PHASE TRANSITIONS IN
THIN IRON-PALLADIUM FILMS

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I. INTRODUCTION

The magnetic behavior of iron is strongly influenced by temperature. Above the Curie temperature, 1043 K, pure iron is paramagnetic, while below 1043 K it is ferromagnetic. Each of these conditions is described by a different equation of state, and the transition is identified with divergent behavior of some thermodynamic functions. It is the purpose of this work to study such phase transitions in thin films of palladium alloys with iron serving as a 1-10% solute.

It is illustrative to compare and contrast magnetic and fluid phase transitions. In the case of gas-liquid transitions it has been experimentally observed that above a certain critical temperature, $T_c$, no amount of pressure can make the gas liquify. This is illustrated in Figure 1a. One sees that for $T<T_c$ there is a sudden drop in the specific volume, $v$, as the pressure increases. For $T>T_c$ the contour lines are smooth functions without the steps that are present when $T<T_c$. So above the critical temperature the system follows a single equation of state.

For magnetic systems one applies an external magnetic field, $H$, rather than pressure, and magnetization, $\overline{M}$, replaces specific volume. Below a critical temperature, $T_c$, a magnetization persists when the external field is turned off. This magnetization will remain to some extent as long as $T<T_c$. At $T=T_c$ the residual
A) PVT ISOTHERMS

B) MAGNETIC ISOTHERMS

FIGURE 1: FLUID AND MAGNETIC PHENOMENA
magnetization disappears, and for $T>T_c$ there can be no magnetization without an external field. Magnetic isotherms are shown in Figure 1b. One can see that there are resemblances between the gas-liquid thermodynamic parameters and the magnetic thermodynamic parameters. The external quantity impressed on the system is the pressure in one case and the magnetic field in the other. The specific volume and the magnetization are the respective 'ordering parameters' that are often denoted by $\sigma$ [1]. In the case of fluids the equation of state is in general

$$V = V(P,T),$$

while for magnetism the general expression is

$$\overline{M} = \overline{M}(H,T).$$

Consider a material placed in a magnetic field $\overline{H}$. The functional relationship between $\overline{M}$ and $\overline{H}$ determines whether the substance is diamagnetic, paramagnetic, or ferromagnetic. The first two types are characterized by a relationship like

$$\overline{M} = X\overline{H}.$$

The quantity, $X$, is called the magnetic susceptibility. A value for $X$ that is positive signifies a paramagnetic material, and a negative value signifies diamagnetism. Ferromagnetic materials
are described by a nonlinear relation denoted

\[ M = F(H). \]

One method of studying ferromagnetic phase transitions is to dilute ferromagnets with a paramagnetic host. Magnetic properties of these alloys can then be studied as functions of magnetic ion concentration as well as of temperature. In particular many studies of iron in a palladium environment have been done [2,3]. The iron atom is capable of polarizing the 4d electrons of the palladium atoms surrounding it. Such polarized regions are called 'giant moments' [4]. The critical temperature of such a system is strongly dependent on the iron concentration in the palladium host, because the mean distance between iron atoms decreases as [Fe] increases.

Recall that in general \( M \) is a function of \( H \) and \( T \). When treating the detailed dependence of pertinent physical quantities on temperature one can advance the concept of the critical exponent [1]. Two pertinent critical exponents are \( \beta \) and \( \gamma \), which are defined by

\[
M \sim |\epsilon|^\beta; \ T \to T^-_C \\
X \sim |\epsilon|^\gamma; \ T \to T^+_C 
\]

where \( \epsilon = T/T_C - 1 \). The values of these critical exponents are model dependent. Curie-Weiss theory (chapter 2), for example,
gives $\gamma=1$ and $\beta=1/2$. Table I gives a compilation of theoretical and experimental values of critical exponents.

An additional point of interest regarding the analogy between magnetic and PVT systems is the concept of universality. Universality is the conjecture that the critical exponents do not depend on whether the system is fluid or magnetic [5]. As long as the proper correspondence is made, the critical exponents of fluids are the same as the critical exponents of magnetism. The analog of susceptibility is the isothermal compressibility. Usually these studies involve noble gases for PVT systems. The data in Table I lends credence to the universality hypothesis. A subsequent experiment, however, has obtained a value of $\beta=0.354$ for He, indicating that universality may not be strictly achieved for all materials [6].

Upon decreasing one dimension of the bulk magnetic system one obtains a film. As the thickness decreases, increasingly important surface atoms have different surroundings than do internal atoms, so one might expect the thermodynamic ordering temperature to be shifted from the bulk value. Seminal work predicting a Curie temperature that decreases with decreasing film thickness was done by Fisher in 1973 [7]. The introduction of the shift exponent $\lambda$ was a result of this work. If $T_C(\infty)$ is the bulk Curie temperature, and $T_C(L)$ is the Curie temperature of the film of thickness $L$ the the shift exponent is defined by
<table>
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<tr>
<th>Critical Exponent</th>
<th>Theory</th>
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<tr>
<td></td>
<td>C-W</td>
<td>2-D</td>
</tr>
<tr>
<td>( \theta )</td>
<td>1/2</td>
<td>1/8</td>
</tr>
<tr>
<td>( \gamma )</td>
<td>1</td>
<td>7/4</td>
</tr>
</tbody>
</table>

(3) E.A. Guggenheim, J. Chem. Phys. 13, 253 (1945)
\[
(T_C(\infty) - T_C(L))/T_C(\infty) = CL^{-\lambda},
\]

where C is a constant of proportionality.

Some experimental tests of these suppositions have been made. Mossbauer detection of Curie temperatures has indicated a downward shift of \(T_C\) with a shift exponent of 0.95±0.15 for Pd(1-x)Fe(x) [4]. Motivated by these findings it was decided to look for this shift in Pd(1-x)Fe(x) using a transport method and a direct magnetic flux measurement.

A variety of methods exists to sense the change in the magnetic disposition of the system as the temperature changes. Detection of the ferromagnetic-paramagnetic transition in bulk Pd(1-x)Fe(x) has been achieved, and the dependence of the Curie temperature on iron concentration has been established from electrical resistivity data [2].

In the resistivity measurement a constant current density is injected into the sample, and an ohmic electric field is measured. The interaction of conduction electrons with the giant moments makes a small contribution to the resistivity. Below \(T_C\) the ferromagnetic alloy presents some order to the conduction electrons, while above \(T_C\) there is nearly total magnetic disorder. So one would expect that the magnetic contribution to the resistivity behaves like
\[
\frac{dp(m)}{dT} = \begin{cases} 
0 & ; \quad T > T_C \\
 f(T) & ; \quad T < T_C
\end{cases}
\]

In bulk samples there is a maximum in the \( dp(m)/dT \) versus \( T \) curves [3]. The maximum is taken to be the Curie temperature. We hoped to detect the shift in \( T_C \) using this method. Details on our measurements are contained in chapter 3.

The second method was a direct measurement of magnetization and a.c. susceptibility. In the absence of an applied magnetic field the magnetization of the sample should be zero above \( T_C \). The sample will, furthermore, spontaneously magnetize below \( T_C \). In a single-domain system the spontaneous magnetization varies like

\[
M(T) = \begin{cases} 
M_\infty(1 - T/T_C)^{\delta = 1/2} & ; \quad T \to T_C^- \\
0 & ; \quad T \to T_C^+
\end{cases}
\]

in a Curie-Weiss approximation (chapter 2). Domain formation, however, can inhibit the sample from reaching the saturation value \( M_\infty \). As \( M(T) \) changes, the flux due to the magnetization will change. Details about how this change is monitored will be discussed in chapter 3.
For the susceptibility one expects a peak, because the behavior is

\[
X = \begin{cases} 
|1 - \frac{T}{T_C}|^{-\gamma} ; & T \rightarrow T^+_C \\
|1 - \frac{T}{T_C}|^{-\gamma} ; & T \rightarrow T^-_C
\end{cases}
\]

Temperature studies on some ferromagnetic alloys, however, have shown double transitions [8]. The transition having the lower temperature is often associated with 'spin glass' behavior. This behavior is due to competition between antiferromagnetism and ferromagnetism in a random spin system. Because the interaction potential between the spins is an oscillatory function of distance, some of the randomly situated spins can be separated by a distance that has the potential negative (antiferromagnetism). The higher-temperature transition is the Curie point. In doing our work one-peak and two-peak curves were also observed in the studies of the a.c. susceptibility. Originally this study was to be dedicated to the shifting of the Curie temperature with the film thickness, but the possibility of spin-glass findings has resulted in a slight expansion of this topic. Preliminary findings showed that the temperature at which the magnetization went to zero increased as the film thickness increased. Because of the double peaks in \(X\), the reliability of \(X\) as an indicator of \(T_C\) was questionable. With the emergence of the unexpected peaks, close attention to sample preparation is very important.
Chapter 2 contains a mean-field treatment (the ions interact with each other via an average internal magnetic field that is proportional to the magnetization) for the comparison of the data with a theory. The third chapter of this work is an extensive discussion of the sample preparation and analysis. The next part deals with instrumentation. Finally the data are analyzed and considered in the light of some statistical mechanical models.
II. THEORY

First-order phase transitions have a latent heat. This is equivalent to a discontinuity in the entropy because

\[ L = T_c \Delta S \]

where \( L \) is the latent heat and \( \Delta S \) is the jump in the entropy. But the specific heat at constant pressure is given by

\[ C_p = T(\partial S/\partial T)_p \]

so that for a first-order transition

\[ C_p = \infty . \]

For second-order phase transitions there is no discontinuity in the entropy with respect to temperature. There is, therefore, no infinite peak in the specific heat. The finite nature of the magnetic specific heat can be easily seen. The magnetic internal energy of an N-atom (per unit volume) lattice having order parameter \( \sigma \cdot M \) in an external field \( H_0 \) is

\[ U(m) = -\text{N}g \langle S_{zj} \rangle (\text{Weiss}/2 + H_0) . \]

where 'u' is the Bohr magneton, 'g' the Lande factor, and \( \langle S_{zj} \rangle \) the average spin. For zero external field one obtains

\[ U(m) \sim \sigma^2 . \]

Then

\[ C(m) = dU(m)/dT \sim \sigma d\sigma/dT . \]
For $T=0$ K, $d\sigma/dT = 0$, giving $C(m) = 0$. For $T>T_C$, the completely randomized system cannot store any more energy magnetically, so $C(m)$ is again essentially equal to zero. Between 0 K and $T_c$ the heat capacity thus rises to a finite value (mean field) before dropping abruptly to zero. This can in general be written

$$C(m) = \begin{cases} g(T) & ; T < T_c \\ 0 & ; T > T_c \end{cases}.$$ 

This result can be compared with the derivative of the resistivity discussed in the introduction.

With the discussion of a simple mean field theory one has a medium for expanding on these features of phase transitions [1,5]. In such a theoretical framework the concepts of ordering parameter, critical exponent, and heat capacity jumps can be qualitatively referenced to experimental results.

Consider the spin which interacts with its $z$ nearest neighbors via the hamiltonian

$$H' = -2J\vec{S}_i \cdot \sum_{j}^{z} \vec{S}_j,$$

where $\vec{S}_j$ is the spin of the jth atom, and $J$ is the coupling constant. The atoms in the neighborhood of the ith site can be said to define an effective magnetic field $\mathcal{H}_W$ at the site. $\mathcal{H}_W$ is given by
Replacing each atomic spin by its average, one obtains

$$\overline{H}_W = (2Jz/gu)\langle S_j \rangle,$$

The magnetization of the crystal is

$$\overline{M} = Ngu\langle S_j \rangle,$$

with \(N\) as the number of atoms per unit volume. The last equation is the basis of the Weiss mean field -- the effective field experienced by each spin is proportional to the crystal magnetization.

An external field \(H_0\) causes each spin to interact with a total field

$$\overline{H}_T = H_0 \overline{K} + \overline{H}_W.$$

But the average alignment must be along \(\overline{K}\), so

$$H_T = H_0 + H_W.$$

The interaction hamiltonian is then

$$H' = -guS_1 \cdot H_T K = -guS_z H_T.$$

The eigenvalues of \(H'\) are

$$E_m = -gmH_T,$$

where \(m\) ranges between \(-S\) and \(+S\) in integer steps. The partition function is then

$$Z = \sum_{-S}^{S} e^{-guH_Tm/kT}.$$
Defining $x = g u H_T S / k T$ the partition function can be simplified to

$$Z = \left( \frac{\sinh x (S + 1/2)}{S} \right) / \sinh (x/2S) .$$

The magnetization can then be expressed in terms of the spin using

$$M = N g u <S_Z> = N g u (1/Z) \sum_m e^{m g u H_T / k T} .$$

Note that the sum is proportional to the derivative of $Z$ with respect to $H_T$ so

$$M = N g u S \left( \frac{2S + 1}{2S} \ coth \left( \frac{2S + 1}{2S} x \right) - \frac{1}{2S} \ coth (x/2S) \right) = N g u S b_S(x).$$

Because $b_S(x)$ has a maximum of one at $x = \infty$, it is convenient to define the order parameter

$$\sigma = M / M_\infty = b_S(x)$$

and let

$$x_0 = g u H_0 S / k T$$

so that

$$x = x_0 + 2 J z S^2 \sigma / k T .$$

For $H_0 = 0$ and small $x$

$$\sigma = b_S(x) = \left( \frac{S + 1}{3S} \right) x$$

so

$$x = 2 z J S^2 (S + 1) x / (3 S k T) ,$$
requiring

\[ T = T_c = \frac{2}{3}(zJ/k)S(S+1) \]

So a spontaneous order develops at some critical temperature. Above \( T_c \) the order parameter, \( \sigma \), is zero in the absence of an applied field.

Once having a model with a Curie temperature, one can look for the explicit temperature dependence of \( M, X, \) and \( C(m) \). For \( S=1/2 \)

\[ M = \left( \frac{N^u}{2} \right) \left( 2\coth(2x) - \coth(x) \right) \]

and using the addition formula

\[ \coth(y + z) = \frac{\coth(y) \coth(z) + 1}{\coth(y) + \coth(z)} \]

one obtains

\[ \coth(2x) = \frac{\coth^2(x) + 1}{2\coth(x)} \]

Substitution gives

\[ M = \left( \frac{N^u}{2} \right) \left( \frac{\coth^2(x) + 1}{\coth(x)} - \coth(x) \right) \]

\[ = \left( \frac{N^u}{2} \right) \tanh \left( \frac{gu}{2kT} (H_0 + H_w) \right) \]

If \( \Theta = T/T_c \)

\[ \sigma = \tanh \left( \frac{guH_0}{2kT} + \frac{\sigma}{\Theta} \right) \]

The identity
\[
\tanh(x + y) = \frac{\tanh(x) + \tanh(y)}{1 + \tanh(x) \tanh(y)}
\]

is now applied to the last equation, giving

\[
\sigma = \frac{\tanh(guH_0/2kT) + \tanh(\sigma/\Theta)}{1 + \tanh(guH_0/2kT) \tanh(\sigma/\Theta)}.
\]

Now let

\[
h = \tanh(guH_0/2kT)
\]

so

\[
\sigma = \frac{h + \tanh(\sigma/\Theta)}{1 + h \tanh(\sigma/\Theta)}.
\]

and

\[
h = \frac{\sigma - \tanh(\sigma/\Theta)}{1 - \sigma \tanh(\sigma/\Theta)}.
\]

Finally

\[
h = \sigma(1 - 1/\Theta) + \sigma^3(1/3\Theta^3 + \frac{1}{\Theta}(1 - \frac{1}{\Theta})) + O(\sigma^5).
\]

To get the zero-field magnetization we set \(h=0\). Neglecting all but the first two terms, and for \(T \leq T_C\)

\[
\frac{1}{\Theta} - 1 = \sigma^2(1/3\Theta^3 + \frac{1}{\Theta}(1 - \frac{1}{\Theta}))
\]

so

\[
\sigma = \frac{-\frac{T - T_C}{T_C}}{T_C(\frac{T_C^2}{3T^3} - \frac{1}{T}(1 - T_C/T))}
\]
The zero-field isothermal susceptibility is given by

\[ \sigma = \left( (T - T_c)/T_c \right)^{1/2} \]

where \( K \) is a constant. Differentiating the series for \( h \) with respect to \( h \) gives

\[ X = \left( \partial M/\partial H_0 \right)_T = \partial M/\partial \sigma \left( \partial \sigma/\partial h \right)_T \left( \partial h/\partial H_0 \right)_T \]

\[ \approx K \left( \partial \sigma/\partial h \right)_T \]

so

\[ \left( \partial \sigma/\partial h \right)_T = (1 - 1/\sigma + 2/\sigma^2) \]

giving

\[ X = \left( K/2T \right) \left( (T_c - T)/T_c \right)^{-1} \quad (T < T_c) \]

Then for \( T \geq T_c \), \( \sigma = 0 \), and

\[ X = K (T - T_c)^{-1} \]

One can thus conclude from mean field theory that \( \gamma = \gamma' = 1 \), and \( \beta = 1/2 \), as stated in the introduction.

It remains to calculate the critical dependence of \( C(m) \).

The magnetic internal energy is

\[ U(m) = -NkT x_0 \]
so

\[ C(m) = -(\partial U/\partial T)_{H_0} = -Nk(\partial \sigma (T_x)/\partial T + (T_x \partial \sigma / \partial T) \ . \]

If \( H_0=0 \), \( x_0=0 \), so

\[ kT_x = zJ\sigma/2 \]

and

\[ C(m) = -(NzJ/4)\partial (\sigma^2)/\partial T \ . \]

For \( T=T_c \)

\[ C(m) = -(NzJ/4)(-3)/T_c = 3Nk/2 \ . \]

For \( T>T_c \), \( \sigma = 0 \), giving

\[ C(m) = 0 \ . \]

Thus, the mean field theory predicts that \( C(m) \) has a finite discontinuity, and the transition is second order. Fisher and Langer, in fact, have shown that the specific heat behaves functionally like the negative first derivative of the logarithm of the resistivity with respect to temperature [9]. The temperature dependences of \( C(m) \), \( M \), and \( X \) just derived will serve as a starting point for the discussion of the data.
III. EXPERIMENTAL

A. Sample Preparation:

The preparation of useful thin films involves the controlled removal and subsequent deposition of material from a reservoir (target) to a support surface (substrate). In the process, atoms that will constitute the film bombard the substrate with momentum components parallel and perpendicular to the substrate surface. Atoms that lose enough energy to the substrate can then remain and roam along the surface. Eventually the mobile atoms collide and coalesce to form islands that grow together into a continuous film. The film's microstructure and chemical composition are significant to this work.

One important microstructural property is thickness. One can control the film thickness by a number of means. An obvious way to produce films of different thicknesses is to vary the time that the deposition is allowed to take place. It is also possible to control the thickness by varying the removal rate of the atoms from the target.

Aside from thickness another important property of the films is their crystallinity. Knowing that substrate temperature and crystallinity affect the surface nucleation conditions, one can generate different degrees of film crystallinity. Very cold substrates can stifle mobility to the extent that amorphous structure is achieved. Heating the substrates causes in situ
annealing and thereby increases crystallite grain size.

There are five phases of sample preparation:

(1) substrate preparation,
(2) substrate analysis,
(3) film deposition,
(4) film analysis, and
(5) storage and handling.

We chose glass substrates for our initial experiments, because glass offered low cost and proven performance in Mossbauer work. The glass substrates were cut in the shape of disks and rectangles from .006 in. thick microscope cover slides.

Starting with a stack of cover slides about .5 in. thick, heated beeswax is poured around the edges of the slides. They are then heated so that the wax flows down in between them. One does this to each edge-face. While hot the slides are squeezed together to get rid of air pockets. The final step is to even out all of the slide edges.

Once the stack is cool disks or rectangles may be cut from it. Using a core drill and a drill press a stack of disks is extracted from the original stack. The disks can be pushed out of the core bit after a light heating. Should rectangles be desired, a standard diamond saw can be used.
In either case the substrates can be separated from each other in boiling water. The substrates are then soaked in ether (C2H5-O-C2H5) to dissolve any wax that remains on the surface. Lastly they are cleaned ultrasonically in alcohol and stored individually in vials. The disks were about 4.5mm in diameter, and the rectangles were about 3mm by 10mm.

Silicon and sapphire were also used as substrates for films. Fabrication of silicon substrates began with the slicing of wafers from a Czokralski-grown ingot. This was done using a Beuhler Ltd. diamond saw. The wafers can be waxed to a microscope slide at different orientations in order to effect the cutting of the substrate.

Substrates now undergo a polishing sequence to restore smoothness. Each is individually mounted in a recess in an aluminum holder. Polishing begins by pressing the holder against a rotating polishing wheel using 1μ grit. The sample should be rotated frequently throughout this step. When all of the pits and scratches are gone a .3μ grit is used. Finally 'figure 8's' are done on a stationary wheel with .05μ grit. By now the surface is mirror smooth.

Prior to storage in a vial each substrate is cleaned ultrasonically in isopropyl alcohol. Sizes were approximately the same as those of the glass rectangles.
Sapphire substrates were cut on a diamond saw from a continuous ribbon 3 in. by 6 in. by .045 in. These were cleaned in the same manner as the other two substrate types.

Just prior to deposition or analysis the substrates were ultrasonically cleaned for the final time. For short-term storage the substrates were kept in vials cleaned ultrasonically. This step was taken to avoid contamination by dust. Such procedures ensure cleaner electron micrographs and films that are free of pinholes. The smoothness of each substrate type was checked on an SEM (scanning electron microscope). Films were stored in vials padded by cotton if they were not to be scanned.

The method of deposition chosen for this work was glow-discharge sputtering. In this technique atoms leave the target (Figure 2) because of bombardment by singly-ionized argon ions whose energy exceeds the threshold removal energy of the target material. The yield of the sputtered atoms depends on the target material as well as on the ion energy. One can see, therefore, that there are several ways of controlling film thickness:

1. duration of exposure,
2. background-gas pressure,
3. target size,
4. substrate-target separation, and
5. incident-ion energy.
Figure 2 shows a typical plasma-target event. The target, a palladium disk with an iron square epoxied to its center, is hit with an argon ion. Coming out with the neutral target atom is a group of electrons. The ejected electrons are important to the deposition process because they ionize the background argon, causing the removal process to be self-sustained. Initiation of the ionization at the start of the process is due to background UV radiation or cosmic rays. The pale blue halo that comes from the photons from the de-exciting argon gives rise to the term glow discharge.

It should be noted that the target is not an alloy. Sputtering from a target that is made up of distinct elemental areas is called cosputtering [10]. This method has the advantage that the iron concentration in the sputtered film can be simply varied by changing the size of the shield opening. Making the shield opening smaller increases the ratio of iron area to palladium area so that higher iron concentrations can be made. This technique, however, results in the migration of atoms to different areas of the target, so time may be required for equilibrium to be established. Thus the chemical composition will depend on time. The fastening together of the different elements may also introduce contaminants.

The target-substrate configuration is housed in a bell jar. During operation the bell jar is pumped on by a turbomolecular
FIGURE 2: SPUTTERING DEPOSITION

- ARGON ION
- TARGET ATOM
- ELECTRON
pump. The background pressure of Ar is kept at 15 mtorr by a constant pressure monitoring device. The palladium disk was 3 in. in diameter (.999 Matthey-Bishop), while the iron foil was a 1 in. square (.9999 Alfa-Ventron). The target is soft-soldered to a copper backing plate. The electronics for plasma production were made by the R. D. Mathis Company.

Analysis of films consists of determination of film thickness, composition, microstructure and topography. Different methods of measuring film thickness were tried.

The first method tried was the microbalance. Using a Mettler balance the substrate is weighed. After deposition it is again weighed. The mass difference is due to the film. Assuming that the bulk density is the same as the film density, the thickness is given by

$$L = \frac{\text{mass}}{(\text{area} \cdot \text{density})}.$$ 

The second method was the stylus technique. A Talysurf stylus was drawn over a masked film that served as a test film. This masked film was made simultaneously with the films that were to be used in the electric and magnetic measurements. A step was thus produced in the film. The Talysurf output is proportional to the height of the step. Measurements were systematically about 700A more than those of the Mettler balance.
Two techniques were tried for determination of chemical composition of the films. Figure 3 shows an electron beam impinging on a sample film. The bulb-shaped region is a representation of the region of interaction of the electron beam with the solid. Both methods of determination used the characteristics of such an electron-solid interaction.

The first method to be discussed utilized the Auger effect. Here the incident electrons cause core electrons to leave the atoms. After ionization at the core there is subsequent cascading to fill the core vacancy. The atom's excess energy can be transferred to another electron, causing it to be ejected. This ejected electron is an Auger electron. Auger electrons are low-energy electrons (<1000eV). Because of this low energy only electrons from atoms near the surface can make the trek from the film to the detector. For this reason region 2 of Figure 3 is called the Auger region.

Ejected electrons have characteristic energies depending on the elements from which they come. Using a retarding-potential analyzer on the Auger electrons and differentiating the signal one obtains a spectrum. Because the yield of Auger electrons increases with decreasing Z, this technique is good for detecting light atoms.

Iron concentration can also be established using electron-probe microanalysis (EPMA). In this technique one uses the
FIGURE 3: THE ELECTRON-SOLID INTERACTION

1. EPMA REGION
2. AUGER REGION
characteristic x-ray spectrum made by the electron beam.

Moseley's law illustrates the strong dependence of wavelength on Z:

$$E_{2s} - E_{1s} = C(Z - 1)^2,$$

where C is a constant. EPMA is especially useful for high Z elements.

Because x-rays can leave the sample much more easily than electrons, the EPMA region is 1 and 2. For thin films the inclusion of the substrate into the excitation volume can be problematic.

From early Auger analysis it was discovered that the oxygen content of some of the films was large. The percentages were too high for any known valence state of iron, so we concluded that at least some of the oxygen was due to trapped O2.

Most analyses of the films were done using Auger Electron Spectroscopy. After milling away many atomic layers using an argon ion beam, an elemental scan was done. In the same spot an atomic percent calculation was done. The calculation considered palladium, iron, and whatever other elements seemed significant from the scan. Finally, we moved to another spot on the film and did a depth profile. This type of analysis monitors
the elements in a concentration-versustime(depth) mode. The signals due to each element are multiplexed as the spot is both excited and milled. Table II contains a summary of the Auger analysis, while Appendix A and Appendix B have the Auger data.

It should be noted from Appendix B that spectra are very different after milling. The oxygen lines at 488eV and 510eV diminish significantly relative to the iron lines at 596, 651, and 703eV. The significance of this reduction will be discussed in Chapter IV.

B. Resistivity measurements:

In doing a resistivity experiment scattering from disordered spins will not be the only contributor to the resistivity. One can expect to find other temperature-dependent and temperature-independent terms. If a Matthiesson's rule expression is written for the resistivity one obtains

\[ \rho = \rho_{\text{res}} + \rho(T) \]

where \( \rho_{\text{res}} \) is the temperature-independent part called the residual resistivity, and where \( \rho(T) \) is the part that depends on temperature [11].

Measurement of \( \rho_{\text{res}} \) is a simple matter. Using liquid helium to lower the temperature of the sample to 4.2 K, one can suppress \( \rho(T) \) to the extent that one obtains
TABLE II: FILM DATA

<table>
<thead>
<tr>
<th>Film</th>
<th>Thickness A</th>
<th>% Fe</th>
<th>% O</th>
<th>$T_c$ K</th>
<th>$T_c$, bulk (3) K</th>
</tr>
</thead>
<tbody>
<tr>
<td>McGrath</td>
<td>440</td>
<td>7.8(1)</td>
<td>0</td>
<td>112-1/X</td>
<td>170</td>
</tr>
<tr>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>105-M</td>
</tr>
<tr>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>114-0</td>
</tr>
<tr>
<td>46</td>
<td>1000</td>
<td>8.0</td>
<td>0</td>
<td>67-M</td>
<td>170</td>
</tr>
<tr>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>&quot;</td>
<td>67-1/X</td>
</tr>
<tr>
<td>39</td>
<td>3000</td>
<td>&quot;</td>
<td>&quot;</td>
<td>70-M</td>
<td>170</td>
</tr>
<tr>
<td>34</td>
<td>5000</td>
<td>&quot;</td>
<td>&quot;</td>
<td>132-M</td>
<td>170</td>
</tr>
</tbody>
</table>

(1) done at Physical Electronics, Minneapolis, Minn.
(2) done at Polyscientific, Blacksburg, VA
(3) G. Nieuwenhuys, Adv. in Physics 24, 515 (1975)
The scattering mechanisms prevalent in this temperature regime are the scattering of conduction electrons from impurity atoms and the scattering of conduction electrons from grain boundaries. It is also possible that a localized group of iron atoms, a cluster, can be frozen into a certain orientation in the crystal. A distribution of these clusters can be randomly frozen in the lattice. The distribution is called a cluster-glass because of the local order and long-range disorder. So in a film that is primarily Pd atoms the iron atoms can serve as multifold scattering centers. Conduction electrons can sense lattice disorder due to impurities and due to magnetic disorder.

When dealing with thin films the grain size can depend on film thickness and substrate crystallinity. A multitude of substrates exists for use in this type of study. Sapphire, silicon, and glass will be discussed later regarding their relative merits.

The scattering mechanisms are:

1. phonon-electron scattering due to lattice vibrations,
2. boundary scattering from the film surface,
3. scattering from individual magnetic ions,
4. scattering from clusters,
5. scattering from impurities, and
(6) scattering from grain boundaries.

The first four depend on temperature. So

\[ \rho = \rho_{\text{ph}}(T) + \rho_{\text{surf}}(T) + \rho_{\text{mag}}(T) + \rho_{\text{c1}}(T) + \rho_{\text{grain}} + \rho_{\text{imp}} \]

For \( T < T_c \)

\[ \frac{d\rho}{dT} = \frac{d\rho_{\text{ph}}}{dT} + \frac{d\rho_{\text{surf}}}{dT} + \frac{d\rho_{\text{mag}}}{dT} + \frac{d\rho_{\text{c1}}}{dT} . \]

The temperature dependence of the surface scattering derives from the fact that at low \( T \) the mean-free path can be sufficiently long that it is comparable to the film thickness. This phenomenon has been observed experimentally in very crystalline films and has been thoroughly developed theoretically [12]. The dependence of phonon and magnetic contributions on temperature are due to the dependence of magnon and phonon densities on \( T \).

For \( T > T_c \) one loses the magnetic term, because complete magnetic disorder is achieved. Because of the small mean free path, the second term can be neglected. This leaves

\[ \frac{d\rho}{dT} = \frac{d\rho_{\text{ph}}}{dT} . \]

One would expect, therefore, that the resistivity curves have a knee at the Curie temperature where the magnetic order turns off and on.

Early measurements for this work showed anomalies in the voltages. These voltages are proportional to the resistivity. The anomalies took the form of local maxima and minima in the \( V \) vs \( T \) curves. The injected current was typically about 30\( \mu \)A.
Pronounced hysteresis between the heating and cooling runs was observed. Data runs with no injected current showed hysteresis, so we attributed the anomalies to thermoelectric voltages.

To minimize the influence of thermoelectric voltages we put Wakefield grease between the substrate and the copper block. This established a better thermal short between the two. We also increased the injected current so that sample voltages would be orders of magnitude greater than the thermoelectric voltages.

Once this was done \( \rho \) vs \( T \) measurements were again made. Using a pure Pd film the expected linear relationship between resistivity and temperature was achieved.

Initial searches for phase transitions in the films were done using the four-probe technique. Resistivity-versus-temperature curves have been shown to have an inflection point at the Curie temperature [2,3]. Figure 4 shows the experimental setup. The can and flange define the experimental region, which can be attached to the vacuum system. The copper block is able to hold three films. In each of the three films the current is injected in the outer of the four fingers, and the sample voltage is measured across the inner two.

Null measurements provided a very accurate means of obtaining the voltages. Thermocouple voltages could be measured to better than one microvolt using a Keithley 150B microvolt-
FIGURE 4: RESISTIVITY APPARATUS
ammeter as a meter and an ESI 300 potentiometric bridge as a source of counter emf. A Keithley 260 nanovolt source provided a source of counter emf for drops across the films, while a Keithley 148 nanovoltmeter served as a nullmeter. Voltage as a function of temperature could be recorded tabularly or via an X-Y recorder.

A vacuum system was built in order to control the energy flow in the experimentation region. A vacuum stops convective transfer of energy from taking place between the copper block and the can; the latter can be in direct contact with either liquid nitrogen or liquid helium. The only conductive path between the bath and the block is a poorly conducting brass rod. So with a resistive heater one can establish a film temperature and hold it while a voltage is measured. Such a vacuum system also allows a helium exchange gas to be leaked in as a means for cooling the films quickly.

C. Susceptibility and magnetization measurements:

Measurements of magnetization and susceptibility deal directly with magnetic quantities. These quantities were obtained using a SQUID (Superconducting Quantum Undulating Interference Device) magnetometer. It is interesting to note that the SQUID's existence is based on a second-order phase transition—a transition from normal conductivity to superconductivity. In order to better understand the SQUID's operation some of the
discoveries leading to its development will be summarized [13].

First detection of superconductivity dates back to the work of Kamerlingh Onnes in 1911 while studying the resistance of mercury. It is interesting that the detection of the superconducting state was done in a resistivity-versus-temperature experiment. In measuring the resistance of mercury Onnes discovered that at a certain temperature the resistance dropped precipitously to zero.

Two major facets of superconductivity were discovered in 1933 and 1950. In 1933 Meissner and Ochsenfeld showed that a superconductor expels magnetic flux. After passing through the critical temperature a superconductor becomes diamagnetic to the extent that internal currents produce a field that exactly cancels the impinging field. In 1950 Reynolds et al found that the critical temperature showed an isotope effect. This result revealed that phonons play an important role in superconductivity [14].

Major theoretical work on superconductivity was done by J. Bardeen, L. Cooper, and J. Schrieffer in 1957. This BCS theory gives the electrons an attractive pairing interaction at low temperatures in the presence of a lattice. The attraction allows the electrons near the Fermi surface to interact coherently. Four years later B. Deaver and W. Fairbank showed experimentally that the flux trapped by a superconducting ring is quantized in
units of $h/2e$; The 'fluxoid' has value $2 \cdot 10^{-7}$ gauss-cm. The last steps in the development of a SQUID were the postulation (Josephson) and experimental utilization (Zimmerman et al) of the tunneling of electron pairs through a thin barrier in a superconducting ring [15].

The first part of this apparatus discussion will begin with the mechanical aspects of SQUID instrumentation. As in the case of the resistivity measurements, the sample is situated in a vacuum envelope during the SQUID experiment. This vacuum provides isolation for the sample so that the only connection between the helium bath and the sample is a radiative one. Figure 5 depicts the SQUID experimental configuration. This apparatus accesses the same vacuum system and dewar system as the resistivity apparatus.

The sample rests on a sapphire rod, because sapphire is magnetically inert and is a good thermal conductor at low temperatures. With a heater at the top of the rod, the temperature of the sample can be varied. A platinum resistance thermometer monitors the temperature, with the constant current passing through it producing a voltage drop that drives the horizontal axis of a two-channel x-y recorder. So with magnetization and susceptibility on the two vertical channels of the recorder, temperature-dependent studies can be done.

To measure these magnetic quantities the sample is placed
FIGURE 5: SQUID SYSTEM
in one of two astatically-wound niobium coils. These loops are wrapped around the vacuum envelope and fastened into place with GE 7031 varnish. Should the two coils not be perfectly matched, a balance coil can provide the nulling current required. The loop system can, furthermore, be driven at 100, 200, 300, and 1000 rad/sec by a coil surrounding it.

This part of the apparatus is enclosed in a lead shield. Lead superconducts at 7.2 K, providing magnetic isolation. Because a highly permeable metal shield is wrapped around the dewar, the only magnetic field at the sample is the small amount due to flux trapped in the lead when it is cooled.

The niobium-encased SQUID (a single-barrier Josephson junction) sits away from the sample, directly in the helium bath. Evaporated helium takes along some thermal energy from a set of baffles before it leaves the dewar. This shields the liquid helium surface from the copper flange that is at room temperature, thereby increasing the helium hold time. The helium level is monitored with an American Magnetics helium level detector.

The sample enters the stainless steel vacuum envelope through a ball valve that serves as an airlock. This provision enables the region below the valve to be at a vacuum even when the sample is changed.

Adjustment of the sample position is done using a calibrated
This unit raises and lowers the sample tube. The inside of the sample tube carries the thermometry and heater wires, and the end of the tube is sealed with an epoxied feedthrough. The quartz tube at the other end of the sample tube has a glass-to-copper seal with the copper attached to the sample tube with Wood's metal.

The initial nulling of the system is done without the sample in the pickup loop. Figure 6 shows the electronics setup. The sample is lowered into the first loop, and its position is adjusted to get the maximum deviation from the original null. Once this position is found, it is held constant throughout the data run.

The change in sample magnetization due to temperature means a change in the flux in one loop of the astatic pair. This lack of balance causes a supercurrent through $L(1)$, which is inductively coupled to the SQUID. The SQUID is inductively coupled to the tank circuit.

An rf current is supplied to the tank circuit at its resonant frequency. The rf supply adjusts the amplitude of the current to keep the mutual inductance between the tank inductor and the SQUID constant. The change in current is necessitated by a change in the sample’s magnetization. In fact the concomitant change in the amplitude of the tank voltage is proportional to the change in the magnetization. This voltage goes to one channel of the x-y recorder.
FIGURE 6: SQUID SCHEMATIC
The element, $Z$, is an element in a bridge circuit. The sample, pickup loops, and primary coil constitute a temperature dependent mutual inductance that is proportional to the susceptibility. So the off-null in-phase component to $Z$ is used to drive the second vertical channel of the recorder, allowing simultaneous measurement of magnetization and a.c. susceptibility. One can also study resistive effects by means of the quadrature output.
IV. DATA AND DATA ANALYSIS

A. Resistivity:

A recent experimental treatment of the thickness dependence of Pd(1-x)Fe(x) Curie temperatures was done at Johns Hopkins University by R. D. McGrath using the Mossbauer effect [4]. A sample was obtained from McGrath to initiate our study [4]. The sample was 440A in thickness and contained 7.8% Fe. The bulk Curie temperature for a sample of this concentration is about 170 K (Table II).

Resistivity measurements on this film showed a change in the slope of the resistance-versus-temperature graph. The plot of voltage (proportional to resistivity) versus temperature is given in Figure 7. The drop in the derivative of $\rho$ is indicative of the loss of a temperature-dependent scattering mechanism. One expects the temperature dependence of $\rho$ to be

$$\rho = \begin{cases} \frac{c + bT + aT^p}{T} & ; \theta_D < T < T_C \\ \frac{c' + bT}{T} & ; T > T_C \end{cases},$$

where $\theta_D$ is the Debye temperature, and $c' = c + aT_C^p$, where $'p'$ is some power governing the magnetic scattering. The quantities 'a', 'b', and 'c' are constants. The first two of these constants relate to the strengths of the temperature-dependent scattering mechanisms. The third constant is the residual resistivity.

43
FIGURE 7: FOUR-PROBE DATA
A pure palladium film showed linear behavior above 7.7 K. In the alloy case, deviations from this line could then be attributed to magnetic scattering. To extract a critical temperature from the data a line was fitted to the first ten data points for the alloy. The first ten points were chosen, because this kept the fit away from the critical region, while allowing a good line to be generated. Because of the additional term, $aT^0$, the data points were above this line until 114 K. At 114 K the data falls below the line as the temperature dependence of the magnetic scattering is lost.

This behavior is consistent with the general behavior of second-order phase transitions. In second-order phase transitions entropy and resistivity behave qualitatively alike. The temperature derivatives of both show a sharp drop at $T_C$. Such a comparison is reasonable, because both entropy and resistivity are measures of the system's randomness.

Graphs of $(\rho - \rho_{res})/T$ (actually $[V-V_{res}]/T$) were plotted versus the square root of temperature to see if the expected $3/2$ power was obtained [16]. The data indicate that ‘p’ is approximately equal to $3/2$ (Figure 8). A more precise value of ‘p’ might be obtained with digital data acquisition, more accurate temperature measurements, and automated analysis. Smearing out of the transition by short-range interactions, however, still might
FIGURE 8: \( \frac{(V - V_{RES})}{T} \) vs \( T^{1/2} \)
upset an accurate determination of the Curie temperature.

Figure 9 shows spontaneous magnetization versus temperature for the same film. Figure 10 shows model fits of $M$ versus temperature for three theoretical models: Curie-Weiss, 2-D Ising, and 3-D Ising. There are noticeable differences from theoretical predictions at both the high and the low ends of the data. Linear regions of the $M^{1/β}$ graphs were used to generate $M_C$ and $T_C$. Near $T_C$, where the critical exponents are valid, the Curie-Weiss model fits the data the best. At low temperatures, the data is best fit by the 2D-Ising model. More collinear points were present, however, in the C-W case.

The fact that a local maximum appears in the magnetization curve may indicate that cooling in zero-field conditions does not allow all the moments to line up. Instead, there are presumably local regions (clusters) having net local magnetization not aligned with the total magnetization. As the temperature increases, the clusters 'melt' (magnetically), causing the overall magnetization to increase. This local maximum is not present in field-cooled samples [17].

Figure 11 shows the a.c. susceptibility. Two features are obvious from the data. There is a strong maximum at 107 K, and there is apparently a weak maximum (40 K) on the temperature-independent component to $X$. The weak maximum may be due to the melting of the clusters, while the strong maximum occurs at
FIGURE 9: M VS T (JOHNS HOPKINS)
FIGURE 10: MODEL FITS (JH)
FIGURE 11: $X$ VS $T$ (JOHNS HOPKINS)
the Curie point. Below $T_C$, $\gamma'$ is roughly unity (Figure 12), where

$$X \sim |\varepsilon|^{-\gamma'}; \quad T < T_C.$$

Analyses of our magnetic and resistivity data show $T_C$ to be between 105 K and 115 K. This is well below the bulk value of 170 K (Table 2). Mossbauer data gave a prediction of 40 K [4]. In order to verify the chemical composition of the film, a chemical analysis was performed by means of Auger Electron Spectroscopy.

Auger spectra for the Hopkins sample are contained in Appendix A. At 273 eV there is a carbon line, and oxygen lines are at 510, 488, and 471 eV. A silicon line appears at 1619 eV. These elements can be attributed to surface contaminants on the film. After one minute of ion milling, however, the silicon line disappears, and the three main iron lines appear at 596, 651, and 703 eV. Oxygen lines are still present, but they are less pronounced. Palladium lines at 223, 245, 281, and 330 eV are now evident. After one more minute of ion milling only Pd and Fe lines are seen. An important feature is that no oxygen is present. In the same spot a depth profile was done monitoring Pd, Fe, and O. We found that the oxygen concentration is about one-half that of iron. To see if the oxygen line was noise, sodium was included in the profiling. Sodium was chosen because it has only one line (1000 eV). Because sodium would not be in the film, its level would establish the noise level. The sodium presence was the same as the oxygen presence, leading to the
FIGURE 12: RECIPROCAL SUSCEPTIBILITY (JH)
conclusion that the film may indeed be only iron and palladium. An atomic percent measurement showed 9.5% Fe and 90.5% Pd (Table 11). This is in reasonable agreement with the 7.8% Fe obtained by McGrath.

Along with McGrath's film, film 46 (made at Virginia Tech) also showed a very sharp Curie temperature. The Auger data for this film are contained in Appendix B. Films made with the RF sputtering apparatus described in Chapter 3 showed 4.3% oxygen in the interior of the film, while the iron concentration was 8.0%. This oxygen level is about the same as the noise level. This measurement was done on film 46, having a thickness of roughly 1000 A.

Several features characterize the SQUID data on this film. The magnetization curve showed no relative maximum at low temperatures, and the susceptibility curve does not show a local maximum below the Curie temperature. The Curie temperature is extremely sharp. Figures 13 and 14 contain the data on a linear temperature scale, while Figure 15 is the raw data. The Curie temperature, found from the vanishing of the magnetization and from the peak of the susceptibility, was about 65 K.

Thicker films (3000 A and 5000 A) showed well-behaved magnetizations, but their a.c. susceptibilities were broadened and were not even qualitatively like the mean-field predictions. The data for one of these films are contained in Figures 16-19.
FIGURE 13: M VS T -- FILM 46
FIGURE 14: X VS T -- FILM 46
FIGURE 15: RAW DATA -- FILM 46
It should be noted that the magnetization vanishes at successively lower temperatures as thickness decreases (Table II). This progression has several possible explanations:

1) There is indeed a shift due to dimensional effects as predicted by Fisher;

2) The target may change from fabrication to fabrication;

3) There may be some other magnetic phenomenon that is dependent on thickness.

In all cases the Curie temperature measured for films is well below the values obtained from bulk measurements by other workers.

Of films 34, 39, and 46 only film 46 was usable for quantitative analysis. For film 46 an analysis for $\beta$ using the different models shows the Curie-Weiss model to give a value of the critical temperature that is closest to the experiment. At low temperatures, however, the Curie-Weiss curve gives the poorest fit to the data (Figure 20). Figure 21 shows $1/X$ versus temperature. The linear aspect to the susceptibility data confirmed the magnetization data in that the behavior was again of a Curie-Weiss nature.
FIGURE 16: M VS T -- FILM 39
FIGURE 17: \( X \) VS \( T \) -- FILM 39
FIGURE 18: M VS T -- FILM 34
FIGURE 19: \( x \) VS \( T \) -- FILM 34
FIGURE 20: MODEL FITS - FILM 46
FIGURE 21: RECIPROCAL SUSCEPTIBILITY - FILM 46
V. CONCLUSIONS

Resistivity, magnetization, and susceptibility measurements were done on a film supplied R. D. McGrath [4] as well as on films grown at Virginia Tech. Results of the data analysis are consistent with each other and demonstrate the ability of these methods to obtain Curie temperatures for Pd(1-x)Fe(x) films. The data on McGrath's film showed that the measured Curie temperature is less than the corresponding bulk value obtained by other experimenters (Table II). Because the film showed no indication of having undergone physical or chemical change since it was analyzed at Physical Electronics, the shift measured by McGrath [4] appears real. The values obtained here, however, are between 105 and 114 K as opposed to McGrath's prediction of 40 K [4]. The measurement of $X$ gave $T_c = 107$ K. The ratio, 107/170, furthermore, compares favorably to the ratio of nearest neighbors for 2-D and 3-D systems, 4/6=2/3. For this film this result may imply the magnetism is due to giant moments extended over large distances.

Cosputtering has been shown to be a viable method of producing films with clean magnetic phase transitions (film 46). As film 46 was made after films 34 and 39, it could be that the plasma processes at the target were closer to equilibrium. This is an important point if such targets are to be considered for future use.
Some films have even shown evidence of spin-glass behavior (two peaks in $X$). This is a surprising result in view of a recent calculation of a phase diagram that shows no ferromagnetic-spin glass transition above $0.17$% Fe for bulk Pd($1-x$)Fe($x$) [18]. This might be explained by antiferromagnetic interactions between clusters (as opposed to antiferromagnetic interactions between individual atoms). This possibility could expand the phase diagram to include higher iron concentrations.

Magnetization measurements, furthermore, have shown a definite downward progression as film thickness decreases. The data was analyzed according to the assumption that

$$M = M_\infty (1 - T/T_C)^\beta .$$

It was found that the Curie-Weiss value of $T_C$ is closest to the experimental value and slightly lower, while at low temperatures the 2D-Ising model is closest to the experiment. Susceptibility measurements showed good correspondence to Curie-Weiss theory for the critical exponent $\gamma$.

The results of this work can be used as a starting point for a series of experiments. Appendix C contains suggested improvements and new experiments that refine this work and build on it.
REFERENCES


APPENDIX A -- Auger Analysis (Johns Hopkins film)

(1) R. D. McGrath's film--no milling
(2) R. D. McGrath's film--one minute of milling
(3) R. D. McGrath's film--two minutes of milling
AES SURVEY SF = 17.316, 000 DAT = 1.90 08/25/01

EXPANSION FACTOR: 1/35

KINETIC ENERGY, EV

(2)
EXPANSION FACTOR = 1.44

KINETIC ENERGY, EV
APPENDIX B -- Auger Analysis (Virginia Tech film)

(1) Film 46--no milling

(2) Film 46--one minute of milling
EXPANSION FACTOR = 2.50

KINETIC ENERGY, ev
APPENDIX C -- Improvements and Experiments

Improvements:

1) Bake substrates in an ion-pumped vacuum to remove trapped water.

2) Sputter in a system with an ultimate pressure of less than $10^{-8}$ to decrease contamination.

3) Simultaneously produce different thicknesses in the same plasma in order to increase the consistency of preparation.

4) Produce the samples from an alloy target.

5) Use a temperature controller with digital output for automated analysis.

6) Store SQUID and resistivity data digitally for greater precision.

Experiments:

1) Complete magnetic and chemical characterization.

2) Determine the equilibration time for cosputtering of Pd(1-x)Fe(x).

3) Determine the substrate dependence of the shapes of X and M.

4) Find $\beta$, $\gamma$, and 'p'.

5) Use a single element target like Gd for the shift exponent work.

6) Check the influence of an H-field on the magnetization curve.
The vita has been removed from the scanned document
PHASE TRANSITIONS IN
THIN IRON-PALLADIUM FILMS

by

Raymond William Mattozzi

(ABSTRACT)

Sputtered thin films have the capability of producing a very random distribution of alloying elements within a host element. This study demonstrates the ability of the SQUID (Superconducting Quantum Undulating Interference Device) to measure the Curie temperature of thin films of Pd(1-x)Fe(x). The Curie temperatures of these films were found to be significantly less than bulk samples having the same iron concentration. The Curie temperature, furthermore, showed a systematic shift to higher values as the thickness of the film increased. Magnetic structure below the Curie temperature is revealed in magnetization and a.c. susceptibility curves for x=.078. For other samples susceptibility data exhibited more sensitivity than magnetization in revealing magnetic detail below the Curie temperature. We attribute some of this magnetic detail to cluster glass behavior.