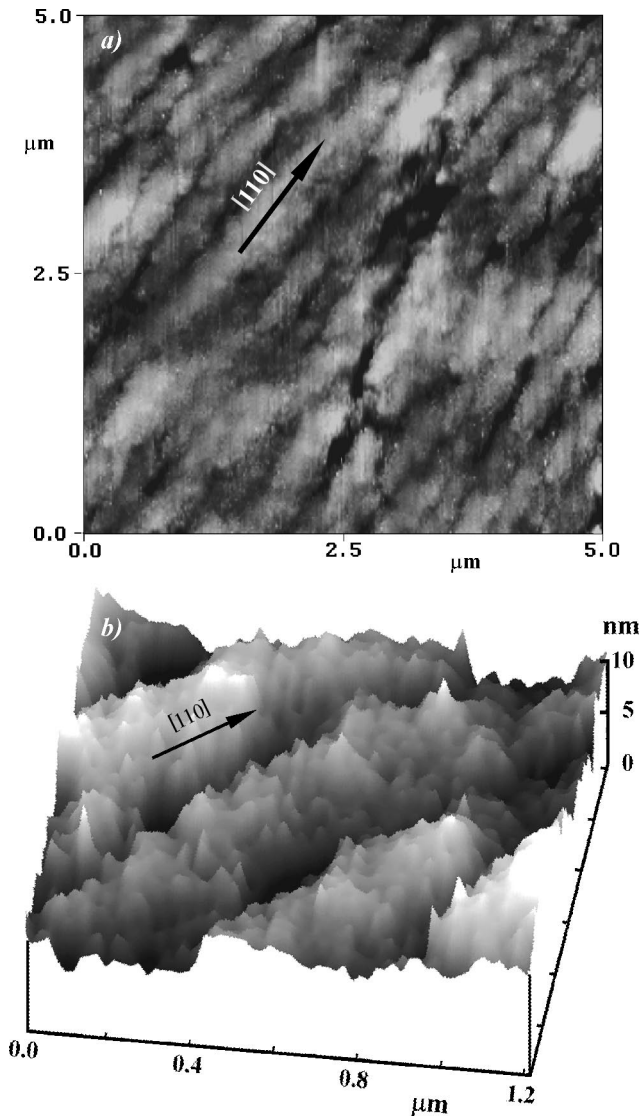
Inverted hysteresis loops, similar to the ones obtained

FIG. 1. Tunneling microcopy images of Fe thin film grown on Si(111); surface scan (a), and three-dimensional image (b). The arrows indicate the [110] direction.

here, have been reported by Chen and Erskine,[1] for thin epitaxial Fe films on stepped W(001), and have been attributed to a magneto-optical effect. Such loops have also been observed in some exchanged-coupled multilayers (see, e.g., Poulopoulos and Flevaris[18] and the references therein). In order to check the influence of the Cr layer on the hysteresis of our sample, we performed magnetic measurements on a 60-Å Fe on Si(111) sample without any protecting layer. Hysteresis loops with negative remanence were observed as well, thus discarding the hypothesis of inverted hysteresis due to exchanged coupling between the Cr and Fe layers in the sample in consideration here.

The experimental dependencies on the field direction of the reduced remanent magnetization M_r/M_s, and the coercive field H_c, are plotted in Fig. 3. The shape of the $M_r/M_s(\phi)$ curve shows that the uniaxial anisotropy predominates over the cubic one. However, the $H_c(\phi)$ curve indicates clearly that the cubic anisotropy still exists. There are six equidistant minima, in accordance with the previous ferromagnetic resonance measurements.[15] The positions of
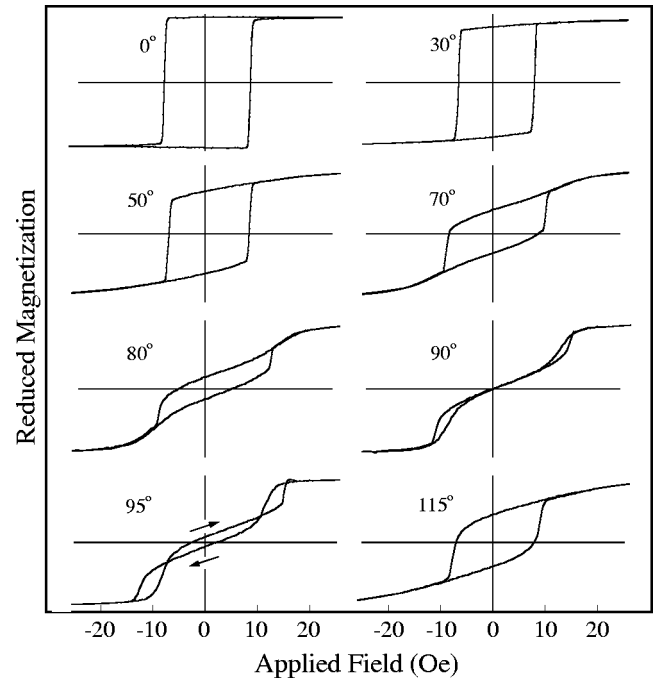

FIG. 2. Experimental in-plane hysteresis loops for several angles (from 0° to 120°) between the field and the easy axis.

the two global minima, related to the uniaxial anisotropy, show that the latter is superimposed upon the cubic one; the uniaxial anisotropy direction coincides with one of the three cubic anisotropy easy magnetization axes in the (111) plane.

Our interpretation of the magnetic behavior of the material studied here is that it is due (i) to competing anisotropy effects and (ii) to the small misorientation of the Fe layer with respect to the Si(111) plane. The phenomenological model used to interpret the experimental data is presented as follows.

Let us consider a film with a cubic intrinsic symmetry whose energy expression contains, apart from the cubic magnetocrystalline anisotropy terms, one uniaxial anisotropy
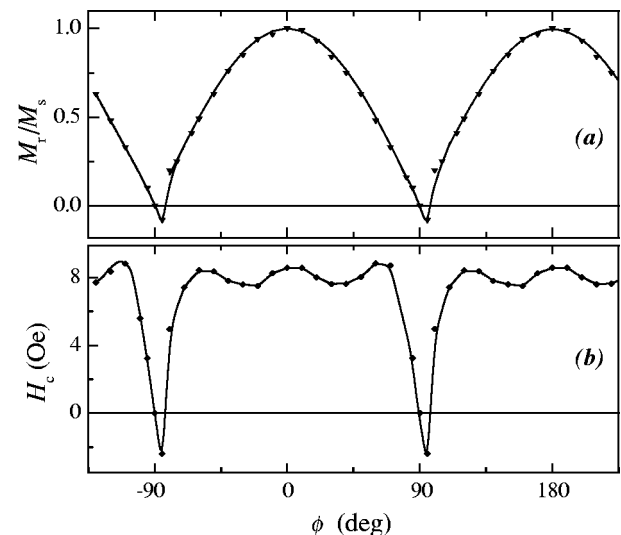

FIG. 3. Reduced remanent magnetization (a), and coercive field (b) as a function of the angle between the in-plane field direction and the easy-axis orientation.

term as well. Let the direction cosines of the magnetization vector $\mathbf{M_s}$ be α_1, α_2, and α_3, referred to the cube axes. Neglecting the thermal activation effects and considering only coherent rotation of the magnetization, the total free-energy density E for fixed magnitude and direction of $\mathbf{H}$, can be written as

$$E = K_1(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2)$$
$$+ K_2\alpha_1^2\alpha_2^2\alpha_3^2 - K_u(\mathbf{M_s}\cdot\mathbf{u}/M_s)^2$$
$$+ 2\pi(\mathbf{M_s}\cdot\mathbf{n})^2 - \mathbf{M_s}\cdot\mathbf{H}, \tag{1}$$

where K_1 and K_2 are the first two cubic anisotropy constants, K_u the uniaxial anisotropy constant, and the last two terms refer to the demagnetization energy and Zeeman energy terms, respectively. The unit vectors $\mathbf{u}$ and $\mathbf{n}$ represent the direction of the uniaxial anisotropy and the normal to the film surface direction.

In zero applied field, for positive K_1 (which is the case of Fe), $K_2 > -\frac{9}{4}K_1$ and $K_u = 0$, there are six minima of E along the $\langle 100 \rangle$ directions. As K_u increases, these minima move towards the uniaxial anisotropy direction. For magnetization rotation in the (111) plane, when $K_u = 0$ and $\mathbf{n}$ is along the [111] direction, it can be easily shown that the anisotropy is given by K_2 term only. The anisotropy is a sixfold one, with easy axes along the $\langle 110 \rangle$ directions for positive K_2, and along the three projections of the cube axes in the (111) plane for negative K_2.

Minimizing E for given M_s, K_1, K_2, K_u, $\mathbf{u}$, $\mathbf{n}$, and $\mathbf{H}$, the equilibrium direction of $\mathbf{M_s}$ can be obtained. Varying the field value, the magnetization curves can be calculated; the two-variable minimization procedure used in the present work is described elsewhere.[19]

The anisotropy in the (111) plane is extremely sensitive to misorientation of the substrate surface with respect to the crystal plane: small tilt is sufficient to completely hide the sixfold anisotropy symmetry.[13] This, however, does not result in anomalous hysteresis loops if the magnetization is considered to rotate in the film's plane only, as it is normally assumed in model studies. In some cases, negative remanence for rotation in a plane can be obtained if, e.g., there is asymmetry of the zero-field energy: for predominant uniaxial anisotropy the point of the maximum energy must not be equidistant from the two minima,[19] but this is not the case for the surface in consideration here. Note that in our model the magnetization vector *is not constrained* to lie in the plane of the film. This, as it will be shown below, is a deciding factor in order to obtain inverted hysteresis loops.

For calculating the model hysteresis loops we used the usual room temperature K_1 and M_s values for Fe, $K_1 = 4.8 \times 10^5$ erg/cm^3 and $M_s = 1708$ emu/cm^3. The best results were obtained for $K_u/K_1 = 0.035$ and $K_2/K_1 = 0.22$, the latter being in good agreement with the value obtained by Chen.[20] Our XRD data show that the sample's surface is tilted by 0.5° with respect to the (111) plane, so that in the calculations the normal $\mathbf{n}$ was chosen to be almost parallel to the [111] direction. This was done by taking its polar and the azimuthal angles to be 55° and 44.6°, respectively, thus assuring a 0.5° tilt. The vector $\mathbf{u}$ is assumed to be along the stripes' direction, i.e., one of the $\langle 110 \rangle$ orientations in the (111) plane, the [1$\bar{1}$0] one, in accordance with the STM data
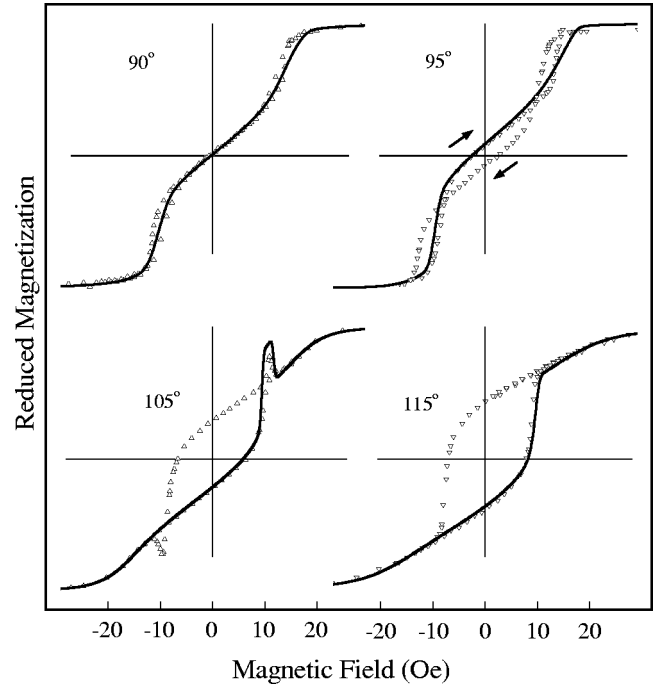

FIG. 4. Experimental hysteresis loops (open symbols) and the corresponding calculated lower branches of the loops for four field orientations.

(Fig. 1). It was estimated that small variations of K_2/K_1 value do not influence significantly on the shape of the magnetization curves. Thus the number of free parameters used is actually reduced to only two: K_u/K_1 and the tilt direction.

Experimental hysteresis loops and the corresponding calculated lower branches of the loops for ϕ between 90° and 115° are plotted in Fig. 4. It is seen that there is an excellent concordance between the model and experimental curves. More attention will be paid to the inverted loop ($\phi = 95°$) and to the one for $\phi = 105°$; the latter has positive remanence but shows rather unusual peaks for $|H| \approx 10$ Oe.

The most important result from the very good fitting of the inverted hysteresis loop is that for low-field values, the magnetization vector *does not lie* in the sample's plane. It was obtained from the model calculations that, due to the small tilt from the (111) plane and due to the competing anisotropy effects, the magnetization vector leaves apparently the sample's plane for $|H| < 15$ Oe, forming an angle of 94.7° with the field direction when $H = 0$, resulting in the negative remanence. This was confirmed by the MOKE data which give the magnetization components parallel $M_\parallel$, and perpendicular $M_\perp$, for fields applied in the sample's plane. The normal to the plane component $M_\mathbf{n}$, given by $M_\mathbf{n} = (M_s^2 - M_\parallel^2 - M_\perp^2)^{1/2}$ is obtained to have nonzero values in the low-field range, in accordance with the calculation results.

In the framework of the model, the anomalous peaks in the hysteresis loop for $\phi = 105°$ (which means that, in the cubic reference system, the polar and the azimuthal angles of the positive field direction are 147.7° and 62.0°, respectively) are explained as follows. For the highest field values, $\mathbf{M_s}$ is along the field direction. When H is increased, starting from the maximum negative field value, up to $+9.2$ Oe, the magnetization rotates reversibly. In the remanent magnetiza-

tion state, $\mathbf{M_s}$ is in the vicinity of the uniaxial anisotropy direction closer, to the negative field direction, and is again out of the film's plane, but now this does not result in inverted hysteresis. Fields slightly higher than 9.2 Oe lead to the irreversible jump indicated by the rapid increase of the magnetization for these fields. Now, $\mathbf{M_s}$ is very close to the $[10\bar{1}]$ direction, which is one of the six easy magnetization directions in the (111) plane given by the cubic anisotropy. This direction is rather close to the positive field direction, accounting for the high M/M_s for this state. The magnetization stays in the vicinity of this direction for fields up to 11.4 Oe. Further increase of H eliminates the corresponding energy minimum and the magnetization jumps to the remaining minimum which is located between the positive field direction, and the uniaxial anisotropy one. This new position, however, is considerably more distant from the $\mathbf{H}$ direction than the previous one, thus resulting in the decrease in the magnetization curve. Further raise of H just approximates $\mathbf{M_s}$ to $\mathbf{H}$, and the magnetization curve follows its normal increase.

There is also a very good agreement between the shapes of the calculated and experimental magnetization curves for field directions close to the easy axis direction. However, the calculated coercive fields are larger than the experimental ones, which can be attributed to other magnetization reversal modes, different from the coherent one, for these configurations.

IV. CONCLUSIONS

Structural and magnetization data for a 60-Å Fe film grown on Si(111) were reported and discussed. STM images showed stripes along one of the $\langle 110 \rangle$ directions in the (111) plane, which favors easy magnetization axis parallel to the stripes. Inverted in-plane hysteresis loops, and loops showing local peaks were observed. Excellent concordance was obtained between the experimental curves and the ones calculated, using a phenomenological model. The negative remanence was explained as caused by competing anisotropy effects and the very small misorientation of the sample's surface from the (111) plane. The calculations showed that the magnetization leaves the sample's plane for small fields, forming obtuse angle with the field for some configurations, thus resulting in negative remanence. This was confirmed by the nonzero values of the normal to the plane magnetization component from the MOKE measurements.

In summary, we have demonstrated that for the thin film studied here the concepts elaborated for the coherent rotation of single-domain particle magnetizations can be applied without any modifications. Regarding the inverted hysteresis loops, since we obtained almost identical loops by using either MOKE or AGM magnetometry, we can assert that the magnetic behavior of our sample is explained solely by proper anisotropy energy considerations, and not by the magneto-optical effect proposed by Chen and Erskine[1] for their Fe films on stepped W(001).

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