RICE UNIVERSITY

MAGNETOSTRICTION IN TbFe$_2$
POLYCRYSTALLINE THIN FILMS

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A THESIS SUBMITTED
IN PARTIAL FULFILLMENT OF THE
REQUIREMENTS FOR THE DEGREE OF

MASTER OF ARTS

Thesis Director's signature: [Signature]

Houston, Texas

(May, 1973)
690 MHz elastic waves have been magnetostrictively generated in polycrystalline thin films of TbFe$_2$ at room temperature. The films are on the order of 100 times more efficient in this respect than is the surface piezoelectric effect. TbFe, TbFe$_3$, and Tb$_{3}$Dy$_{7}$Fe$_2$ films have demonstrated comparable effectiveness. A large magnetic anisotropy whose easy axis is perpendicular to the plane of the film has been observed in the TbFe$_2$ films. A simplistic theory, based on a single-ion molecular-field interaction with the terbium sublattice considered independently, is developed.
To Carol
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I. INTRODUCTION

Scope

This thesis reports on a portion of the investigation into the magnetic properties of the rare earths being conducted by our group under the support of the Advanced Research Projects Agency. More specifically, the magnetostrictive generation of elastic waves in polycrystalline thin films of TbFe, TbFe₂, TbFe₃, and the ternary compound Tb₃Dy₇Fe₂ is studied along with the magnetization of TbFe₂ films in an effort to provide insight into the fundamental magnetoelastic properties of rare earth materials. Major emphasis is placed on the room temperature characteristics of the various materials, with TbFe₂ being the composition of principal interest, although a temperature study of TbFe₂ is also included. The films (with one exception) are two microns thick, and the ultrasonic-generation work was conducted primarily at 670 MHz (although some cursory examinations were made at lower frequencies, they were not included in this report as they were inadequately documented and contained no anomalous results to indicate the immediate need for further detailed frequency-dependence studies). Ultrasonic generation properties were studied as a function of applied dc magnetic field between zero and 18 kOe directed normally to the plane.
of the film (fields lying in the plane of the film having been determined to be ineffectual).

The principal results of these investigations are presented and include the verification of the anticipated high efficiencies in the generation of elastic waves by magnetostriction and the discovery of a very large coercive force in TbFe$_2$ thin films with the uniaxial anisotropy easy axis normal to the plane of the film.

Finally, the beginning of a theory, incomplete at present but under development, is presented in the form of a simplistic, theoretical model accounting for the nonresonant generation of elastic waves in a manner similar to that observed in the case of TbFe$_2$ and some other pure rare earth thin films (Tb and Dy at low temperatures).  

Motivation

It is hoped that this work, along with projected investigative efforts, will provide sufficient information on the fundamental, magnetoelastic properties of rare earth materials to permit the development of a more complete theoretical description of these materials. A complete understanding of any phenomenon, too, is highly desirable in practical applications of that effect, and there are immense technological implications present here.

The practical potential of an highly efficient, room temperature, ultrasonic transducer in the higher frequency regions (UHF and higher) is obvious. It was to this end that, after marked success in elastic wave generation at
cryogenic temperatures with pure rare earth films, $^3$TbFe$_2$ was selected for evaluation of its potential since Clark and Belson$^4$ had recently reported a room temperature, static magnetostriction in bulk polycrystalline TbFe larger than that known for any other material. Not only were our hopes for efficient ultrasonic generation rewarded but the existence of the uniaxial nonplanar easy axis was revealed. Inasmuch as a large anisotropy had also been reported$^5$ for bulk TbFe$_2$, other compositions, for which more modest coercive forces had been reported, were tried. The anisotropic aspect of these materials extends their technological potential to the area of bubble domain memory devices.

**Magnetism Background**

Magnetostriiction, in the case of the rare-earth metals, derives primarily from the large strain dependence of the single-ion magnetocrystalline anisotropy energy. This strain dependence constitutes a coupling mechanism between the elastic strain and the magnetization whereby an oscillatory rf magnetization excited by an applied rf magnetic field is accompanied by an oscillatory strain at the same frequency. As single crystals of the Tb-Fe materials have not yet been grown, there is not sufficient information at present to permit the complete description of magnetostrictive generation. It is assumed, however, that the situation is similar to that of the rare earth garnets wherein the Fe-Fe exchange is strong, leading to high Curie temperatures, the
RE-RE exchange is weak, and the Fe-RE exchange is strong enough to force the ferromagnetic alignment of most of the RE ion moments at temperatures much higher than normal for the pure rare earths. The magnetostriction is almost entirely due to the RE ions, which behave essentially like free ions subject to crystalline fields and the effective molecular field of the neighboring iron atoms. It is on this basis that our simplistic model is constructed.

**Thesis Organization**

Following the introduction, Chapter II presents a description of the experimental apparatus and procedures employed in this investigation. Chapter III contains a description of the experimental results, while a simple theoretical model is developed in Chapter IV, and an effort is made to explain the experimental results insofar as the theory is applicable. Conclusions based on the experimental results and the limited analysis are made in Chapter V, and further avenues of experimental and theoretical exploration are speculated upon.
The experiments reported here were concerned with the magnetic properties of polycrystalline thin films of various rare-earth-iron compounds. The major portion of effort in this study has been directed toward the investigation of TbFe$_2$ films at room temperature although other rare earth-iron compounds were explored in less detail, and some temperature dependence observations on TbFe$_2$ have been conducted. This chapter deals with the experimental techniques and equipment employed. In the first section the vacuum deposition apparatus and the substrate preparation and film deposition processes are discussed, while the second section contains a description of the ultrasonic pulse spectrometer and its operation, and the third section describes the measurement of magnetization.

Thin Film Production

All the thin films included in this study were produced by vacuum deposition. The vacuum chamber in our deposition system (Fig. 11-1) consists of an 18 inch diameter bell-jar (approximate volume 100 liters) on a 20 inch diameter stainless steel baseplate. A 15 cubic feet per minute Duo Seal model 1397 mechanical pump and a CVC (Consolidated Vacuum Corp.) model PCMS-6B diffusion pump comprise the pumping system. A CVC model BCN-61B liquid-nitrogen baffle
DEPOSITION SYSTEM

FIGURE II-1
is incorporated to minimize backstreaming of the diffusion-pump vapor. Vacuum monitoring is accomplished with a CVC model GC-110B Ionization Vacuum Gauge modified to accommodate two GTC-004 thermocouple vacuum-gauge tubes. The GIC-017 ion tube is mounted in the 6-Inch main vacuum passage just below the baseplate, while one thermocouple gauge tube is in the foreline of the diffusion pump and the other is in the system roughing line. The various feedthroughs in the baseplate and main vacuum line include high-current water-cooled, medium- and low-current ambient cooled, mechanical, miniature coax and water (for the thickness monitor sensor), and an octal connector for instrumentation. Substrate temperatures are measured with a Leeds and Northrup model 8686 Millivolt Potentiometer and a chromel/alumel thermocouple. Substrate heater current and the currents for the evaporant heaters are supplied by variable autotransformers and high-current stepdown transformers, although a recent modification of the external control system employs solid-state thyristor (Triac) current control in place of the bulkier, more expensive, autotransformers. The external control system also contains sequence and safety interlocks to ensure proper cycling of the pumps and heater controls. Real-time thickness monitoring is provided by an Inficon model 321 Film Thickness Monitor.

There have been two configurations of substrate holder employed in this study. The body of the holder is made of stainless steel and has six substrate positions. In the
first model the heater was separate and consisted of six to eight turns of #22 tungsten wire placed in the heater cavity of the holder but not in contact with it (to avoid shorting out the heater) thereby radiantly heating the substrate holder (Fig. 11-2a). The heater had to be remounted and adjusted prior to each run. The fact that the filament was brittle and somewhat unmanageable made this adjustment procedure unnecessarily tedious. Consequently a heater of about one foot of #22 nichrome wire was wound on a ceramic form and embedded in alundum cement in the cavity (Fig. 11-2b). A current of approximately 5 amps at 5 volts will heat the substrate holder to about 300°C in one hour. The temperature will stabilize at 320-330°C in two to three hours.

The thickness monitor consists of a control unit, an oscillator (remotely located from the control unit to be as near the sensor head as possible), and a sensor head located within the vacuum chamber. The sensor head consists of an AT-cut quartz crystal mounted in a water-cooled holder positioned such that one surface of the sensor crystal is normal to and in the direct path of the evaporant. The simplest situation, of course, would be for the sensor to be mounted next to the film substrates and at the same distance from the sample source. We experienced repeated failures of the thickness monitor in this configuration on deposition runs of any significant duration, but these failures were attributed to heating of the sensor due to its proximity to
TUNGSTEN WIRE HEATER

SUBSTRATE HEATER ASSEMBLY

FIGURE II-2

INTEGRAL HEATER

SEPARATE HEATER

NICHROME HEATER LEADS

ALUMINUM CEMENT

DETAIL "A"

MOUNTING HOLE

DETAI A (TYP)

(6) SUBSTRATE HOLES

THERMOCOUPLE MOUNT

TUNGSTEN WIRE HEATER
the boat (about 6 inches) and the abnormally high temperatures of the boat required to achieve the desired deposition rate (an explanation for which will follow). This problem was alleviated by positioning the sensor head twice as far from the source as the substrates and, assuming $1/r^2$ dependence in the evaporant flux and correcting the substrate film thickness by a factor of four over that on the sensor crystal.

The resonant frequency of the sensor crystal is a function of its mass; thus the sensor oscillator changes frequency relative to the mass of the material evaporated onto it. The control unit, which is programmed with the evaporant density, measures the change in frequency of the sensor oscillator and computes from that the film thickness and displays it in digital form. Inasmuch as the thickness (or frequency shift) varies linearly with the mass (or density) of the film, one may program the control unit with an appropriately adjusted density (say one fourth the true density) and it will indicate a correspondingly adjusted (by a factor of four) thickness. This permits the automatic compensation of the error acquired in relocating the sensor.

All the substrates used in this investigation were three millimeter diameter single-crystal X-cut quartz rods varying in length from one centimeter to 1.8 centimeters. Single-crystal substrates are used primarily because of the large losses encountered in fused materials at the frequencies of interest here (e.g. 690 MHz). The use of
X-cut quartz also permits the ready identification of the polarization and mode of the generated acoustic waves by transit-time measurement and, further, the coarse qualitative estimation of the relative efficiencies of the ultrasonic generation properties of the various films by comparison of the signals generated by the films with those obtained by surface piezoelectric generation.

The substrates were cleaned by a multiple ultrasonic bath process and introduced into the substrate holder immediately prior to pumpdown to minimize contamination. The standard process used on all the rods is a degreasing process beginning with a 5-minute bath in 95% pure hexane followed with two successive 5-minute baths in 99% pure hexane. The substrates are then kept in the last hexane bath until they are ready for insertion in the holder. Substrate rods which already have films on them or have an uncertain history regarding their surface condition are first put through a stripping process comprised of a 15-minute 5% nitric-acid bath (longer if visual inspection so indicates) followed by a series of three 5-minute rinses in distilled water and a 5-minute dehydrating bath in ethanol. They then go through the standard degreasing process just delineated. During the early part of the experiment the substrate holder was also run through the stripping and degreasing procedures. This was discontinued, however, when the holder was modified to include the substrate heater, as the alundum cement in which the heating element is embedded is quite
porous and would absorb substantial quantities of the cleaning agents, necessitating lengthy pumping times and outgassing. Consequently the holder was cleaned thoroughly once, and subsequent cleaning consisted of wiping exposed surfaces with acetone impregnated tissues. No apparent adverse effects have been observed since relaxing the cleaning standards to that extent.

Similarly, the entire system is cleaned prior to each deposition run with acetone impregnated tissues. The dampened tissues loosen and pick up the bulk of the material deposited previously as well as removing most of the oil and gas film. The bell-jar surfaces usually require steel-wool rubbing prior to the acetone wipe as it is desirable that there be no residual films for visibility purposes and the evaporants adhere to the glass surfaces better than the other surfaces involved.

The procedure used for the vacuum deposition of the films used in this study is the standard one consisting of the evacuation of the deposition chamber followed by the heating of the evaporant until it vaporizes and subsequently condenses in a thin film on the exposed surfaces. After ensuring that the deposition chamber is clean, all mechanical adjustments and electrical connections in order, a good sensor crystal in the sensor head, the substrates in place, the silicon monoxide boat charged, about three to five grams of the sample material in the source boat, and the bell-jar seated, the chamber is ready to be evacuated.
At this time the vacuum system should have the mechanical pump pumping on the diffusion foreline, all the other vacuum line valves closed, liquid nitrogen in the cold trap, and the diffusion pump hot (implying that the coolant pump and heat exchanger water are on). The foreline vacuum gauge should be indicating less than $25 \times 10^{-3}$ torr. The pumpdown cycle is begun by closing the foreline valve and opening the roughing-line valve. If all is in order the roughing-line thermocouple vacuum gauge should indicate less than 0.1 torr in a few minutes at which point the roughing-line valve should be closed, the foreline valve opened again, and the high-vacuum valve opened. The roughing-line gauge should indicate a rapid drop in pressure below the sensitivity of the thermocouple gauge (about 1 millitorr). The ionization gauge may now be turned on and should soon be indicating a pressure on the order of $10^{-5}$ torr. The substrate heater and the bell-jar heat tapes should now be energized and the system allowed to bake and outgas for several hours. If there are no leaks or residual deposits of high vapor pressure materials the pressure should reach a pressure of about $4 \times 10^{-7}$ torr and pressures of $2 \times 10^{-7}$ torr are not unusual.

After verifying that the vacuum is adequate, the substrate temperature is sufficient (e.g. 300°C), the shutter is covering the substrates and, and that the thickness monitor is on and operating properly, the sample material is outgassed by slowly increasing the heater
current until the evaporant just melts and maintaining that current for a minute or two then reducing it to zero. The system is then allowed to pump for several minutes to clear the impurity vapor from the chamber, after which the pressure should return to as good a reading as was obtained prior to sample outgassing.

At this point it seems appropriate to mention that many of our early attempts to produce usable films were failures. This was attributed for the most part to contaminants in the film. At that time we were obtaining pressures on the order of $10^{-6}$ torr. At that time, too, the sample was heated in a boron nitride crucible by a tungsten heater, leading to a much lower deposition rate, and the substrate heater was less efficient, with substrate temperatures some 20-30°C lower. All these effects probably contributed to film impurities and possibly altered the relative proportions of the film constituents; efforts were therefore made in all three areas to improve the situation. The system was tightened up and baked out more thoroughly thereby improving the pressure by an order of magnitude, and the substrate heater and holder were redesigned, permitting slightly higher substrate temperatures. The boron nitride crucible was eliminated and the evaporant placed in a tungsten boat in direct thermal contact with the heated tungsten resulting in a higher deposition rate. The deposition rate is increased further by going from the melting point current to the maximum current as rapidly as possible rather than
gradually as had been the practice.

It should be pointed out here that there are other motives for the attainment of high deposition rates in the case of compounds or alloys which, in this instance, consist of terbium and iron. The vapor pressure of the terbium is higher than that of iron and the vapor tends to be rich in terbium to the extent that there is free terbium available. Also, the boiling point of terbium is lower than that of iron, and while the source temperature is increasing from the terbium boiling point to the iron boiling point the free terbium tends to evaporate much more rapidly. In a slowly heated environment this effect is augmented, of course, by the cooling effect of the evaporating terbium which slows the the heating thereby increasing the duration of this transition. In addition, a detrimental side effect of going to directly heated boats is the proclivity of the free iron to interact with the tungsten. Thus a rapidly heated boat and and a correspondingly high deposition rate are used to minimize these effects. Our data, in comparison with some obtained by Benningfield, seem to indicate these steps have been effective. Somewhat surprisingly, the post-deposition appearance of our source boats indicate, at least for well constituted compounds of TbFe, TbFe₂, TbFe₃, and Tb₃Dy₇Fe₂, that there is little or no free iron available to attack the boat at any time during the deposition implying that the molecular binding energy is greater than the thermal energy at the temperatures reached.
To continue the deposition procedure, when the system has pumped down sufficiently, subsequent to the preliminary sample material outgassing, the heater current is again gradually increased until the evaporant is just melted. At this point, after zeroing the thickness monitor, the current is rapidly increased to maximum and the shutter is opened, exposing the substrates and thickness sensor. When the desired thickness is attained the shutter is immediately closed and the heater current cut off. The system is again allowed to pump down, the shutter retracted, and the silicon monoxide source current slowly increased to that required for a moderate rate of evaporation. The shutter is closed after depositing one-half to one micron of SiO (taking care to observe thickness/density considerations) and the current reduced to zero. This completes the film, and the substrate heater is turned off and the substrates allowed to cool. Often a small amount of helium is introduced into the chamber to expedite cooling.

Films produced in this manner have been durable and have produced reasonably consistent results. We have films well over six months old which still display characteristics similar to those they possessed when first made.

Ultrasonic Generation

The ultrasonic generation of acoustic waves in thin films in an ultrasonic pulse spectrometer provides a convenient method of investigating the magnetoelastic interaction term of the Hamiltonian. A high-frequency time-
varying (or rf) magnetic field is applied to the sample and, if conditions are such that the magnetoelastic interaction is significant, magnetic energy is transformed to elastic energy in the form of acoustic waves of the same frequency as that of the rf magnetic field. Some parameters which affect the interaction are: magnitude of an applied static (or dc) magnetic field, relative orientation of the film, rf field, and dc field, and temperature. Since the film is bonded to a substrate, these acoustic waves couple into the substrate material and are propagated through the substrate. At the opposite end of the substrate rod, which is highly polished and parallel to the film, the acoustic energy is reflected and propagated back through the substrate into the film, and, through the magnetoelastic interaction, back into magnetic energy in the form of an rf magnetic field. The rf field is applied in a burst, or pulse, whose duration is sufficiently short so that the first of the reflected acoustic energy or echo arrives at the film after the applied pulse, and the subsequent acoustically generated rf pulses or signals may be detected and analyzed. The transit time of the acoustic waves in the X-cut substrate is a function of their polarization (along the fast or slow axes) and mode (transverse or longitudinal). Too, if the film is an effective one-half acoustic wavelength thick at the frequency of the exciting rf field, there is a large increase in efficiency due to the resonance condition this creates.
The pulse spectrometer used in this experiment (Fig. 11-3) consists of a Matec model 6000 rf Pulse Generator and Receiver with a model 770 rf Plug-in (300- 700 MHz range) which generates and detects the rf pulse and echos. The detected signal (video) output of the receiver is displayed on a scope and monitored by a Matec model 1235A Pulse Amplitude Monitor. The pulse-amplitude monitor is a boxcar integrator which provides a time averaged dc signal, logarithmically proportional to a selected echo amplitude, to the vertical or Y input of a Moseley model 7001AM X-Y Recorder. /synchronizing signal from the pulse generator is connected to the "SYNC IN" jack of the pulse amplitude monitor and to the external trigger input of the oscilloscope. The Z-axis or cathode input of the scope is also connected to "STROBE OUT" on the amplitude monitor which causes that portion of the video signal which is selected by the monitor gate to be brightened on the oscilloscope display. dc magnetic field monitoring is furnished by a Systron Donner model 3102 Gaussmeter utilizing a Hall-probe detector. The gaussmeter provides a dc voltage linearly proportional to the applied dc field to the X Input of the X-Y recorder. The magnet which provides the dc field is a conventional Harvey Wells electromagnet with 12-Inch poles which can provide about 18 kilogauss with pole pieces affording a 2 inch gap and about 25 kilogauss in a 1 inch gap. The magnet power supply is a Harvey Wells model HS-1050 50 amp supply modified with an external motor
ULTRASONIC GENERATION SYSTEM

FIGURE II-3
driven, selective-speed sweep control. For temperature dependence studies an Andonian dewar with a continuous flow cryostat is available. A PAR (Princeton Applied Research) model 152 Cryogenic Temperature Controller and a TG-100P gallium arsenide diode provide temperature monitoring and control.

A vital element of any spectrometer is that device which constitutes the interface between the instrumentation and the test specimen. It must efficiently couple energy, of whatever form the response of which is being determined, between the spectrometer and the sample; and, at the same time, provide for the effective application and control of whichever parameters are of interest, such as electric or magnetic field, orientation, stress, temperature, and many others. In our case, of course, we are investigating the interaction between an rf magnetic field and a thin film, and the parameters of principal concern are the applied dc magnetic field and the relative orientation of the dc and rf fields and sample as well as the film temperature.

The applied rf field must lie in the plane of the film, and we wish to vary the angle between the dc field and the rf field by at least 180 degrees, both in the plane of the film and in a plane normal to the film. A resonant length of microstrip transmission line or stripline "cavity" was selected as being a device which would adequately satisfy these criteria and yet is easily constructed. One may see from the mode diagram for such a stripline device (Fig.
that the magnetic field is strongest and of the desired planar shape in the region between the microstrip and ground plane at the shorted end. The sample film is positioned in this region by inserting the film end of the substrate through a small hole in the ground plane and dielectric and placing the film next to the microstrip; thus the rf magnetic field lies in the plane of the sample and perpendicular to the direction of the microstrip. Energy is inductively coupled between the resonant segment of microstrip and the nonresonant segment which in turn is connected through a stripline/coax transition (or launcher) and coaxial line to the generator/receiver. The cavity also contains a hole in the ground plane at the E-field maximum, similar to the one for the sample and displaced from it by a quarter wavelength, to provide for the generation of signals in the quartz substrate by the surface piezoelectric effect.

The design of our electromagnet provides for the rotation of the magnetic field in the horizontal plane about an axis midway between the pole pieces. Thus the straight quarter wavelength configuration (Fig. 11-5a) positioned along this vertical rotation axis allows the dc magnetic field to be oriented anywhere in a plane perpendicular to the film, while the folded half wave cavity (Fig. 11-5b) results in the dc field lying in the plane of the sample and at any angle relative to the rf field. A brass cylinder having a three millimeter hole in it is soldered on the ground plane such that the hole in it and the ground plane
Figure II-4

Stripline Mode Diagram

Electrical Field (E Field)

Ground Plane

Dielectric

Stripline Conductor

Shorted End — Maximum H Field
are aligned. The holder has a heater consisting of about six feet of #30 constantan wire wound on it with a TG-100P diode fixed to it with Wood's metal to provide temperature sensing. The stripline is suspended from a header on a section of rigid coaxial transmission line which positions the sample at the center of the magnet pole pieces. This assembly may also serve as part of a cryostat and be inserted into the dewar when appropriately installed between the pole pieces.

After preliminary adjustments have been made on the spectrometer such as proper oscilloscope presentation, X-axis sensitivity of the plotter (e.g. 1 kOe per centimeter), and approximate frequency settings of the Matec gen/rec; the generator, receiver, and preamp sections are tuned to the exact frequency of whichever stripline cavity is being used. Alignment signals are generated by the surface piezoelectric effect on a quartz rod inserted in the hole located at the E-field maximum (to facilitate this operation the cavity should be removed from between the magnet pole pieces and connected by a flexible coax to the Matec). The sample film and substrate are then inserted into the holder at which time magnetostrictively generated signals should be observed (it will quite likely be necessary to hand hold the cavity in a magnetic field to obtain these signals); once signals are observed the substrate rod is rotated to maximize either the fast or slow transverse signals and minimize the other. After the desired
polarization is selected the exposed end of the substrate rod is taped to secure it against accidental rotation and, of course, against its falling out, and the cavity is restored to its normal operating position. The dc magnetic field and its orientation are adjusted for maximum signal amplitude, while the coaxial attenuator in the receiver input is adjusted so that the pulse amplitude monitor, whose gate width has been adjusted to accommodate a full echo as determined by the brightened segment of the video display and the reference level then set to indicate 0 db for noise, registers 20 db on an echo selected by the delay control (usually the first, and again as defined by the accented scope display). With zero insertion loss in the receiver input line there are typically two or three echos which saturate the receiver, followed by a long train of echos which exhibit the familiar exponential decrease in amplitude, but the signals are reduced not only to avoid saturating the receiver but also to stay within the dynamic range of the pulse amplitude monitor. The Y-axis sensitivity of the X-Y plotter is calibrated at this time (usually 1 db signal plus noise to noise per centimeter).

With the completion of these preparations we are in a position to collect data on the applied dc magnetic field magnitude and angular dependence of ultrasonic generation in the selected specimen. These data are recorded as signal plus noise to noise time averaged echo amplitude as a function of a variable parameter with other parameters of a
fixed, selected value.

For temperature-dependence studies the dewar system is utilized. In preparation, the dewar jackets are evacuated, and the sample chamber and reservoir flushed, dried, and pumped down with the cryostat assembly installed (sample in place). The outer dewar is filled with liquid nitrogen, and either liquid nitrogen or helium is put in the reservoir depending on what minimum temperature is desired. In the case of the TbFe study reported here, only one temperature run was made and that only down to liquid nitrogen. A low backpressure is maintained in the reservoir and the needle valve between the reservoir and sample chamber is opened to provide a slow stream of the coolant vapor across the sample holder and cavity. The temperature controller, when properly adjusted, Automatically provides the correct heater current to maintain the sample at the selected temperature. Temperature dependence data are taken as temperature being a series of fixed values while echo amplitude as a function of dc field is recorded.

**Magnetization**

Magnetization measurements of such a small mass as our samples (e.g. 568 micrograms for an 8 micron film of TbFe) demand a magnetometer of the highest sensitivity, while the requirement to measure the magnetization in a large magnetic field virtually excludes all but a vibrating sample configuration. Our magnetometer (Fig. 11-6) was constructed along the lines of one described by Foner and a similar
one built at the Naval Ordnance Laboratories by Clark's group. A vertical oscillatory motion is imparted to the sample via a coupling rod by a 60 watt University speaker driver powered by a Dynaco 70 watt power amplifier whose input is derived from the reference output of the PAR model 124 Lock-In Amplifier. The vibrating dipole field of the sample induces a voltage in a pair of pickup coils, of 17,000 turns each of #48 wire, mounted beside the sample on the faces of the housing. This voltage is connected to the signal input of the PAR model 116 plug-in preamp at the lock-in amplifier. This signal is filtered, amplified, and compared phasewise with the driving or reference signal, and the results are time integrated then in turn drive the Y-axis of the X-Y recorder whose X-axis is controlled as it was in the pulse spectrometer. Synchronous vibration of the pickup coils at the driving frequency constituted a major problem which was first attacked by insulating the transducer mounting plate from the rest of the system by supporting it on a slab of rubberized hair packing material. Contact between the driving rod and the pickup coil housing was eliminated and lateral alignment of the rod provided by bushings in heavy plates mounted to the yoke of the magnet immediately above and below the housing. Flexing of the housing walls on which the coils were mounted was minimized by potting the walls and coils with epoxy and at the same time embedding all the very light wires in the epoxy and using larger, more rigid wire for internal connections in the
housing. Calibration of the magnetometer is accomplished by comparison with the magnetization curve obtained for a small nickel sample whose magnetization characteristics are well known and documented.\(^9\)

The coil configuration used here is most sensitive to sample magnetization parallel to the applied field and insensitive to components normal to it. Earlier results from ultrasonic generation experiments, however, indicated that the most interesting possibilities lay in measuring the sample field normal to the plane of the disc, (though parallel to the applied field), and the space between the coils could not accommodate the substrate rods lengthwise (it should be pointed out that high-field pole pieces with a one inch gap were being used). This difficulty was circumvented by depositing a film on a 10-MHz quartz transducer of the same diameter as the rods at the same time films were being deposited on regular substrates and of the same thickness. This thin quartz disc could then be inserted in the magnetometer with its plane (and the plane of the sample film) normal to the applied field.

Consistent results were eventually obtained though we only had one quartz transducer available and only obtained data for one film in the work encompassed by this thesis.
III. EXPERIMENTAL RESULTS

This chapter presents the data obtained from a series of experiments conducted as described in the preceding chapter with particular attention to those features of apparent significance. The first section describes the results of the investigation into the ultrasonic generation properties of the various terbium-iron compounds as well as one ternary compound specimen of terbium, dysprosium, and iron; and the second section shows the outcome of the magnetization measurements on TbFe$_2$.

Ultrasonic Generation

As was remarked earlier, the bulk of the experimental effort included in this thesis was concentrated in the area of ultrasonic generation in TbFe thin films. Our interest in this specific material, as mentioned in the Introduction, was stimulated by the large, room temperature, static, bulk magnetostriction it exhibited.\(^4\) Figure III-1 is a curve showing the echo amplitude of acoustic waves generated by a TbFe$_2$ film as a function of the magnitude of the dc magnetic field applied normal to the plane of the film and with an rf field of 670 MHz at room temperature. The curve begins at zero applied field (recalling that a substantial field was applied to the sample film for alignment purposes prior to
TbFe$_2$ on QUARTZ

FIGURE III-1

H$_{\text{APPLIED}}$ (kOe)

ECMO AMPLITUDE (dB)
the sweep and in the same direction as the initial sweep) from where it is slowly increased to about 20 kOe and decreased back to zero, at which point the magnet leads are reversed by means of a reversing switch and the field then increased in the opposite direction and again returned to zero. This field sweeping procedure is typical of all the field magnitude dependence data presented here. A number of TbFe films of various thicknesses (but primarily two microns) were tested in this manner with qualitatively consistent results. There were some quantitative variations, but, given the irreproducibility of thin films in general, they were judged to fall within the scope of the term "consistent".

The TbFe$_2$ films fulfilled our fondest expectations regarding their ability to magnetostrictively generate acoustic waves in the UHF region at room temperature by being more than 100 times as efficient as quartz. There are in addition, however, two other features of major significance evident in this curve which were entirely unexpected. One first notes that there is an appreciable signal present at zero applied field (dependent, of course, upon the prior magnetization of the sample) and, secondly, that the signal vanishes at a reversed field on the order of 4 kOe. This implies a remanent magnetization at zero applied field that requires a reversed field of appreciable magnitude to be removed, which in turn suggests that the film possesses a magnetic anisotropy axis. That a film whose
bulk material has a noticeable anisotropy\textsuperscript{5} should also exhibit anisotropy is not altogether surprising, but that it should be so large and with the easy axis normal to the plane of the film was entirely unexpected (and unexplained to this point). That this was indeed an acceptable description of the phenomena involved remained to be verified by magnetization measurements, the results of which appear in the next section.

The temperature dependence of the ultrasonic generation properties of a TbFe\textsubscript{2} film was determined in a single temperature run beginning at 77°C. The coercive field, or the reverse field required to reduce the remanent magnetization to zero, is inferred as being that field at which the magnetostrictively generated signals vanish and is plotted as a function of temperature in Figure III-2a; while the correlation between temperature and echo amplitude at zero applied field is shown in Figure III-2b. These data, too, are unexplained as yet and are, at any rate, insufficient to warrant conclusions. In addition, the TbFe\textsubscript{2} study included an evaluation of the ultrasonic generation relative to the angle between the dc field and the rf field with the dc field in a plane perpendicular to the plane of the film (Fig. III-3).

The discovery of this large, nonplanar anisotropy increased the motivation to investigate some of the other rare earth-iron compounds beyond just that of comparing overall generation efficiencies, for, again according to
TbFe$_2$ TEMPERATURE DEPENDENCE

FIGURE III-2
TbFe$_2$

$\theta = \Delta H_{bc}$ and normal to film plane

$H_{bc} = 12.3$ kOe

FIGURE III-3

ANGULAR DEPENDENCE OF ECHO AMPLITUDE

$\theta$ (degrees)

(8P) $N/(N+S)$
Clark and Belson's studies of the magnetic properties of various rare earth-iron compounds in bulk form, TbFe$_2$ exhibited the largest anisotropy of the materials included in their study. Ultrasonic generation data on some of these compounds might then be used to evaluate the anisotropy characteristics of the films as well as their relative generation efficiencies. Two films, two microns thick, of each of those materials tried was made and tested. The data for each type were qualitatively consistent between films and, again, within reasonable limits, quantitatively.

The material in Clark and Belson's report exhibiting the least degree of magnetic anisotropy yet with a large bulk magnetostriction is the ternary Laves phase compound Tb$_3$Dy$_7$Fe$_2$. The ultrasonic-generation dc field dependence of this type film (made from a bulk sample obtained from Clark) is given in Figure III-4. The efficiency is quite large, and there is no zero-field signal present though there is a very small hysteresis in the response, implying a correspondingly small coercive force. The response is quite sharp and resonant like. Figure III-5 depicts the response of a TbFe$_3$ film. There is a very small but detectable zero field signal and an appreciable hysteresis indicating a moderate coercive force in the response. The only extraordinary feature here is the smaller maximum signal obtained on the first upward sweep following a field reversal than on subsequent sweeps. The TbFe film exhibits the most anomalous and puzzling behaviour of all the films.
\[ \text{Tb}_3 \text{Dy}_{7/2} \text{Fe}_2 \]
There is no detectable remanent field signal although the hysteresis is substantial, yet the amplitude of the response during the first sweep following a field reversal is quite startling. Once the sample had been saturated the response seen on the right could be obtained repeatedly until the field again reversed from which the next sweep invariably resulted in the very large increase in signal.

**Magnetization**

The need for magnetization data on at least some of our thin films became apparent when it was deemed advisable to substantiate the less conclusive evidence pointing to the large, nonplanar magnetic anisotropy of the TbFe$_2$ films; and the magnetometer was constructed as described in Chapter II. The magnetization curve for an eight micron film of TbFe$_2$ is shown in Figure III-7. The trace was started at saturation and the field swept down to zero, reversed and increased to saturation in the opposite direction, then decreased to zero and reversed again, and finally increased to saturation once more. The curve for the case of the applied field perpendicular to the film indicates a saturation field of 12-13 kOe and a coercive field of about 5 kOe, while the saturation field is about 20 kOe and the and the coercive field only 3 kOe in the case of the field parallel to the plane of the film. The magnetization curve for a bulk sample of TbFe$_2$ is given in Figure III-8 and agrees well with that published by Clark and Belson. These magnetization data are
BULK TbFe₂

FIGURE III-8
then used to determine the magnetization dependence of the echo amplitude in the TbFe$_2$ case the results of which are given in Figure III-9.
SIGNAL AMPLITUDE vs MAGNETIZATION

**FIGURE III-9**
IV. THEORY

In this chapter, a simplistic theory is developed which leads to a qualitative explanation of the non-resonant generation of elastic waves in polycrystalline thin films of TbFe$_2$ through the magnetoelastic interaction. TbFe$_2$ is a cubic, Laves phase compound with a diamond lattice structure, and is ferrimagnetic at room temperature. The iron sublattice, whose Curie temperature is much higher than room temperature, is credited with causing the magnetic ordering of the terbium sublattice antiparallel to the iron sublattice at room temperature. Indirect exchange is considered to be the principal mechanism responsible for this ordering. The magnetostriction, on the other hand, is considered to result, predominantly, from the single-ion, crystal-field interaction. Review articles by Cooper$^{10}$ and Rhyne$^{11}$ on magnetic ordering and bulk magnetic properties provide an outline of the present level of understanding in these areas.

Our development will be based on a simple, hybrid Hamiltonian consisting of the three terms

$$H = H_m + H_e + H_{me}.$$  

$H_m$ is the magnetic energy term. For simplicity, we assume a molecular field model and include the exchange interaction using a presumably uniform magnetization. Anisotropy
contributions are excluded as being considered of negligible significance to the magnetoelastic interaction. $H_m$ is of the form

$$H_m = N g \mu_B (\vec{H} + \Gamma \vec{M}) \cdot \vec{J},$$

where $N$ is the number of terbium ions per unit volume, $g$ is the Landé $g$-factor, $\mu_B$ the Bohr magneton, and $\vec{J}$ is the angular momentum. $\vec{H}$ is the externally applied magnetic field, while the molecular field constant, $\Gamma$, and the magnetization, $\vec{M}$, constitute the internal magnetic field.

The elastic energy term, $H_e$, includes the classical potential and kinetic energies, and is given by

$$H_e = \frac{1}{2} C_{ij} e_i e_j + \frac{1}{2} \sum \rho \left( \frac{\partial u_i}{\partial x} \right)^2.$$

Here, the $C_{ij}$, $e_i$'s, and $\rho$ are the traditional elastic constants, strain, and density, respectively, and the $u_i$ are cartesian coordinates.

Assuming cubic symmetry at the terbium ion sites, the magnetoelastic energy term, $H_{me}$, may be written

$$H_{me} = N \left\{ B_1 \left( (e_x - e_z)(J_x^2 - J_y^2) + \frac{1}{3} (e_z - \frac{1}{3} (e_x + e_y + e_z)) \right) (3J_z^2 - J(J+1)) \right\} + B_2 \left[ e_4 (J_y J_z + J_z J_y) + e_6 (J_x J_z + J_z J_x) \right] + e_8 (J_x J_z + J_z J_x) + e_6 (J_x J_y + J_y J_x) \right\}$$

where the $B_i$ are the static magnetoelastic coupling coefficients.

Magnetostriction involves the exchange of energy between the purely magnetic and elastic energies through the
magnetoelastic interaction term. The magnetic energy dynamic variable is, of course, the rf magnetic field, and the elastic energy's time dependence may be expressed in terms of the strains. Our procedure shall be to evaluate the average energies of each of these components using perturbation methods and determine the coupling between the magnetic and elastic terms.

The static portion of the magnetic term is much larger than the dynamic term and has well known solutions, making it a suitable unperturbed Hamiltonian. The dynamic segment of the magnetic energy, $H_{dc}$, and the magnetoelastic term comprise the perturbing Hamiltonian, $H'$; the elastic energy term being a classical expression. Thus the Hamiltonian is rewritten

$$H = H_0 + H', $$

where

$$H_0 = H_{dc},$$

and

$$H' = H_{rf} + H_{me}. $$

Since we are seeking only a qualitative determination of the dynamic magnetostriction at this time, we shall, in the course of finding solutions to the perturbing system, isolate all the terms in $H_{af}$ and $\mathcal{E}$ and discard all others. We shall also find it necessary to go to second order calculations because in the first order solution

$$E_m' = \langle m | H' | m \rangle ,$$
the $J_x$ operator has no non-zero diagonal elements, and no $H_{hf}$ terms appear. The second order solutions are given by

$$E_m^{\prime\prime} = \sum_n \frac{\langle m|N|l_m^n\rangle \langle n|N|l_m^n\rangle}{E_m - E_n}$$

$$= \sum_n \frac{\langle m|N_{me} l_m^n\rangle \langle n|N_{me} l_m^n\rangle}{E_m - E_n}$$

$$+ \sum_n \frac{\langle m|N_{me} l_m^n\rangle \langle n|H_{hf} l_m^n\rangle + \langle m|H_{hf} l_m^n\rangle \langle n|N_{me} l_m^n\rangle}{E_m - E_n}$$

$$+ \sum_n \frac{\langle m|H_{hf} l_m^n\rangle \langle n|H_{hf} l_m^n\rangle}{E_m - E_n}.$$

Now again, there is no $H_{hf}$ dependence in the $\langle N_{me} X N_{me} \rangle$ segment and the $\langle N_{me} X H_{hf} \rangle$ term leads to a non-linear, dynamic susceptibility containing no elastic terms. The only possible contributing combinations in the cross product terms are those of $J_x$ with $J_x J_y + J_y J_x$ and $J_z J_z$, and it happens that the $J_x J_y$ terms are also zero, so that the net effective second order solution is

$$E_m^{\prime\prime} = \left( N B_e e_x (N g \mu_b H_{hf}) \right) \right.$$
We want an averaged form of this to manipulate in conjunction with the classical elastic energy term, so we rewrite it

\[ \langle E_m \rangle = \frac{NB_3 \varepsilon_s H_{\mathrm{eff}}}{H_{\mathrm{dc}} + \Gamma M} \langle J_{z_0} \rangle , \]

where \( \langle J_{z_0} \rangle \) is the statistically averaged, second order solution of the \( J_x, J_x \) cross terms. 12

This solution demonstrates that the only strain component which couples to the dynamic magnetic field is the strain \( \varepsilon_5 \). This indicates that any magnetostrictively generated elastic waves will be polarized in the \( x \) direction which is supported by experimental evidence. We may, therefore, expand the elastic energy, \( \mathcal{H}_e \), keeping only the \( \varepsilon_5 \) terms, obtaining

\[ \mathcal{H}_e = \frac{1}{2} c_{44} \varepsilon_5^2 + \frac{1}{2} \rho \left( \frac{\partial u_x}{\partial t} \right)^2 , \]

where

\[ \varepsilon_5 = \frac{\partial u_x}{\partial x} . \]

We now have an expression for the total free energy containing the magnetic field and strain dynamic variables which is written

\[ \mathcal{H} = \frac{1}{2} \rho \left( \frac{\partial u_x}{\partial t} \right)^2 + \frac{1}{2} c_{44} \varepsilon_5^2 + \frac{NB_3 \varepsilon_s H_{\mathrm{eff}}}{H_{\mathrm{dc}} + \Gamma M} \langle J_{z_0} \rangle , \]

which, through the equations of motion, leads to the wave equation
\[
\rho \frac{\partial^2 u_x}{\partial t^2} = c_{44} \frac{\partial^2 u_x}{\partial z^2} - \frac{1}{\delta} \left( \frac{NB_z H_{sf} \langle J_{so} \rangle}{H_{dc} + \Gamma M} \right).
\]

We note that if \( H_{sf} \) is uniform, the \( B_z \) term will vanish, destroying the magnetostriction theory with it. We are rescued from this, though, by considering the skin depth of \( H_{sf} \) in the conducting film. Therefore, if

\[
H_{sf} = H_{sf}^0 e^{-i \omega t - z/\delta}
\]

the wave equation becomes

\[
\rho \frac{\partial^2 u_x}{\partial t^2} = c_{44} \frac{\partial^2 u_x}{\partial z^2} + \frac{NB_z H_{sf} e^{-i \omega t - z/\delta} \langle J_{so} \rangle}{\delta (H_{dc} + \Gamma M)}.
\]

If one assumes a linear relationship between and so that their time dependence is equivalent, within a phase factor, then \( u_x \) will be of the form

\[
U_x(z,t) = U_x(z) e^{-i \omega t}
\]
This permits the reduction of the second order differential equation to the ordinary differential equation

\[-\omega^2 \rho u_x = c_{44} \frac{d^2 u_x}{dz^2} + \frac{NB_2 H_{Af} e^{-i\omega t - z/\delta}}{\delta (H_{dc} + \Gamma M)},\]

or

\[\frac{d^2 u_x}{dz^2} + k^2 u_x = \frac{-NB_2 H_{Af} e^{-i\omega t - z/\delta}}{\delta (H_{dc} + \Gamma M)},\]

\[= c e^{-z/\delta}.\]

This equation has a solution of the form

\[u_x = Ae^{i k z} + Be^{-i k z} + \frac{c \delta^2}{1 + k^2 \delta^2} e^{-z/\delta},\]

where

\[A = \frac{c \delta^2}{1 + k^2 \delta^2}.\]

We may assume, for simplicity, a solution involving unidirectional waves (i.e. an infinite medium) which eliminates the coefficient B. The second boundary condition is established by the free surface of the film at z=0. The stress-free surface implies the strain is also zero, or

\[e_s = \left(\frac{\partial u_x}{\partial z}\right)_{z=0} = 0.\]
These conditions lead to a solution for $\mathbf{U}_x$ of

$$
\mathbf{U}_x = \frac{c \delta e^{ikz}}{i k (1 + k^2 \delta^2)} + \frac{c \delta^2 e^{-2i/\delta}}{1 + k^2 \delta^2}.
$$

For small skin depths the transient term may be neglected, and we obtain

$$
\mathbf{U}_x = \frac{c \delta e^{ikz}}{i k (1 + k^2 \delta^2)},
$$

or

$$
\mathbf{e}_s = \frac{\partial \mathbf{U}_x}{\partial z} = -B_z H_{n_z} e^{-i \omega t} \langle J_{z_0} \rangle \frac{(1 + k^2 \delta^2)}{(H_{dc} + \Gamma M)} \gamma
$$

$$
= \gamma \frac{\langle J_{z_0} \rangle}{H_{dc} + \Gamma M} e^{-i \omega t}.
$$

We should point out here, that $\Gamma M$ is much larger than $H_{dc}$, thus the applied static field dependence is indirectly realized through the magnetization, and the strain, in effect, goes like

$$
\mathbf{e}_s \sim \frac{\langle J_{z_0} \rangle}{M}.
$$

A normalized plot of $\mathbf{e}_s$ is shown in Figure IV-1.
FIGURE IV-1
V. CONCLUSION

The investigation of TbFe$_2$ polycrystalline thin films was undertaken in anticipation of finding them to be highly efficient generators of UHF elastic waves at room temperature. Experimental results confirmed these expectations and also revealed the existence of a large, non-planar anisotropy in these films. Thin films of differing compositions were also studied and were found to be similarly capable of ultrasonic generation and to possess anisotropies of varying but lesser degrees. The appreciable remanent magnetization of the TbFe$_2$ films permits ultrasonic generation with no applied static field and enhances their potential as high frequency transducers. The non-planar anisotropy, particularly in those materials having a smaller coercive force, opens the possibility of their application in bubble-domain memory devices.

A theory has been developed which qualitatively accounts for the dynamic magnetostriction in materials of this class. The polarization of the elastic waves is successfully predicted, and the static field dependence is in reasonable agreement with experimental results. Theory does not yet account for the anisotropies observed nor the anomalous behavior of the magnetostriction in TbFe.

It is anticipated that the current program will go far
toward resolving many of these questions. Experiments are planned to systematically investigate the ultrasonic generation and magnetization properties of thin films of several compositions while controlling various parameters of film production such as film thickness, substrate temperature, and a magnetic field applied during the deposition process. Temperature and frequency dependence studies are also planned as well as some ferromagnetic resonance experiments.
ACKNOWLEDGEMENTS

I wish to express my appreciation for my research advisor, Dr. P. L. Donoho, who has been a capable and cheerful source of guidance and ideas, and for his patience when things were going slowly. I thank my colleagues in the Rare-Earth Group, both former and present, for their many contributions; particularly Dr. W. C. Hubbell who built the magnetometer when time was such a vital element. I am indebted to Rice University and the Advanced Research Projects Agency for their financial support. I am especially grateful to my wife, Carol, without whose unending patience and encouragement this effort would have been impossible.
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