Ferromagnetic resonance of Fe(111) thin films and Fe(111)/Cu(111) multilayers

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Ferromagnetic resonance at 9.4 GHz has been used to characterize several samples of Fe single-crystal films and Fe/Cu multilayers prepared by electron-beam deposition on HF-etched, hydrogen-terminated Si(111). Resonance field data as a function of the field orientation in the film plane confirmed that Fe films and Fe/Cu multilayers grow epitaxially on Si(111) with excellent crystallinity in the [111] orientation. Moreover, we have found that the (111) plane is unique among the principal planes in cubic crystals, because it allows precise measurements of the small second-order and in-plane anisotropies as well as misorientations from the crystal plane.

INTRODUCTION

Magnetocrystalline anisotropy plays an important role in the magnetic properties of single-crystal materials. In ultrathin films the anisotropy is largely influenced by the surfaces, interfaces, and the distortions of the lattice induced by strains from the growth process. It has been known for decades[1-5] that ferromagnetic resonance (FMR) is one of the most powerful techniques for determining the crystallinity of a material and for measuring the phenomenological anisotropy constants. With the recent interest in magnetic thin films, FMR has been increasingly used in the study of single-crystal films.[6-11] The basic procedure consists of placing the film in a microwave resonator with the rf magnetic field perpendicular to a static field **H**, and measuring the field for resonance at a certain frequency as a function of the direction of **H** in some chosen plane. This yields a direct measurement of the symmetry and amplitude of variation of the magnetic energy in that plane. When the plane of scan is perpendicular to the film, the variation is dominated by demagnetizing and perpendicular anisotropy energies. On the other hand, when the field scans the plane of the film, those energies are constant so that one can measure the magnetocrystalline anisotropy.

The FMR technique has been used[6-11] to investigate the magnetic anisotropy of thin Fe films, deposited on various substrates. Usually the films are grown either in the (100) or (110) planes, where the variation of the resonance field is on the order of several hundred oersteds and totally dominated by the first-order cubic crystal anisotropy. This large variation overwhelms smaller contributions to the anisotropy energy, which may also be important for understanding the physics of magnetic films. In this paper we use FMR to study Fe films and Cu/Fe multilayers grown on Si(111) and on Si(111)/Cu(111). The results not only confirm earlier evidence[12,13] that the Fe layers are crystalline on the (111) plane but also reveal very interesting features of the (111) orientation. Since the first-order cubic anisotropy energy does not change as the magnetization rotates in the (111) plane, the FMR technique allows precise measurements of the second-order cubic anisotropy and the strain-induced in-plane uniaxial anisotropy, and detection of small misorientations from the (111) plane.

THEORETICAL CONSIDERATIONS

As in many other resonance phenomena, the magnetic resonance frequency is a measure of the energy surface curvature at the equilibrium point. Since the magnetization has two degrees of freedom, the expression for the resonance frequency ω_0 must involve derivatives with respect to two coordinates. In spherical coordinates it was shown long ago[2,3] that

$$\omega_0=\frac{\gamma}{M\sin\theta}\left[\frac{\partial^2 E}{\partial\theta^2}\frac{\partial^2 E}{\partial\phi^2}-\frac{\partial^2 E}{\partial\theta\,\partial\phi}\frac{\partial^2 E}{\partial\phi\,\partial\theta}\right]^{1/2}, \quad (1)$$

where γ is the gyromagnetic ratio ($\gamma=1.4g$ GHz/kOe), E is the total free energy per unit volume, and θ and ϕ are the polar and azimuthal angles of the magnetization **M** with respect to some coordinate system. We will consider the following form for the free-energy density:

$$E=-\mathbf{H}\cdot\mathbf{M}+K_1(\alpha_1^2\alpha_2^2+\alpha_2^2\alpha_3^2+\alpha_3^2\alpha_1^2)+K_2\alpha_1^2\alpha_2^2\alpha_3^2$$
$$+2\pi(\mathbf{M}\cdot\hat{\mathbf{n}})^2+K_u\sin^2(\alpha-\alpha_u)-K_n(\mathbf{M}\cdot\hat{\mathbf{n}}/M)^2\,. \quad (2)$$

The six terms are, respectively, the Zeeman energy; the first-order magnetocrystalline cubic anisotropy, with α_1, α_2, and α_3 being the direction cosines of the magnetization vector with respect to the cubic axes [100], [010], and [001]; the second-order anisotropy, with a constant K_2 normally on the order of $K_1/10$; the demagnetizing energy for a film, $\hat{\mathbf{n}}$ being the unit vector normal to the film plane; the uniaxial in-plane anisotropy, with α representing the direction of the magnetization in the film plane and α_u denoting the angle of the uniaxial field; and finally the perpendicular anisotropy with constant K_n.

In order to calculate the resonance frequency one must first determine the equilibrium direction of the magnetization for each orientation of the applied field, which is done by minimizing the free energy (2). When **H** is either in (100) or (110) it is possible to find an analytical expression, in first order in K_1/MH, for the angle between **M** and **H** and also for ω_0.[4] However, this is not true for (111), where the equilibrium position of **M** and ω_0 must be obtained numerically.[5] In this study, we have numerically computed the equilibrium orientation of **M** and the resonance field for a given ω_0, constraining the magnetic field to the film plane. The calculation is made for an arbitrary crystal plane, changing the coordinate axes in the energy expression (2) so that the z direction is perpendicular to the film plane.

Figure 1 demonstrates the unique features of the (111) plane regarding the magnetocrystalline anisotropy. The anisotropy energy and the resonance field for $\omega/2\pi=9.4$ GHz as a function of the orientation of the external field in the film plane are shown for the three principal cubic planes for an Fe film, with $4\pi M_{\text{eff}}=4\pi M-2K_n/M=18$ kG, $2K_1/M=500$ Oe, and $K_2=K_u=0$. Here and in the remainder of the paper we use $g=2.1$. As seen in Fig. 1(a), contrary to what happens in (100) and (110), the first-order anisotropy does not vary in the (111) plane. As a result, the variation of the resonance field in (111) is on the order of only 10 Oe (due to the nonzero derivative of the anisotropy energy in the direction perpendicular to the plane). This contrasts with the variation of almost 1 kOe occurring when the film plane is either (100) or (110). Of course, the plane of the film can be immediately recognized from the symmetry, four-, two-, or sixfold for (100), (110), or (111), respectively.

It was the apparent lack of sixfold symmetry in the FMR data of an Fe film deposited on a nominally (111) Si substrate that drew our attention to the features of (111) Fe films. Due to the small variation of the resonance field in (111), smaller contributions to the energy play an important role in the FMR when the film is in this plane. To illustrate this we show in Fig. 2 the anisotropy energy and the resonance field versus field angle in the (111) plane when a small uniaxial in-plane anisotropy is present. Remarkably, as seen in Fig. 2, a uniaxial field of only $2K_u/M=20$ Oe is sufficient to completely hide the sixfold symmetry variation due to the 500-Oe large first-order anisotropy.

Another feature of the (111) plane is the extreme sensitivity to misorientation of the substrate surface with respect to the crystal plane. This is significant because it is a current practice to cut Si ingots in planes misoriented by a few degrees from (111), in order to speed up the liquid-phase epitaxial growth used in the final preparation of wafers for microelectronics. If such a wafer is used as substrate to grow Fe films, the magnetization is constrained to a plane tilted with respect to (111). Figure 3 shows the behavior of the energy and the resonance field when the film plane is rotated by an angle β from (111) about a ⟨110⟩ axis. Small misorientations are sufficient to reduce the sixfold symmetry to twofold, resulting in variations of the resonance field much larger than in (111).

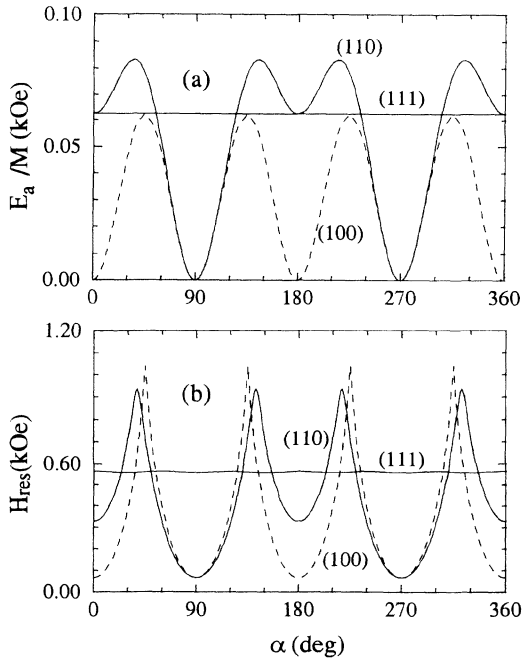

FIG. 1. In-plane angular dependence for the three principal cubic planes of (a) the anisotropy energy; (b) the 9.4-GHz FMR resonance field. The parameters considered for the Fe films are $4\pi M_{\text{eff}}=18k$ G, $2K_1/M=500$ Oe, and $K_2=K_u=0$.

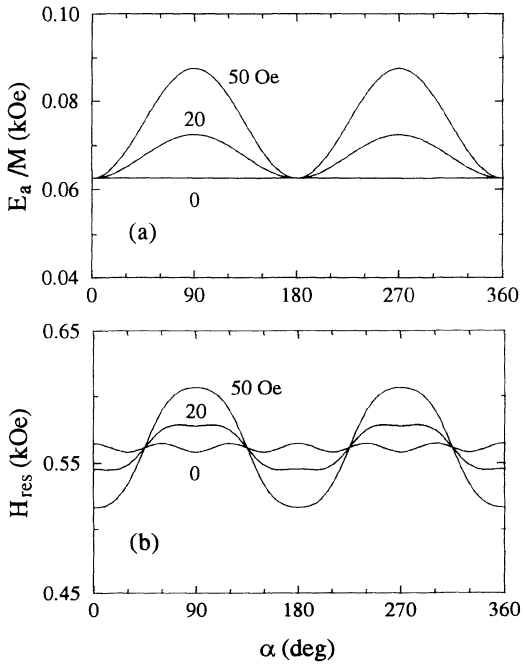

FIG. 2. Angular dependence in the (111) plane of the anisotropy energy (a) and of the 9.4-GHz resonance field (b) for an Fe film with $4\pi M_{\text{eff}}=18k$ G, $2K_1/M=500$ Oe, and $K_2=0$, with three values of the in-plane anisotropy, $2K_u/M=0$, 20, and 50 Oe.

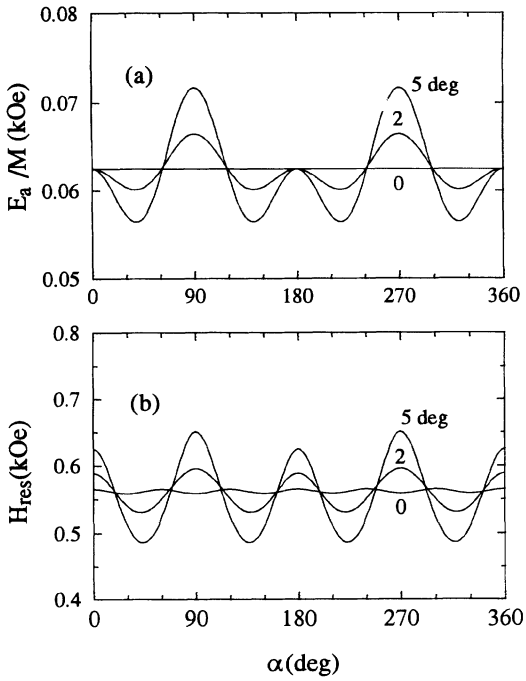

FIG. 3. Demonstration of the effect of small misorientations of the film plane with respect to the (111) crystal plane in an Fe film with $4\pi M_{eff}=18$ kG, $2K_1/M=500$ Oe, and $K_2=K_u=0$. $\beta(=0,\ 2°,$ or $5°)$ is the angle between the film plane and (111).

SAMPLES AND EXPERIMENTAL PROCEDURE

The thin-film samples were grown on HF-etched, hydrogen-terminated, Si(111) wafers at room temperature. Deposition was made in an ultravacuum system (Balzers UMS 500P) using two independent quartz-crystal-controlled electron beams. The base pressure was better than 2×10^{-8} mbar and the deposition rate was 0.5 Å/s for Cu (99.9999%) and Fe (99.99%).

Four samples were fabricated for the studies reported in this paper. Sample 1 is a single 100-Å-thick Fe film grown directly on a Si wafer nominally in the (111) plane. After the initial results revealed that the Fe film was in a plane tilted with respect to (111), more careful analysis of the x-ray data confirmed that the wafer surface was tilted by 1.5° from (111). Sample 2 is a 50-Å-thick single Fe film grown directly on a Si wafer obtained from a different supplier, having no measurable misorientation from (111). Sample 3 consists of a 50-Å Fe film deposited on a 15-Å Cu layer grown on the (111) Si substrate. Finally sample 4 is a multilayer with structure Si (111)/{Cu 15 Å/Fe 15 Å}$\times$10 with a final Cu 15-Å capping layer.

All samples were characterized by x-ray diffraction in the θ-2θ Debye-Scherrer geometry, employing Cu $K\alpha$ radiation. Figure 4 shows the high-angle spectrum from sample 4, the Cu/Fe multilayer. Two weak peaks are seen besides the three strong ones from the Si substrate. The peak at $2\theta=137.2°$ can be assigned to bcc Fe(222) or to bcc Cu(222) planes lying within the substrate plane. Since the bcc structure is not a stable phase of Cu we rule out the latter interpretation. The peak at $2\theta=44.8°$ can be due to fcc Cu(111) or bcc Fe(110). The FMR results

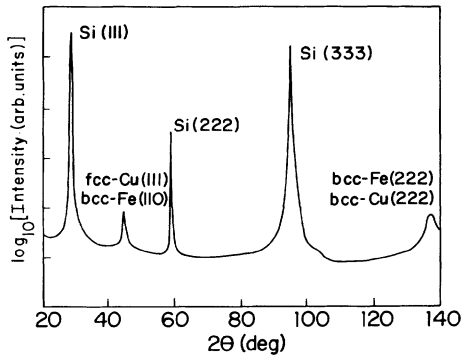

FIG. 4. High-angle x-ray-diffraction scan (Cu $K\alpha$) for sample 4, a Cu/Fe multilayer grown on Si(111).

presented in the next section rule out the possibility of the Fe film being in the (110) plane. Hence, the conclusion from the x-ray and FMR analyses is that sample 4 consists of fcc Cu(111) and bcc Fe(111) layers. This interpretation is consistent with previous studies[12,13] done with x-ray and electron diffraction in a similarly prepared sample. The x-ray spectrum for sample 3 is similar to that in Fig. 4. In the data for samples 1 and 2 the peak at $2\theta=44.8°$ corresponding to fcc Cu(111) is absent, but the one at $2\theta=137.2°$ is present. Thus, the x-ray data for all samples show that the Fe layers have the bcc (111) structure.

The FMR spectrometer is homemade, employing a sweep oscillator with frequency stabilized at the microwave cavity resonance. The cavity used in this study is rectangular, operating at 9.4 GHz with $Q=2500$ in the TE_{102} mode, and is kept fixed relative to the poles of a Varian 9″ magnet. Helmholtz coils on the cavity walls modulate the field at 100 kHz so that we observe the derivative of the absorption lines. The samples are cut in disk shape, glued to the end face of a phenolic rod, and located at the center of the cavity. The rod is mounted on a goniometer to allow measurement of the angular dependence of the FMR spectra, maintaining the static field in the film plane. The angle between the sample and the rod face was determined by sending a laser beam toward the film and measuring the deviation of the reflected beam as the rod rotates around its axis. Initially we took data only after making sure that the tilting was smaller than 0.5°. However, later we verified that larger tiltings, up to 5°, did not affect the data. This is so because the strong demagnetizing field keeps the magnetization in the film plane. In fact, it can be shown that for an external field making angle β_H with the film plane, the deviation of the magnetization from the plane is $\beta_H H/(H+4\pi M)$. This is $\sim(3\times10^{-2})\beta_H$ for Fe in FMR at the X band, so that the effect of a misalignment of 5° would be a change in the resonance field of less than 2 Oe. This is much smaller than all other anisotropy contributions, so that the alignment of the film in the static field is not critical at all.

RESULTS AND DISCUSSION

The in-plane angular dependence of the 9.4-GHz FMR resonance field for a 100-Å Fe film, sample 1, is shown in

Fig. 5. The absorption derivative shown in the inset can be very well described by a Lorentzian line. The observed peak-to-peak width of 25 Oe is among the smallest linewidths observed for Fe and other 3*d* metal films,[6-11] indicating the good crystallinity of the sample. The solid line in Fig. 5 is a least-squares fit obtained with the calculation outlined earlier, yielding the following parameters: $4\pi M_{\rm eff}=17.13$ kG, $2K_1/M=430$ Oe, $K_2/K_1=0.05$, $2K_u/M=29$ Oe, and $\beta=1.6°$. Two angles also enter in the data fitting, the direction of the uniaxial field α_u and the angle δ between the direction [110] and the axis about which the film plane is rotated away from (111). In Fig. 5 the fit was obtained with $\alpha_u=-13°$ and $\delta=12°$. The values of the effective magnetization and first-order anisotropy constant are consistent with those previously reported for Fe films.[6-8] Note that they are smaller than in bulk Fe, 21.45 kG and 560 Oe respectively, due to the contributions from surface anisotropy.[14] Notice that the angle $\beta=1.6°$ between the film plane and (111) obtained from the fitting is in excellent agreement with the value of 1.5° measured by x-ray diffraction for the misorientation of the Si wafer. It is remarkable that this small misorientation, combined with the in-plane uniaxial anisotropy, completely destroys the sixfold symmetry of the field variation expected for a (111) film.

Figure 6 shows the angular dependence of the resonance field for a 50-Å Fe film deposited on a Si wafer with no measurable misorientation from the (111) plane, sample 2. In this case the sixfold variation is apparent, mixed with a twofold variation due to the in-plane uniaxial field. The solid curve is the theoretical fit obtained with $4\pi M_{\rm eff}=14.5$ kG, $2K_1/M=340$ Oe, $K_2/K_1=-0.03$, $2K_u/M=12$ Oe, $\alpha_u=15°$, $\beta=-0.2°$, and $\delta=-12°$. The reduction in $4\pi M_{\rm eff}$ and K_1 compared to sample 1 is expected because this is a thinner film, and

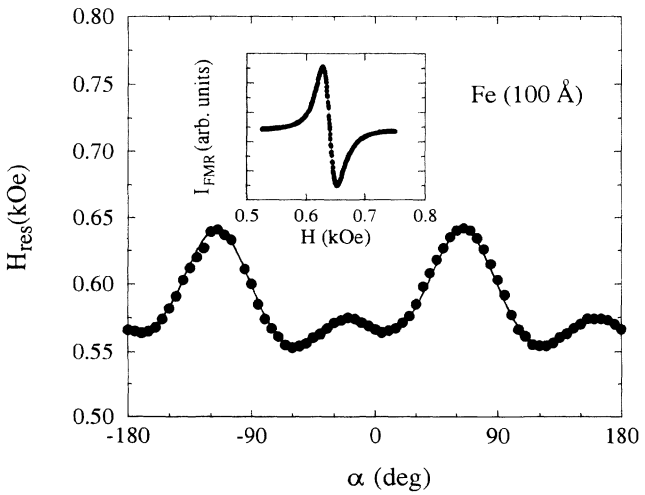

FIG. 5. Angular dependence of the 9.4-GHz FMR resonance film for a 100-Å Fe film misoriented with respect to the (111) plane. The solid line is a fit obtained with $4\pi M_{\rm eff}=17.13$ kG, $2K_1/M=430$ Oe, $K_2/K_1=0.05$, $2K_u/M=29$ Oe, $\alpha_u=-13°$, and an angle $\beta=1.6°$ between the film and the (111) plane, about an axis making an angle $\delta=12°$ with the [110] direction. The inset shows the 25-Oe peak-to-peak wide spectrum.

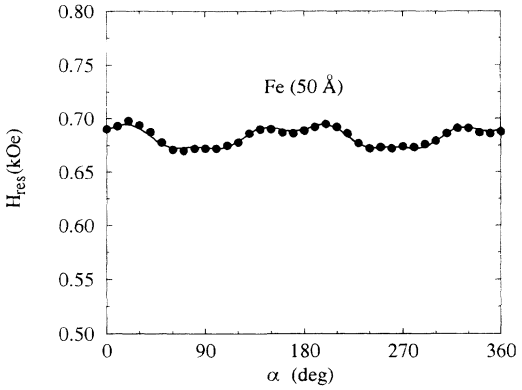

FIG. 6. Resonance field vs in-plane angle for a 50-Å Fe film on a Si wafer with no misorientation from the (111) plane. The parameters obtained from the fit (solid line) are $4\pi M_{\rm eff}=14.5$ kG, $2K_1/M=340$ Oe, $K_2/K_1=-0.03$, $2K_u/M=12$ Oe, $\alpha_u=15°$, $\beta=-0.2°$, and $\delta=-12°$.

thus presents a larger effect of surface anisotropy.[14] The small value of the in-plane uniaxial field indicates that Fe(111) films grow with very small strain on Si(111).

The data of Fig. 7 were obtained in sample 3, a 100-Å-thick Fe film grown on Si(111)/15 Å Cu(111). The data and the solid-line fit demonstrate that the Fe film is crystalline, on the (111) plane, except for a small misorientation. The parameter values obtained from the fit are $4\pi M_{\rm eff}=17.9$ kG, close to sample 1, $2K_1/M=580$ Oe, $K_2/K_1=-0.1$, $2K_u/M=22$ Oe, $\alpha_u=91°$, $\beta=1.2°$, and $\delta=47°$. Notice that K_1 and K_2 are larger than in Fe deposited directly on Si, and that K_2 is negative. This is certainly a result of the Fe/Cu interface anisotropy.

Finally, in Fig. 8 we show the data and the theoretical fit for a multilayer Si(111)/{Cu 15 Å/Fe 15 Å}×10. The best fit was obtained with $4\pi M_{\rm eff}=13.0$ kG, $2K_1/M=680$ Oe, $K_2/K_1=-0.13$, $2K_u/M=42$ Oe, $\alpha_u=71°$, $\beta=-0.4°$, and $\delta=-68°$. These values are consistent with those for sample 3, considering that the Fe layers are thinner and hence have larger surface anisotropy. The somewhat larger value of the in-plane anisotropy indicates that the Fe layers are more strained than in single films deposited on Si(111) or on Si(111)/Cu(111).

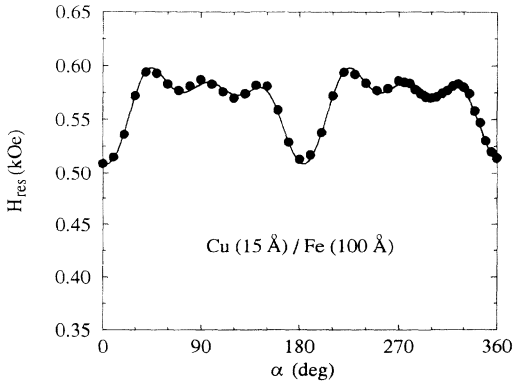

FIG. 7. Resonance field vs in-plane angle for a 100-Å Fe film on Si(111)/15 Å Cu(111). The parameters for the fit are $4\pi M_{\rm eff}=17.9$ kG, $2K_1/M=580$ Oe, $K_2/K_1=-0.1$, $2K_u/M=22$ Oe, $\alpha_u=91°$, $\beta=1.2°$, and $\delta=47°$.

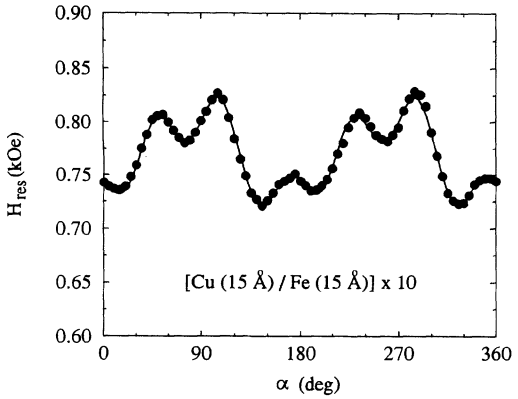

FIG. 8. Resonance field vs angle for a multilayer Si(111)/{Cu 15 Å/Fe 15 Å}×10 and solid line with fitting parameters $4\pi M_{eff}=13.0$ kG, $2K_1/M=680$ Oe, $K_2/K_1=-0.13$, $2K_u/M=42$ Oe, $\alpha_u=71°$, $\beta=-0.4°$, and $\delta=-68°$.

Examination of Figs. 5–8 shows that quite different resonance field vs angle patterns are obtained with relatively small changes in the anisotropy parameters. This results from the fact that in the (111) plane the first-order, the second-order, and the in-plane anisotropies, as well as the effect of misorientation, compete with each other with similar magnitudes. A consequence of this is that small changes in a given parameter produce visible change in the fitting curve, allowing a precise determination of all parameters. Nevertheless, the data for all samples confirm earlier evidence[12,13] that Fe(111) films and Fe(111)/Cu(111) multilayers grow epitaxially on oxide-free Si(111). Note, finally, that the FMR technique by itself cannot distinguish between different phases of the cubic structure; it only serves to determine the crystal plane of the film. However, the combined FMR and x-ray-diffraction data can rule out some possible phases. In the case of the samples investigated here there is no question about the Fe films having the bcc structure. We plan to extend this investigation to other Fe/Cu multilayers apparently having the unstable fcc Fe phase.[12,13,15]

CONCLUSION

The FMR technique has been used to confirm that thin Fe films and Fe/Cu multilayers deposited by electron beam on Si(111) have excellent crystallinity with the [111] orientation. This study has also revealed that the (111) plane is unique among the principal planes in cubic crystals, in the sense that the variation of the first-order anisotropy as the orientation of the field rotates in the film plane is greatly reduced as compared to other planes. A consequence of this is that a data-fitting analysis yields precise measurement of smaller contributions to the anisotropy, such as second-order and uniaxial in-plane anisotropies.

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