

REPORT DOCUMENTATION PAGE

1a. REPORT SECURITY CLASSIFICATION

UNCLASSIFIED

1b. RESTRICTIVE MARKINGS

2a. SECURITY CLASSIFICATION AUTHORITY

3. DISTRIBUTION/AVAILABILITY OF REPORT

Approved for public release,
distribution unlimited

AD-A213 551

(S)

5. MONITORING ORGANIZATION REPORT NUMBER

AFOSR-TR-89-1280

6a. NAME OF PERFORMING ORGANIZATION

University of Minnesota

OFFICE SYMBOL
(If applicable)

7a. NAME OF MONITORING ORGANIZATION

SAME SA & 8a

6c. ADDRESS (City, State and ZIP Code)

Physics - 116 Church St. S.E.
Minneapolis, MN 55455

7b. ADDRESS (City, State and ZIP Code)

SAME AS 8c

8a. NAME OF FUNDING/SPONSORING
ORGANIZATION

AFOSR/NE

8b. OFFICE SYMBOL
(If applicable)

NE

9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER

AFOSR-86-C201

8c. ADDRESS (City, State and ZIP Code)

Bolling AFB
Washington, D.C. 20332-6448

10. SOURCE OF FUNDING NOS.

PROGRAM
ELEMENT NO.PROJECT
NO.TASK
NO.WORK UNIT
NO.

11. TITLE (Include Security Classification)

Epitaxial Iron Films

C11162F

2306

C1

12. PERSONAL AUTHOR(S)

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13a. TYPE OF REPORT

FINAL

13b. TIME COVERED

FROM 7/15/86 TO 7/15/89

14. DATE OF REPORT (Yr., Mo., Day)

15 September 1989

15. PAGE COUNT

22

16. SUPPLEMENTARY NOTATION

Final

17. COSATI CODES

FIELD GROUP SUB. GR.

18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)

19. ABSTRACT (Continue on reverse if necessary and identify by block number)

SEE ATTACHED SHEET

89 10 24 091

20. DISTRIBUTION/AVAILABILITY OF ABSTRACT

UNCLASSIFIED/UNLIMITED ☒ SAME AS RPT. ☐ DTIC USERS ☐

21. ABSTRACT SECURITY CLASSIFICATION

UNCLASSIFIED

22a. NAME OF RESPONSIBLE INDIVIDUAL

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22b. TELEPHONE NUMBER
(Include Area Code)

(212) 761-4933

22c. OFFICE SYMBOL

NE

Block 19.

The research program focused on the growth and the magnetic properties of epitaxial iron films grown on GaAs/InAs alloy substrates. Pseudomorphic growth of magnetic films on semiconductor substrates provides the potential for tuning of the magnetic properties of thin film magnets by varying the lattice constant and the morphology of the growth surface; this type of research has both device and fundamental research applications. Our studies of Fe films grown on GaAs have revealed a number of interesting phenomena including a dynamic Fe-FeO coupling, and a correlation between surface morphology and the coercivity of the films. In the growth studies reflection high energy electron diffraction determines the quality of the material (surfaces and interfaces) and the growth mechanisms of the metal films. The magnetic studies relied on the transport properties of the films supplemented by magnetization measurements. The transport measurements included the anisotropic magnetoresistance, the anomalous and regular Hall voltage and the planar Hall effect. The transport properties and some of the other magnetic studies were studied as a function of film thickness and crystallographic orientation.



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FINAL REPORT FOR
AFOSR GRANT NO. AFOSR-86-0201

SUBMITTED TO
AIR FORCE OFFICE OF SCIENTIFIC RESEARCH
BOLLING AFB, DC 20332-6448

Grant Title: Epitaxial Iron Films

Starting Date: 15 June 1986

Report Date: 30 July, 1988

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I. Statement of Work

The research program focused on the growth and characterization of epitaxial iron films grown on GaAs/InAs alloy substrates. Pseudomorphic growth of magnetic films on semiconductor substrates provides the potential for tuning of the magnetic properties of thin film magnets by varying the lattice constant and the morphology of the growth surface. This ability to tune the magnetic properties of materials and this type of research in general has both device and fundamental research applications. When grown on GaAs the Fe lattice is compressed from its equilibrium value by roughly 1.5%. With 100% InAs substrates, the Fe lattice would be expanded by roughly 7% (that an iron film would grow with this magnitude strain is unlikely however smaller values are not unreasonable). In general, the lattice constant of InGaAs alloys depends upon the mole fraction of In to Ga. Therefore by altering the mole fraction of In in a buffer layer, the magnetic properties of pseudomorphic Fe films can be modified and studied as a function of the Fe lattice constant. Our studies of Fe films grown on GaAs have revealed a number of interesting phenomena including a dynamic Fe-FeO coupling, and a correlation between surface morphology and the coercivity of the films. These studies, although interesting in their own right, have only scratched the surface of the potential of this research area.

In the growth studies reflection high energy electron diffraction determines the quality of the material and the growth mechanisms of the metal films. This part of the research provided information related to epitaxial processes in general and also provided the information to insure high quality surfaces and interfaces of the materials.

The magnetic studies relied primarily on the transport properties of the films supplemented by magnetization measurements using both a superconducting susceptometer and a magneto-optic magnetometer. The transport measurements included the anisotropic magnetoresistance, the anomalous and regular Hall voltage and the planar Hall effect. The transport properties and some of the other magnetic studies were studied as a function of film thickness and crystallographic orientation. The

magnetic studies were also used also as exploratory studies for potential device applications based on epitaxial magnetic films.

This sponsored research has resulted in eight publications (four published, two accepted for publication, and two to be submitted) involving the magnetics of the epitaxial magnetic films. Five of these publications involve the use of films kindly provided by Dr. G. Prinz of the Naval Research Laboratory. Much of the research was limited by the lack of samples, however, by extending the research to high quality sputtered films and with an increase in the manufacture of epitaxial films it is anticipated that the productivity will be increased in the new contract.

II. Magnetic Properties of Epitaxial Iron Films

The initial research on the magnetic properties of the epitaxial magnetic films focused on the transport properties of films grown on the (110) surface of GaAs substrates (the films were prepared by the group of Dr. G. Prinz at the Naval Research Laboratory in Washington D.C.). A limited study magnetic properties of films grown on (100) GaAs was accomplished (these latter films were grown by Prof. Cohen's group). We will first discuss the results of the transport studies of the (110) films and then the other studies of the (100) films by listing and discussing the publications which are either accepted for publication or are planned and last the areas of current research investigations.

A. "Saturation Magnetization and Perpendicular Anisotropy of Fe/GaAs (110) Epitaxial Films Studied by the Extraordinary Hall Effect," K.T. Riggs, Jan Dahlberg, and G. Prinz, Jour. of Magn, and Magn. Mat. 73, 46 (1988).

This paper involves measuring the extraordinary Hall effect (EHE) in (110) iron films. The study utilized eight films of varying thickness (from 20 nm). The study of the EHE provides a measure of the sum of the magnetization of the films and the surface anisotropy energy. This study provides two important results for the understanding of the magnetic

properties of epitaxial iron films. The first is that for films grown on the (110) surface of GaAs the magnitude of the surface anisotropy energy is not inordinately large. This result indicates the importance of the growth symmetry in epitaxial magnetic films since in (100) grown films the magnitude of this energy is sufficient to rotate the magnetization out of the plane of the film. The second important result included in this work is the variation of the results between transport measurements and ferromagnetic resonance which measures the same quantity. We have postulated that on the thinnest films the resonance measurement is observing a dynamic coupling of the metallic iron and the oxide on the surface whereas the transport is only sensitive to the metallic form. The implications of this result are that multilayered magnetic systems or magnetic systems where interfacial phenomena are important may have unique frequency dependent properties which could aid certain technologies.

B. "Magnetotransport: An Ideal Probe of Anisotropy Energies in Epitaxial Films," E. Dan Dahlberg, K.T. Riggs and G.A. Prinz, Invited paper to the 1987 Magnetism and Magnetic Materials Conference, J. of Appl. Phys. 63, 4270 (1988).

This invited paper stems from our use of transport to determine the magnetic properties of epitaxial magnetic films. In general the transport measurements provide an ideal tool to study the magnetic properties of epitaxial magnetic films. More traditional techniques such as SQUID magnetometry require the subtraction of any magnetic contribution of the substrate materials and although the substrate magnetism is small per unit of volume or mass, because the amount of substrate present is large it typically creates a difficult subtraction problem. The transport, on the other hand, measures only the metallic portion of the substrate/film combination and therefore for epitaxed metals on semiconductor/insulator substrates frees one of the usual subtraction problems.

C. "Magnetic Properties of MBE Grown (100) Iron Films," D.K. Lottis, E. Dan Dahlberg, S. Batra, A.M. Wowchak, and P.I. Cohen, J. of Appl. Phys. 63, 3662 (1988).

We have observed that the magnetic properties of (100) epitaxial iron films are dependent upon the surface preparation of the substrate. The iron films studied in this publication were grown on smoothed GaAs (epitaxed GaAs surface layer), chemically polished GaAs, and lattice matched InGaAs substrates. The measured coercivities were found to vary by a factor of four for the various substrates with the smooth GaAs sample having the smallest value. Although we do not presently have a detailed model to explain the origin of the effect it appears that both the lattice match and the smoothness of the substrate are very important in controlling the coercivity. By understanding exactly what the effects are and how they are controlled would allow us to grow iron films with prespecified coercivities for specific applications.

D. "Study of a Magnetic Field Induced First Order Phase Transition," K.T. Riggs, E. Dan Dahlberg, and G. Prinz, to be submitted to the Phys. Rev.

This manuscript is almost in a final version. It is anticipated that the manuscript will be submitted for publication by Aug., 1989. This study focuses on the rotation of the saturation magnetization in the plane of the epitaxial iron films and utilizes the anisotropic magnetoresistance to follow the rotation of the magnetization in the presence of magnetic fields applied parallel to the plane of the films. As mentioned in A. above, the surface anisotropy energy in the (110) films is insufficient to rotate the magnetization out of the plane of the films. With the magnetization pinned in the plane of the film then the rotation of the magnetization in the plane of the films as a function of the magnitude and direction of a magnetic field applied in the plane of the film can be modeled as a first order transition. The easy way to understand this behavior is to consider two easy axes separated by a hard axis in the plane of the film. If the

magnetization is required to rotate from one easy axis to the other by the application of a magnetic field, the applied magnetic field must be of sufficient strength that the magnetization can pass by the hard axis. Once this occurs the magnetization then abruptly (in a first order sense) makes the transition to the other easy axis. In the analysis of this behavior the uniaxial and fourth order anisotropy energies of the epitaxial films can be determined.

E. "Magnetic Domains in Epitaxial (100) Fe Thin Films," Jeffrey M. Florczak, P.J. Ryan, J.N. Kuznia, A.M. Wowchek, P.I. Cohen, R.M. White, G.A. Prinz, and E. Dan Dahlberg, accepted for publication as part of the proceedings of the 1989 MRS meeting.

This work shows how the surface morphology of the semiconductor substrate can influence the magnetic properties of the epitaxial magnetic films. It was found that the misfit dislocations which penetrate the surface of the InAs alloyed substrates can dominate the magnetic properties of the Fe films. The magnetics were investigated with both a magneto-optic magnetometer technique we developed (see below in other research) and with imaging Kerr microscopy. The most interesting feature of this research is the result that by correct preparation of the substrate the magnetization can be made almost isotropic in the plane of the film and controlled with modest applied magnetic fields (20 Oe).

F. Magnetic Anisotropy Constants of Epitaxial (110) Fe/GaAs Films From 77K to 293K Studied by Magneto-resistance," Daniel K. Lottis, G.A. Prinz, and E. Dan Dahlberg, accepted for publication as part of the proceedings of the 1989 MRS meeting.

Because the iron films are locked to the substrate the question arises as to the temperature dependence of various magnetic properties. In this paper we studied the temperature dependence of the anisotropy energies K_1

and K_u . A K_1 energy is also found in bulk iron whereas the K_u energy is unique to the epitaxial iron films. This paper determined that the K_u energy is consistent with a uniaxial strain arising from the growth of the metal film on the semiconductor at elevated temperatures. A comparison of the K_1 energy shows differences from that of bulk iron but the origin of the difference is uncertain.

G. Other research areas: The two other manuscripts which will be submitted and were funded in part by this contact (the remainder funded by the new contract) involve the exchange coupling of the FeO_x on the surface of the films with the underlying magnetic iron film and the use of magneto-optics to provide a simultaneous measure of the magnetization in two orthogonal directions in a magnetic film. The research on the exchange coupling between the surface oxide and the iron film is potentially very exciting. The Neel temperature of the oxide is roughly 90K and by studying the hysteresis loops of the iron films we have observed both an increase in the coercivity of the iron films and a shift in the hysteresis loops due to the exchange coupling to the antiferromagnetic oxide. Our investigation of this phenomena has focused on the temperature dependence of both of these quantities. We have thus far been utilizing a SQUID magnetometer in these studies but are switching the research to make use of a VSM for better efficiency (one M-H loop on the SQUID system requires 3 hours as compared to the 5 minutes on the VSM). At the present time a need for a detailed understanding of exchange coupling is necessary for several technologies from magneto-optic recording media to bias elements in thin film magnetometers.

The work on the use of magneto-optics to simultaneously measure the magnetization of two orthogonal components of the magnetization in a thin film is also very exciting. We have used this technique, which we developed, to study how the magnetization in the epitaxial films evolves in the presence of an applied magnetic field. It is this technique which provided the data we used to model the effects of surface morphology on the magnetism of the epitaxial films described earlier. A manuscript is in

draft form and it is expected to be submitted for publication by the middle of August, 1989. This technique has utility other than for epitaxial films and can be used as a general technique to probe for in plane preferred growth directions in polycrystalline films.

III. The Growth of Pseudomorphic Epitaxial Iron Films

Our goal is to tailor the magnetic properties of thin magnetic films by control of substrate morphology and lattice constant. The interface is crucial. Prinz [Pr87] has speculated that interfacial reaction reduces M_s for very thin films. Farrow [Fa88] tried to study the role of the interface by using MBE to prepare surfaces and by varying the strain. But he found few differences in the magnetic properties of Fe either grown on lattice mismatched GaAs or lattice matched InGaAs. He was also concerned with the interfacial reaction, but found no evidence for an FeAs antiferromagnetic layer [Fa88]. Later work examined the efficacy of diffusion barriers. Thin Fe films grown on Ag/ZnSe/GaAs show perpendicular remanence [Ko87], while those grown on Ag crystals do not [St87]. For Fe on Ag on GaAs, Farrow [Fa88] found the interface to dominate the perpendicular anisotropy. In all of these the nucleation and growth of the films are still only partly understood. In our work we have prepared films of Fe on III-V substrates both with and without diffusion barriers. We have tried to combine quantitative measurements of epitaxy with measurements of magnetic properties.

The main result is that we have succeeded in growing Fe films in two different states of strain. This was accomplished by using GaAs(100) and InP(100) as templates, by preventing interdiffusion of In by first growing FeAl and then AlAs films, and by growing Fe near liquid nitrogen temperatures.

The starting point was the observed differences in growth that we had observed between Fe on GaAs and Fe on iron whiskers. In Fig. 1, a plot of the diffracted intensity vs time during the growth of Fe on an Fe(100) whisker is shown. The period of the observed oscillations corresponds to one monolayer of Fe. The growth is therefore layer-by-layer with an exceedingly sharp diffraction pattern (sharper than MBE grown GaAs). However growth of Fe on GaAs is by a three dimensional mode. We presented work at the National AVS meeting in Atlanta

showing that at one monolayer of Fe deposition on GaAs(100), there was a complete loss of long range order. Subsequent deposition gave a diffraction pattern in which the order was regained, but in which there was a preferred two dimensional island size of 2nm. A plot of the diffracted intensity vs parallel momentum transfer is shown in Fig. 2. The circle of diffracted intensity corresponds to nucleated islands at random positions. The island size was found to depend on the surface Fe coverage and substrate temperature. For subsequent Fe growth, the surface became rough. Since Fe grows layer by layer on Fe whiskers, we assume that As impurities and strain are the main difference. Similar results were reported at the same meeting by Stroscio (NIST) in which STM results showing the small three dimensional growth on GaAs(110). He reported some elongation of the clusters but not in the direction of the easy axis. Because of the rough initial growth, it is not possible to use diffraction to confirm whether the Fe is pseudomorphic. The clustering indicates that Fe is mobile at room temperature and above. With three dimensional growth, we expect defects and impurities to segregate at the juncture of islands. We do not expect to be able to understand to be able to characterize the interface. Some films were prepared for magnetic measurements, but the growth studies were not pursued.

To minimize the interfacial reaction that occurs at the initial deposition of Fe we studied the growth on InGaAs films. Like the work of Farrow, the goal is to grow In(Ga,Al)As on GaAs past critical thickness, to serve as the starting point for Fe deposition. We examined the role of AlAs and FeAl buffers. From thermodynamic arguments these are expected to have no driving force for reaction. Fig. 3 shows a sequence of the relevant Auger measurements. In the top panel the first derivative of the ejected electrons is plotted vs electron energy for an InAlAs film. A small In peak is observed. Upon deposition of Fe at 200C the curve (b) shows a much larger In peak, perhaps indicating the Fe has uncovered or replaced In. After heating to 550C in (c) no Fe is observed. By contrast if Al is codeposited along with the Fe, as in (d), little additional In is observed even if annealed to 550C. The addition of Al to Fe results in an exceedingly smooth surface. The diffraction beams are nearly as sharp as those from an Fe whisker. One can grow Fe_xAl_y with a range of mole fractions. These all grow in a layer by layer mode, as evidenced by oscillations in the RHEED

intensity, and appear to be stable to 550C. If FeAl is grown, the monolayer time corresponds to the pair of Fe and Al alternating planes expected in the CsCl structure. Or if Fe₃Al is deposited, a growth period corresponding to the BiF₃ structure is observed. The latter is a magnetic phase. These films have so far been grown on AlAs or on InAlAs lattice matched to InP substrates.

When Fe is deposited onto these smooth FeAl layers, layer by layer growth is observed. Some Al segregates to the surface but a Ag film will likely stop prevent this. (Ludeke has shown that Ag has a 3D growth mode on GaAs but Heinrich sees 2D on Fe.) This was presented at the Electronic Materials Conference in Boston and at the conference on the Physics and Chemistry of Semiconductor Interfaces in Bozeman. (A.M. Wowchak, J.N. Kuznia, and P.I. Cohen, J. Vac. Sci. Technol., in press, 1989). Previously this had only been observed on Fe whiskers. Fig. 4 shows RHEED measurements of growth at 200C. About 9 layers can be put down at this temperature before a three dimensional growth mode is observed. The films have a 2.5% strain. We speculate that this change in growth mode corresponds to the formation of dislocations. Before the relaxation occurs, the pattern is very sharp. Fig. 5 shows the MH loop for a 10 nm film, but containing dislocations.

By depositing at about 150K, the bulk growth mode is not observed and the diffraction pattern remains quite sharp. The MH data is shown in Fig. 6. Notice that the loop is more rectangular, indicating fewer defects to pin domains. Finally to check whether Al segregates to the surface, we show a Auger measurement in Fig. 7 of the top of the 10nm Fe film. This film is as clean as Fe whiskers prepared in the same chamber. For the future we need to check for surface segregation of the components as a function of film thickness.

The results are exciting. We can now grow Fe films that are smooth enough that they can be characterized. The diffusion of In or As into the epilayer no longer seems to be a difficulty. We have changed the lattice constant of Fe. Films need to be grown on InGaAs in which the strain is minimized. We need to check to see whether the FeAl phases and Fe do not intermix. We need to measure the magnetic anisotropy of Fe₃Al. We need to measure the critical thicknesses for

the formation of dislocations.

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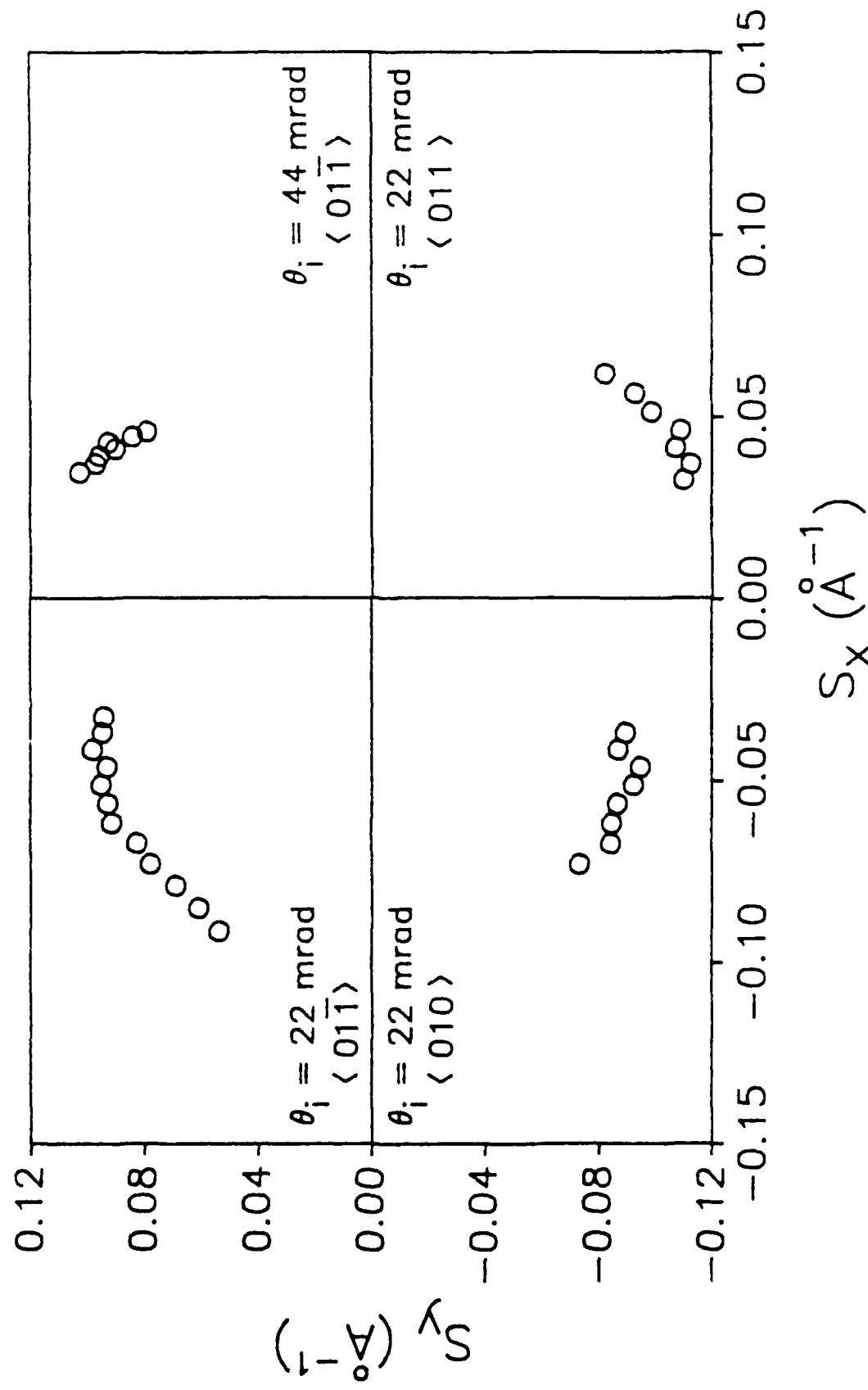
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FIGURE CAPTIONS

1. The specular diffracted intensity during the growth of Fe on Fe(100) whiskers.
2. The (00) diffracted intensity vs parallel momentum transfer for several glancing angles of incidence and azimuthal directions. This indicates that islands have formed during the deposition of Fe on GaAs(100).
3. dN/dE Auger signal comparing surface concentration of In and Fe after the deposition of Fe and FeAl on the pseudomorphic AlAs film on an InP substrate. (a) Initial AlAs film showing a few percent of In. (b) Between 3nm and 7nm of Fe was deposited on the AlAs film (c) Annealing to 820K (d) Addition of a small fraction of Al reduces the intermixing
4. RHEED intensity oscillations during the growth of Fe pseudomorphic on InP. The lattice mismatch is about 2.5%. The decrease in oscillations at 9 layers is interpreted as the thickness for dislocation formation. This is inhibited by growth at lower temperatures.
5. MH loop of 10nm of Fe grown on InP above the critical thickness
6. MH loop of 10nm of Fe grown on InP at low temperatures to inhibit dislocation formation.
7. Auger plot of the surface of the 10nm of Fe used above.

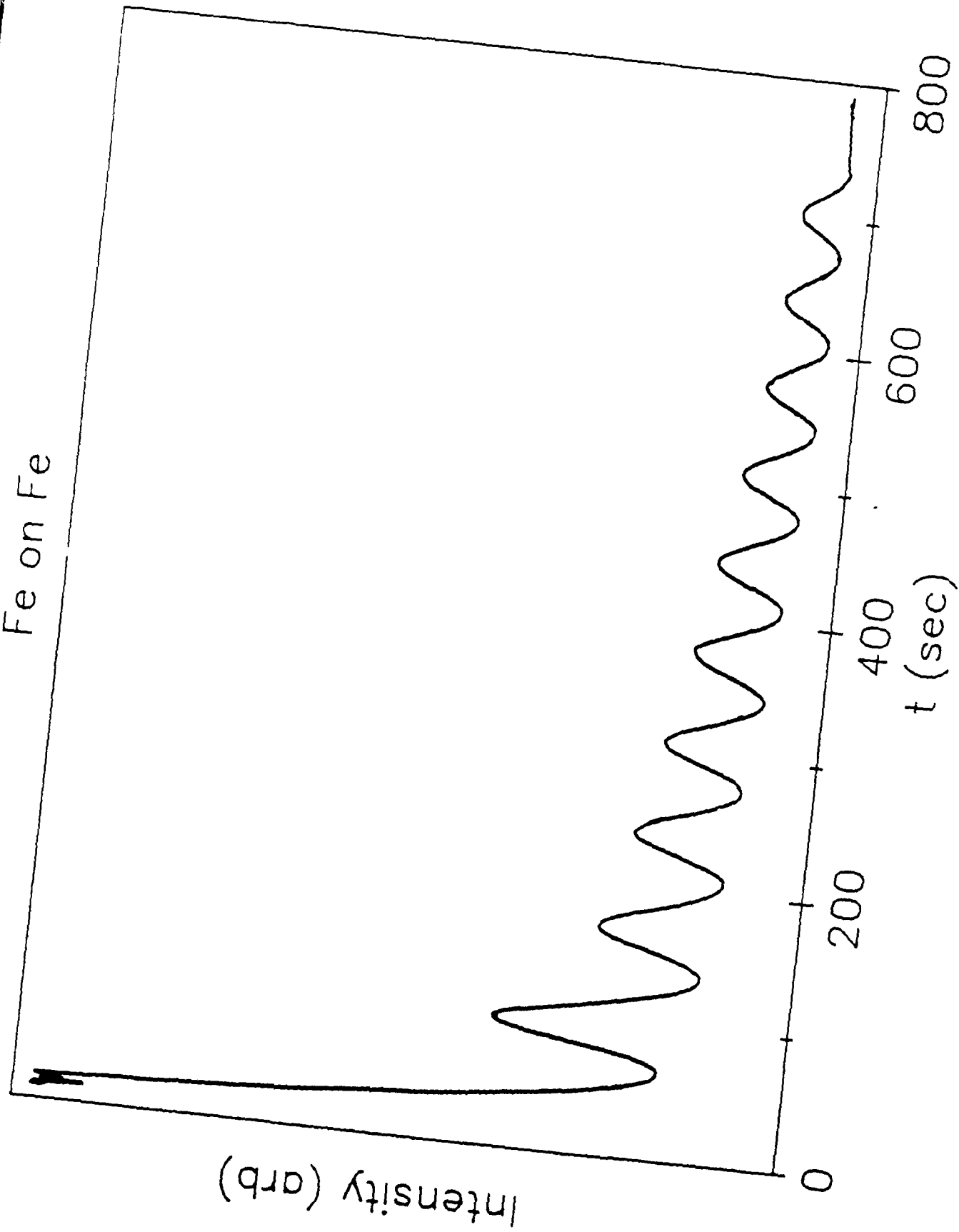
DIFFRACTION PROJECTION

3 Fe layers on GaAs (100); $T = 150^\circ\text{C}$

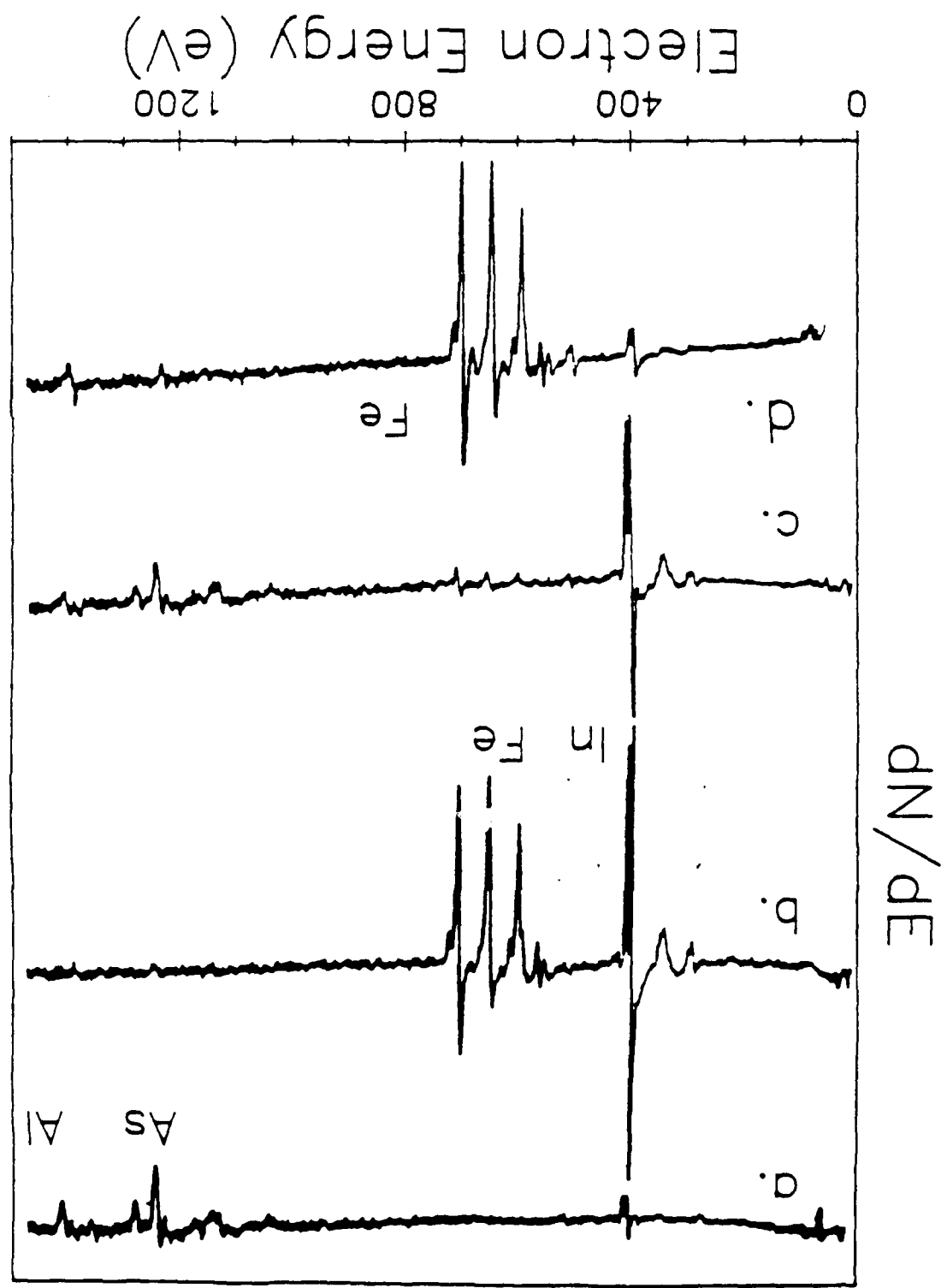


Fe on Fe

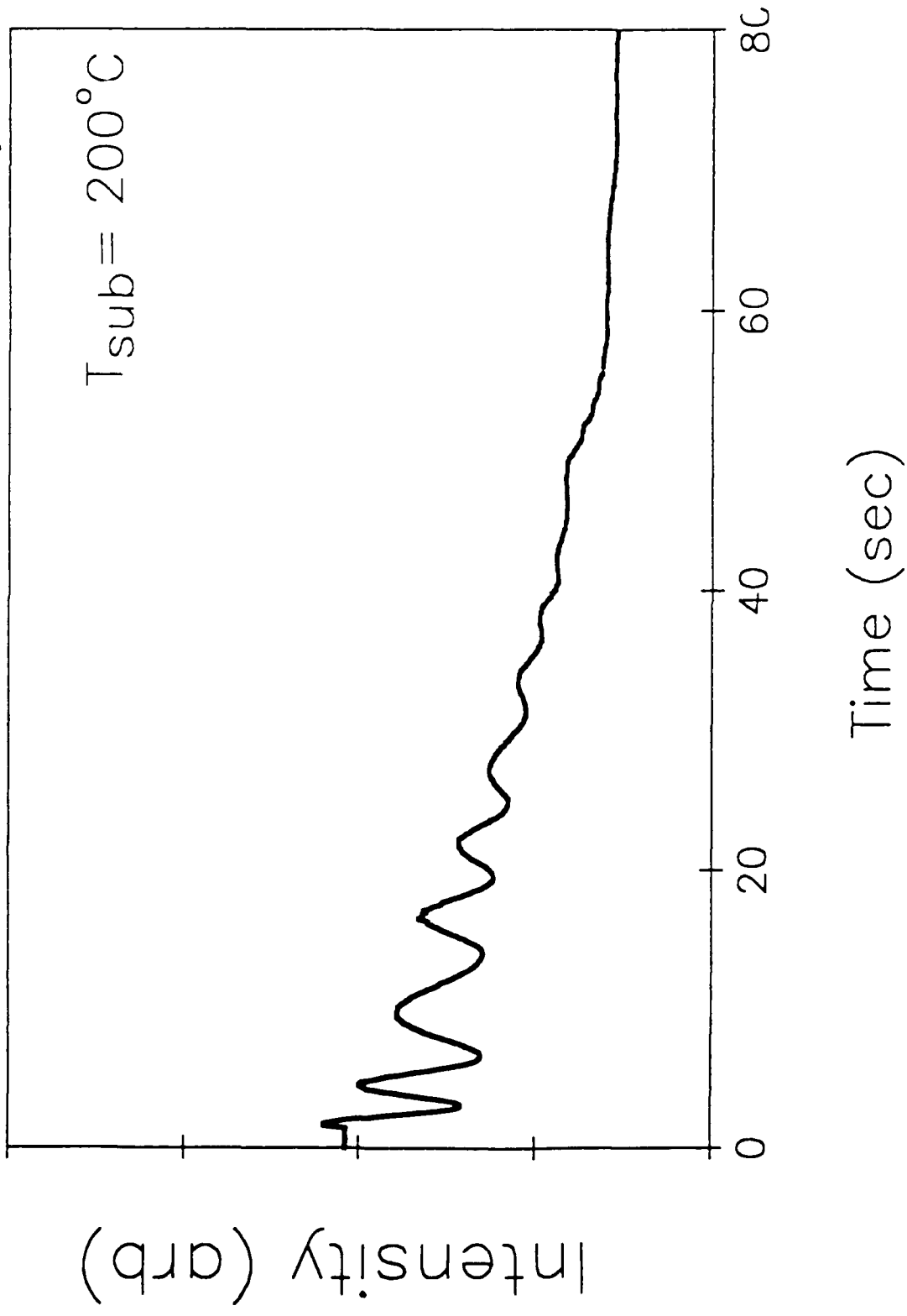
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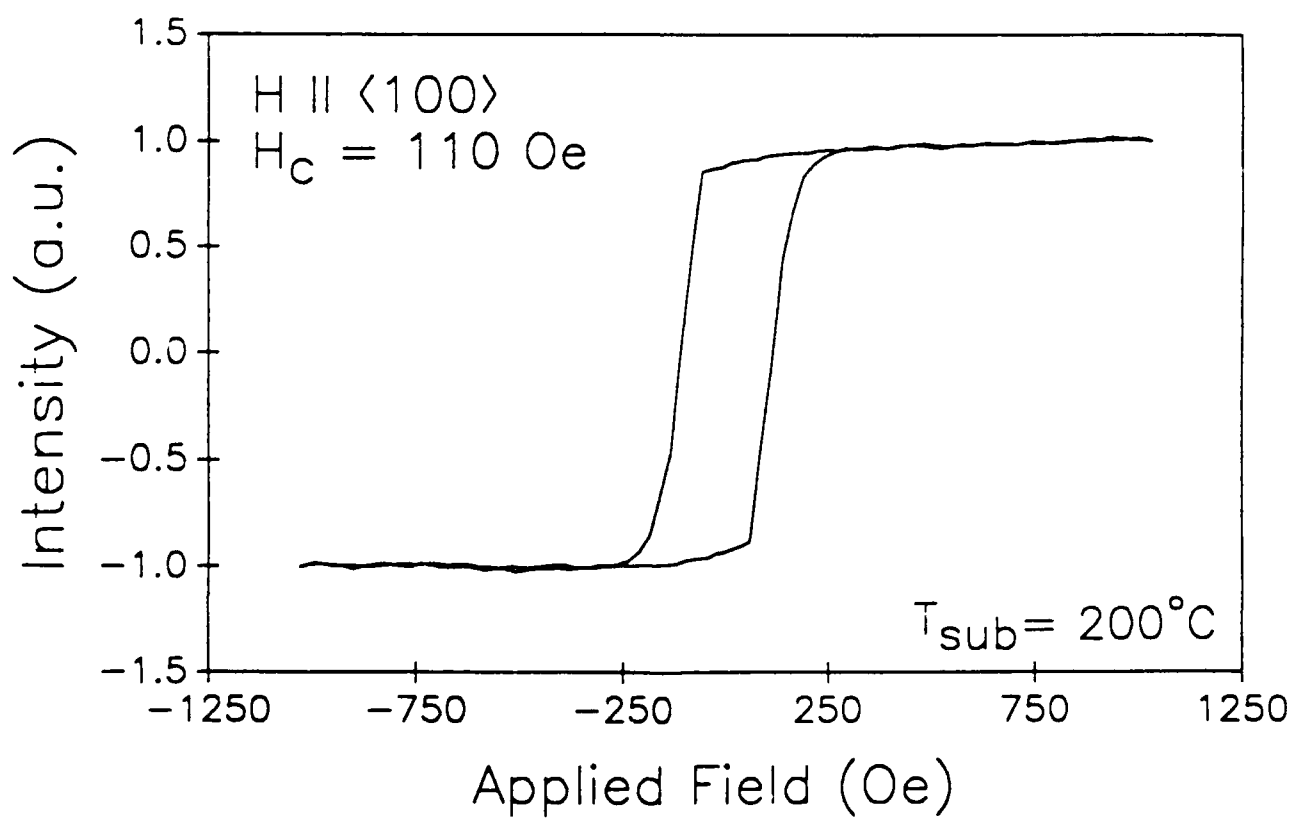
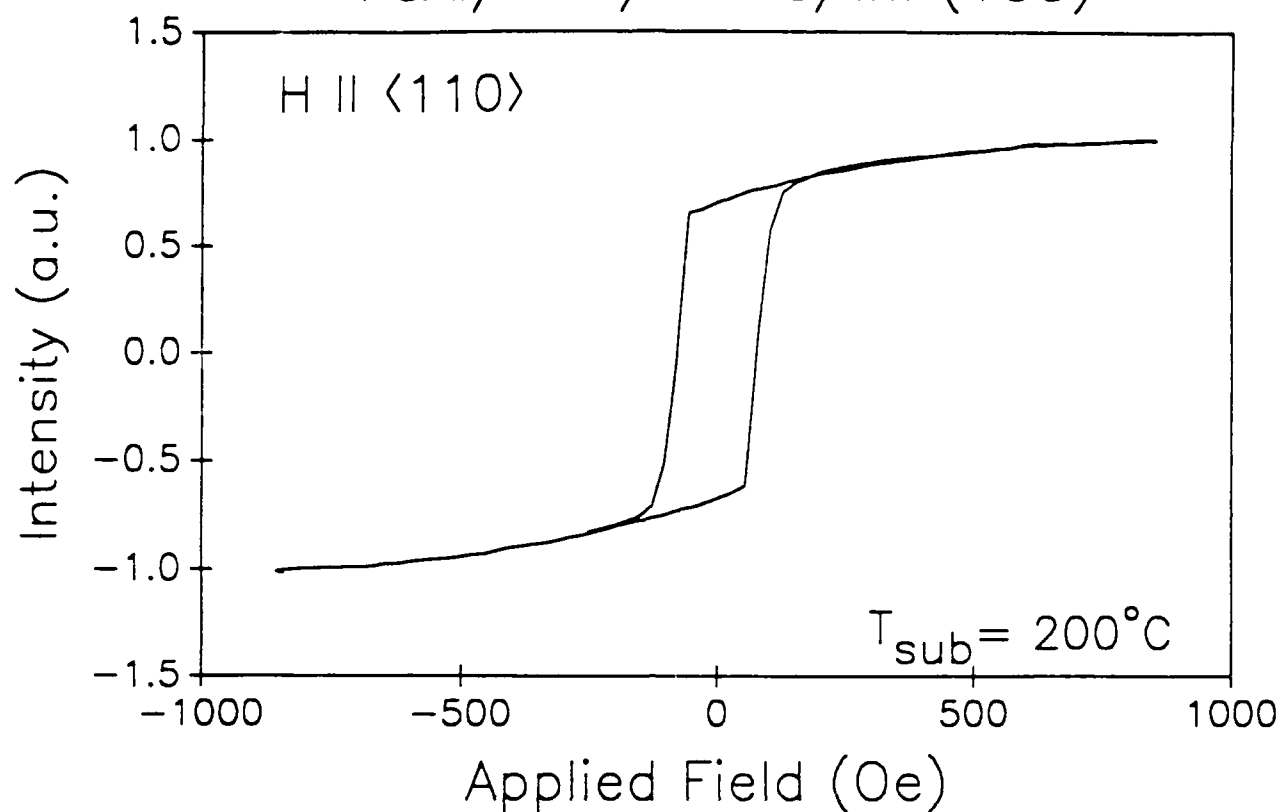
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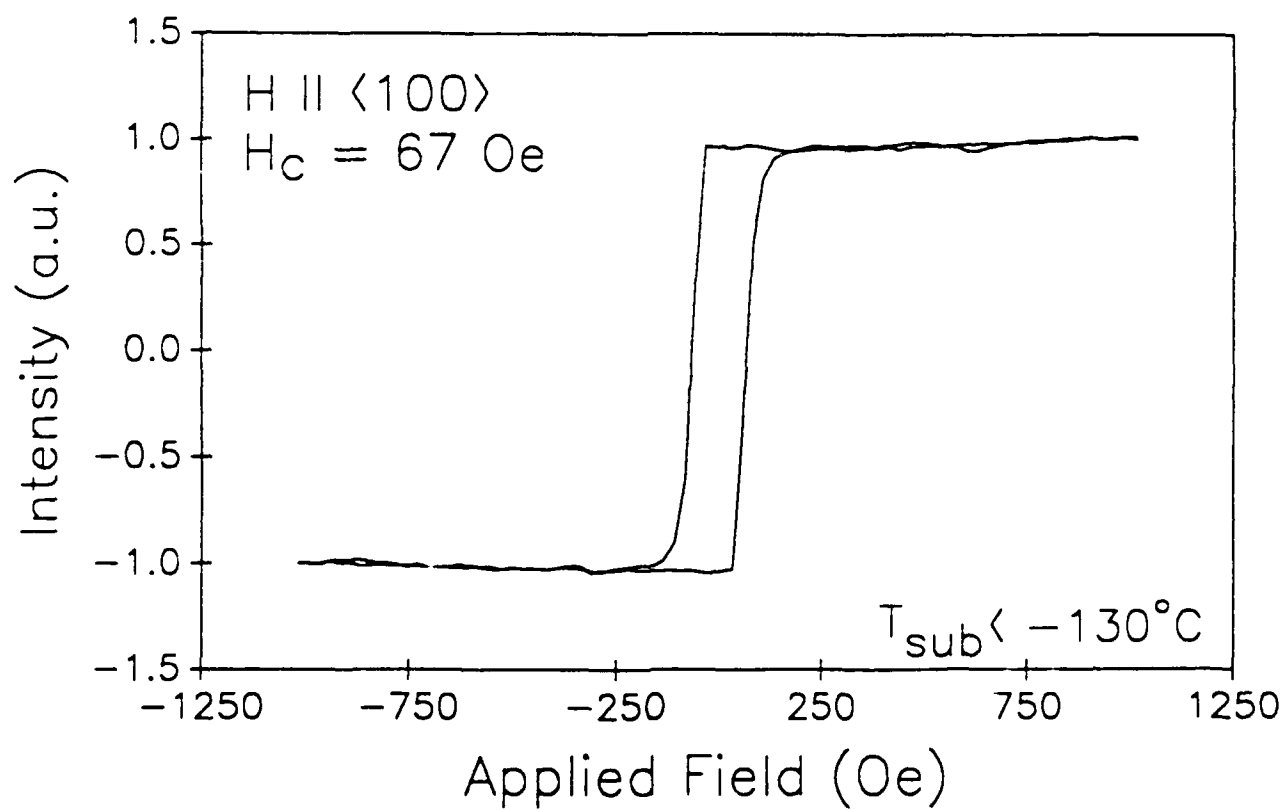
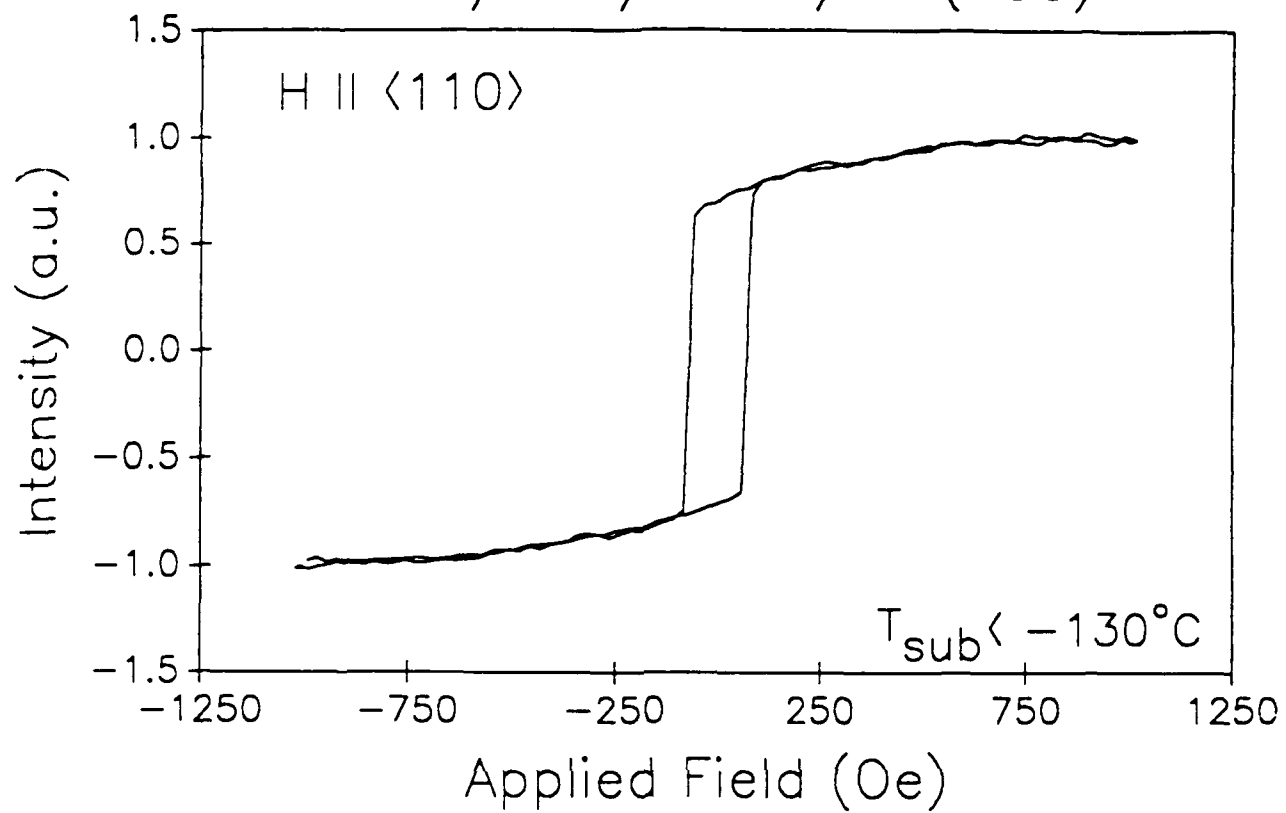
Fe/FeAl/AlAs/InAlAs/InP(100)
Critical Layer thickness: 9 monolayers



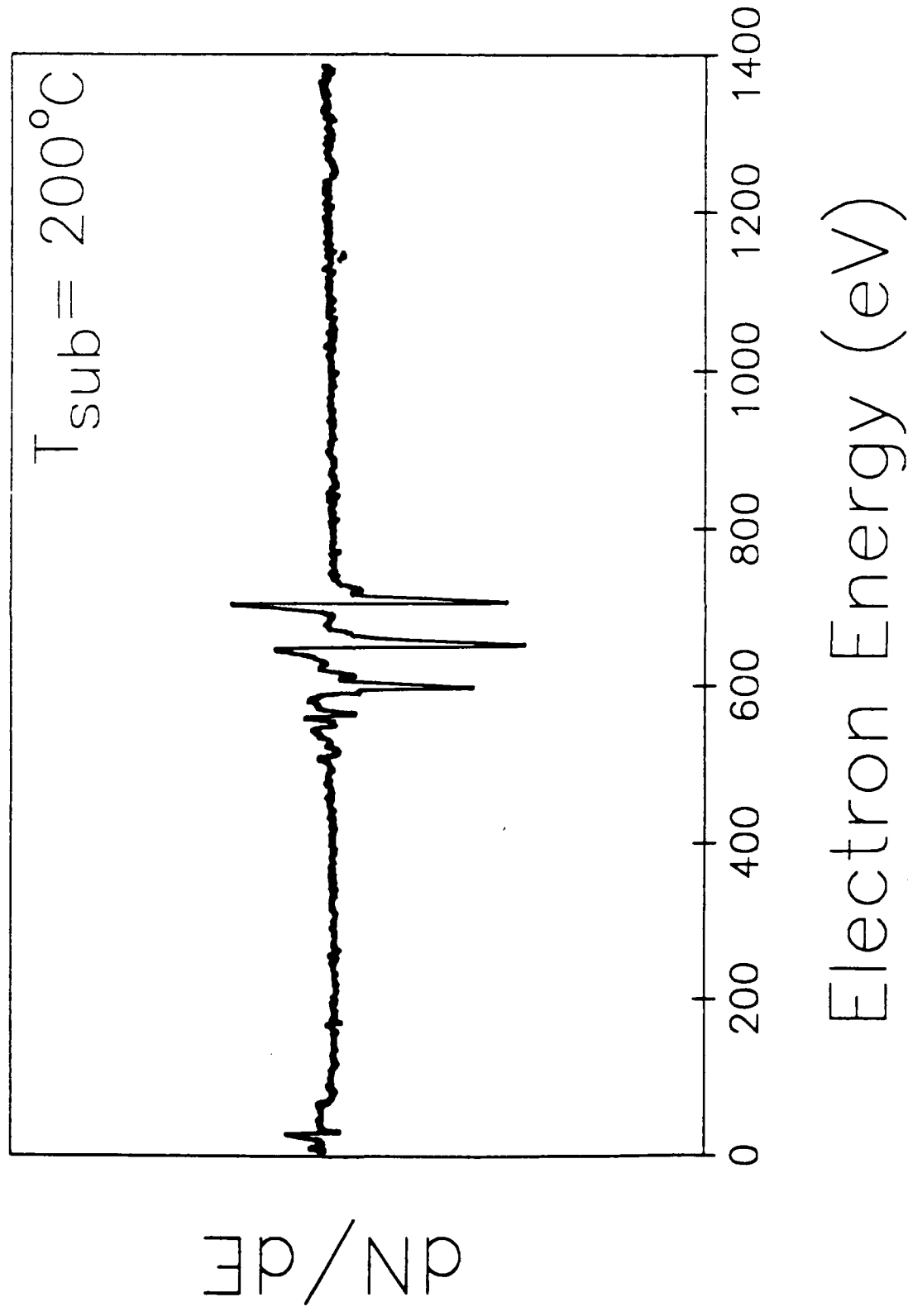
100 Å of Fe on annealed
FeAl/AlAs/InAlAs/InP(100)



100 Å of Fe on annealed
FeAl/AlAs/InAlAs/InP(100)



100 Å Fe on 50 Å of FeAl on
AlAs/InAlAs/InP(100)



IV. Personnel

A. Batra, S., Post-doctoral appointment has now left the group and is presently employed by Digital Equipment Co.

B. Jamison, K.D., Post-doctoral appointment is now presently employed by University of Houston.

C. Kuznia J.N., Graduate Student in Electrical Engineering

D. Wowchak, A.M., Graduate Student in Electrical Engineering

E. Riggs, K.T., Graduate Student in Physics (PhD received in 1989
presently an Assistant Professor at Stetson College in Florida)

F. Lottis, D.K., Graduate Student in Physics

G. Florczak, J.T., Graduate Student in Physics

H. Chen, You-Jun, Graduate Student in Physics