CONF-951101--5

UCRL-JC- 121930
PREPRINT

Magnetic Properties of Epitaxial and Polycrystalline Fe/Si Multilayers

A. Chaiken
R. P. Michel
C. T. Wang

This paper was prepared for submittal to
Magnetism and Magnetic Materials Conference,
November 6-9, 1995, Philadelphia, Penna., and published in the
Journal of Applied Physics

August 1995

Lawrence Livermore National Laboratory

DISCLAIMER

Magnetic Properties of Epitaxial and Polycrystalline Fe/Si Multilayers

A. Chaiken* and R.P. Michel†

Materials Science and Technology Division

Lawrence Livermore National Lab

Livermore, CA 94551

C.T. Wang‡

Department of Materials Science and Engineering

Stanford University

Palo Alto, CA 94305

Abstract

Fe/Si multilayers with antiferromagnetic interlayer coupling have been grown via ion-beam sputtering on both glass and single-crystal substrates. X-ray diffraction measurements show that both sets of films have crystalline iron silicide spacer layers and a periodic composition modulation. Films grown on glass have smaller crystallite sizes than those grown on single-crystal substrates and have a significant remanent magnetization. Films grown on single-crystal substrates have a smaller remanence. The observation of magnetocrystalline anisotropy in hysteresis loops and (*hkl*) peaks in x-ray diffraction demonstrates that the films grown on MgO and Ge are epitaxial. The smaller remanent magnetization in Fe/Si multilayers with better crystallinity suggests that the remanence is not intrinsic.

75.50.Bb,61.10.-i,68.65.+g,81.15.Cd,75.30.E

I. INTRODUCTION

Considerable experimental evidence supports the existing theories of interlayer exchange coupling across metallic spacer layers. While many details remain to be worked out, the basic picture of interlayer exchange interaction modulated by quantum well states in the metallic spacer layer is reasonably clear.[1] This picture would have to be extensively modified for insulating or semiconducting spacer layers, where the spacer does not possess a Fermi surface.[5,6] In particular the temperature dependence of the exchange coupling might be significantly different in ferromagnet/semiconductor multilayers where the exchange is mediated by thermally activated carriers.[5]

Unusual temperature-dependent magnetic properties have been reported for Fe/Si multilayers. For example, a large increase in the remanent magnetization has been observed at low-temperature in Fe/Si multilayers.[7,8] If the interlayer antiferromagnetic (AF) coupling increased with decreasing temperature, as in multilayers with metal spacers, one might expect the remanence instead to *decrease* at low temperature. Proper interpretation of the remanent magnetization in Fe/Si multilayers may therefore be important to understanding the origin of the interlayer coupling in this system.

One way to explore the origin of the remanence is to compare films of different crystalline quality. If the remanence has an intrinsic origin, like biquadratic coupling which causes orthogonal alignment of moments in Fe layers at low field,[2] then it should be only weakly dependent on film morphology.

II. EXPERIMENTAL METHODS

The films used in this study were grown using ion-beam sputtering (IBS) in a chamber with a base pressure of 2×10^{-8} torr. The deposition system is described in more detail elsewhere.[3] All samples used in this study were grown at a substrate temperature of 200° which was monitored by a thermocouple. All comparisons between films grown on glass and single-crystal substrates will be made on samples which were deposited *simultaneously* so as to eliminate any reproducibility issues.

The substrates used in this study were glass coverslips, MgO (001), Ge(001) and $Al_2O_3(0\bar{2}11)$. The MgO and Al_2O_3 substrates were cleaned according to a recipe reported by Farrow and coworkers.[4] The glass and Ge substrates were rinsed in solvents before loading into the vacuum chamber. All films are capped with a 200Å Ge oxidation barrier. The magnetic and structural properties of the films are stable for at least one year.

Most of the x-ray diffraction characterization has been performed using a 18kW rotating anode system outfitted with a graphite monochromator. The asymmetric and ϕ scans were taken using an x-ray tube source and a four-circle goniometer. Magnetization curves are obtained using SQUID and vibrating sample magnetometers.

III. STRUCTURAL CHARACTERIZATION

Epitaxial growth of Fe films on MgO and Al_2O_3 substrates is well-known.[9] One might therefore expect to be able to grow high-quality Fe/Si superlattices on these substrates. Figure 1a) shows high-angle x-ray diffraction spectra for a purely (001)-oriented (Fe40Å/SiÅ)x60 multilayer grown on MgO(001). The spectrum in Figure 1b) is data for a highly (011)-oriented (Fe40Å/Si14)x46 multilayer grown on Al_2O_3. Figure 2 shows a ϕ scan of the MgO and Fe [110] peaks for the film on the MgO substrate. These peaks are offset by 45° in ϕ, confirming the well-known epitaxial relation Fe(001) ∥ MgO(001) and Fe(110) ∥ MgO(100).[9] The ϕ scans for the Al_2O_3 substrate show that this film is only weakly oriented in-plane. Rocking curves width for both films are about 1° wide. Small-angle x-ray scattering data for these multilayers typically show three peaks, indicating a moderate degree of composition modulation. (Fe40Å/Si14Å) multilayers grown at lower substrate temperatures have as many as 7 low-angle x-ray peaks, but have poorer crystallinity and lower AF coupling.[3]

High-angle x-ray diffraction data for a (Fe40Å/Si14Å)x60 multilayer grown on glass are shown in Figure 1c). The observed peak is at an angle corresponding to (011)-textured growth. A single superlattice satellite is typically observed in multilayers on glass on the low-angle side in keeping with previous observations.[10] Rocking curves for these multilayers are typically 10° to 15° wide, indicating a large mosaic. Small-angle x-ray spectra on films grown on glass are similar to those for the films grown on the single-crystal substrates. These low-angle results indicate that the quality of layering is similar for films grown on different kinds of substrates. Therefore inferior crystallinity in the films grown on glass must be the reason for the absence of high-angle superlattice satellites.

The shape of the high-angle peaks and their superlattice satellites are described by a theory due to Fullerton *et al.*[13] Application of this theory to the Fe/Si multilayers is difficult because the silicide lattice constant, the thickness of the remaining pure Fe and the thickness of the silicide spacer can be estimated only roughly. A precise determination of the silicide lattice constant should make a quantitative analysis of these satellite features possible.

IV. MAGNETIC CHARACTERIZATION

Figure 3a shows magnetization curves for 40-repeat (Fe40Å/Si14Å) multilayers grown simultaneously on glass and MgO(001). The magnetization data is normalized to the highest measured value in order to facilitate comparison of the shape of the two curves. The high-field slopes of the two curves are close, suggesting that they will have similar saturation fields H_s. On the other hand, the remanent magnetization is 58% for the film on glass and 7% for the film grown on MgO. A remanence as low as 1% has been observed for other multilayers grown on MgO substrates. SQUID magnetometer data taken up to higher fields gives a saturation field of 9.75 kOe for the multilayer on MgO at room temperature. Assuming for a moment that the interlayer coupling is purely bilinear in nature, a well-known formula relates the saturation field to the AF coupling strength A_{12}

$$A_{12} = H_s M_s t_{Fe}/4$$

where M_s is the saturation magnetization and t_{Fe} is the thickness of an individual Fe layer.[14] Use of this equation with $H_s = 9.75$ kOe and the measured magnetization $M_s = 1271$ emu/cm^3 gives $A_{12} = 1.2$ erg/cm^2. Other films have had A_{12} at room temperature as large as 2.5 erg/cm^2. These AF coupling values are among the largest ever measured at room temperature, second only to 5 erg/cm^2 for Co/Ru multilayers[15] and a 2 erg/cm^2 biquadratic coupling found in CoFe/Mn multilayers.[16]

The magnetization of the Fe/Si multilayers is significantly reduced from the bulk Fe value of 1710 emu/cm^3. A saturation magnetization equal to 75% of the bulk value is typical for Fe/Si multilayers, and is explained on the basis of the significant interdiffusion that occurs in this system.[3] Saturation magnetization values do not depend on the choice of substrate.

Figure 3b shows magnetization curves for Fe100Å/Si14Å/Fe100Åtrilayer films grown on Al_2O_3 $(0\bar{2}11)$, Ge(001), and MgO(001) substrates. All three of the magnetization curves in Fig. 3b were taken with the field applied along an Fe(100) easy direction. Significant in-plane anisotropy of the magnetization curves occurs for the films on the Ge and MgO substrates, similar to what has been observed previously for Fe/Cr/Fe trilayers.[14] The observation of magnetocrystalline anistropy in the film on MgO but not in the film grown on Al_2O_3 is consistent with expectations from the ϕ scans, which show that the in-plane orientation of the film on MgO is much stronger. Up to this point the degree of in-plane orientation of the films on Ge has not been checked with x-ray diffraction, but the magnetization data predict a high degree of order.

The data in Figure 3b once again demonstrate that the degree of remanence in Fe/Si multilayers is strongly related to crystalline order. While the remanent magnetization of the epitaxial trilayers on Ge and MgO is only about 5% of the saturated value, the remanence of the polycrystalline trilayer on Al_2O_3 is close to 50%. The remanent magnetization of the trilayers on Ge and MgO is about 5% in the in-plane hard direction (H $\parallel$ Fe(110)) as well.

A SQUID magnetometer has been used to measure the magnetization curves of the IBS-grown Fe/Si multilayers at lower temperatures.[11] The temperature dependence of the remanent magnetization of these films is similar to that reported by other authors.[7,8]

V. DISCUSSION

The films grown on MgO and Ge are the only purely (001)-textured Fe/Si multilayers produced by IBS so far. Dekoster et al. have grown epitaxial Fe/FeSi multilayers on room-temperature MgO(001) by MBE, but they do not present any x-ray diffraction or magnetization data.[12] Mattson and coworkers have grown Fe/FeSi multilayers on Al_2O_3 by magnetron sputtering, but they do not specify the orientation of the substrate. At the moment it is not possible to tell why the crystalline ordering of the films grown on $Al_2O_3(0\bar{2}11)$ is inferior to that grown on the (001) MgO and Ge substrates. The difficulty with the Al_2O_3 growth may have to do with the 6° miscut of the substrates, or it may be due to an intrinsic difficulty with (011) growth of the Fe/Si multilayers. Previous work has shown that AF coupling in Fe/Si multilayers is dependent upon formation of a metastable iron silicide spacer layer phase.[3] The possibility exists that the spacer silicide does not grow well on Fe in the (011) orientation. This question can be answered only by further growth studies on better (011) substrates and careful structural characterizations.

A related question is whether the larger remanent magnet moment in the (011)-textured films might be due to a fundamental difference in magnetic properties from the (001)-textured films. Because the 46-repeat Fe/Si multilayer grown on Al_2O_3 has a remanence of only about 10%, this is unlikely. Undoubtedly the trilayer on Al_2O_3 has a higher remanence than the multilayer because the thinner film is more greatly impacted by the poor substrate surface quality. The staircase morphology caused by the 6° miscut of this Al_2O_3 substrate may lead to wavy interfaces between the Fe and iron silicide films or to pinholes through the silicide layers. Wavy interfaces can cause increased magnetostatic coupling or even biquadratic coupling,[17] both of which would tend to increase the remanence.

The issue of possible biquadratic coupling is also raised by the shape of the magnetization curves at high field. A small concave-downward curvature can be seen in the MgO data of Figure 3a, but often in samples grown at nominal room temperature, the curvature is considerably larger, and the high-field data can be fit to a parabola. This curvature is observed on several types of substrates and is not a diamagnetic correction. Slonczewski has proposed that the effect of pinholes may be to contribute higher-order terms to the energy expression, thereby mimicking biquadratic exchange coupling.[18] This pinhole-based explanation is an appealing one for the IBS-grown Fe/Si multilayers since the high-field concavity is consistently larger in films with higher remanence and broader high-angle x-ray diffraction peaks. Polarized neutron reflectometry experiments which are already underway may help to answer questions about non-collinear moment orientations in these films.[11]

Pinhole-influenced coupling may also explain the unusual temperature dependence of the remanence. The Fe atoms in the bridges through the silicide spacer layers may well have a reduced Curie point. A larger remanence at low temperature therefore makes sense if the remanence is derived from pinhole coupling and is not an intrinsic effect.

VI. CONCLUSIONS

By growing on a number of substrate materials and by using different deposition conditions, Fe/Si multilayers have been prepared with a varying degree of crystalline perfection. A large amount of accumulated evidence demonstrates that smaller crystallite size in Fe/Si multilayers is associated with higher remanence and larger high-field curvature of the magnetization curves. The remanence and high-field curvature are therefore not likely to be related to unusual exchange coupling but instead to originate from defects, perhaps pinholes through the silicide spacer layer. Since the remanent magnetization is caused by extrinsic effects, future studies should concentrate on other aspects of the magnetic behavior in order to learn more about the interlayer coupling.

We would like to thank E.E. Fullerton and Y. Huai for helpful discussions, and B.H. O'Dell and S. Torres for technical assistance. Part of this work was performed under the auspices of the U.S. Department of Energy by LLNL under contract No. W-7405-ENG-48.

REFERENCES

* chaiken@llnl.gov

† michel@cmsgee.llnl.gov

‡ ctwang@leland.stanford.edu

[1] K.B. Hathaway, in *Ultrathin Magnetic Structures I*, J.A.C. Bland and B. Heinrich eds., Springer-Verlag, New York (1994).

[2] Z. Celinski, B. Heinrich, and J.F.Cochran, J. Magn. Magn. Matls. **145**, L1 (1995).

[3] A. Chaiken, R.P. Michel, and M.A. Wall, Phys. Rev. **B**, submitted.

[4] R.F.C. Farrow, G.R. Harp, R.F. Marks, T.A. Rabedeau, J. Cryst. Growth **133**, 47 (1993).

[5] P. Bruno, Phys. Rev. **B49**, 13231 (1994).

[6] Z.-P. Shi, Rajiv R.P. Singh, and B.M. Klein, Europhys. Lett. **29**, 585 (1995).

[7] J.E. Mattson, S. Kumar, E.E. Fullerton, S.R. Lee, C.H. Sowers, M. Grimsditch, S.D. Bader and F.T. Parker, Phys. Rev. Lett. **71**, 185 (1993).

[8] K. Inomata, K. Yusu, and Y. Saito, Jpn. J. Appl. Phys. **33**, L1670 (1994).

[9] N. Metoki, M. Hofelich, T. Zeidler, and T. Muhge, J. Magn. Magn. Matls. **121**, 137 (1993).

[10] C.L. Foiles, M.R. Franklin, and R. Loloee, Mat. Res. Soc. Proc. **382**, to be published (1995).

[11] J.F. Ankner, H.J. Kaiser, R.P. Michel and A. Chaiken, to be published.

[12] J. Dekoster, A. Vantomme, S. Degroote, R. Moons, and G. Langouche, Mat. Res. Soc. Proc. **382**, to be published (1995).

[13] E.E. Fullerton, I.K. Schuller, H. Vanderstraeten, and Y. Bruynseraede, Phys. Rev. **45**, 9292 (1992).

[14] J.J. Krebs, P. Lubitz, A. Chaiken, and G.A. Prinz, Phys. Rev. Lett. **63**, 1645 (1989).

[15] S.S.P. Parkin, Phys. Rev. Lett. **67**, 3598 (1991).

[16] M.E. Filipkowski, C.J. Gutierrez, J.J. Krebs, and G.A. Prinz, J. Appl. Phys. **76**, 7090 (1994).

[17] J.C. Slonczewski, Phys. Rev. Lett. **67**, 3172 (1991).

[18] J.C. Slonczewski, J. Magn. Magn. Matls., to be published.

FIGURES

FIG. 1. High-angle x-ray diffraction spectra from Fe/Si multilayers grown on single-crystal substrates. a) Data for a (Fe40Å/Si14Å)x60 multilayer grown on MgO(001). The Fe(002) peak is shown with 5 satellites centered at 64.77°. b) Data for a (Fe40Å/Si14Å)x46 multilayer grown on $Al_2O_3(0\bar{2}11)$. Visible in the spectrum are the Al_2O_3 $(0\bar{2}11)$ peak at 37.79° and the Fe(011) peak centered at 44.99° with its 4 satellites.

c) Data for a (Fe40Å/Si14Å)x60 multilayer grown on glass at the same time as the multilayer grown on MgO.

FIG. 2. ϕ scans plotted on a logarithmic scale for the MgO (110) and Fe(110) peaks of the (Fe40Å/Si14Å)x60 multilayer grown on MgO(001). The Fe (110) direction makes an angle of 45° with the MgO (110) direction, as expected, but small amounts of secondary orientations are visible.

FIG. 3. Magnetization curves for Fe/Si multilayers grown on various substrates. a) Hysteresis loops for the (Fe40Å/Si14Å)x60 multilayers simultaneously grown on MgO (001) and glass. The remanence is lower for the film grown on MgO. b) Hysteresis loops for (Fe100Å/Si14Å)x2 multilayers (trilayers) grown on Al_2O_3 $(0\bar{2}11)$, Ge(001), and MgO(001). All data are taken with the applied magnetic field along the Fe (100) easy direction.

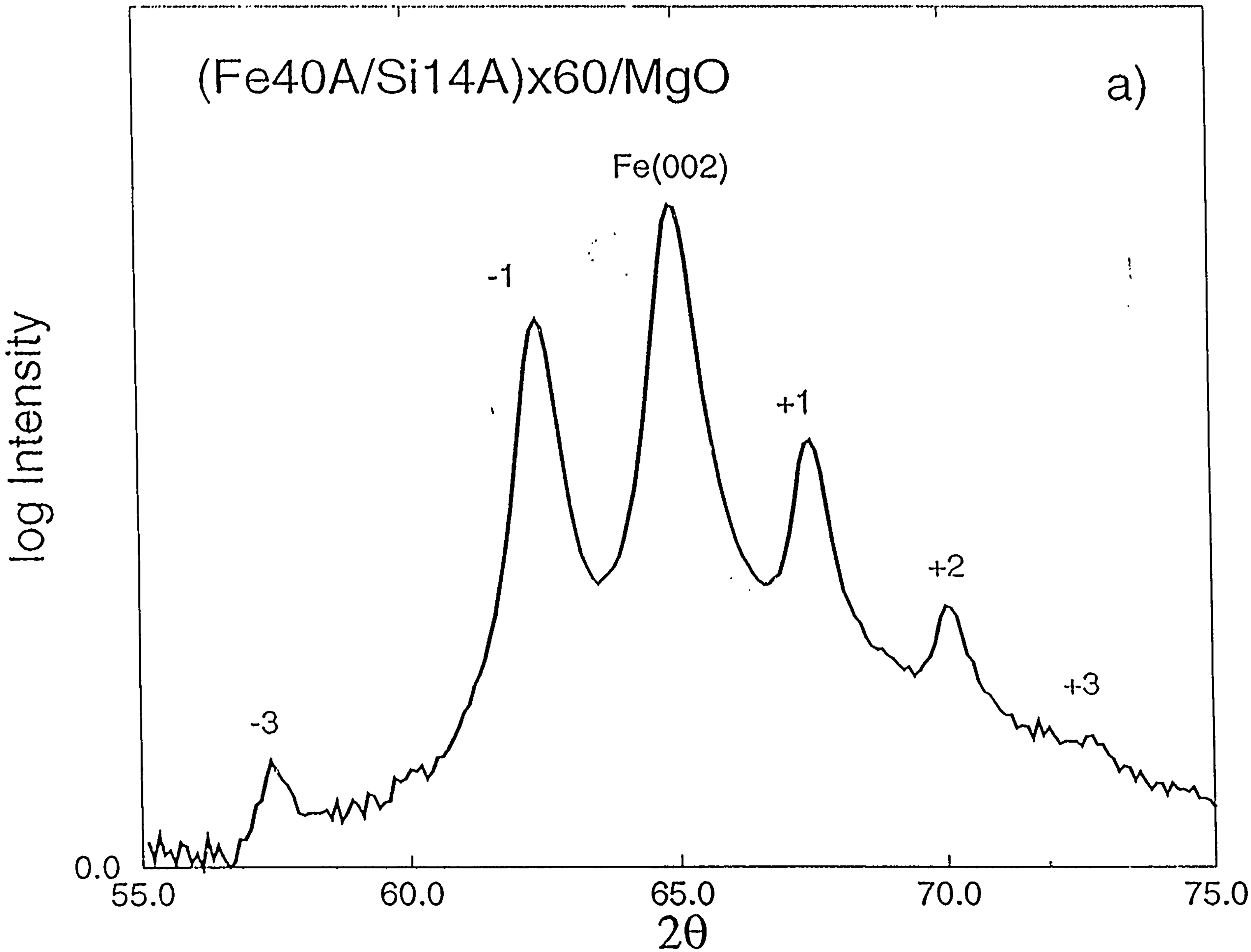
(Fe40A/Si14A)x60/MgO
a)
Fe(002)
-1
+1
+2
-3
+3
log Intensity
0.0
55.0
60.0
65.0
70.0
75.0
2θ

Figure 1a

X Graph

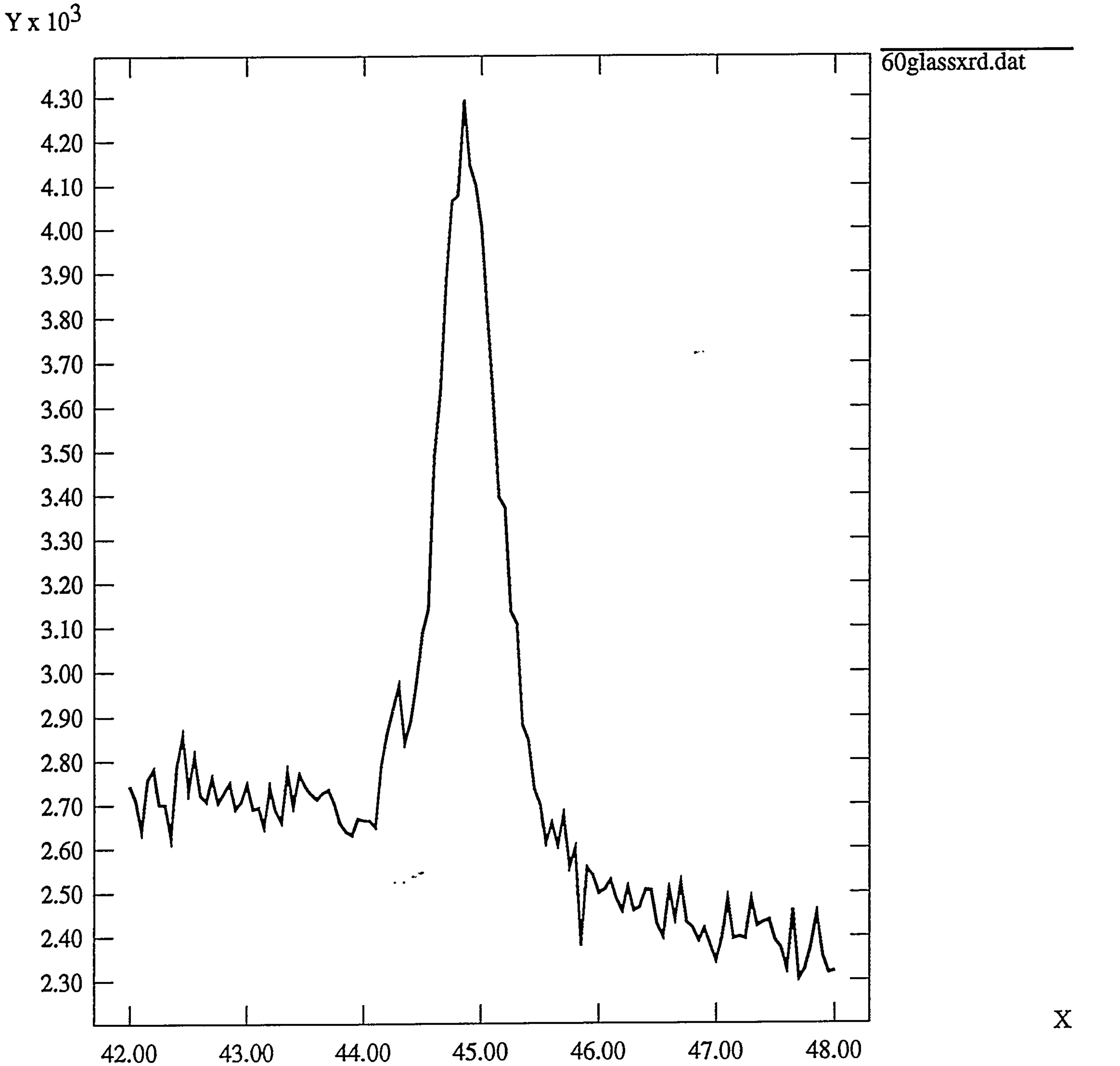

Figure 1(c)

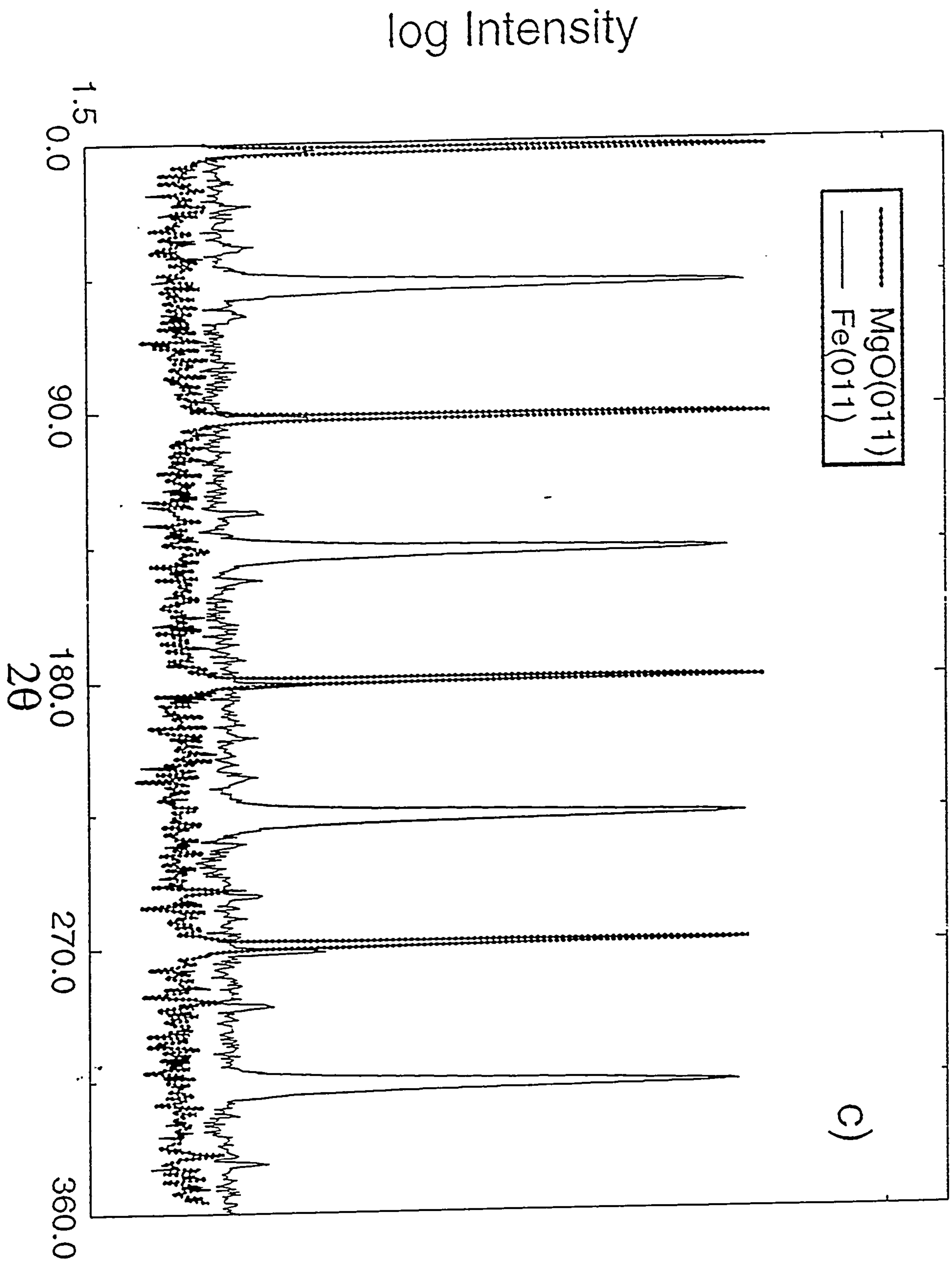

Figure 2

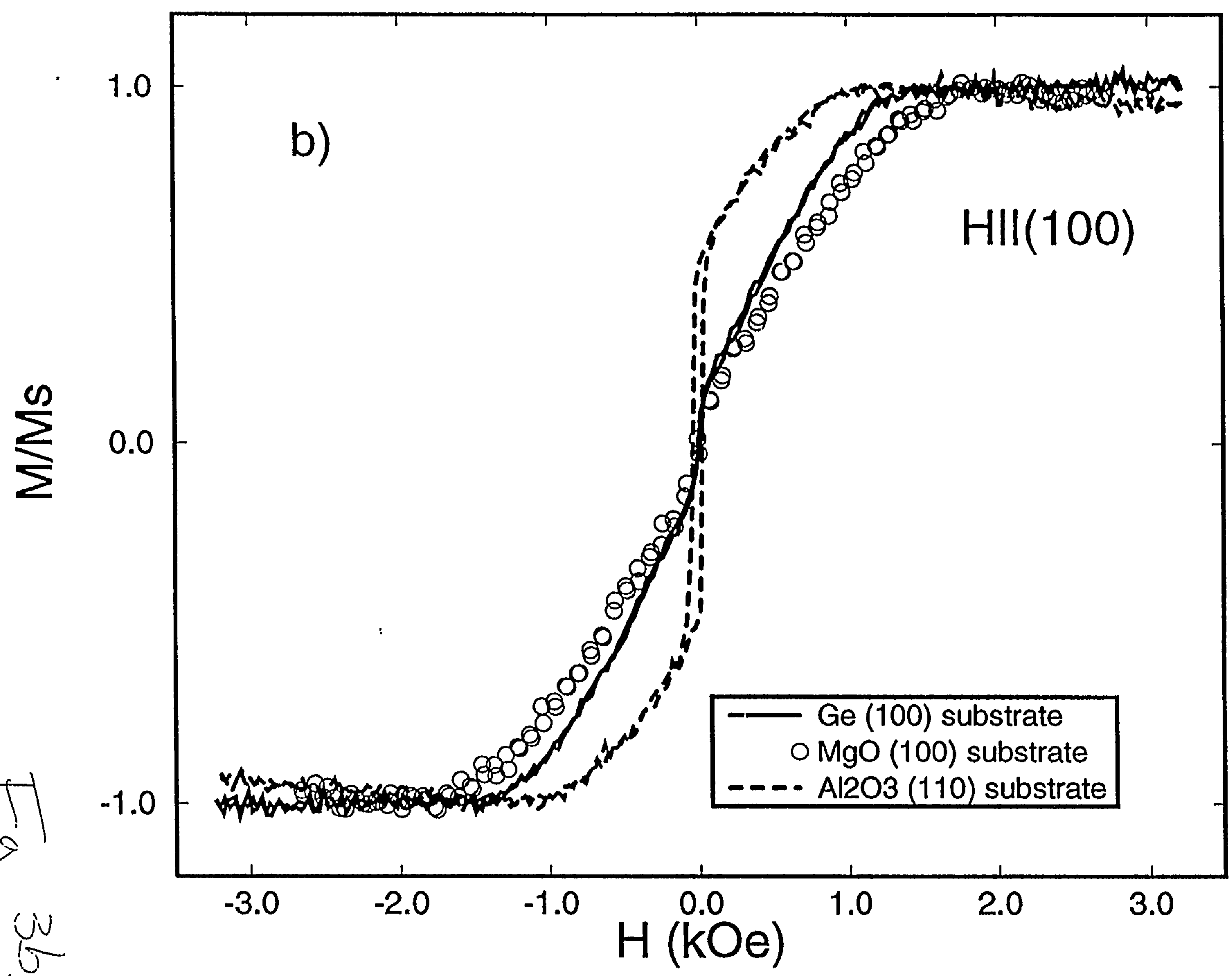
b)
HII(100)
M/Ms
1.0
0.0
-1.0
-3.0
-2.0
-1.0
0.0
1.0
2.0
3.0
H (kOe)
Ge (100) substrate
MgO (100) substrate
Al2O3 (110) substrate

Fig 3b)

Technical Information Department · Lawrence Livermore National Laboratory
University of California · Livermore, California 94551

CONF-951101--5

UCRL-JC- 121930
PREPRINT

Magnetic Properties of Epitaxial and Polycrystalline Fe/Si Multilayers

A. Chaiken
R. P. Michel
C. T. Wang

This paper was prepared for submittal to
Magnetism and Magnetic Materials Conference,
November 6-9, 1995, Philadelphia, Penna., and published in the
Journal of Applied Physics

August 1995

Lawrence Livermore National Laboratory

DISCLAIMER

Magnetic Properties of Epitaxial and Polycrystalline Fe/Si Multilayers

A. Chaiken* and R.P. Michel†

Materials Science and Technology Division

Lawrence Livermore National Lab

Livermore, CA 94551

C.T. Wang‡

Department of Materials Science and Engineering

Stanford University

Palo Alto, CA 94305

Abstract

Fe/Si multilayers with antiferromagnetic interlayer coupling have been grown via ion-beam sputtering on both glass and single-crystal substrates. X-ray diffraction measurements show that both sets of films have crystalline iron silicide spacer layers and a periodic composition modulation. Films grown on glass have smaller crystallite sizes than those grown on single-crystal substrates and have a significant remanent magnetization. Films grown on single-crystal substrates have a smaller remanence. The observation of magnetocrystalline anisotropy in hysteresis loops and (hkl) peaks in x-ray diffraction demonstrates that the films grown on MgO and Ge are epitaxial. The smaller remanent magnetization in Fe/Si multilayers with better crystallinity

suggests that the remanence is not intrinsic.

75.50.Bb,61.10.-i,68.65.+g,81.15.Cd,75.30.E

Typeset using REVTEX

I. INTRODUCTION

Considerable experimental evidence supports the existing theories of interlayer exchange coupling across metallic spacer layers. While many details remain to be worked out, the basic picture of interlayer exchange interaction modulated by quantum well states in the metallic spacer layer is reasonably clear.[1] This picture would have to be extensively modified for insulating or semiconducting spacer layers, where the spacer does not possess a Fermi surface.[5,6] In particular the temperature dependence of the exchange coupling might be significantly different in ferromagnet/semiconductor multilayers where the exchange is mediated by thermally activated carriers.[5]

Unusual temperature-dependent magnetic properties have been reported for Fe/Si multilayers. For example, a large increase in the remanent magnetization has been observed at low-temperature in Fe/Si multilayers.[7,8] If the interlayer antiferromagnetic (AF) coupling increased with decreasing temperature, as in multilayers with metal spacers, one might expect the remanence instead to *decrease* at low temperature. Proper interpretation of the remanent magnetization in Fe/Si multilayers may therefore be important to understanding the origin of the interlayer coupling in this system.

One way to explore the origin of the remanence is to compare films of different crystalline quality. If the remanence has an intrinsic origin, like biquadratic coupling which causes orthogonal alignment of moments in Fe layers at low field,[2] then it should be only weakly dependent on film morphology.

II. EXPERIMENTAL METHODS

The films used in this study were grown using ion-beam sputtering (IBS) in a chamber with a base pressure of 2×10^{-8} torr. The deposition system is described in more detail elsewhere.[3] All samples used in this study were grown at a substrate temperature of 200° which was monitored by a thermocouple. All comparisons between films grown on glass and single-crystal substrates will be made on samples which were deposited *simultaneously* so as to eliminate any reproducibility issues.

The substrates used in this study were glass coverslips, MgO (001), Ge(001) and $Al_2O_3(0\bar{2}11)$. The MgO and Al_2O_3 substrates were cleaned according to a recipe reported by Farrow and coworkers.[4] The glass and Ge substrates were rinsed in solvents before loading into the vacuum chamber. All films are capped with a 200Å Ge oxidation barrier. The magnetic and structural properties of the films are stable for at least one year.

Most of the x-ray diffraction characterization has been performed using a 18kW rotating anode system outfitted with a graphite monochromator. The asymmetric and ϕ scans were taken using an x-ray tube source and a four-circle goniometer. Magnetization curves are obtained using SQUID and vibrating sample magnetometers.

III. STRUCTURAL CHARACTERIZATION

Epitaxial growth of Fe films on MgO and Al_2O_3 substrates is well-known.[9] One might therefore expect to be able to grow high-quality Fe/Si superlattices on these substrates. Figure 1a) shows high-angle x-ray diffraction spectra for a purely (001)-oriented (Fe40Å/SiÅ)x60 multilayer grown on MgO(001). The spectrum in Figure 1b) is data for a highly (011)-oriented (Fe40Å/Si14)x46 multilayer grown on Al_2O_3. Figure 2 shows a ϕ scan of the MgO and Fe [110] peaks for the film on the MgO substrate. These peaks are offset by 45° in ϕ, confirming the well-known epitaxial relation Fe(001) || MgO(001) and Fe(110) || MgO(100).[9] The ϕ scans for the Al_2O_3 substrate show that this film is only weakly oriented in-plane. Rocking curves width for both films are about 1° wide. Small-angle x-ray scattering data for these multilayers typically show three peaks, indicating a moderate degree of composition modulation. (Fe40Å/Si14Å) multilayers grown at lower substrate temperatures have as many as 7 low-angle x-ray peaks, but have poorer crystallinity and lower AF coupling.[3]

High-angle x-ray diffraction data for a (Fe40Å/Si14Å)x60 multilayer grown on glass are shown in Figure 1c). The observed peak is at an angle corresponding to (011)-textured growth. A single superlattice satellite is typically observed in multilayers on glass on the low-angle side in keeping with previous observations.[10] Rocking curves for these multilayers are typically 10° to 15° wide, indicating a large mosaic. Small-angle x-ray spectra on films grown on glass are similar to those for the films grown on the single-crystal substrates. These low-angle results indicate that the quality of layering is similar for films grown on different kinds of substrates. Therefore inferior crystallinity in the films grown on glass must be the reason for the absence of high-angle superlattice satellites.

The shape of the high-angle peaks and their superlattice satellites are described by a theory due to Fullerton *et al.*[13] Application of this theory to the Fe/Si multilayers is difficult because the silicide lattice constant, the thickness of the remaining pure Fe and the thickness of the silicide spacer can be estimated only roughly. A precise determination of the silicide lattice constant should make a quantitative analysis of these satellite features possible.

IV. MAGNETIC CHARACTERIZATION

Figure 3a shows magnetization curves for 40-repeat (Fe40Å/Si14Å) multilayers grown simultaneously on glass and MgO(001). The magnetization data is normalized to the highest measured value in order to facilitate comparison of the shape of the two curves. The high-field slopes of the two curves are close, suggesting that they will have similar saturation fields H_s. On the other hand, the remanent magnetization is 58% for the film on glass and 7% for the film grown on MgO. A remanence as low as 1% has been observed for other multilayers grown on MgO substrates. SQUID magnetometer data taken up to higher fields gives a saturation field of 9.75 kOe for the multilayer on MgO at room temperature. Assuming for a moment that the interlayer coupling is purely bilinear in nature, a well-known formula relates the saturation field to the AF coupling strength A_{12}

$$A_{12} = H_s M_s t_{Fe}/4$$

where M_s is the saturation magnetization and t_{Fe} is the thickness of an individual Fe layer.[14] Use of this equation with $H_s = 9.75$ kOe and the measured magnetization $M_s = 1271$ emu/cm^3 gives $A_{12} = 1.2$ erg/cm^2. Other films have had A_{12} at room temperature as large as 2.5 erg/cm^2. These AF coupling values are among the largest ever measured at room temperature, second only to 5 erg/cm^2 for Co/Ru multilayers[15] and a 2 erg/cm^2 biquadratic coupling found in CoFe/Mn multilayers.[16]

The magnetization of the Fe/Si multilayers is significantly reduced from the bulk Fe value of 1710 emu/cm^3. A saturation magnetization equal to 75% of the bulk value is typical for Fe/Si multilayers, and is explained on the basis of the significant interdiffusion that occurs in this system.[3] Saturation magnetization values do not depend on the choice of substrate.

Figure 3b shows magnetization curves for Fe100Å/Si14Å/Fe100Åtrilayer films grown on Al_2O_3 $(0\bar{2}11)$, Ge(001), and MgO(001) substrates. All three of the magnetization curves in Fig. 3b were taken with the field applied along an Fe(100) easy direction. Significant in-plane anisotropy of the magnetization curves occurs for the films on the Ge and MgO substrates, similar to what has been observed previously for Fe/Cr/Fe trilayers.[14] The observation of magnetocrystalline anistropy in the film on MgO but not in the film grown on Al_2O_3 is consistent with expectations from the ϕ scans, which show that the in-plane orientation of the film on MgO is much stronger. Up to this point the degree of in-plane orientation of the films on Ge has not been checked with x-ray diffraction, but the magnetization data predict a high degree of order.

The data in Figure 3b once again demonstrate that the degree of remanence in Fe/Si multilayers is strongly related to crystalline order. While the remanent magnetization of the epitaxial trilayers on Ge and MgO is only about 5% of the saturated value, the remanence of the polycrystalline trilayer on Al_2O_3 is close to 50%. The remanent magnetization of the trilayers on Ge and MgO is about 5% in the in-plane hard direction (H $\|$ Fe(110)) as well.

A SQUID magnetometer has been used to measure the magnetization curves of the IBS-grown Fe/Si multilayers at lower temperatures.[11] The temperature dependence of the remanent magnetization of these films is similar to that reported by other authors.[7,8]

V. DISCUSSION

The films grown on MgO and Ge are the only purely (001)-textured Fe/Si multilayers produced by IBS so far. Dekoster et al. have grown epitaxial Fe/FeSi multilayers on room-temperature MgO(001) by MBE, but they do not present any x-ray diffraction or magnetization data.[12] Mattson and coworkers have grown Fe/FeSi multilayers on Al_2O_3 by magnetron sputtering, but they do not specify the orientation of the substrate. At the moment it is not possible to tell why the crystalline ordering of the films grown on $Al_2O_3(0\bar{2}11)$ is inferior to that grown on the (001) MgO and Ge substrates. The difficulty with the Al_2O_3 growth may have to do with the 6° miscut of the substrates, or it may be due to an intrinsic difficulty with (011) growth of the Fe/Si multilayers. Previous work has shown that AF coupling in Fe/Si multilayers is dependent upon formation of a metastable iron silicide spacer layer phase.[3] The possibility exists that the spacer silicide does not grow well on Fe in the (011) orientation. This question can be answered only by further growth studies on better (011) substrates and careful structural characterizations.

A related question is whether the larger remanent magnet moment in the (011)-textured films might be due to a fundamental difference in magnetic properties from the (001)-textured films. Because the 46-repeat Fe/Si multilayer grown on Al_2O_3 has a remanence of only about 10%, this is unlikely. Undoubtedly the trilayer on Al_2O_3 has a higher remanence than the multilayer because the thinner film is more greatly impacted by the poor substrate surface quality. The staircase morphology caused by the 6° miscut of this Al_2O_3 substrate may lead to wavy interfaces between the Fe and iron silicide films or to pinholes through the silicide layers. Wavy interfaces can cause increased magnetostatic coupling or even biquadratic coupling,[17] both of which would tend to increase the remanence.

The issue of possible biquadratic coupling is also raised by the shape of the magnetization curves at high field. A small concave-downward curvature can be seen in the MgO data of Figure 3a, but often in samples grown at nominal room temperature, the curvature is considerably larger, and the high-field data can be fit to a parabola. This curvature is observed on several types of substrates and is not a diamagnetic correction. Slonczewski has proposed that the effect of pinholes may be to contribute higher-order terms to the energy expression, thereby mimicking biquadratic exchange coupling.[18] This pinhole-based explanation is an appealing one for the IBS-grown Fe/Si multilayers since the high-field concavity is consistently larger in films with higher remanence and broader high-angle x-ray diffraction peaks. Polarized neutron reflectometry experiments which are already underway may help to answer questions about non-collinear moment orientations in these films.[11]

Pinhole-influenced coupling may also explain the unusual temperature dependence of the remanence. The Fe atoms in the bridges through the silicide spacer layers may well have a reduced Curie point. A larger remanence at low temperature therefore makes sense if the remanence is derived from pinhole coupling and is not an intrinsic effect.

VI. CONCLUSIONS

By growing on a number of substrate materials and by using different deposition conditions, Fe/Si multilayers have been prepared with a varying degree of crystalline perfection. A large amount of accumulated evidence demonstrates that smaller crystallite size in Fe/Si multilayers is associated with higher remanence and larger high-field curvature of the magnetization curves. The remanence and high-field curvature are therefore not likely to be related to unusual exchange coupling but instead to originate from defects, perhaps pinholes through the silicide spacer layer. Since the remanent magnetization is caused by extrinsic effects, future studies should concentrate on other aspects of the magnetic behavior in order to learn more about the interlayer coupling.

We would like to thank E.E. Fullerton and Y. Huai for helpful discussions, and B.H. O'Dell and S. Torres for technical assistance. Part of this work was performed under the auspices of the U.S. Department of Energy by LLNL under contract No. W-7405-ENG-48.

REFERENCES

* chaiken@llnl.gov

† michel@cmsgee.llnl.gov

‡ ctwang@leland.stanford.edu

[1] K.B. Hathaway, in *Ultrathin Magnetic Structures I*, J.A.C. Bland and B. Heinrich eds., Springer-Verlag, New York (1994).

[2] Z. Celinski, B. Heinrich, and J.F.Cochran, J. Magn. Magn. Matls. **145**, L1 (1995).

[3] A. Chaiken, R.P. Michel, and M.A. Wall, Phys. Rev. **B**, submitted.

[4] R.F.C. Farrow, G.R. Harp, R.F. Marks, T.A. Rabedeau, J. Cryst. Growth **133**, 47 (1993).

[5] P. Bruno, Phys. Rev. **B49**, 13231 (1994).

[6] Z.-P. Shi, Rajiv R.P. Singh, and B.M. Klein, Europhys. Lett. **29**, 585 (1995).

[7] J.E. Mattson, S. Kumar, E.E. Fullerton, S.R. Lee, C.H. Sowers, M. Grimsditch, S.D. Bader and F.T. Parker, Phys. Rev. Lett. **71**, 185 (1993).

[8] K. Inomata, K. Yusu, and Y. Saito, Jpn. J. Appl. Phys. **33**, L1670 (1994).

[9] N. Metoki, M. Hofelich, T. Zeidler, and T. Muhge, J. Magn. Magn. Matls. **121**, 137 (1993).

[10] C.L. Foiles, M.R. Franklin, and R. Loloee, Mat. Res. Soc. Proc. **382**, to be published (1995).

[11] J.F. Ankner, H.J. Kaiser, R.P. Michel and A. Chaiken, to be published.

[12] J. Dekoster, A. Vantomme, S. Degroote, R. Moons, and G. Langouche, Mat. Res. Soc. Proc. **382**, to be published (1995).

[13] E.E. Fullerton, I.K. Schuller, H. Vanderstraeten, and Y. Bruynseraede, Phys. Rev. **45**, 9292 (1992).

[14] J.J. Krebs, P. Lubitz, A. Chaiken, and G.A. Prinz, Phys. Rev. Lett. **63**, 1645 (1989).

[15] S.S.P. Parkin, Phys. Rev. Lett. **67**, 3598 (1991).

[16] M.E. Filipkowski, C.J. Gutierrez, J.J. Krebs, and G.A. Prinz, J. Appl. Phys. **76**, 7090 (1994).

[17] J.C. Slonczewski, Phys. Rev. Lett. **67**, 3172 (1991).

[18] J.C. Slonczewski, J. Magn. Magn. Matls., to be published.

FIGURES

FIG. 1. High-angle x-ray diffraction spectra from Fe/Si multilayers grown on single-crystal substrates. a) Data for a (Fe40Å/Si14Å)x60 multilayer grown on MgO(001). The Fe(002) peak is shown with 5 satellites centered at 64.77°. b) Data for a (Fe40Å/Si14Å)x46 multilayer grown on $Al_2O_3(0\bar{2}11)$. Visible in the spectrum are the Al_2O_3 $(0\bar{2}11)$ peak at 37.79° and the Fe(011) peak centered at 44.99° with its 4 satellites.

c) Data for a (Fe40Å/Si14Å)x60 multilayer grown on glass at the same time as the multilayer grown on MgO.

FIG. 2. ϕ scans plotted on a logarithmic scale for the MgO (110) and Fe(110) peaks of the (Fe40Å/Si14Å)x60 multilayer grown on MgO(001). The Fe (110) direction makes an angle of 45° with the MgO (110) direction, as expected, but small amounts of secondary orientations are visible.

FIG. 3. Magnetization curves for Fe/Si multilayers grown on various substrates. a) Hysteresis loops for the (Fe40Å/Si14Å)x60 multilayers simultaneously grown on MgO (001) and glass. The remanence is lower for the film grown on MgO. b) Hysteresis loops for (Fe100Å/Si14Å)x2 multilayers (trilayers) grown on Al_2O_3 $(0\bar{2}11)$, Ge(001), and MgO(001). All data are taken with the applied magnetic field along the Fe (100) easy direction.

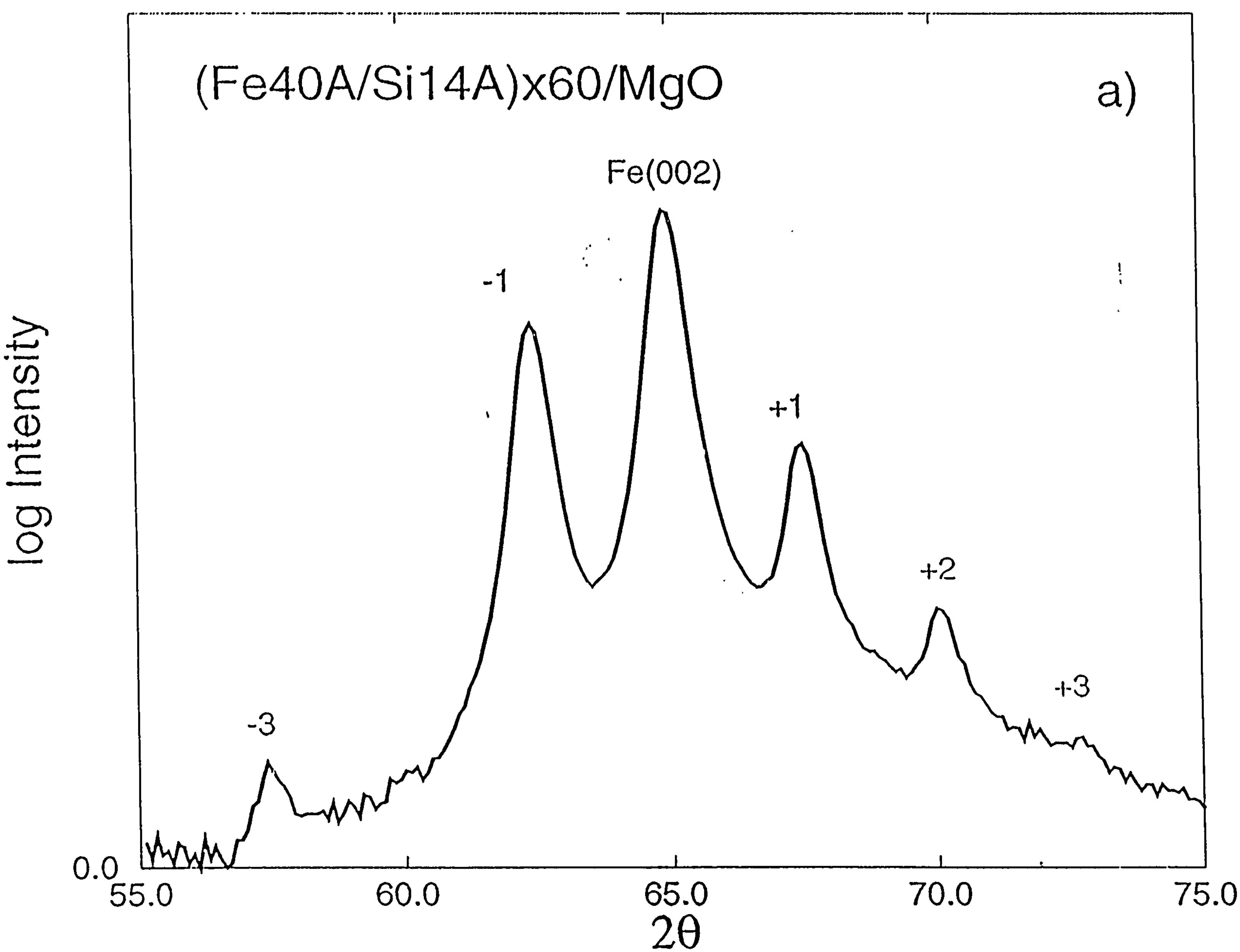
(Fe40A/Si14A)x60/MgO
a)
Fe(002)
-1
+1
+2
-3
+3
log Intensity
0.0
55.0
60.0
65.0
70.0
75.0
2θ

Figure 1a

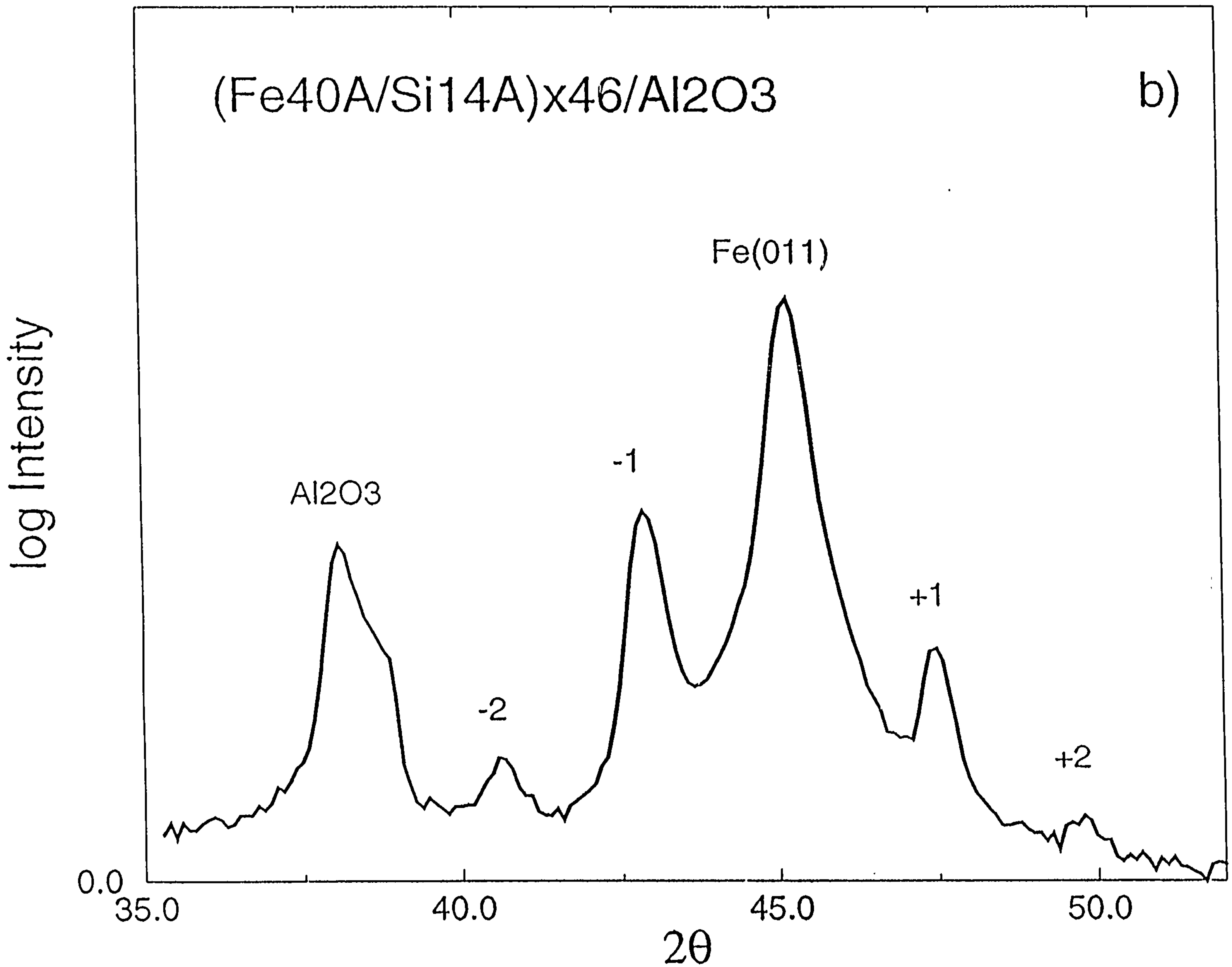
(Fe40A/Si14A)x46/Al2O3
b)
log Intensity
Al2O3
-2
-1
Fe(011)
+1
+2
0.0
35.0
40.0
45.0
50.0
2θ

Figure 1b)

X Graph

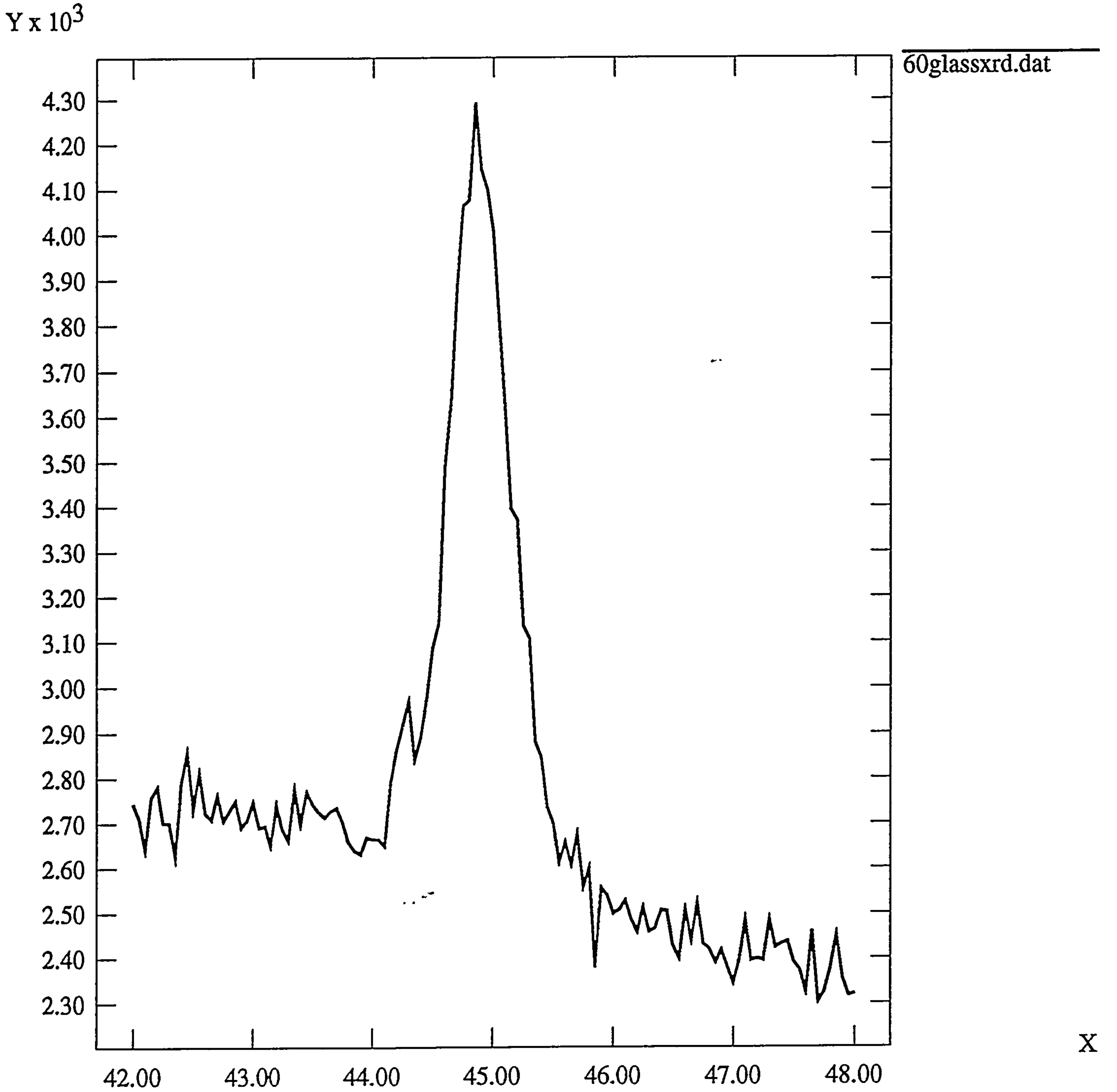

Figure 1(c)

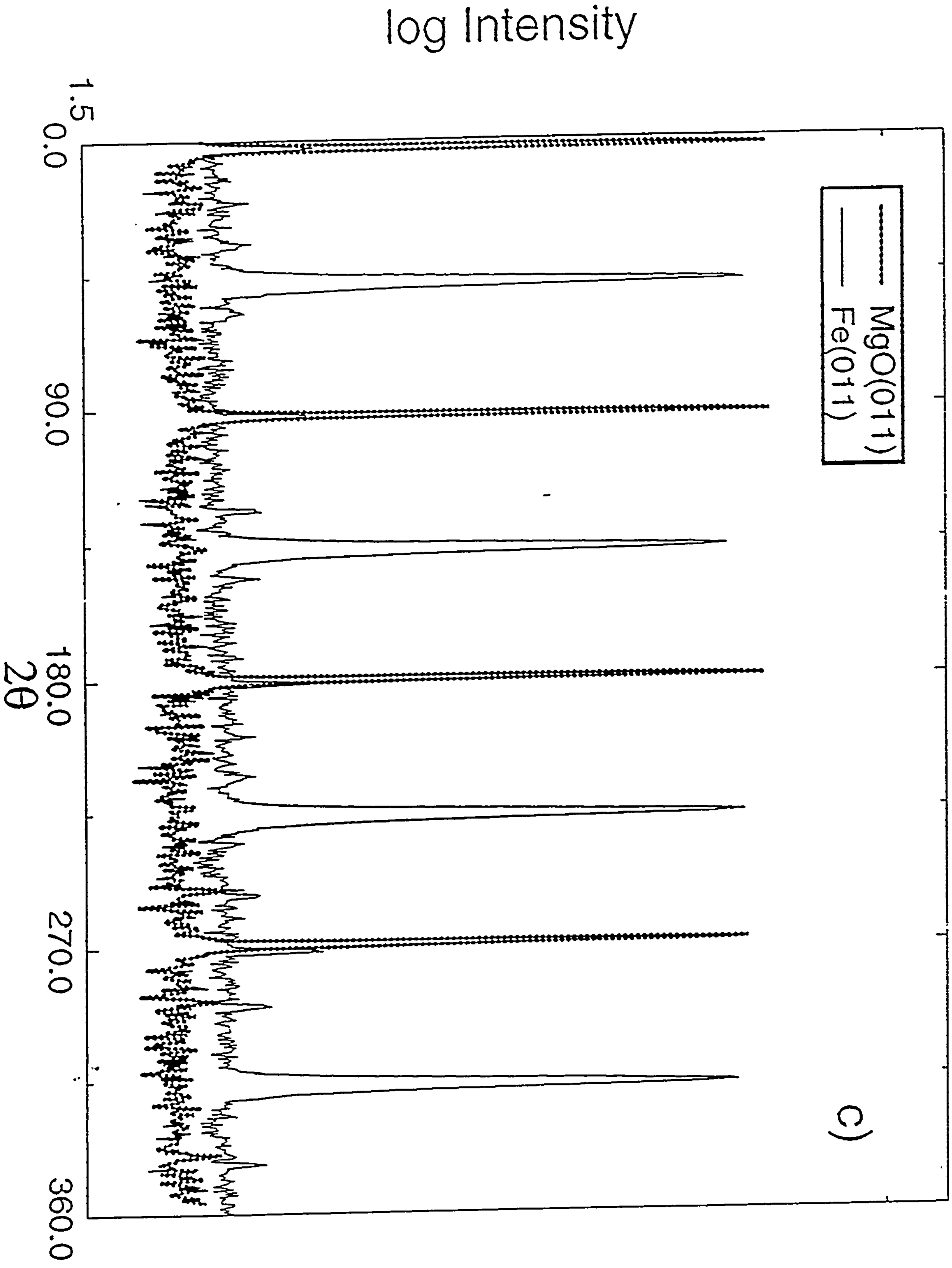

Figure 2

X Graph

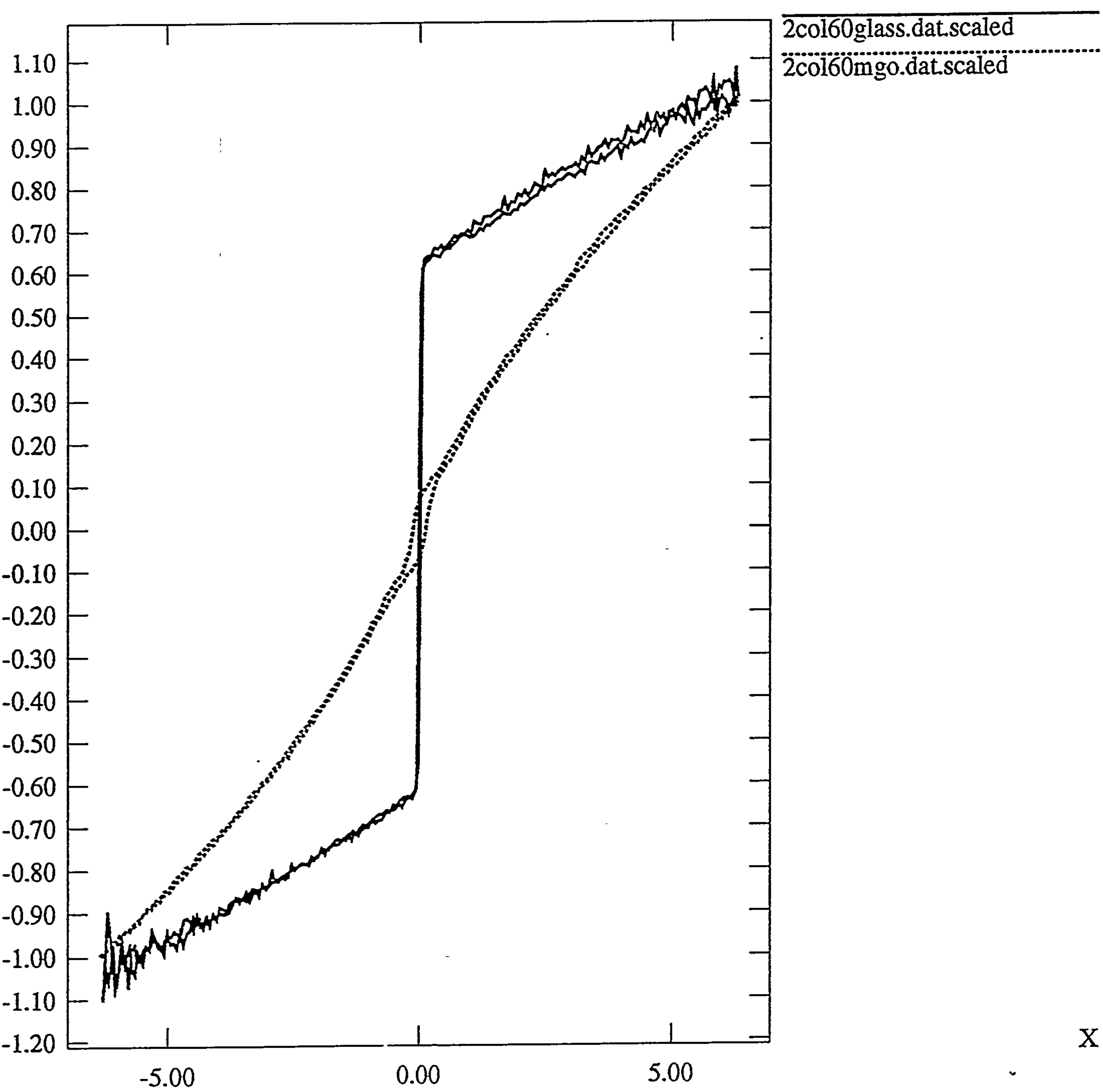

Fig 3a

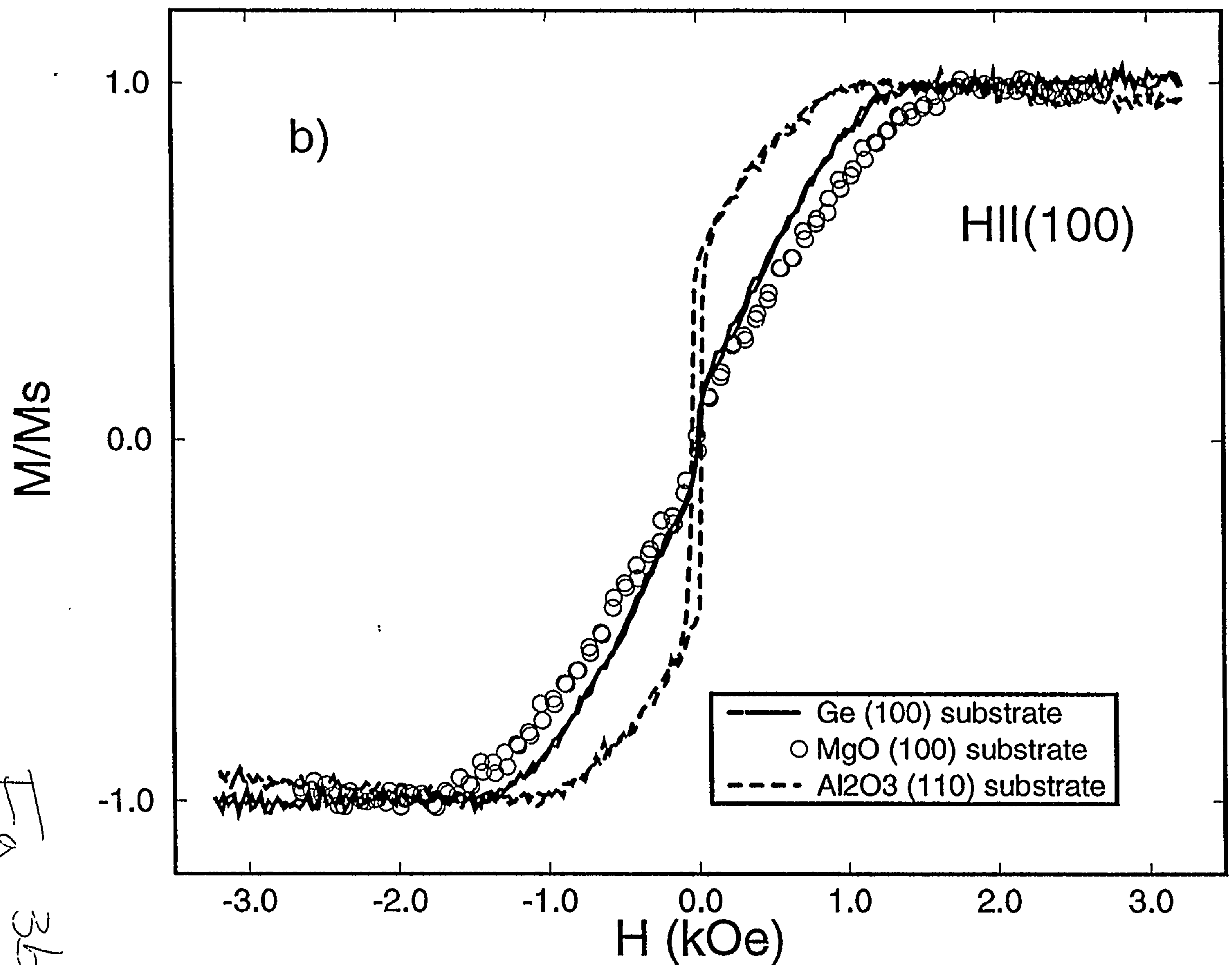
b)
H||(100)
M/Ms
1.0
0.0
-1.0
-3.0
-2.0
-1.0
0.0
1.0
2.0
3.0
H (kOe)
Ge (100) substrate
MgO (100) substrate
Al2O3 (110) substrate

Fig 3b)

Technical Information Department · Lawrence Livermore National Laboratory
University of California · Livermore, California 94551