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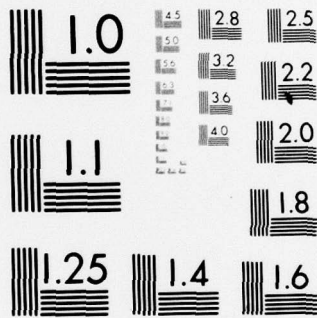
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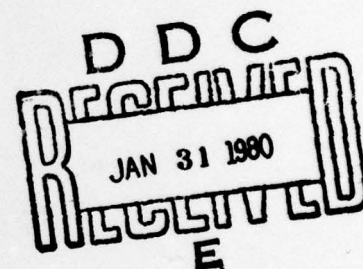
**MAGNETOMECHANICAL COUPLING AND MAGNETO-
STRICTION IN VERTICALLY ZONED $Tb_{.27}Dy_{.73}Fe_2$**

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1 NOVEMBER 1979

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FOREWORD

The magnetic and magnetomechanical properties reported here are developments of a program to develop magnetostrictive materials for high power sonar. The materials are an alloy of terbium-iron and dysprosium-iron Laves phase compounds. The magnetoelastic strains are very large and very anisotropic.

This report is a compilation of magnetostriction, magnetomechanical coupling and grain structure data of textured samples prepared by a vertical free standing zone technique. Unlike previous texturing techniques we have reported on, this technique yields full sized transducer elements in the shape of a rod. The magnetomechanical properties are considerably better than random polycrystal materials. Part of this work was reported at the 1979 International Conference on Magnetism in Munich Germany.

This study was carried out in the Solid State Branch of the Radiation Division. The materials development was sponsored by the NRL Material Block Program (Howard Lessoff). Magnetic measurements and the fabrication of prototype transducer components were carried out under the sponsorship of the NOSC Transducer Block Program (R. Smith). Research on the magnetoelastic properties of highly magnetostrictive rare earths is sponsored by the Office of Naval Research (R. Pohanka) and the NSWC Independent Research Fund (J. di Rende).

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INTRODUCTION

The cubic Laves-phase rare-earth Fe_2 compounds have huge values of magnetostriction constants (λ) and anisotropy energy (E_k). However, with an appropriate alloying of binary compounds, ternaries of the form $\text{R}_x\text{R}_{1-x}\text{Fe}_2$ may be obtained with large values of λ but with E_k reduced by two orders of magnitude.¹ Large values of the magnetomechanical coupling factor, k_{33} , have been observed in $\text{Tb}_{0.27}\text{Dy}_{0.73}\text{Fe}_2$ (Terfenol)³. The magnetostriction is quite anisotropic with $\lambda_{100} \ll \lambda_{111} = 1.6 \times 10^{-3}$. Because the large, anisotropic magnetostriction introduces large inhomogeneous strains in random polycrystals (RPC), the permeability is quite low.² This restricts magnetomechanical activity.

We previously reported efforts in grain orienting to enhance magnetomechanical activity.³ Bridgman growth and horizontal zoning methods were examined during FY78. During FY79, methods were developed to prepare high coupling oriented samples by a vertical zoning technique. This report describes this effort.

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1. A. E. Clark, AIP Conf. Proc. 18, 1015 (1974).
 2. H. T. Savage, A. E. Clark and J. M. Powers, IEEE Trans. on Magnetics, MAG-11, 1355 (1975).
 3. H. T. Savage, R. Abbundi, A. E. Clark, "Permeability, Magnetomechanical Coupling and Magnetostriction in Grain-Oriented Rare Earth-Iron Alloys," NSWC/WOL TR 78-197.

SAMPLE FABRICATION AND CHARACTERIZATION

In the FY78 study cited above,³ the permeability and magnetomechanical coupling were reported on grain oriented samples prepared by both Bridgman and horizontal zone techniques. The samples prepared by the Bridgman techniques, while overall single crystal, contained substantial quantities of Widmanstätten precipitate. These planar defects provided pinning points for magnetic domains, which inhibited easy domain wall motion and high magnetostrictivity. Only in quaternary (Tb-Dy-Ho)Fe₂ alloys, which are nearly congruently melting, is the amount of precipitate sufficiently low to obtain high magnetomechanical coupling.³ In these alloys, however, the saturation magnetostriction is relatively low. On the other hand, the horizontal zone technique, although resulting in Widmanstätten free samples, yielded only thin samples of high quality. Only the upper part of the samples were useful. The presence of the supporting "cold finger" altered the grain structure at the bottom of the boule in such a manner that the magneto-mechanical characteristics were deleteriously altered.

We have thus proceeded to develop a crucibleless vertical free standing zone method. With this technique a uniform rod can be obtained. The starting materials are arc-cast cylinders approximately 2 1/2" long. Two or three cylinders are subsequently joined end to end to yield 1/4" diameter rods 4" to 6" long. This is an ideal size for magnetostrictive elements intended for low frequency sonar projectors (both toroid and Tonpilz type). The long cylinders are then carefully r.f. zoned at a relatively high speed (~50 cm/hr) to inhibit single crystal growth. Two magnetostrictive rods are shown in Fig. 1. A typical photo micrograph is shown in Fig. 2. Because of the high growth rate employed, the growth is dendritic, not "plane front". Microprobe studies show compositional changes due to the dendritic growth. The rare-earth/iron ratio remains constant while the Tb/Dy ratio fluctuates. We find the composition to be Tb_{27±.025}Dy_{73±.025}Fe₂. The "wavelength" of the fluctuation is about 150 μm. The fluctuation in Tb/Dy ratio implies a change in anisotropy energy from 2×10^5 to $\approx 10^6$ ergs/cm³ over the range of the fluctuation.⁴ See Fig. 3. The change in anisotropy energy over a domain wall width does not seem to be large enough to impede domain wall motion. The fluctuation in Tb/Dy ratio also causes a fluctuation in λ_{111} of about 35×10^{-6} .

4. Private communication, Conrad Williams and N. C. Koon, Naval Research Laboratory, Washington, DC.

MAGNETOMECHANICAL MEASUREMENTS

Measurements were completed on two cylindrical samples approximately 7.5 cm long and .5 cm in diameter. Peak coupling factors measured by the method described in reference 2 were found to be .63 and .69. The peak coupling factors occurred at a field of about 240 Oe. See Fig. 4. The above parameters can be compared to random polycrystal (RPC) values of .55 for the peak coupling factor which occurs at a field of ≈ 150 Oe. RPC were prepared by arc melting. The magnetomechanical coupling in the vertically zoned material is not quite as high as selected samples of grain oriented material previously prepared. However, one should note that the whole rod is useful here. Coupling factors for our various preparation methods are summarized in Table I.

Table I. Peak coupling factors for different preparation techniques.

Preparation Method	Vertical Zone	Horizontal ^a Zone	Bridgman ^a Technique	Arc-melting (Random Polycrystal)
Coupling Factor	.63-.69	.74	.73	.55

a. Selected samples.

We define $d_{500} = \lambda/500$ where λ is the parallel magnetostriction at 500 Oe. Strain gages of 1 mm^2 in area were placed at five different positions on the boule. The area of the strain gage is comparable to the area of a crystallite. A summary of the values of $\lambda_{\parallel}$, $\lambda_{\perp}$ and d_{500} are shown in Table II. $\lambda_{\parallel}$ and $\lambda_{\perp}$ are the saturation values of the magnetostriction with the field parallel and perpendicular to the axis of the sample. The larger value ($\sim 20\%$) of $\lambda_{\parallel}$ in this material compared with the value found in RPC is proof that favorable grain orientation has been obtained. The crystallites have a growth axis near $\langle 111 \rangle$. d_{500} is an important figure of merit in applications. Most of the locations show a considerable improvement in the value of d_{500} over RPC.

Table II. Strain measurements at various positions on the boule. All values are multiplied by 10^{-6} .

Position	$\lambda_{\parallel}$	$\lambda_{\perp}$	$\lambda_s \equiv \frac{2}{3}(\lambda_{\parallel} - \lambda_{\perp})$	$d_{500}(\text{Oe}^{-1})$
1	1520	-360	1250	1.9
2	1390	-250	1090	1.7
3	1380	-540	1280	1.8
4	1450	-500	1300	1.4
5	1010	-430	960	.85
Average			1180	1.53
Random Polycrystal	1000	-500	1000	.8

RECOMMENDATIONS

The goal of this program is to develop mass production techniques to inexpensively produce large samples of highly magnetostrictive rare earth-Fe alloys. These alloys should possess " d_{500} " constants greater than $2.0 \times 10^{-6}/\text{Oe}$ over the entire length. In our judgment two methods show high promise: (1) rapid vertical zoning, and (2) Bridgman growth in a high temperature-gradient.

The rapid zoning method should be refined to yield transducer rods of magnetomechanical coupling $k \approx .70$ and $d_{500} \approx 2.0 \times 10^{-6}/\text{Oe}$ reproducibly. Such a goal should be attainable within the next year. Subsequently refinements should be examined to minimize cost. For example, the performance of rods prepared from less pure starting materials should be tested.

Widmanstätten-free Bridgman growth single crystals offer the highest promise. These samples will possess the highest possible saturation strain (2400×10^{-6}). The " d " constants will be limited by the defect structure. Here methods to grow plane front single crystals in high gradient furnaces should be explored. First, methods must be found to eliminate second phase Widmanstätten precipitates and twins.

A second low frequency transducer, has been designed and constructed at Raytheon Corporation, using RPC rods.⁵ While designed to project 50-100 W, outputs as high as 340 W have been reached.⁶ Performance, in particular efficiency, would be substantially improved utilizing materials prepared by either of the above methods. It is recommended that refinements in both methods be pursued. Neither method should be dropped at this time.

ACKNOWLEDGEMENTS

We wish to thank R. Marinenko of the National Bureau of Standards and F. Veltry of the International Nickel Corporation for their contribution to the microprobe studies.

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5. J. Butler and S. Ciosek, "Rare Earth Magnetostrictive Transducer" Final Report, Raytheon Company, Portsmouth, RI, October, 1978.
 6. J. Butler and S. Meeks, unpublished.

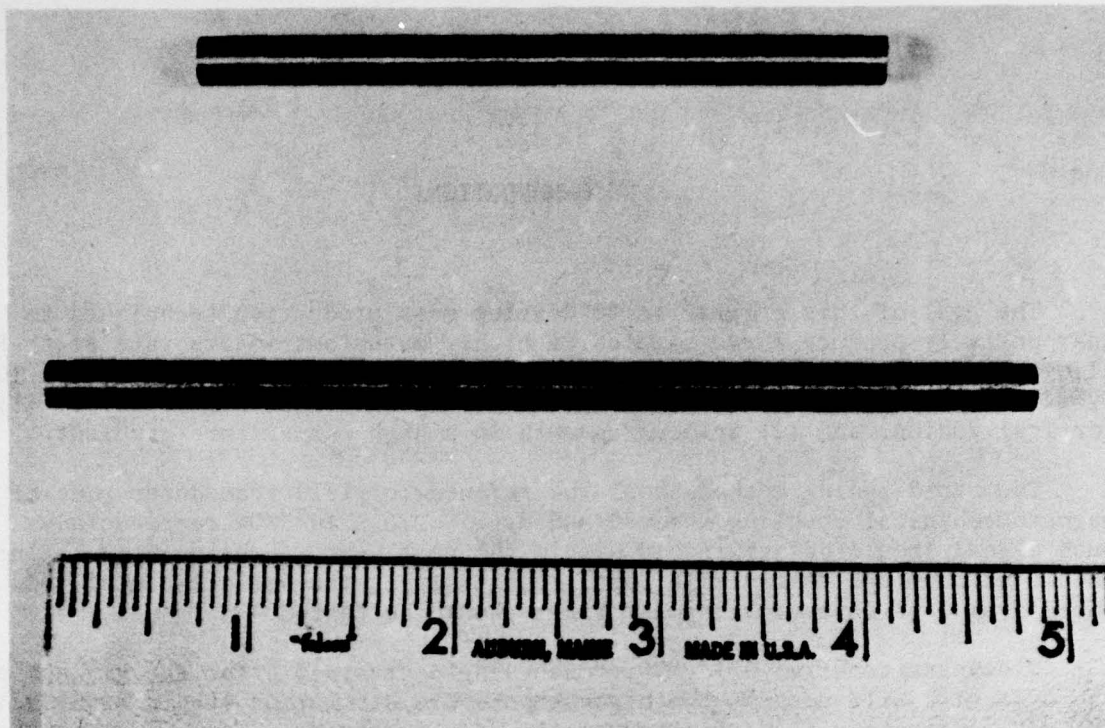


Figure 1. Two cylindrical transducer rods. The rods are .235 inches in diameter. The ruler is calibrated in inches.

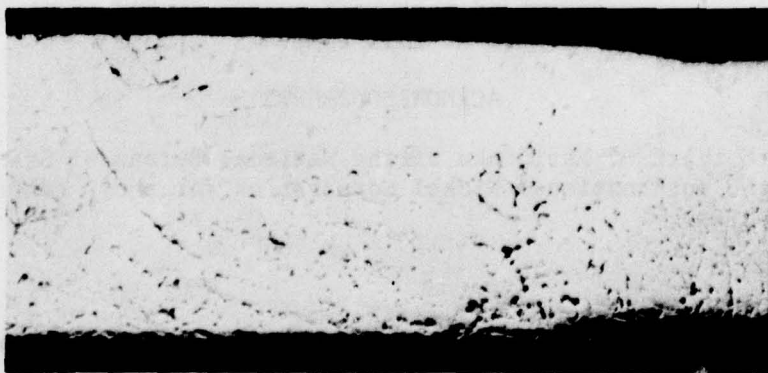


Figure 2. Photo micrograph of a sample of vertically zoned $\text{Tb}_{.27}\text{Dy}_{.73}\text{Fe}_2$. 10 x magnification.

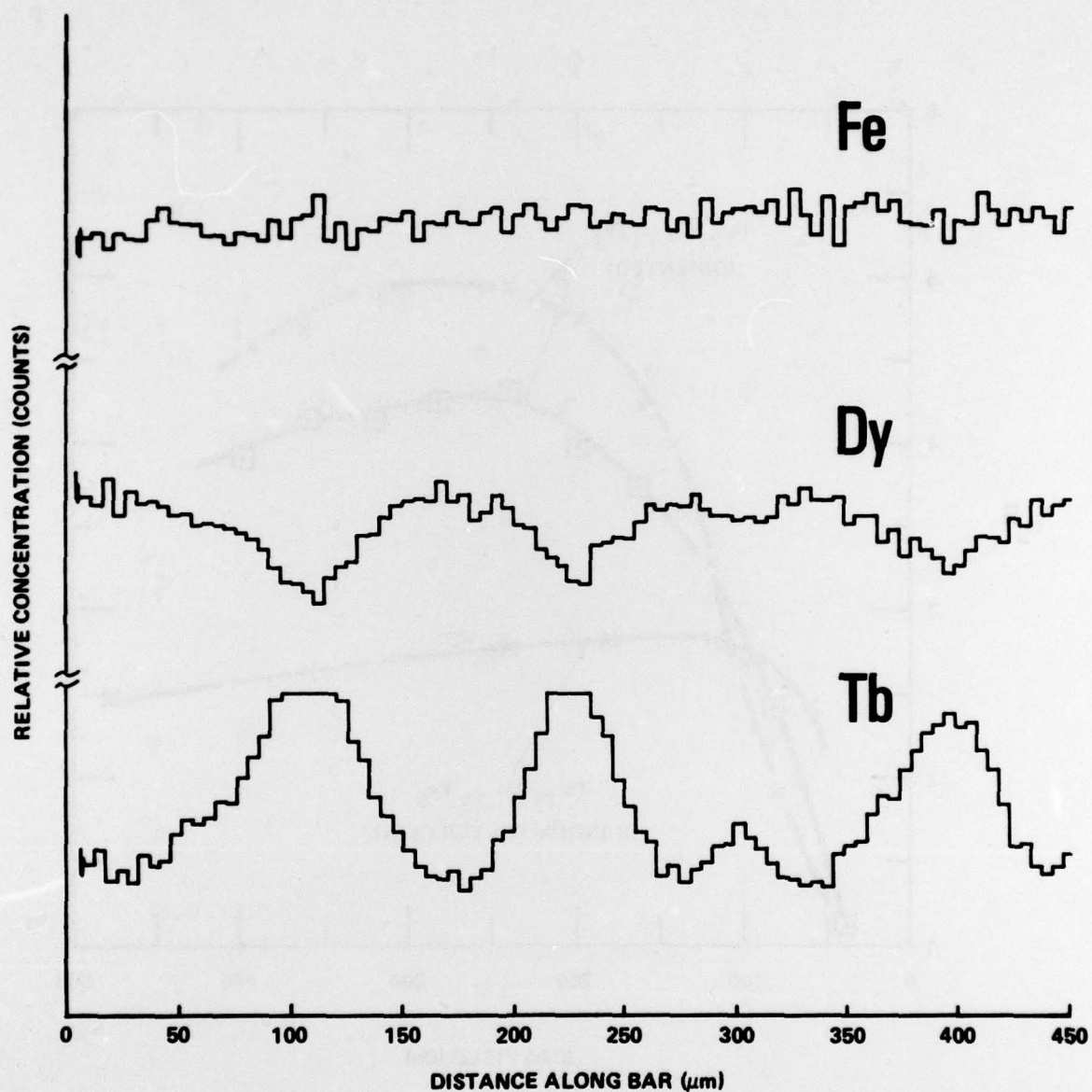


FIGURE 3 FLUCTUATION IN Tb/Dy RATIO ALONG THE SAMPLE AS MEASURED BY AN ELECTRON MICROPROBE. THE Tb & Dy ARE MONITORED BY INDEPENDENT ANALYSERS. NOTE THAT THE Tb INCREASES IN CONCENTRATION WHEN THE Dy DECREASES. THE FLUCTUATION CORRESPONDS TO A COMPOSITION OF $\text{Tb}_{.27 \pm .025} \text{Dy}_{.73 \pm .025} \text{Fe}_2$.

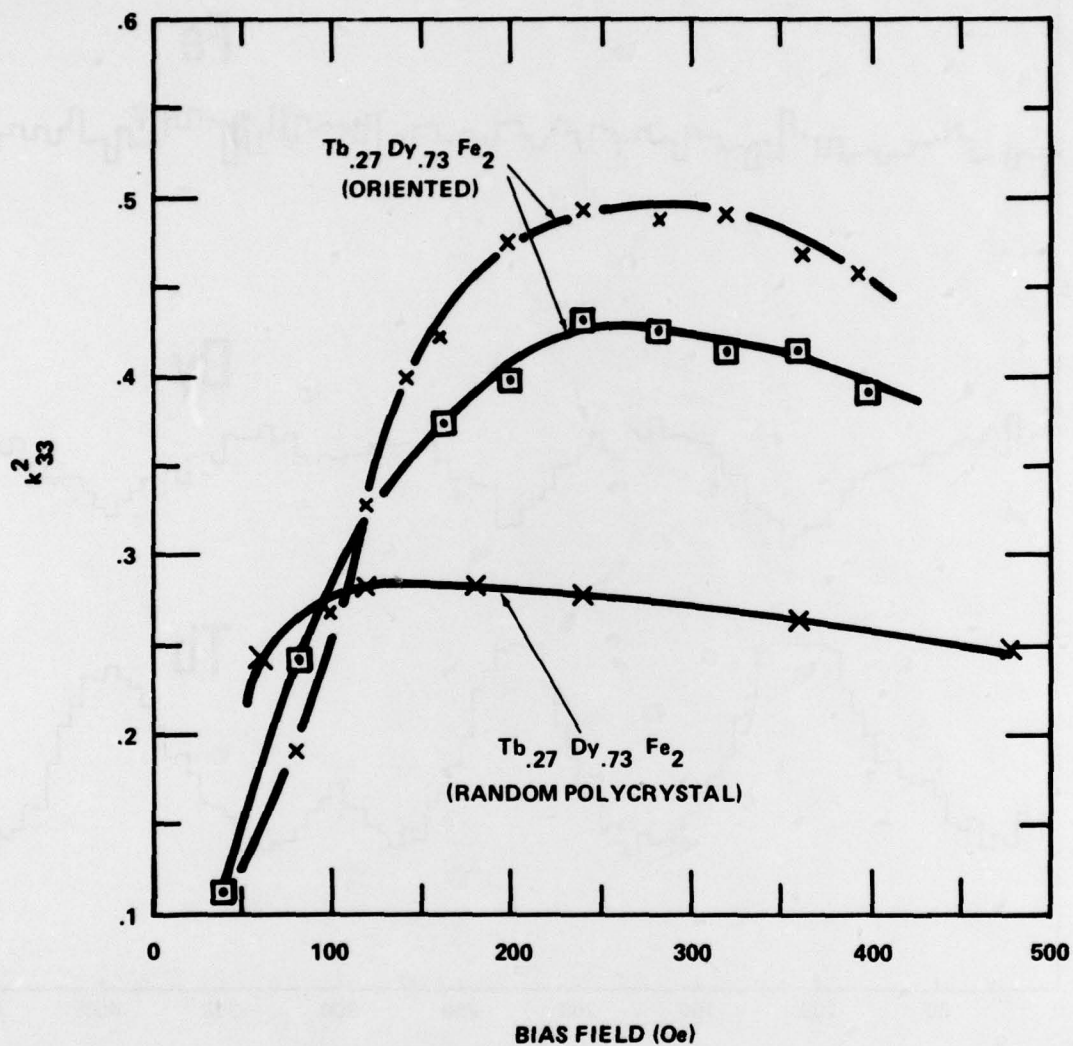


FIGURE 4 A COMPARISON OF THE MAGNETOMECHANICAL COUPLING FACTOR AS A FUNCTION OF APPLIED BIAS FOR PARTIALLY GRAIN-ORIENTED Tb_{0.27}Dy_{0.73}Fe₂ AND A RANDOM POLYCRYSTAL OF Tb_{0.27}Dy_{0.73}Fe₂.

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