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RICE UNIVERSITY

Ultrasonic Generation in Thin Films of TbFe$_2$

by

W. L. Wimbush, Jr.

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ABSTRACT

Ultrasonic Generation in Thin Films of TbFe$_2$

by

Walter Leslie Wimbush, Jr.

Ultrasonic elastic waves have been generated in thin films of Terbium Iron. The thin films of TbFe$_2$ were prepared by vacuum evaporation onto crystalline quartz substrates. The films were observed to possess a strong magnetic anisotropy with the easy axis of magnetization perpendicular to the plane of the film. Hysteresis curves were obtained using a P.A.R. vibrating sample magnetometer. The magnetization data indicated magnetic remanence as high as 90% saturation and coercive forces as large as 10 kOe. These films retained 90% of their maximum ultrasonic production in the absence of an external dc biasing field.

An investigation into the effects of a perpendicular magnetic field during deposition is discussed as well as the effects of an externally applied stress on the film.

A theoretical fit to the experimental data was obtained using a Green's Function approach to the magnetoelastic generation within the film.
To
Joan
Acknowledgement

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Introduction

While the general features of ferromagnetism in rare-earth iron alloys are fairly well understood, the actual interactions involved have yet to be fully explained. It is known that rare earth ions combine with iron ions to form intermetallic compounds of simple stoichiometry. Examples are: $\text{RB}_2$, $\text{RB}_3$, $\text{R}_2\text{B}_{17}$, and $\text{RB}_5$, where $\text{R} =$ Rare Earth ion and $\text{B} =$ Mn, Fe, Co, or Ni. The stoichiometry common to all the systems is $\text{RB}_2$. Hence, it is not unreasonable that the cubic Laves Phase $\text{RB}_2$ compound has received extensive investigation. One particular compound, $\text{TbFe}_2$, has received singular treatment in our lab due to its enormous room temperature magnetostriction$^1$ as well as the largest negative anisotropy$^2$ known. Focusing our efforts on the magnetostrictive properties of $\text{TbFe}_2$, an effort was made to incorporate the dependence of the magnetostrictive properties on other intrinsic properties along with those obtained from the method of fabrication. The $\text{TbFe}_2$ samples were in the form of thin films.

The films used in this study were grown by vapor deposition in high vacuum onto the end of quartz rod substrates. The films ranged in thickness from 1.5 $\mu$m to 8.0 $\mu$m. They were grown on substrates whose ambient temperature was varied between 190°C and 350°C with an
orienting magnetic field normal to the surface of the substrates.

A Princeton Applied Research Vibrating Sample Magnetometer was used to measure the magnetization of the sample as a function of the applied field.

The phonon generation by ferromagnetic resonance was observed using an ultrasonic Pulse Spectrometer. The magnetoelastic interaction is responsible for the microwave generation of the phonons. The magnetic film bonded to the end of a quartz rod was placed in a static magnetic field and driven by a microwave field of 690 MHz, parallel to the film surface. The phonon generation is the result of the interdependence of exchange, spin-orbit, and dipole-dipole interactions on the separation distance. As the spin system is altered by the microwave magnetic field, the lattice deforms so as to minimize the total energy. The oscillating deformation is the phonon, which is transmitted to the quartz rod. After reflection from the opposite end, the shear wave couples again to the spin system to produce an oscillating magnetic field which is detected in the stripline cavity.

It is the main intent of this research program to gain further insight into the microwave generation of phonons. By examining the intensity of phonon generation as a function of the applied field and comparing to the magnetization of the sample as a function of the applied
field, the dependence of the phonon generation on the magnetization can be studied.

A second area of research was the design and construction of a magnetic system capable of supplying a field normal to the substrate surface. The entire system was to be placed in a high vacuum and cooled by forcing air passed the coils of the solenoid.

A third area of research involved the evaluation of the effect of the deposition field on the generation of microwave phonons.

**Magnetic Interaction**

The Rare Earth ions form a sublattice which is antiparallel to the iron ions' sublattice. This alignment can be accounted for by hypothesizing an exchange field, as given by the Weiss Molecular Field Theory. The origin of this exchange field was found in the quantum mechanical treatment of the spins on the ions by Heisenberg. It can be seen to be electrostatic in nature as the Pauli Exclusion Principle favors the separation of parallel spins which would lower the coulomb repulsion energy. Although electrostatic in nature, the exchange interaction between the 4f spin of the Rare Earth ion localized at \( R_1 \) and an electron in the conduction band at \( r \) is given by the Heisenberg expression \( J(r-R_1)S \cdot s \) where \( S \) is the spin of the ion and \( s \) is the conduction electron spin. The conduction electron, having been polarized by the ion at \( R_1 \),
interacts with another ion at \( R_j \); thus the spin of the ion at \( R_i \) interacts with the spin of the ion at \( R_j \) via the conduction electron. This type of exchange interaction is called the Indirect Exchange or the RKKY Interaction, named after the men who developed it, Ruderman and Kittel (1954), Kasuya (1956) and Yosida (1957). The Indirect Exchange can be written as

\[
H_{ij} = -J(R_i - R_j) \mathbf{S}_i \cdot \mathbf{S}_j
\]

and the total exchange is given as the sum over all pairs of ions. The conduction electron polarization is negative with respect to the spin on the Rare Earth ion and positive with respect to the spin on an iron ion. Since the distance between the Rare Earth ion and another Rare Earth ion is approximately the same as the distance between a Rare Earth ion and an iron ion, this suggests, qualitatively, that the Rare Earth ions are polarized with their spins aligned antiparallel to those of the iron ions.

**Crystal Structure**

The set of general principles governing the structure of the Rare Earth alloys are embodied in the rules established by Hume and Rothery. Semiquantitative considerations, such as ionic size, electronegativity, and valence and electron concentration influence the alloying behavior of the Rare Earths.
The major factor influencing the solid solubility is the difference between the size of the solvent and the solute atoms; if the solute atom is larger, strain will develop around it. As a general rule, if the solvent and solute atoms differ by more than 15%, restricted solubility results. Hence, most metallic elements will have restricted solubility with the Rare Earth elements.

The structural sequences observed in phase diagrams have a fixed ratio of valence electrons to atoms. Each of these phases will be referred to as intermetallic compounds. The RB₂ phases depend primarily on definite atom size ratios which lie between 1.10 and 1.30. Furthermore, the MgCu₂ structure (C15) Cubic Laves Phase requires valence electron concentrations between 1.33 and 1.75, as was shown by Laves and Witte.⁴ The arrangement of the atoms in the Cubic Laves Phase structure is seen in figure 1 after Taylor⁵ and Berry and Raynor.⁶ The iron ions form tetrahedral structures oriented symmetrically about the [111] axial direction. With a Terbium ion situated at the origin and the [111] axis as the polar axis, the symmetry can be visualized as in figure 2. There are twelve nearest neighbor iron ions, each at a distance of 0.414 \(a_o\) and four next nearest neighbor Terbium ions each at a distance of 0.433 \(a_o\), where \(a_o\) is the dimension of the unit cell. The threefold axis is the axis of crystal symmetry and thus the axis of easy magnetization.
Figure 1: The Cubic Laves Phase Structure of TbFe₂ after Taylor.

Figure 1b: The two sublattices of TbFe₂ shown independently.
Figure 2: The nearest and next nearest neighbors relative to the Terbium ion (note the symmetry about the [111] axis).
Crystal Field

The Rare Earth ion is situated in an inhomogeneous electric field produced by neighboring ions within the crystal. Approximating the neighboring ions as point charges, the electrostatic potential at the site of the Rare Earth ion would be

\[ V(r) = \sum_i \frac{e Z_i}{|r-R_i|} . \]

\( V(r) \) satisfies Laplace's equation and can be written in a spherical harmonic expansion:

\[ V(r) = \sum_{\lambda} \sum_{|m| \leq \lambda} a_\lambda^m r^\lambda Y_\lambda^m(\theta, \phi) = \sum_{\lambda, |m| \leq \lambda} v_\lambda^m \]

where

\[ a_\lambda^m = \sum_j \frac{4\pi}{2\lambda+1} \frac{Z_j}{R_j^{\lambda+1}} \frac{(-1)^m}{(\lambda+1)} Y_{\lambda+1}^{-m}(\theta, \phi) . \]

The crystal field associated with cubic symmetry is expressed as

\[ V(r) = V_4^0 + V_4^4 + V_6^0 + V_6^6 . \]

The crystal field causes a mixing of some of the ground states, thus removing some of the degeneracy. The matrix elements between multiplet states \( |JM_j\rangle \) can be evaluated using the method of operator equivalents. For example, in \( V_4^0 \)
\[(35z^4-30r^2z^2 + 3r^4) - \beta\langle r^4 \rangle [35J_z^4-30J(J+1)J_x^2
\]
\[+ 25J_z^2-6J(J+1)+3J^2(J+1)^2]\]
\[= \beta\langle r^4 \rangle 0_4^0.\]

Hence, the equivalent operator for the cubic crystal field is

\[V_c = E_4^0(0_4^0 + 5 0_4^4) + E_6^0(0_6^0 + 21 0_6^4)\]

where the terms are defined in the Appendix.

This crystal field tends to align the magnetic moments along particular crystallographic directions. For the Laves Phase compound, TbFe$_2$, the direction of easy magnetization is along the [111] axis.\textsuperscript{7,8}

**Anisotropy**

Magnetic anisotropy represents the tendency of the magnetization to resist turning from its preferred direction. The energy associated with rotating the magnetization from this preferred direction is expressed as a polynomial in direction cosines multiplied by phenomenological anisotropy constants. The form of these polynomials is dictated by the symmetry of the crystal under consideration. The magnetic anisotropy energy for a crystal of cubic symmetry is

\[E_a = K_1(\alpha_1^2 \alpha_2^2+\alpha_2^2 \alpha_3^2+\alpha_3^2 \alpha_1^2) + K_2(\alpha_1^4+\alpha_2^4+\alpha_3^4)\]

where \(\alpha_2\) are the direction cosines of the magnetization and \(K_1, K_2\) are the anisotropy constants.
The origin of this anisotropy may be pseudodipolar or pseudoquadrupolar or single ion interactions resulting from crystal field splitting.\textsuperscript{9} Callen and Callen\textsuperscript{10} express the single ion anisotropy as

\[ H_a = \sum_{\lambda} k_{1}^{\alpha, \lambda} \]

where \( k_{1} \) is defined as "intrinsic anisotropy coefficient" and \( K_{\alpha, \lambda} \) is a Cubic Tensor Operator. The similarity between these operators and the crystal field operators used previously can be seen from

\[ K_{\alpha, \lambda} = \sum_{m} Q_{\alpha, \mu, \lambda} \nu_{1}^{m}. \]

In another paper, Callen and Callen\textsuperscript{11} show that the temperature dependent anisotropy constants are functions of the magnetization in the sample, i.e.,

\[ K_{1}(T) = K_{1}(0) \hat{I}_{9/2}(L^{-1}(m_{r}(T))) \]

where \( \hat{I}_{9/2} \) is the \( l = 4 \) reduced hyperbolic Bessel function and \( L^{-1}(m_{r}) \) is the inverse Langevin function with \( m_{r} \), the reduced magnetization \( (m(t)/m(0)) \), as its argument. \( K_{1}(0) \) was recently reported by Clark\textsuperscript{12} to be \(-7.6\times10^{8}\) ergs/cc.

For metallic films bonded to substrates there may be an additional contribution to the anisotropy due to thermal stress induced into the film by a difference in thermal expansion of the film and substrate. This anisotropy will be treated in greater detail in later sections.
Magnetostriction

Magnetostriction is the change in the shape of a ferromagnetic specimen due to a change in the magnetization. It is a result of the distance dependence of the exchange integral, spin orbit interaction or the dipole-dipole interaction. As might be expected, the magnetostrictive strains which are related to the magnetic anisotropy through the elastic energy are very large for TbFe$_2$ (2630 ppm) as compared with YFe$_2$ (2.6 ppm).

The fractional change in length of a cubic crystal has been determined by Chikazumi$^9$ as

$$\frac{\delta l}{l} = \frac{3}{2} \lambda_{100} (\alpha_1^2 \beta_1^2 + \alpha_2^2 \beta_2^2 + \alpha_3^2 \beta_3^2 - 1/3)$$

$$+ 3 \lambda_{111} (\alpha_1 \alpha_2 \beta_1 \beta_2 + \alpha_2 \alpha_3 \beta_2 \beta_3 + \alpha_1 \alpha_3 \beta_1 \beta_3)$$

where $\alpha, \beta$ are the direction cosines of the magnetization and the measured elongation, respectively. The temperature dependence of the magnetostriction has been given in the general treatment of Callen and Callen,$^{11}$ who used a single ion model to describe the strain dependence of the exchange energy and a two ion model to describe the dependence on the dipole-dipole energy. Their single ion theory predicts

$$\lambda(T) = \lambda(0) \hat{i}_{5/2} \left(L^{-1}(m_\text{r}) \right)$$

where $\hat{i}_{5/2}$ is the reduced hyperbolic Bessel function with $l = 2$. 
Magnetization

The obvious applications of the Rare Earth-Iron Compounds can be seen as a direct result of their magnetostrictive and magnetic properties. The latter property has prompted extensive investigations of them as permanent magnets.\textsuperscript{13} Materials combining high anisotropy with a high value of saturation magnetization would make permanent magnets of outstanding technical properties.

The magnetic properties of a material can be evaluated by an examination of its M-H loop. The M-H loop in figure 3 describes the magnetization process in a sample. In the illustration in 3(a) it is seen that the initial magnetization takes place by the growth of the domain nearest the field direction until only one domain occupies the sample. The direction of the single domain then rotates toward the direction of the applied field. Once the material becomes a single domain it has achieved its maximum magnetization. For a polycrystal each grain may be saturated, but at a slight angle to the applied field. With greater applied fields, the direction of magnetization within each grain can be rotated into alignment with the direction of the applied field. When the applied field is decreased to zero, each grain remains magnetized but returns to its original direction with respect to the field direction. Chikazumi\textsuperscript{9} has shown the resultant magnetization or remanent
Figure 3: Schematic illustration of domain structure at various stages of the magnetization process.

Figure 4: A hysteresis curve for a ferromagnetic material.
magnetization for a cubic crystalline material with random
grain orientation to be 0.866 of the saturation value. By
prealignment of the grains, the remanent magnetization may
be increased. This was attempted by growing the crystals
within a biasing field. Reversing the applied field, the
magnetization first rotates then, at a value of the applied
field called the nucleating field, domains form and dis-
placement of the domain walls take place until each
particle is again a single domain with its magnetization
toward the reversed field. The value of the reversed field
required to bring the magnetization to zero is called the
coercive field or coercive force.

If magnetization reversal could take place only by
coherent rotation, then the samples possessing large
magnetocrystalline anisotropy should also have large coer-
cive forces approaching a theoretical upper limit of
\( H_c = 2K/M_s \) which for \( \text{TbFe}_2 \) would be 175 kOe.\(^1\)

Since the reversal does take place by the nucleation
of domains and the subsequent motion of the domain walls
through the crystal, the coercive force can be increased
by the presence of small imperfections in the material.
The imperfection impedes the domain wall motion; when a
domain wall passes through an imperfection, spike domains
form lowering the total magnetic energy.\(^9\)

Becker\(^{14}\) has verified that large scale magnetization
rotation is of less importance than nucleation and domain
wall motion for small samples. Domain boundaries are not always entirely removed, and the field normally required to nucleate domain walls is lessened. Magnetizing a sample in a larger field will increase the coercivity of this class of samples. Zijlstra\textsuperscript{15} has emphasized the role of imperfections acting as pinning sites. A pinning field, $H_p$, is required to detach a wall from its pinning sites. These elementary processes would be averaged out for a polycrystalline substance containing many domains. The existence of pinning sites would result in an increase in coercive fields.

The maximum magnetic moment in the sample is a function of the net spin within the unit cell. In the case of Rare Earth-Iron alloys, the spin of the Rare Earth ion couples antiparallel to the spin of the Iron ion. Hence, in the light Rare Earths ($J = |L-S|$) the moment of the Rare Earth ion is parallel to the Iron ion; whereas, in the heavy Rare Earths ($J = |L+S|$) the moment of the Rare Earth ion couples antiparallel to the Iron ion. Thus, TbFe$_2$ has a saturation moment of 4.72/mole\textsuperscript{16} which Buschow and Van Stapele report as 82 emu/g. A more recent estimate by Clark using a larger magnetic field states that the saturation magnetization is greater than 90 emu/g.\textsuperscript{12}

Becker\textsuperscript{13} points out that in Rare Earth-Iron compounds, the Curie temperatures increase with decreasing Fe content, opposite to the behavior of Rare Earth-Cobalt Compounds.
Figure 5: Illustrating Zijlstra's concept of the effect of nucleating and pinning sites on the hysteresis curve.
For example, the Curie temperatures for TbFe$_2$, TbFe$_3$, TbFe$_{23}$, and Tb$_2$Fe$_{17}$ are 711°K, 648°K, 574°K, and 408°K, respectively. Although SmCo$_5$ is under investigation by several researchers,$^{17}$ the possession of both the high Curie Temperature and the largest negative cubic anisotropy of the Rare Earth alloys, make TbFe$_2$ an outstanding candidate for permanent magnet material.
Phonon Generation

The generation of microwave phonons by ferromagnetic resonance or spin wave resonance in thin ferromagnetic films has been investigated by several researchers.\textsuperscript{18-22} The geometry of the system, figure (6), is that of a magnetic thin film bonded to one end of a quartz rod, placed in a static magnetic field and driven by a microwave field in the stripline cavity.

Using a microwave cavity to magnetically couple with the film, an incident microwave pulse can be observed to produce microwave echo pulses on resonance. The ferromagnetic resonance transmission is a function of the relative impedances of the rod and the film and the DC magnetic field which determines the resonance. On resonance, the film generates phonons that travel across the film-substrate boundary into the rod. The phonons are reflected from the opposite end of the rod and propagate back across the film-substrate boundary into the film, where they re-excite the resonance and regenerate a microwave field. These phonon echoes repeat the process, until they are attenuated in the rod. During the pulse of microwave magnetic field, an acoustic standing mode is set up within the film of finite thickness. Kobayashi\textsuperscript{18} purposely chose the acoustic impedance of the rod to be 0.5 that of the film, so that only part of the acoustic power is transmitted into the rod.
Maximum acoustic power is attained when the maximum acoustic power is transmitted out of the film without destroying the acoustic standing wave resonance. In order to minimize conductivity losses, the film thickness must be less than the skin depth.

Since the acoustic wave is generated via the magnetic system, it is desirable to have a material with a large magnetostriction such as TbFe$_2$.

An expression for the acoustic power transmitted into the rod by the magnetostrictively generated phonon will be derived following a theory developed by Seavey.$^{21}$

Fundamental to all theories is the magnetoelastic interaction between the magnetization and the elastic strains within the metal film. This interaction can be expressed as:

$$U = B \left( \frac{M_x^2}{M^2} e_{xx} + \frac{M_y^2}{M^2} e_{yy} + \frac{M_z^2}{M^2} e_{zz} + \frac{M_x M_y}{M^2} e_{xy} + \frac{M_x M_z}{M^2} e_{xz} + \frac{M_y M_z}{M^2} e_{yz} \right).$$

(1)

The magnetostrictive stress produced in the plane of the film is obtained by differentiating (1) with respect to the appropriate strains. Thus

$$X_z = \frac{\partial u}{\partial e_{xz}} = B \frac{M_x M_z}{M^2} \quad Y_z = \frac{\partial u}{\partial e_{yz}} = B \frac{M_y M_z}{M^2}$$

(2)

where $X_z$ represents a force in the x direction on a plane whose normal is in the z direction.
Phonons are generated by the variations of this strain in the z direction. Assuming an isotropic medium, the equations of motion for the phonons would be

\[
\frac{\partial^2 u}{\partial z^2} - \frac{1}{v_t^2} \frac{\partial^2 u}{\partial t^2} = - \frac{BM_z}{\mu M^2} \frac{\partial M_x}{\partial z}
\]

\[
\frac{\partial^2 v}{\partial z^2} - \frac{1}{v_t^2} \frac{\partial^2 v}{\partial t^2} = \frac{BM_z}{\mu M^2} \frac{\partial M_y}{\partial z}
\]

A similar equation of motion was used by McLane in his quantum mechanical treatment of the magnetoelastic interaction where he determined that only the $e_{xz}$ and $e_{yz}$ strains would contribute to the microwave generation of phonons.

Since the driving term is a derivative of the microwave magnetization component, a gradient in the microwave magnetization is required for phonon generation. An expression for the microwave magnetization is developed from a consideration of the ferromagnetic resonance in the film.

A Hamiltonian consisting of simple dipole, magnetoelastic and anisotropy interactions has the form

\[
\mathcal{K} = - \vec{M} \cdot \vec{H} - KM_z^2 + B[e_{xz} M_x M_z + e_{yz} M_y M_z].
\]

The magnetoelastic interaction will be small compared to either $\vec{M} \cdot \vec{H}$, the dipole interaction, or $KM_z^2$, the anisotropy interaction, so it will be treated as a perturbation. The microwave component of the magnetization can be determined
as a ferromagnetic resonance. The local fields acting on a dipole are described as

\begin{align*}
H_x &= H_{rf} + \Gamma M_x \\
H_y &= \Gamma M_y \\
H_z &= H + \Gamma M_z + K M_z
\end{align*}

where \( H \) is the externally applied field, \( \Gamma M_i \) is the molecular field in the \( i \) direction, \( H_{rf} \) is the microwave field in the cavity, and \( KM_z \) is the anisotropy field.

Since the rate of change of the angular momentum (\( \hbar J \)) is equal to the torque on the dipole,

\begin{align*}
\hbar \frac{dJ}{dt} &= \mu \times \vec{H} \\
\frac{dM}{dt} &= \gamma \vec{M} \times \vec{H}
\end{align*}

or

\begin{align*}
\frac{d\vec{M}}{dt} &= \gamma \vec{M} \times \vec{H}
\end{align*}

where \( \mu = \gamma \hbar J \) and \( \vec{M} = N \vec{\mu} \) were used.

Including the relaxation terms, the familiar Block Equations result

\begin{align*}
\dot{M}_x &= \gamma (M_y H_z - M_z H_y) - \frac{M_x}{T} \\
\dot{M}_y &= \gamma (M_z H_x - M_x H_z) - \frac{M_y}{T} \\
\dot{M}_z &= \gamma (M_x H_y - M_y H_x) - \frac{(M_z - M_0)}{T_1}
\end{align*}

where \( T \) and \( T_1 \) are the transverse and longitudinal relaxation terms respectively.
Considering $M_x$, a concise expression can be determined

\[
\dot{M}_x = \gamma (M_y H z - M_z H y) - \frac{M_x}{T}
\]

\[
\dot{M}_x = \gamma [M_y (H + T M_z + K M_z) - M_z T M_y] - \frac{M_x}{T}
\]

\[
\dot{M}_x = \gamma [H + K M_z] M_y - \frac{M_x}{T}
\]

\[
\dot{M}_x = \omega_o M_y - \frac{M_x}{T}
\]

(8)

where \( \omega_o = \gamma [H + K M_z] \).

In a similar manner, expression for $M_y$ and $M_z$ can be shown to be

\[
\dot{M}_y = - \omega_o M_x + \omega_l M_z - \frac{M_y}{T}
\]

\[
\dot{M}_z = - \frac{(M_z - M_o)}{T l}
\]

(9)

where \( \omega_l = \gamma H_{rf} \). If $H_{rf}$, $M_t \propto e^{-i \omega t - \frac{z}{\delta}}$

\[
(i \omega + \frac{1}{T}) \vec{M}_x = \omega_o \vec{M}_y
\]

\[
(i \omega + \frac{1}{T}) \vec{M}_y = -\omega_o \vec{M}_x + \omega_l \vec{M}
\]

(10)

Letting $\Omega = (i \omega + \frac{1}{T})$, the solutions become

\[
\vec{M}_x = \frac{\omega_o M}{\omega_o^2 + \Omega^2}, \quad \vec{M}_y = \frac{\omega_l \Omega M}{\omega_o^2 + \Omega^2}
\]

(11)

A further simplification can be made utilizing spin wave resonance data on Rare Earth Garnets\(^{23}\) which indicated an approximate line width of 9000 MHz compared to $\omega$ which
was approximately 700 MHz. Hence, \( \omega \) will be neglected. Setting \( \Omega = \frac{1}{T} = \gamma H_R \),

\[
M_x = H_{\text{rf}}^O \frac{(H+KM_z)M}{(H+KM_z)^2 + H_R^2}, \quad M_y = \frac{H_{\text{rf}}^O H_R M}{(H+KM_z) + H_R^2}
\]

(13) and \( M_x = \overline{M}_x e^{-i\omega t - z/\delta} \) where \( \delta \) is the skin depth given by \( \delta^2 = c^2/2\pi\sigma\omega \).

Returning to equation (3), the solution can be expressed in terms of the Green's function as

\[
u = \sum_{z'} G(z, z') \left[ B \frac{\partial M_x}{\partial z'} \right] dz'
\]

The Green's function is a sum of two parts, one propagating in the +z direction and another in the −z direction. Total reflections occur at \( z = 0 \), an antinode position, without changing phase. This satisfies one of the boundary conditions, that the free surface should be stress free.

Summing the series of reflections within the film for each part of the Green's function, the following expression is obtained: (a detailed derivation is given in the Appendix)

\[
G(z, z') = \frac{i}{k_1} \left( \frac{e^{-ik_1L}}{1 - r e^{-2ik_1L}} \right) e^{-ik_2(z-L)}
\]

where \( t \) is the transmission coefficient and \( r \) is the reflection coefficient at the film-substrate interface. \( k_1 \)
and $k_2$ are the phonon wave vectors in the film and substrate respectively. The transmission and reflection coefficient are given by

\begin{equation}
(16) \quad r = \frac{Z_1 - Z_2}{Z_1 + Z_2} \quad t = \frac{2Z_1}{Z_1 + Z_2} \quad Z_1 = \rho_1 v_1 \quad Z_2 = \rho_2 v_2
\end{equation}

$Z_1$ and $Z_2$ are the acoustic impedances of the film and rod, respectively.

The elastic energy density in the rod is

\begin{equation}
(17) \quad U_e = \frac{1}{2} \rho_2 v_2^2 \left| \frac{du}{dz} \right|^2
\end{equation}

\[= \frac{1}{2} \rho_2 v_2^2 |G(z,z')\left[ \frac{B}{\mu M} \frac{\partial X}{\partial z'} \right]|^2\]

and the acoustic power transmitted is given by

\begin{equation}
(18) \quad P_{AC} = U_e v_2 A_f
\end{equation}

where $A_f$ is the surface area of the film.

Substituting (15) into (14)

\begin{equation}
(19) \quad u = \int_0^\infty \frac{i}{k_1} \left( \frac{te^{-2ik_1L}}{1-r e^{-ik_1L}} \right) e^{-ik_2(Z-L)} \left[ \frac{B}{\mu M} \frac{\partial X}{\partial z'} \right] dz'
\end{equation}

\begin{equation}
(20) \quad u = \frac{B}{\mu M} \frac{i}{k_1} \left\{ \frac{te^{-2ik_1L}}{1-r e^{-ik_1L}} \right\} \int_0^L \left[ \frac{\partial X}{\partial z'} \right] \cos k_1 z' dz'
\end{equation}

\begin{equation}
(21) \quad u = A \int_0^L \left[ \frac{\partial X}{\partial z'} \right] \cos k_1 z' dz'
\end{equation}
Substituting the expression for the microwave magnetization from (13) into (21) results in

\[ u = A \bar{M}_x \int_0^L -\frac{1}{\delta} e^{-z'/\delta} \cos k_1z' \, dz' \]

\[ u = A \bar{M}_x [1 - B' \cos k_1L] \]

where \( B' = \frac{e^{-L/\delta}}{\delta} \).

The elastic energy density in the rod is given by (17)

\[ U_e = \frac{1}{2} \rho_2 v_2^2 \left[ \frac{k_2 B}{\mu M k_1} \left( e^{-ik_1 L} - e^{-ik_2 (L - L)} \right) \left( e^{2ik_1 L} - e^{2ik_2 (L - L)} \right) \right] \bar{M}_x [1 - B' \cos k_1L]^2 \]

Using \( \rho_2 = \frac{1}{v_2^2} \), the acoustic power in the rod is

\[ P_{AC} = \frac{B^2 A_f}{2M^2 Z_1} \frac{(Z_2/Z_1)}{\sin^2 k_1L + (Z_2/Z_1)^2 \cos^2 k_1L} [1 - B' \cos k_1L]^2 \bar{M}_x^2 = \frac{P_d}{8\pi Qu} \left( \frac{\lambda_g}{\lambda_0} \right)^2 \]

The power dissipated in the cavity is given by Seavey as

\[ P_d = \frac{\omega h^2 V}{8\pi Qu} \left( \frac{\lambda_g^2}{4\lambda_0^2} \right) \]

where \( h \) is the maximum microwave field strength in the cavity, \( \lambda_g \) is the guide wavelength, \( \lambda_0 \) is the free space wavelength, \( Qu \) is the \( Q \) of the cavity, \( V = ab(\lambda_g/2) \) is the cavity volume, and \( a \) and \( b \) are the effective length and width of the cavity.

The ratio of the acoustic power transmitted into the rod to the microwave power dissipated in the cavity walls is given by \( P_{AC}/P_d \). If the assumption is made that the
process of generating phonons by the microwave field is as efficient as generating a microwave field by phonons, then

\[ \frac{P_{\text{out}}}{P_{\text{ac}}} = \frac{P_{\text{ac}}}{P_{\text{d}}} \]

multiplying by \( \frac{P_{\text{ac}}}{P_{\text{d}}} \), the ratio of the power out of the cavity to the power into the cavity is given as

\[ \frac{P_{\text{o}}}{P_{\text{d}}} = \left( \frac{P_{\text{ac}}}{P_{\text{d}}} \right)^2 \]

Due to the dependence of the acoustical power on the magnetization component in the x direction, a further simplification can be achieved through an assumption that the domains are small. If the domains are sufficiently small that the magnetization within each domain is either in the positive or negative z direction, then the anisotropy term will be \( K|M| \) and the demagnetization term will be \(-D|M|\). Hence, the microwave magnetization would be

\[ |\vec{M}_x| = P \frac{H_{\text{ref}}(H-D|M|+K|M|)M}{(H-D|M|+K|M|)^2+H_R^2} + (1-P) \frac{H_{\text{ref}}(H+D|M|-K|M)M}{(H+D|M|-K|M)^2+H_R^2} \]

The anisotropy field will have the same direction as the magnetization within the domain, and the demagnetizing field will have the opposite direction to the domain magnetization. \( P \) is the fraction of the sample with domains in the positive direction.

The phonon generation is seen to be strongly dependent on the applied field and the magnetic moment of the sample.
in the z direction. As Seavey pointed out one of the disadvantages of magnetic thin film microwave transducers is the requirement of strong biasing magnetic fields. One of the advantages of TbFe$_2$ is that it has a high Curie Temperature, 711°C, with a large remanence in zero applied field. Thus phonon generation is a reality without the need of strong D.C. Magnets. Prototype transducers have been designed at both government and industrial laboratories using TbFe$_2$ and the ternary alloy Tb$_{.3}$Dy$_{.7}$Fe$_2$ with high efficiency. These materials promise an attractive future for magnetostrictive transducers.
Experimental Considerations

It is the main interest of this research program to investigate the parameters influencing the microwave generation of phonons within thin films of a Terbium Iron Compound, TbFe$_2$. Several parameters are suggested from the previous theoretical section: those influencing the magnetization of the film and those affecting the anisotropy of the film. Examples of the former are: prealignment of the grains within the film, nucleation and pinning sites due to defects induced during deposition, and temperature. Examples of the latter are: stress due to defects, thermal stress and applied stress. McLane studied the dependence of phonon generation on the various deposition parameters: substrate temperature, deposition rate, film thickness and composition.

By depositing the films in a magnetic field in order to prealign the crystallites, the dependence of the magnetization on this parameter is determined within the limits of the aligning magnet. By comparing the magnetization curves (magnetic moment of the sample vs. applied field) determined by a vibrating sample magnetometer to the "ultrasonic curves" (phonon pulse amplitude vs. applied field) determined by a pulse spectrometer, the functional dependence on the magnetization suggested by the theory was verified.
Since the temperature affects both the magnetization and the anisotropy of the sample, its overall effect on the phonon generation within the film was determined using "ultrasonic curves" produced at temperatures between 77°K and room temperature.

Films of TbFe$_2$ possess an uniaxial easy axis perpendicular to the plane of the film with a large perpendicular anisotropy. K, the perpendicular anisotropy constant, has two components: a shape demagnetization factor given by $2\pi M^2$ plus a structural contribution due to thermal expansion and intrinsic strain resulting from defects induced during deposition. The intrinsic strain will depend on the quality of the vacuum during deposition and may vary from sample to sample. The thermal stress which is due to the difference in the thermal expansion of the film relative to the substrate will change with the temperature. The strain-magnetoelastic anisotropy is produced by the crystal trying to minimize the magnetocrystalline anisotropy energy arising from the strain. The anisotropy constant, K, could be determined by a torque magnetometer, however, the expected anisotropy is greater than $10^3$ ergs per cc. which is too large for the existing torque magnetometers. The anisotropy constant may also be determined through the relation $H_c = 2K/M$ if fields on the order of $H_c$ could be obtained. The magnetometer we had access to was capable of 14 k0e using a small air gap; this necessitated going to
quartz disks for substrates in place of the quartz rods we were using. However, 14 k0e was still an order of magnitude too small. Measurement was therefore restricted to the variation of the anisotropy calculated from the predicted variation in the measured coercive field. Further investigation into the effect of stress on the phonon intensity was made by creating a strain within a ruby rod substrate directly above the film which in turn would produce a stress on the film.

The dependence of phonon generation and the coercive field on the deposition parameters impel us to reevaluate their overall effect on the films. The possible mechanisms through which these parameters could exercise their influence were believed to be: (a) the substrate temperature which affects the grain size and thermal stress induced in the film upon cooling to room temperature; (b) the deposition rate which determines the composition ratio of the metals within the film; (c) the duration of the bake-out period which affects the final atmosphere of residual gases inside the chamber during deposition; and (d) the final pressure during deposition which affects the purity of the films which in turn determines the number of nucleation and pinning sites present within the film.

Elaborating further, the substrate temperature would cause thermal strain within the film as described earlier. The effect of the substrate temperature on the grain size
would be due to the increase mobility of the evaporant ions (molecules) on the warmer substrate surface.\textsuperscript{28}

The deposition rate would influence the composition of the film due to the difference of the melting points and the vapor pressure of the constituents: (Tb: M.P. 1356°C and V.P. 6x10\textsuperscript{-3} torr; Fe: M.P. 1535°C and V.P. 4x10\textsuperscript{-2} torr). Since the temperature of the boat was measured to be 1675°C by a Leeds & Northrup optical pyrometer, the consistency of the proper stoichiometric ratio was attributed to the high binding energy of the compound, TbFe\textsubscript{2}, which was commercially prepared before evaporation. Examination of the films by X-ray fluorescence gave an indication that there was consistency in the overall composition of the films, but was unable to yield any information as to the composition of the individual grains throughout the film. Micro X-ray analysis could have shed some light on this problem.

The quality of the vacuum present during deposition is an important parameter and a difficult one to control. The vacuum system used was capable of pressures as low as 10\textsuperscript{-7} torr after lengthy bake-out periods. The pressure invariably increased to 10\textsuperscript{-5} torr during deposition and decreased to 10\textsuperscript{-7} or 10\textsuperscript{-8} torr after deposition. Modern vacuum systems are capable of pressures as low as 10\textsuperscript{-11} torr before deposition and 10\textsuperscript{-9} torr during deposition. Since a monolayer of oxygen can form in 2 seconds at a pressure of 10\textsuperscript{-6} torr (300°C)K, compared to 20 seconds at a pressure
of $10^{-7}$ torr, every effort was made to achieve the best possible vacuum. Moreover, the maximum deposition rate was used to minimize the possibility of impurities within the film. An increased deposition rate would increase the number of structural defects within the film; these structural defects would act as nucleation and pinning sites which could be removed by annealing the films at elevated temperatures ($> 350^\circ$C). Clark has shown a marked increase in the coercive force of amorphous TbFe$_2$ films after annealing.

A full theoretical treatment of the effect of an applied magnetic field during deposition on the alignment of the crystallites has not been carried out. However, it is believed that fields greater than 3 kOe are required to prealign the crystallites. Unfortunately, fields greater than 3 kOe could not be maintained for long periods of time; thus we were forced to decrease the field to 2 kOe after deposition. Again substituting a quartz disk in place of the quartz rod, allowed us to decrease the air gap thus increasing the field during deposition.
Experimental Method and Apparatus

The investigations reported here are an outgrowth of the investigations of McLane$^{22}$ into the magnetostrictive ultrasonic generation in Rare Earth-Iron Thin Films. The basic technique for production of the films was developed by McLane, and thoroughly described in his thesis. The major contribution of this investigation is in the effect of stress and of a biasing magnetic field during deposition on the magnetic and transducer properties of the films.

Deposition Apparatus

The films used in this investigation were grown by vapor deposition in a high vacuum onto the end of either a quartz rod or a quartz disk. The vacuum obtained within the 100 liter bell jar was produced by a diffusion pump system capable of pressures on the order of $10^{-7}$ torr. The final vacuum was measured using a CVC Ionization Gauge. The vacuum in the foreline and roughing line was measured by thermocouple vacuum gauge tubes. The entire vacuum system is shown in figure 7. The external structure consists of the bell jar mounted on a 20 inch diameter baseplate. The internal superstructure consists of a 3/8 inch thick circular aluminum plate supported by three aluminum columns. Attached to this plate are the thickness monitor sensor, the substrate holder (by means of thermally insulating standoffs), the remote controlled shutter system and the electromagnet used during deposition.
Figure 6. An illustration indicating the orientation of the film on a quartz substrate in the perpendicular DC Applied Field and the in-plane microwave field.
Figure 7: DEPOSITION SYSTEM
Substrate temperatures were monitored by a solid state Weather Measure Time-Proportional Controller using a chromel-alumel thermocouple and commercial cylindrical heaters of various power ratings (50-150 watts). These heaters were inserted into the substrate holders which were machined out of aluminum and copper. The original design of the substrate holder-shutter system (figure 8) proved inefficient and costly; the revised system (figure 9) keeping the shutter stationary and moving the substrate holder proved to be better adapted to depositions within a magnetic field. The temperature for all depositions was maintained within 1°C of the desired temperature for at least one hour prior to deposition. The shutter system isolated the surface of the substrates from the residual gases inside the chamber until the substrate was moved into position a few seconds before deposition.

The thickness of the films was monitored by an Inficon Film Thickness Monitor. The unit monitors the resonant frequency of the sensor crystal mounted near the substrate. The resonant frequency of the crystal varies linearly with the mass of the film deposited on the crystal. The density of the film is programmed into the control unit which then computes the thickness of the film and displays it in digital form. It was assumed that the source radiates the evaporant uniformly over a hemispherical surface so that the density of the flux could be assumed to obey a $\frac{1}{R^2}$
Figure 8: The original design for the substrate-holder system incorporates a moveable shutter with a stationary substrate holder.
Figure 9 The revised design for the substrate holder system incorporating the moveable substrate holder with a stationary shutter.
dependence in a small solid angle above the source boat. Since the sensor was twice as far from the source as the substrate, the film thickness on the sensor would be one fourth the thickness of the film on the substrate. By programming a density of one fourth the actual value of the material into the control unit, the calculated thickness would be corrected for the difference in the distance from the source boat to the sensor crystal.

The substrates used for this investigation were three millimeter diameter single crystal x-cut quartz rods, 1.8 cm long, and x-cut quartz disks, 1 mm thick. Fused quartz rods and disks of the same dimensions as the single crystal substrates were used to check the effect of single crystal substrates on induced epitaxial growth of the films. Unfortunately, due to the high frequency losses in amorphous material, the reflected ultrasonic pulses could not be detected if they were generated. All substrates were cleaned by a multiple ultrasonic bath process developed by McLane\textsuperscript{22} terminating in a final bath of 99.9\% pure Hexane.

The magnet used for prealignment of the crystallites during deposition was designed by the author to produce a field perpendicular to the surface of the substrates while allowing perpendicular access of the evaporant to the substrate surface as well. The magnet consisted of a solenoid, encased within a stainless steel housing with feed-throughs
to permit cooling the coils inside the vacuum chamber, and a soft iron core (figure 10). From the solenoid the insulated copper wire was threaded through the copper tubing to supply the current to the coils. With forced air cooling the coil, a current of 25 amps corresponding to a field of 2.5 kOe could be maintained without damage to the coil. The need for larger fields meant larger currents and a more efficient cooling system. First, the air was forced through a cooling coil immersed in liquid Nitrogen. When still higher fields were desired, the coils were cooled directly by the liquid Nitrogen vapors. This had the added feature of providing a liquid Nitrogen cold trap inside the deposition chamber which improved the vacuum by half an order of magnitude. The increased current was supplied by a second power supply placed in series with a 10 v. 100 amps Hewlett Packard Power Supply. An inside cone was machined out of the lower pole piece of the core to permit the evaporant to enter between the poles perpendicular to the substrate. The core and cooling system for the solenoid were designed and fabricated by the author. A second solenoid was designed for the core but was never fabricated.

The interior of the vacuum chamber was cleaned prior to each deposition run. All surfaces were left clear of any visible residue by wiping first with dilute Nitric Acid then rinsing with Acetone.
Figure 10: The electromagnet used for prealigning the crystallites during deposition. Note: the inside cone hollowed out of the lower pole.
Deposition Technique:

All preparations having been completed the system was pumped down to approximately $10^{-7}$ torr while the internal structures were baked out at 150°C by a quartz heater. The system remained stable under these conditions for six to eighteen hours prior to deposition. It was previously believed that long bake-out periods would increase remanence; this, however, was not observed. Flushing the system with argon gas prior to the final pump-down was also tried in an effort to create a controlled atmosphere during deposition. The substrate temperature controller was set for the desired temperature after the pressure dropped to $10^{-7}$ torr.

The actual deposition procedure consisted of first melting the sample material in the boat allowing it to outgas for approximately 30 seconds. It would then take approximately five minutes after the current in the boats was turned off for the pressure to return to its previous value. If the deposition was to occur with the magnetic field, the power supplies would have been set for the desired current at this time. The substrate was brought into position directly above the boat containing the TbFe$_2$ source; the source material was again melted and quickly evaporated by turning the power on full. When the thickness monitor indicated the desired thickness, the current was quickly reduced to zero. A one micron thick coating
of Si0 was then deposited over the metal film to protect it from oxidation. The procedure was repeated for the remaining substrates allowing five minutes between each deposition for the pressure to return to its previous value. The current in the magnet was reduced to 25 amps to protect the solenoid, and the substrate temperature controller was turned off. The temperature of the substrate holder required about one hour to drop below 200°C. The magnet was completely deenergized at this temperature because no effect had been observed for annealing inside a magnetic field below this temperature. If it were possible, we would have annealed the films immediately after deposition for several hours before lowering the substrate temperature (here we are assuming annealing the film immediately following deposition would require less time for the Tb and the Fe ions to diffuse through the film removing any inhomogeneities in the structure). The pressure was maintained below $10^{-5}$ torr until the substrates reached room temperature.

After removal from the Vacuum Chamber, the films went through a series of tests. The grain structure was examined under an American Optical Metalograph, and a textured axis was observed, i.e., the grain boundaries formed ellipsoidal shapes directed along a common axis. A scanning electron microscope was used to examine the film for structural defects such as voids, inclusions or peeling from the sub-
strates; most samples were relatively free of such defects. The samples were then tested for ultrasonic generation and magnetization. Further investigation into the effect of textured grain growth and etching on the magnetic and ultrasonic properties might produce very interesting results.

The samples were then returned to the vacuum chamber and annealed with a large magnetic field perpendicular to the film surface. The annealing apparatus (figure 11) consisted of an aluminum block with holes to accommodate the substrate rods, a cylindrical heater and a thermocouple. The aluminum block was then inserted between the poles of a permanent magnet having a field of approximately 2.0 kOe. A second apparatus (figure 12) was designed for annealing in an argon atmosphere with a larger field of approximately 20 kOe.
Figure 11: Original annealing apparatus consisting of an aluminum substrate holder thermally insulated by asbestos sheets between the poles of a permanent magnet. The entire apparatus is placed in an evacuated chamber.
Figure 12: High field annealing apparatus shown in position between the poles of a large electromagnet. The substrate holder is supported by two stainless steel rods in an Argon atmosphere.
Magnetization:

The films used in this investigation ranged from 1 to 3 microns in thickness or from 71 to 213 micrograms in mass. A magnetometer of the highest sensitivity was required to measure the magnetic moment of these samples. An attempt was made to renovate an existing model, but these efforts failed. Fortunately, R. Morris was willing to permit us to use the Princeton Applied Research vibrating sample magnetometer at NASA. It is based on the basic principles outlined by Foner (\textsuperscript{34}) (figure 13). A vertical oscillatory motion is imparted to the sample by a driving mechanism. The sample produces a vibrating dipole field which induces a voltage in a set of pickup coils mounted on the pole pieces of the magnet. The oscillatory voltage from the coils is filtered, amplified, and compared phasewise with the driving frequency. The oscillatory voltage is compared to a reference voltage, produced by a sample of Nickel of known moment; the unknown moment of the sample is computed and displayed in emu. Since the moment of the sample holder and quartz substrate was comparable to that of the metal film, it was necessary to determine the background moment as a function of the applied field. The background moment was measured in 500 Oe intervals starting at 12 kOe and decreasing to -12 kOe then increasing back to 12 kOe. This procedure was repeated for a substrate with a film. The background was subtracted for each
Figure 13: Illustration showing basic features of a vibrating sample Magnetometer,"VSM". Also shown are some sample holders used with the VSM.
value of the applied field from the moment of the substrate with film and the results plotted as a function of the applied field (figure 14). It was observed that the samples were not saturating in these fields, but it is believed that a comparison with the ultrasonic data would still be valid.

Ultrasonic Generation

The generation of microwave phonons by ferromagnetic resonance was investigated using an ultrasonic pulse spectrometer (figure 15). The film bonded to a nonmagnetic rod (quartz, ruby, or sapphire) was placed in a stripline cavity (figure 16) described by S. P. Maxwell\textsuperscript{32} and R. A. Waldron.\textsuperscript{33} The electromagnetic fields within the cavity are shown in figure 17. The film was then oriented inside the air gap of a large electromagnet with the plane of its surface perpendicular to the field direction. A high frequency (690 MHz) magnetic field was applied in the plane of the film perpendicular to the static field. Through the magnetoelastic interaction, transverse elastic waves of the same frequency as the magnetic field are generated in the film. These elastic waves are transmitted across the film-substrate boundary into the substrate rod. At the opposite end of the substrate rod, the elastic wave is reflected and propagated back through the substrate, across the substrate-film boundary and into the film. Through a reciprocal process the magnetoelastic interaction transforms
Figure 14: Magnetization vs. applied field perpendicular to the plane of the film, TaFe$_2$ on quartz.
Figure 15: The Ultrasonic Pulse Spectrometer (after McLane)
Figure 16: Basic design for the stripline cavity (after McLane)
Figure 17: Field disposition for a \% Wave Stripline Cavity after McLane. Note: only half of the stripline is shown and the view is from the underside.
the elastic wave back into a magnetic wave in the cavity which is detected and analyzed. The high frequency magnetic field was applied in the form of a short burst or pulse, much shorter than the transit time for the elastic wave in the substrate. The returning pulses or echoes were analyzed by feeding them into a boxcar integrator which provided a time averaged dc signal logarithmically proportional to a selected echo amplitude. The time-averaged dc signal from the Pulse-Amplitude Monitor was fed into the y-input of the x-y recorder. The dc magnetic field was measured by a Hall Probe Gaussmeter which provided a dc signal linearly proportional to the magnetic field. This signal was then fed into the x-input of the x-y recorder. Monitoring the first echo, the power output was plotted as a function of the applied field.

For temperature dependent studies, a heater composed of 25 turns of resistance wire wrapped clockwise around the brass cylinder followed by 25 turns of resistance wire wrapped counterclockwise around the brass cylinder so as to cancel their contribution to the applied field in the vicinity of the film. The temperature was monitored by a chromel-alumel thermocouple using a commercial potentiometer. For low temperatures, an Andonian dewar with a continuous flow cryostat was used.

For the investigation into the effect of an applied strain, some modifications were required. First, to obtain
an easy access to the cavity for adjustment of the strain, a support structure was designed for room temperature measurements giving free access to the stripline cavity. Second, a stripline cavity was modified (figure 18) so as to accommodate the ruby rod which was used as the substrate in this investigation. A rectangular hole was machined in the brass cylinder to accommodate the ruby rod with special attention being paid to the back surface to assure a smooth plane surface against which the crystal would be strained. The stress was applied by means of a stainless steel screw acting on a stainless steel pressure plate which pressed against the ruby rod less than one millimeter above the film. The strain produced in the rod was measured by a standard strain gauge which was glued to the surface of the ruby crystal as close as possible to the film. A Baldwin-Lima-Hamilton Strain Indicator produced a digital reading of the strain in microinches/inch. For each strain setting, the microwave output was plotted as a function of the applied field. The effect of the strain on the coercive field was determined from the variation in the applied field necessary to produce a pulse one half of its maximum value. A discussion of this and the previous investigations is presented in the following section.
Figure 18: Stripline cavity for the application of external stress. Also shown is the ruby rod substrate with the strain gauge attached; the orientation of the substrate is shown in the cutaway section of the cavity.
Discussion of Results

This section reports on the results obtained from the experiments described earlier in this text. The initial discussion concerns the magnetization data taken on films deposited on both rod and disk substrates. This is followed by a discussion of the microwave generation of phonons with respect to deposition parameters. Finally, a correlation is made between the magnetization and the ultrasonic data. Magnetization data deduced from the ultrasonic data are then presented in Table 3. All of the data were taken on thin films of TbFe$_2$.

Magnetization Measurements

Magnetization data on TbFe$_2$ were necessary to estimate its capacity as a permanent magnet material, to substantiate our crude theoretical interpretation of the microwave generation of phonons and to substantiate the deductions made from previous ultrasonic data.

The magnetization measurements presented in Table 1 illustrates the scatter that may accompany thin film investigations. Magnetic moments of the films varied from 77 emu/g to 10 emu/g. Moments measured on rod substrates were seen to be greater than 20 emu/g while the moments measured on the disk substrates were found to be less than 20 emu/g. We attribute this discrepancy to the positioning of the sample between the pick-up coils. The requirement of
Table 1. Magnetization Data Obtained with a Vibrating Sample Magnetometer on Thin Film Samples of TbFe₂

<table>
<thead>
<tr>
<th>Substrate</th>
<th>Film Thickness (microns)</th>
<th>Deposition Field (kOe)</th>
<th>Substrate Temperature (°C)</th>
<th>Remanence (emu/g)</th>
<th>Saturation Magnetization (emu/g)</th>
<th>Coercive Field (kOe)</th>
<th>BH_{max} (MG0e)</th>
</tr>
</thead>
<tbody>
<tr>
<td>rod</td>
<td>2.8</td>
<td>5.0</td>
<td>320</td>
<td>8</td>
<td>17</td>
<td>2.0</td>
<td>0.44</td>
</tr>
<tr>
<td>rod</td>
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<td>5.5</td>
<td>350</td>
<td>55</td>
<td>86</td>
<td>4.6</td>
<td>6.9</td>
</tr>
<tr>
<td>rod</td>
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<td>5.0</td>
<td>320</td>
<td>20</td>
<td>23</td>
<td>5.6</td>
<td>2.75</td>
</tr>
<tr>
<td>rod</td>
<td>2.0</td>
<td>0.0</td>
<td>350</td>
<td>9</td>
<td>40</td>
<td>2.0</td>
<td></td>
</tr>
<tr>
<td>rod</td>
<td>2.0</td>
<td>2.0</td>
<td>320</td>
<td>8</td>
<td>40</td>
<td>1.1</td>
<td>0.25</td>
</tr>
<tr>
<td>rod</td>
<td>2.0</td>
<td>0.0</td>
<td>320</td>
<td>8</td>
<td>24</td>
<td>1.0</td>
<td>0.25</td>
</tr>
<tr>
<td>rod</td>
<td>1.5</td>
<td>0.0</td>
<td>350</td>
<td>12</td>
<td>38</td>
<td>0.7</td>
<td>0.05</td>
</tr>
<tr>
<td>disk</td>
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<td>5.0</td>
<td>320</td>
<td>8</td>
<td>10</td>
<td>4.2</td>
<td>1.5</td>
</tr>
<tr>
<td>disk</td>
<td>2</td>
<td>5.0</td>
<td>320</td>
<td>8</td>
<td>17</td>
<td>4.2</td>
<td>2.7</td>
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<tr>
<td>disk</td>
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<td>5.0</td>
<td>320</td>
<td>27</td>
<td>28</td>
<td>4.2</td>
<td>2.5</td>
</tr>
<tr>
<td>disk</td>
<td>2</td>
<td>5.0</td>
<td>320</td>
<td>11</td>
<td>11</td>
<td>5.5</td>
<td></td>
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<tr>
<td>disk</td>
<td>4</td>
<td>5.0</td>
<td>320</td>
<td>8.8</td>
<td>8.8</td>
<td>10.5</td>
<td>10.5</td>
</tr>
<tr>
<td>disk</td>
<td>8</td>
<td>5.0</td>
<td>320</td>
<td>1</td>
<td>10</td>
<td>1.0</td>
<td></td>
</tr>
</tbody>
</table>
Table 2. Magnetic Properties of Permanent Magnets in Commercial Use

<table>
<thead>
<tr>
<th>Material</th>
<th>( B_r ) (G)</th>
<th>( H_c ) (Oe)</th>
<th>( (BH)_{max} ) (MG-Oe)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.5% Cr steel</td>
<td>9500</td>
<td>66</td>
<td>0.29</td>
</tr>
<tr>
<td>40% Co steel</td>
<td>10000</td>
<td>242</td>
<td>1.03</td>
</tr>
<tr>
<td>Alnico 1</td>
<td>6600</td>
<td>540</td>
<td>1.40</td>
</tr>
<tr>
<td>Alnico 2</td>
<td>7000</td>
<td>650</td>
<td>1.70</td>
</tr>
<tr>
<td>Alnico 3</td>
<td>6400</td>
<td>560</td>
<td>1.35</td>
</tr>
<tr>
<td>Alnico 4</td>
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<td>1.35</td>
</tr>
<tr>
<td>Alnico 5</td>
<td>12000</td>
<td>720</td>
<td>5.0</td>
</tr>
<tr>
<td>Alnico 5 DG</td>
<td>13300</td>
<td>685</td>
<td>6.5</td>
</tr>
<tr>
<td>Alnico 8</td>
<td>7100</td>
<td>2000</td>
<td>5.50</td>
</tr>
<tr>
<td>Alnico 9</td>
<td>10400</td>
<td>1600</td>
<td>8.50</td>
</tr>
</tbody>
</table>
Table 3. Magnetization Data Obtained from Ultrasonic Curves of Thin Films of TbFe$_2$

<table>
<thead>
<tr>
<th>Film Thickness (microns)</th>
<th>Substrate Temperature (°C)</th>
<th>Deposition Field (kOe)</th>
<th>Coercive Field (kOe)</th>
<th>Annealing Temperature (°C)</th>
<th>Annealing Field (kOe)</th>
<th>Duration of Annealing (hrs.)</th>
<th>Coercive Field (kOe)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.3</td>
<td>310</td>
<td>2.5</td>
<td>4.5</td>
<td>300</td>
<td>2</td>
<td>8</td>
<td>9</td>
</tr>
<tr>
<td>3.3</td>
<td>310</td>
<td>3.5</td>
<td>4.0</td>
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<td>8</td>
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<tr>
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<td>310</td>
<td>4.0</td>
<td>4.3</td>
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<td>8</td>
<td>5</td>
</tr>
<tr>
<td>3.4</td>
<td>310</td>
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<td>5.4</td>
<td>300</td>
<td>2</td>
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<td>7</td>
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<tr>
<td>3.4</td>
<td>310</td>
<td>5.0</td>
<td>4.3</td>
<td>300</td>
<td>2</td>
<td>8</td>
<td>10</td>
</tr>
</tbody>
</table>
accurate positioning of the sample between the pick-up coils is an important one. If the sample is not in the exact position as the reference Nickel material which was used for calibration of the apparatus, the measurements made with the apparatus will be in error. Another source of error in quantitative measurements of the magnetization was discovered in the sample holder used in earlier measurements; the present sample holder, machined from Kel-F plastic, insured repeatability of the data to within 5 emu/g. Another important factor was that the magnetometer was calibrated with a relatively large sample of Nickel which produced a moment three orders of magnitude larger than the moments of the thin films we were measuring. To prevent this source of error a Nickel film should have been made as a reference.

Although the magnitude of the moment of some of the early samples may be in doubt, the general features of the hysteresis curves are believed to be accurate. The percent of the saturated moment that remains in a zero applied field is seen to be greater than 80%. In some samples the remanence exceeds 95% of the saturation magnetization. For a polycrystalline material, remanence should be approximately 85% of the saturation magnetization. This is a good indication of the effect of the prealigning magnetic field during deposition as the samples grown without the field have a remanence-to-saturation-magnetization-ratio of less than .85 while those grown with the field have a ratio greater than .85.
The coercive force, tabulated in Table 1, has a range from zero to 10 kOe. The effect of the field during deposition can again be seen, as the coercive force for the films with a deposition field exceed 3 kOe whereas the coercive force for the films grown without the deposition field is less than 3 kOe. Films deposited with a magnetic field show a tendency toward larger coercive fields and greater remanence. Unfortunately, magnetization data were not taken on annealed films where, as will be deduced later from the ultrasonic data, the effect of the prealigning field was most pronounced in producing large coercive forces (see Table 3).

The energy product for some of the films was included for comparison of TbFe₂ as a permanent magnet material with some commercially available magnetic material²⁶ (Table 2). Energy products in the MGOe range are considered respectable for modern permanent magnetic materials. As can be seen from Table 1 several films of TbFe₂ had energy products in the MGOe range. Moreover, if it can be assumed that the moments measured on the disk substrates were less than the true moments, then letting them have an average moment of 30 emu/g would yield energy products as large as 30 MGOe for samples with rectangular hysteresis loops (figure 19).
Ultrasonic Measurements

The major investigations described within this thesis were involved with the generation of phonons by a high frequency rf magnetic field of 690 MHz. A typical curve shown in figure 20 illustrates the basic features of this phenomenon. The curve begins with the film (previously magnetized in the opposite direction to the initial sweep) at zero applied field. The applied field is increased to 18 kOe and the magnitude of the power output from the first echo (referred to as the pulse) decreases to zero near the coercive field, increases reaching a peak value before tapering off. Upon decreasing the field to zero, the pulse increases reaching a second peak at a field different from the first peak and decreasing to the remanent value of the pulse at zero applied field. A symmetric curve is produced upon sweeping the applied field to 18 kOe in the reverse direction. The magnitude of the maximum applied field is important as mentioned earlier for magnetization considerations (small fields do not completely remove the domain walls from the crystallites and thus nucleating fields are not required before magnetization reversal takes place). Moreover, the samples are not completely saturated at low fields; even fields as large as 14 kOe show a noticeable difference in the coercive force and the peak amplitude of the pulse (figure 21). The coercive force is obtained from an extrapolation of the ultrasonic curve as shown in figure 22.
Figure 20: An Ultrasonic Curve produced by a thin film deposited with a deposition field of 3 kOe. The initial path is labeled "1" and the second path is labeled "2".
Figure 21a: Two Ultrasonic Curves run on the same sample film; a) had a maximum field of 18 kOe whereas b) had a maximum field of 14 kOe. Note: the degree of remanence and the coercive force difference between the two runs.
Figure 21b: Ultrasonic curve produced with a maximum field of 14 kOe.
Figure 22: An Ultrasonic curve produced by a thin film of TbFe$_2$ after Annealing the sample. Remanent Pulse ($P_r$), Maximum Pulse ($P_{max}$) and The Coercive Field are indicated.
Judging from the theory, the magnitude of the maximum peak is strongly dependent on the magnitude of the spin-lattice relaxation time. As the temperature decreases the spin-lattice relaxation time should decrease; $H_T \approx 1/T$ should increase making the pulse decrease as observed (figure 23).

The coercive force is dependent on the anisotropy constant which increases with decreasing temperature, due to the thermal strain contribution to the anisotropy. Therefore, it is to be expected that the coercive force should increase with decreasing temperature. Figure 24 indicates that the theory is in general agreement with the data. Another method of observing the effect of the strain on the anisotropy is by a direct application of stress to the film. The stress on the film was increased by increasing the strain on the substrate to which it was attached. The coercive force was again seen to increase with an increase in the stress on the film (figure 25) supporting the contention that the stress is a major component in the perpendicular anisotropy.

**Correlation of Magnetization and Ultrasonic Data**

The theory described in this thesis was used to reproduce the Ultrasonic Curve from the M-H Hysteresis Loop obtained from measurements made with a vibrating sample magnetometer. The Ultrasonic curves so generated were then compared to the Ultrasonic Curves obtained from the pulse
Figure 23: The variation of the Maximum Peak Amplitude with Temperature
Figure 24: The variation of the coercive force with temperature.
Figure 25: The variation of the coercive force with applied stress.

The coercive force was determined from the Ultrasonic data.
spectrometer described earlier. As can be seen from a comparison of figure 26 and figure 27, the theory compares quite favorably with experiment. A curve generated by Vittoria\textsuperscript{19} using the spin wave theory development of Kobayashi\textsuperscript{18} is shown for comparison (figure 28).

There is one major flaw in the theory that is the Ultrasonic Curves produced by experiment do not have their minimum at the same field as the magnetization curves (coercive field). In the past it has always been assumed that the coercive field and the minimum in the Ultrasonic Curve would coincide. From an observation based on the few samples for which both curves exist, it would appear that the coercive force corresponds to a field for which the ultrasonic pulse has a magnitude of half of its maximum amplitude. One explanation for the disparity between the two curves could come from the size of the maximum fields used for the two measurements: the magnetization measurements were performed with a maximum field of 12 kOe whereas the ultrasonic measurements were performed using a field of 18 kOe. The theory does produce peaks for the Ultrasonic Curve at the proper fields and does show the correct temperature dependence and does show the minimum to be at fields less than the coercive field but larger than the minimum of the ultrasonic curve produced by experiment.

Magnetic data can now be deduced from the ultrasonic data. Magnetization data so obtained are shown in Table 3.
Figure 26: An Ultrasonic curve generated from theory with $K = 4$ and $H_r^2 = 10^9$ Oe$^2$. 
Figure 27: An ultrasonic curve generated from theory with $K = 1$ and $H_r^2 = 10^8$ Oe$^2$. 
Figure 28: Signal vs. Field, TbFe₂ on Quartz; this is the experimental curve for the film used in generating figures 26 and 27.
Figure 28: An Ultrasonic curve generated from the spin wave Theory of Kobayashi by Vittoria.
Annealing

After the samples were tested for ultrasonic generation and magnetic properties, the samples were placed in the annealing apparatus. Heating a sample within a vacuum removes some of the inhomogeneities present within the film. Applying a magnetic field to the sample while it is heating removes some of the inhomogeneities in the alignment of the crystallites.

Magnetically annealing the crystallites increases the coercive force as seen from Table 3. Ultrasonic curves showing the before and after effect can be seen in figures 29 and 30. There is a marked increase in the remanence, an increase in the coercive force and an increase in the maximum pulse amplitude. This is independent of the deposition field although the deposition field seems to enhance this effect.

Magnetic hysteresis loops were taken on only one set of samples before and after annealing and it appears that fields much greater than 12 k0e would be necessary to saturate them after annealing. Hysteresis curves for a representative sample are shown in figure 31.
Figure 30: Ultrasonic Curve for a thin film of TbFe$_2$ before annealing.
Figure 31: Ultrasonic curve for a thin film of TbFe$_2$ after annealing.
Figure 32 (a) Hysteresis curve for a thin film of TbFe$_2$ on a quartz disk substrate before annealing.
(b) Hysteresis curve for the same film after annealing; nucleation sites appear to have been removed but the sample was not saturated.
Relaxation Constant

A value for the relaxation constant for polycrystalline TbFe$_2$ has been determined from the best fit of the theoretical curve to the experimental Ultrasonic Generation curve. The strong dependence of the theoretical curve on the value of the relaxation field, $H_R$, provides an excellent tool for obtaining the relaxation constant for thin films of polycrystalline TbFe$_2$. The relaxation constant is obtained through the relation

$$\gamma H_R = \frac{1}{T}$$

where $\gamma$ is the gyromagnetic ratio. Figures 26 and 27 demonstrate the effect of increasing the relaxation field on the shape of the curve. The maximum peak amplitude decreases, the minimum occurs at higher field and is broader, and the absolute value of the slope of the decreasing applied field portion of the curve is decreased. At room temperature, $H_R \approx 10^4$ Oe, $\gamma \approx 1.73 \times 10^8$, and near liquid Nitrogen temperature, $H_R \approx 10^5$ Oe. These values are larger than those reported for amorphous TbFe$_2$ films by a factor of six. It should not be expected that the amorphous value would agree with the polycrystalline value of the relaxation constant. We believe that the increase in the linewidths are a result of inhomogeneous broadening by the large magnetostrictive strains present within the film. Further research is necessary before this method can be refined for measuring the relaxation constant.
Unexplained Phenomena

During the course of this investigation, several interesting phenomena were observed. The textured axis present in the thin films of TbFe$_2$, the differences between the major and minor loops of the ultrasonic curves, the effect of the different substrates on the Ultrasonic Generating properties of the thin films, the shape of the ultrasonic curve generated by the thin film when the applied magnetic field was in the same direction as the oscillating magnetic field, i.e., parallel to the surface, were among the most notable.

More investigation is needed before more can be said about the grain structure of the film and its influence on the magnetic properties of the film. Etching the film will relieve some of the strain present; hence etching the film will affect the anisotropy constant. Observing the film free of the substrate might prove very informative as the film will be free from most of the strain. Magnetization data taken on a strain free film could also prove to be enlightening.

During the investigation into the effect of externally applied strain on the film, it was noted that the minor loops of the Ultrasonic curve produced by the film on the ruby substrate differed quite drastically from those of a film on a quartz substrate. (See the data in Appendix I.) Upon returning to zero applied field the amplitude of the
Ultrasonic signal is larger than its previous value. This appears to be dependent on the magnitude of the applied field before returning to zero applied field. This continues until at some value of the maximum applied field the curve for the film on a ruby substrate appears the same as the curve for a film on a quartz substrate. More information is needed on the effect of the substrate on the ultrasonic generating properties of these films before a model can be formulated.

Both McLane and I have observed that some films are capable of generating ultrasonic phonons with the applied field in either a perpendicular or parallel orientation to the surface. Moreover, I observed that the shape of the curves are quite different from those capable of generating phonons from one orientation of the applied field. Although the magnitude of the signal would indicate that this is of little practical use still it remains a puzzling and interesting phenomenon.

Summing up, thin films of TbFe₂ can be of commercial value both as permanent magnetics and as ultrasonic transducers in the high frequency range. Further investigation may uncover future applications as bubble memory and microwave devices.
Conclusion

This investigation has probed into some of the magnetic properties of the Rare Earth-Iron Compound, TbFe$_2$. Its capacity as an ultrasonic transducer in the high frequency range is by now well established. Although Lithium Niobate, a piezoelectric transducer material, is operable at comparable frequencies, the large remanent pulse produced without the need of a biasing magnet has made these magnetostrictive transducers competitive with the Lithium Niobate transducers.

Moreover the large coercive forces observed on the film after they were annealed compare favorably with modern permanent magnetic materials. These films offer high energy products along with high Curie temperatures. The feasibility of these films as thin-film permanent magnets is becoming a reality.

A theory capable of reproducing the gross features of the microwave generation of phonons has been developed. The perpendicular anisotropy playing a major role in the determination of the ultrasonic curve is seen to be related to the thermal stress within the film.

Several factors are seen to influence the magnetic properties possessed by these films. Besides the deposition parameters investigated by McLane, the strength of the deposition is seen to increase both the remanence and the coercive field.
With lower pressures, investigations into the thin film magnetic properties of other Rare Earth-Iron Group compounds might prove to be very profitable especially as thin film permanent magnets.
References

Appendix A

Derivation of the Green's Function for multiple reflections within the source material:

The illustration above shows the propagation paths for the Green's function $G(z,z')$; the direct and reflected parts are combined to give the total contribution of the source at $z'$ to the element at $z$. The solution to the one-dimensional inhomogeneous wave equation having a point source at $z'$ oscillating sinusoidally in time has been described by Morse and Feshback. The wave equation is:

$$\nabla^2_x G(z-z'|w) - \frac{1}{\nu^2} \left( \frac{\partial^2}{\partial t^2} \right) G(z-z'|w) = -\delta(z-z')e^{-i\omega t}$$

The solution for this equation is of the form:

$$G(z-z'|w) = \frac{i}{2k} e^{-i\omega t + k(z-z')}$$

(-) for $z > z'$

(+) for $z < z'$

The displacement is given by a traveling wave with the origin at $z'$. This solution is valid for a medium which is unbounded. For the case of multiple reflections within the film, consideration must be given to the reflection-
transmission at the interface of the rod and film. If
\( G(z, z') \) is an incident wave on the interface, \( rG(z, z') \) will
be reflected while \( tG(z, z') \) will be transmitted. \( r = \)
reflection coefficient and \( t = \) transmission coefficient.
After reflection the wave travels a distance \( 2L \) inside the
film being reflected once from the free end without change
of phase before impinging on the boundary a second time.
The reflected Green's function would be \( r^2G(z-z'+2L) \) and
the transmitted Green's function would be \( rtG(z-z'+2L) \).
The transmitted Green's functions are then summed to give
the total Green's function inside the rod. The origin
will be shifted to the film-substrate boundary at \( L \). The
Green's function for the direct paths (due to a propagation
initially to the right) is
\[
G_D(z-z') = \frac{i}{2k_1} e^{\left[-i\left(wt-k_2(z-L)-k_1(z'-L)\right)\right]}
\]
The Green's function coming from the reflected paths (due
to a propagation moving initially to the left) is
\[
G_R(z+z') = \frac{i}{2k_1} e^{\left[-i\left(wt-k_2(z-L)+k_1(z'+L)\right)\right]}
\]
The multiple transmissions for the direct paths of the
Green's functions are summed as
\[
tG(z-z')+rtG(z-z'+2L)+rrtG(z-z'+4L)+\ldots+rt^n(e^{-ik_12L})^n \]
The total Green's function is obtained by adding the two
summation series for the direct and reflected paths:
\[ G_{total}(z,z') = \frac{it}{k_1} e^{-ik_2(Z-L)} \left( e^{\frac{ik_1z' + -ik_1z'}{2}} \right) e^{-ik_1L \sum (x^n)} e^{-ik_1 2L} \]

Since \[ \sum x^n = \frac{1-x^n}{1-x} \], \( x = re^{-ik_1 2L} < 1 \)

\[ = \frac{1}{1-re^{-ik_1 2L}} \]

\[ G_{total}(z,z') = \frac{i}{k_1} \left( \frac{te^{-ik_1 L \cos k_1 z'}}{1-re^{-i2k_1 L}} \right) e^{-ik_2(Z-L)} \]
Parameterization of the Equation

Determination of the proper choice of the value of \( K \) in the expression for the Ultrasonic Pulse Amplitude can be made from a consideration of the anisotropy constant \( K_\perp \). The energy associated with a rotation of the magnetization from the film normal by an angle \( \theta \) is given by

\[
E_\perp = -K_\perp \sin^2 \theta
\]

where \( K_\perp \) is the perpendicular anisotropy constant. If \( K_\perp \) is negative, the normal to the film surface is the easy axis. If the only contribution to the anisotropy constant was the shape demagnetization factor, the perpendicular anisotropy constant would be \( 2\pi M^2 \); \( K_\perp \) would be positive and the magnetization would prefer to remain in the plane of the film. However, these films have been observed to be extremely stressed. This stress believed to be of thermal origin is a result of the difference in thermal-expansion coefficients of the substrate and the film. The contribution of the stress to the energy is expressed as

\[
E^S_\perp = -\frac{3}{2} \lambda s \sin^2 \theta
\]

Hence, \( K_\perp \), the perpendicular anisotropy constant has two components: a shape demagnetization factor given by \( 2\pi M^2 \) and a contribution due to the thermal strain given by

\[
K^S_\perp = \frac{3}{2} \lambda [\alpha_F - \alpha_s] \Delta T E_F .
\]
λ is the magnetostriction coefficient for the film, \( \alpha_f, \alpha_s \) are the thermal expansion coefficients for the film and substrate respectively, and \( E_f \) is Young's modulus for the film.

Using values for the coefficients obtained from experiment and literature, the perpendicular anisotropy can be determined. This can be used to determine the coercive force of the film which will in turn determine the value of \( K \) to be used in our expression for the Ultrasonic Pulse.

Sample #3 used in the discussion of results will be used as an example for the calculation.

\[
M_s = 756 \ \text{emu/cc}
\]

\[
2\pi M_s^2 = 3.59 \times 10^6 \ \text{ergs}
\]

\[
\lambda = -1000 \times 10^{-6}
\]

\[
\alpha_s = 12 \times 10^{-6}
\]

\[
\alpha_f = 0.55 \times 10^{-6}
\]

\[
E_f = 9.4 \times 10^{11}
\]

\[
\Delta T = -350 \ ^\circ\text{C}
\]

\[
K_s^s = -5.65 \times 10^6 \ \text{ergs}
\]

\[
K_s^s = 3.59 \times 10^6 - 5.65 \times 10^6
\]

\[
K_s^s = 2.06 \times 10^6 \ \text{ergs}
\]

The coercive field is given by \( H_C = 2K_s^s/M_s \)

\[
H_C = 5.4 \ \text{kOe}
\]
The K used in our expression from the Ultrasonic pulse is given as \( K = H_0 / 4\pi M_s \) which for a coercive field of \( 5.4 \times 10^3 \) Oe would be 0.6.

\[
H_R = \gamma \frac{1}{T}
\]

\[
H_R \approx 10^4
\]
Appendix C

Parameterization of the Equation for Ultrasonic Generation Relaxation Constant

A value for the relaxation constant was determined by the best fit to the data. Critical features of the data curve to which careful attention was paid are the minimum points, maximum points, and the slope of the curve for decreasing field with saturation magnetization.

Relaxation was introduced into the equation as a relaxation field: \( \gamma H_R = \frac{1}{T} \) where \( \gamma \) is the gyromagnetic ratio. Although there are no published results for relaxation constants in crystal \( \text{TbFe}_2 \), P. Lubitz, J. Schelleng, and C. Vittoria\(^{36} \) have published the following data for amorphous \( \text{TbFe}_2 \):

\[
\gamma \geq 1.73 \times 10^7 \text{ (Oe sec)}^{-1}
\]

\[
H_R > 1500 \text{ Oe.}
\]

However, the data reported indicate a relaxation field on the order of 10,000 Oe which is approximately six times the amorphous value. The most probable explanation of the large linewidth is that it is a result of inhomogeneous broadening due to the high magnetostrictive strain present in the film.
Appendix D

Crystal Field Terms

B. Bleaney, F.R.S.\textsuperscript{37} has shown that the crystal field interaction energy with the 4f electrons of the Lanthanide ion may be written as

\[ B_4 (0^c_4 + 50^4_4) + B_6 (0^o_6 - 210^4_6) \]

where

\[ B_4 = \frac{7e^2}{36R_2^5} \left[ Z_2 - 1.20Z_1 \right] \langle r^4 \rangle \langle J|J \rangle \]

\[ B_6 = \frac{-e^2}{18R_2^7} \left[ Z_2 + 0.303Z_1 \right] \langle r^6 \rangle \langle J|J \rangle \]

Here, \( R_2 \) is the distance between the Lanthanide ions, \( Z_2 e \) is the charge on the Terbium ion and \( Z_1 e \) is the charge on the iron ion. The value of \( \langle r^4 \rangle \), and \( \beta_j \) can be found in the appendix of Taylor and Darby.\textsuperscript{4}

B. Bleaney argued that the \( B_6 \) term is negligible when compared with the \( B_4 \) term; thus, only the \( B_4 \) term need be used in calculations of the crystal field.
Film Thickness.: 2 μm
Substrate Temperature: 550°C
Run Temperature: 20°C
Deposition Field: 0 kOe
Annealing Field: 0 kOe
Annealing Temperature: 0°C

Figure E-1 Signal vs. Field, TbFe₂ on Quartz
Figure E-2  Signal vs. Field, TbFe₂ on Quartz
Figure E-3 Signal vs. Field, TbFe$_2$ on Quartz
Figure E-5 Signal vs. Field, TbFe$_2$ on Quartz
Figure E-6 Signal vs. Field, TbFe$_2$ on Quartz
Figure E-7 Signal vs. Field, TbFe$_2$ on Quartz
Film Thickness 2 \mu m
Substrate Temperature 350°C
Run Temperature -120°C

Deposition Field 0 kOe
Annealing Field - kOe
Annealing Temperature -- °C

Figure E-8 Signal vs. Field, TbFe$_2$ on Quartz
Figure E-9  Signal vs. Field, TbFe$_2$ on Quartz
Figure E-10 Signal vs. Field, TbFe$_2$ on Quartz
Figure E-11 Signal vs. Field, ThFe$_2$ on Quartz

Film Thickness 2 $\mu$m
Substrate Temperature 350°C
Run Temperature -180°C

Deposition Field $0$ kOe
Annealing Field $-$ kOe
Annealing Temperature $-$ °C
Film Thickness $2 \mu m$
Substrate Temperature $350^\circ C$
Run Temperature $-190^\circ C$

Deposition Field $0$ kOe
Annealing Field $-$ kOe
Annealing Temperature $-$ $^\circ C$

Figure E-12 Signal vs. Field, TbFe$_2$ on Quartz
Film Thickness $2 \mu m$
Substrate Temperature $275^\circ C$
Run Temperature $-11^\circ C$

Deposition Field $5$ kOe
Annealing Field $0$ kOe
Annealing Temperature $0^\circ C$

Figure E-13 Signal vs. Field, ThFe$_2$ on Quartz
Figure E15 Signal vs. Field, ThFe₂ on Quartz

Film Thickness 2 µm
Substrate Temperature 275 °C
Run Temperature -80 °C
Deposition Field 5 kOe
Annealing Field - kOe
Annealing Temperature - °C
Film Thickness: $2 \mu m$
Substrate Temperature: $275 \, ^\circ C$
Run Temperature: $-100 \, ^\circ C$

Deposition Field: $5 \, kOe$
Annealing Field: $- \, kOe$
Annealing Temperature: $- \, ^\circ C$

Figure E.16 Signal vs. Field, ThFe$_2$ on Quartz
Figure E.17 Signal vs. Field, TbFe$_2$ on Quartz
Figure E-18 Signal vs. Field, ThFe$_2$ on Quartz
Film Thickness $\frac{2}{\mu m}$
Substrate Temperature $275^\circ C$
Run Temperature $-160^\circ C$

Deposition Field $5$ kOe
Annealing Field $-$ kOe
Annealing Temperature $-$ $^\circ C$

Figure E-19 Signal vs. Field, ThFe$_2$ on Quartz
Film Thickness $2\,\mu$m
Substrate Temperature $275^\circ C$
Run Temperature $180^\circ C$

Deposition Field $5\,\text{kOe}$
Annealing Field $-\,\text{kOe}$
Annealing Temperature $-\,\text{C}$

Figure E-20 Signal vs. Field, TbFe$_2$ on Quartz
Figure E.21 Signal vs. Field, TbFe$_2$ on Quartz
Figure E-22: Signal vs. Field, TbFe₂ on Quartz; Temperature during run was 25°C.
Film Thickness 2 \( \mu \text{m} \)
Substrate Temperature 275\(^\circ\text{C}\)

Deposition Field 5 kOe
Annealed in Argon Atmosphere
Annealing Temperature 350\(^\circ\text{C}\)
Annealing Field 19 kOe

Ultrasonic Signal Amplitude (dB)

APPLIED FIELD (kOe)

Figure E.23 SIGNAL VS. FIELD, TbFe\(_2\) ON QUARTZ: Temperature during run was -20\(^\circ\text{C}\)
Figure 5.4 SIGNAL VS. FIELD, TbFe$_2$ ON QUARTZ: TEMPERATURE DURING RUN WAS -40°C
Film Thickness 2 μm
Substrate Temperature 275°C
Deposition Field 5 kOe
Annealed in Argon Atmosphere
Annealing Temperature 350°C
Annealing Field 19 kOe

FIGURE 7.5 SIGNAL VS. FIELD, TbFe₂ ON QUARTZ: TEMPERATURE DURING RUN WAS -60°C.
Figure 2: Signal vs. Field, TbFe$_2$ on Quartz: Temperature during run was -30°C.
Film Thickness 2 µm
Substrate Temperature 275°C
Deposition Field 5 kOe
Annealed in Ar/02 Atmosphere
Annealing Temperature 350°C
Annealing Field 19 kOe

Ultrasonic Signal Amplitude (db)

APPLIED FIELD (kOe)

FIGURE 7: SIGNAL VS. FIELD, TbFe2 ON QUARTZ: TEMPERATURE DURING RUN WAS -100°C.
FIGURE E-28 SIGNAL VS. FIELD, ThFe₂ ON QUARTZ: TEMPERATURE DURING RUN WAS -120°C.
Film Thickness 2 μm
Substrate Temperature 275°C
Deposition Field 5 kOe
Annealed in Argon Atmosphere
Annealing Temperature 350°C
Annealing Field 19 kOe

Ultrasonic Signal Amplitude (db)

APPLIED FIELD (kOe)

FIGURE 1: SIGNAL VS. FIELD, TbFe₂ ON QUARTZ: TEMPERATURE DURING RUN WAS -140°C.
Film Thickness 2 μm
Substrate Temperature 275°C

Deposition Field 5 kOe
Annealed in Argon Atmosphere
Annealing Temperature 350°C
Annealing Field 19 kOe

**Figure E-30** Signal vs. Field, ThFe₂ on Quartz: Temperature during run was -180°C.
Figure F-2: Signal vs. Field, TbFe₂ on Quartz
Figure F-3: Signal vs. Field, ThFe₂ on Quartz
Figure F-4: Signal vs. Field, TbFe₂ on Quartz
Figure F-5: Signal vs. Field, FeCo on Quartz
Figure F-6: Signal vs. Field, TbFe$_2$ on Quartz after annealing. This is the same film used in Figure F-1.
Figure F-7: Signal vs. Field, TbFe$_2$ on Quartz after annealing. This is the same film as used in Figure F-2.
Figure F-8: Signal vs. Field, TbFe2 on Quartz after annealing. This is the same film as used in Figure F-3.
Figure F-9: Signal vs. Field TbFe$_2$ on Quartz after annealing. This is the same film as used in figure F-4.
Figure F-10: Signal vs. Field, TbFe$_2$ on Quartz after annealing. This is the same film used in Figure F-5.
FIGURE G-2: MAGNETIZATION VS. APPLIED FIELD PERPENDICULAR TO THE PLANE OF THE FILM, TaFe₂ ON Quartz.
Figure G-4: Magnetization vs. Applied Field perpendicular to the plane of the film, TaFe₂ on quartz.
FIGURE G - 5: MAGNETIZATION VS. APPLIED FIELD PERPENDICULAR TO THE PLANE OF THE FILM, TbFe$_2$ ON QUARTZ.
**Figure G-6:** Magnetization vs. Applied Field perpendicular to the plane of the film, TbFe$_2$ on quartz.
FIGURE G-7: MAGNETIZATION VS. APPLIED FIELD PERPENDICULAR TO THE PLANE OF THE FILM, TaFe$_2$ ON QUARTZ.
Figure G-8: Magnetization vs. Applied Field perpendicular to the plane of the film, TaFe₂ on quartz.
Filmen Thickness 2 μm  
Substrate Temperature 350 °C  
Deposition Field 0 kOe  

FIGURE G-9: MAGNETIZATION VS. APPLIED FIELD PERPENDICULAR TO THE PLANE OF THE FILM, TbFe₂ ON QUARTZ.
Figure G-10: Magnetization vs. Applied Field perpendicular to the plane of the film, TbFe₂ on quartz.
Figure G-11: Magnetization vs. applied field perpendicular to the plane of the film, TbFe$_2$ on quartz.
FIGURE G-12: MAGNETIZATION VS. APPLIED FIELD PERPENDICULAR TO THE PLANE OF THE FILM, TbFe₂ ON QUARTZ.
Film Thickness 2 μm
Substrate Temperature 350°C
Run Temperature 0°C

Deposition Field 4 kOe
Annealing Field 0 kOe
Annealing Temperature 0°C

Figure H-1 Signal vs. Field, TbFe₂ on Quartz
Figure H-2: Signal vs. Field, ThFe$_2$ on Quartz
Figure H-3: Signal vs. Field, TbFe$_2$ on Quartz

- Film Thickness: 2 $\mu$m
- Substrate Temperature: 350°C
- Run Temperature: -60°C
- Deposition Field: 4 kOe
- Annealing Field: 0 kOe
- Annealing Temperature: 0°C

APPLIED FIELD (kOe)
Figure H-4: Signal vs. Field, ThFe$_2$ on Quartz
Film Thickness: 2 μm
Substrate Temperature: 250 °C
Run Temperature: 190 °C

Deposition Field: 4 kOe
Annealing Field: 0 kOe
Annealing Temperature: 0 °C

Figure H-6: Signal vs. Field, ThFe₂ on Quartz
Figure H-7: An ultrasonic curve produced by a thin film of TbFe₂ on a Ruby substrate. The film was previously magnetized to the left prior to this run.
Figure H-8: A minor Ultrasonic loop for the thin film of TbFe$_2$ on a Ruby substrate shown in figure I-7. Note the remanent ultrasonic pulse present at zero field.
Figure H-9: A minor ultrasonic loop for the film shown in I-8 with a greater maximum field of 3.5 kOe. Note the change in the horizontal scale.