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MAGNETIZATION AND MAGNETIC ANISOTROPY OF TBFE2, DYFE2, TB0.27DY--ETC(U)
JAN 79 A E CLARK, R ABBUNDI, W R GILLMOR
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MAGNETIZATION AND MAGNETIC ANISOTROPY OF
TbFe₂^M, DyFe₂^M, Tb_{0.27}Dy_{0.73}Fe₂^M AND TmFe₂^M

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BY A. E. CLARK, R. ABBUNDI W. R. GILLMOR
RESEARCH AND TECHNOLOGY DEPARTMENT

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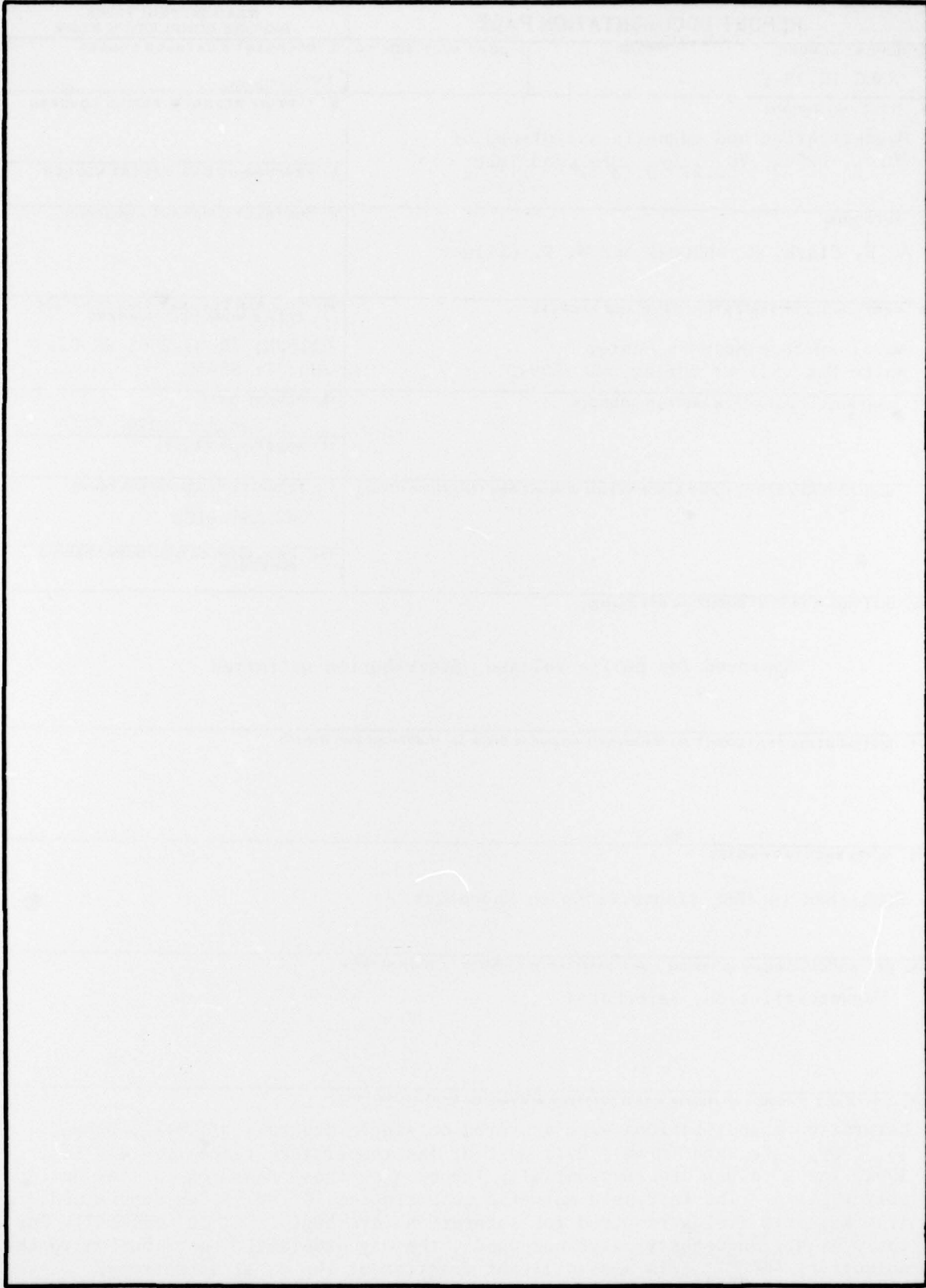
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SUMMARY

In this report are presented saturation magnetization measurements on single crystal TbFe_2 , DyFe_2 , $\text{Tb}_{0.27}\text{Dy}_{0.73}\text{Fe}_2$ and TmFe_2 . These compounds are strongly magnetic, highly magnetostrictive, and possess a wide range of cubic magnetocrystalline anisotropy, K_1 - ranging from the largest known ($5 \times 10^8 \text{ erg/cm}^3$) for TbFe_2 to a small value ($<10^6 \text{ erg/cm}^3$) for the compensated $\text{Tb}_{0.27}\text{Dy}_{0.73}\text{Fe}_2$ compound. From the magnetization vs. field measurements along the principal axes, the magnetic anisotropy was calculated as a function of temperature for DyFe_2 and TmFe_2 . It was discovered that, even in the noncompensating compounds, the magnetoelastic contribution to the magnetic anisotropy ($\Delta K_1 = -\frac{1}{2}C_{44} \lambda_{111}^2$) is a significant portion (~ 20%) of the intrinsic anisotropy constant, K_1 . For the case of the compensated $\text{Tb}_{0.27}\text{Dy}_{0.73}\text{Fe}_2$ compound, the temperature dependence of the magnetomechanical coupling factor was correlated to the temperature dependence of the anisotropy field.

This study was carried out in the Solid State Branch of the Radiation Division as part of the research program on magnetostrictive materials. The research was sponsored by the Office of Naval Research (PO 4-0081, NR 039-110) and the NSWC Independent Research Funds.

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MAGNETIZATION AND MAGNETIC ANISOTROPY OF $TbFe_2$, $DyFe_2$, $Tb_{0.27}Dy_{0.73}Fe_2$ and $TmFe_2$ †

A. E. Clark, R. Abbundi and W. R. Gillmor *

ABSTRACT

Saturation magnetizations were measured on single crystals of $TbFe_2$, $DyFe_2$, $Tb_{0.27}Dy_{0.73}Fe_2$, and $TmFe_2$. Over most of the temperature range from 4 K to 300 K, these values are substantially larger than those measured earlier on polycrystals. The intrinsic magnetic anisotropies, $K_1(0)$'s, as determined from magnetic fields required for saturation, are huge ($\sim 5 \times 10^6$ erg/cm³). For these highly magnetostrictive compounds, the magnetoelastic contribution to the anisotropy $-\frac{1}{2}c_{44}\lambda_{111}$ is a significant fraction of the total anisotropy.

INTRODUCTION

The rare-earth-Fe₂ Laves phase compounds possess the largest known cubic anisotropies. At low temperatures, anisotropies of $DyFe_2$ and $ErFe_2$ exceed 5×10^6 erg/cm³; at room temperature, anisotropies of $TbFe_2$ and $DyFe_2$ exceed 2×10^7 erg/cm³. The RFe₂ compounds also possess huge magnetostrictions, which also extend to room temperature for $TbFe_2$ and $SmFe_2$. In this paper we report magnetization measurements on single crystals of $TbFe_2$, $DyFe_2$, $Tb_{0.27}Dy_{0.73}Fe_2$ and $TmFe_2$. From measurements along the hard magnetic directions, magnetic anisotropy energies are calculated. In magnetostrictive materials, the magnetic anisotropy depends upon the state of strain—a clamped sample possessing the conventional intrinsic anisotropy, described e.g. by K_1 ; a sample allowed to freely strain possessing an additional contribution ΔK_1 arising from the magnetoelastic energy. Even though a large number of materials possess relatively large magnetostrictions, only in a few materials, e.g. Tb_3Fe_{10} , $\Delta K_1 > K_1$. As part of this paper we report the magnetic properties of $Tb_{0.27}Dy_{0.73}Fe_2$. This pseudobinary compound, synthesized from highly magnetostrictive $TbFe_2$ and $DyFe_2$, has the proper rare earth ratio to compensate the total magnetic anisotropy constant (K_1^{tot}) near room temperature while at the same time maintaining a large magnetostriction. Hence near room temperature, ΔK_1 and the intrinsic contribution K_1^{int} individually are large, but their sum $K_1^{tot} = K_1^{int} + \Delta K_1 = 0$.

MAGNETIZATION AND MAGNETIC ANISOTROPY

The magnetizations of polycrystal $TbFe_2$, $DyFe_2$ and $TmFe_2$ have been known for some time.^{4,5,6} Measurements on single crystal $DyFe_2$ at low temperatures were reported earlier.⁷ Because of the huge magnetic anisotropies of the RFe₂ compounds, magnetization measurements made on polycrystals using conventional laboratory fields do not yield the true saturation magnetic moments. Fields greater than 100 kOe are necessary to saturate the heavy rare earth compounds $TbFe_2$ through $TmFe_2$.

Single crystals of $TbFe_2$, $DyFe_2$, $Tb_{0.27}Dy_{0.73}Fe_2$ and $TmFe_2$ were grown by O. D. McMasters by horizontal zoning and Czochralski techniques. The magnetic moments of these crystals were measured from 4 K to above their Curie temperatures (except for $TbFe_2$) utilizing a vibrating sample magnetometer. In Fig. (1-4) the magnetic moments measured along their respective easy axes are compared with the earlier values of Burzo measured on polycrystals. In all cases, the moments of the single crystals are substantially higher. In the heavy RFe₂ series, the rare earth-iron exchange energy de-

creases with increasing rare earth atomic number approximately according to $[(g-1)J(J+1)]^{1/2}$.⁸ Thus for $TmFe_2$, with a weak R-Fe exchange, the rare earth sublattice magnetization decreases rapidly with increasing temperature, falling below that of the iron sublattice near 235 K. For $TbFe_2$ and $DyFe_2$, the exchange interaction is much stronger, leading to much higher moments, anisotropies and magnetostrictions at room temperature. In Table I we list the saturation values of the moment at 0 K and 300 K, the theoretical densities and the iron moments calculated assuming $\mu_R = gJ\mu_B$.

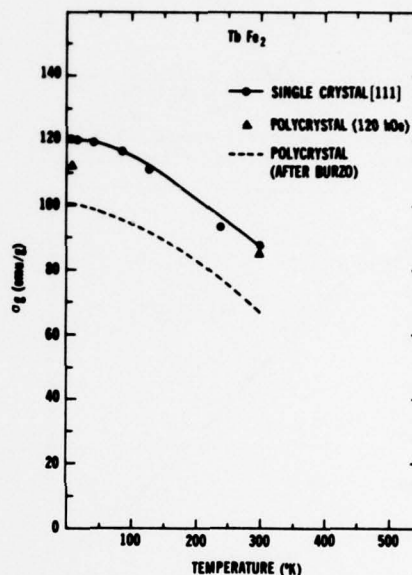


Fig. 1 Spontaneous magnetic moment of a $TbFe_2$ single crystal as a function of temperature.²

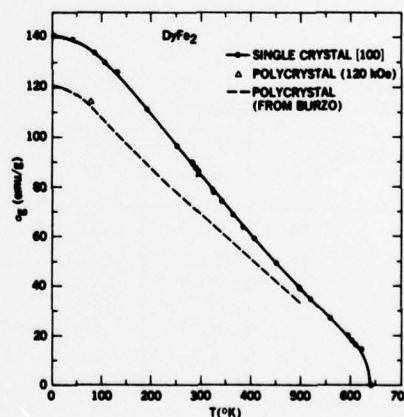


Fig. 2 Spontaneous magnetic moment of a $DyFe_2$ single crystal as a function of temperature.

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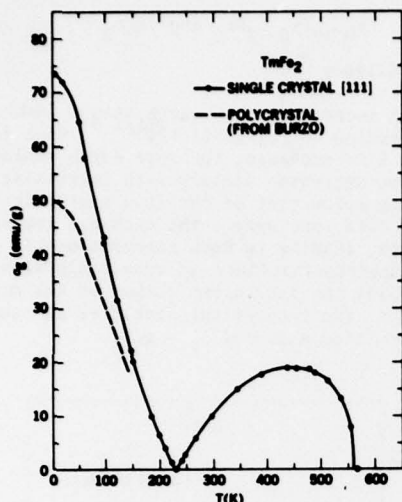


Fig. 3 Spontaneous magnetic moment of a TmFe_2 single crystal as a function of temperature.

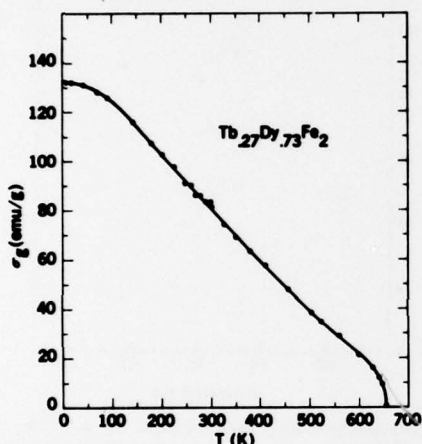


Fig. 4 Spontaneous magnetic moment of a $\text{Tb}_{0.27}\text{Dy}_{0.73}\text{Fe}_2$ single crystal as a function of temperature.

TABLE I. MAGNETIC MOMENTS, THEORETICAL DENSITIES AND CURIE TEMPERATURES OF RFe_2

RFe_2	σ (emu/gm) 0 K*	300 K	ρ_t gm/cm ³	$n_{b\text{Fe}}^{(d)}$ 0 K	$n_{b\text{Fe}}^{(e)}$ 0 K	T_C K
TbFe_2	120	88	9.06	1.60	1.75	697,711 ^a
DyFe_2	140	87	9.28	1.57	1.60	635
HoFe_2 ^b	135	64	9.44	1.65	1.55	597,612 ^a
ErFe_2 ^c	116	29	9.62	1.61	1.60	590,597 ^a
TmFe_2	74	10	9.79	1.64	1.55	560

* Extrapolated

- Taken from polycrystal data of K. H. J. Buschow and R. P. Van Staple, *J. de Physique* **32**, C1-672 (1971), and E. Burzo, *Z. Angew. Physik* **32**, T27 (1971).
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- See A. E. Clark, *AIP Conf. Proc.* **18**, 1015 (1974).
- Magnetic moment per iron atom in Bohr magnetons calculated from single crystal data assuming $n_{b\text{R}} = gJ$.
- Magnetic moment per iron atom in Bohr magnetons determined from Mössbauer spectra. W. E. Wallace, *Prog. Rare Earth Sci. Tech.* **3**, 1 (1968).

For DyFe_2 , TmFe_2 and $\text{Tb}_{0.27}\text{Dy}_{0.73}\text{Fe}_2$, the magnetic anisotropy constant K_1 was calculated from the fields required for magnetic saturation along the [100], [110] and [111] directions. The anisotropy constants K_1 for DyFe_2 and TmFe_2 are plotted in Fig. (5). Except at the very low temperatures, the anisotropy of DyFe_2 is much larger than that of TmFe_2 . The single-ion theory has been successful in fitting the temperature dependence of the magnetostriction of the RFe_2 compounds as well as the anisotropy of ErFe_2 . Here we apply that theory to DyFe_2 and TmFe_2 . It is important to realize that even in the non-compensating RFe_2 compounds, because of the high magnetostrictions, ΔK_1 is not negligible. Taking the magnetoelastic contribution to be $\Delta K_1 = -9c_{44}\lambda_{111}^2/2$ and utilizing the single crystal magnetostriction data published earlier, we arrive at the values shown in Table II. The elastic constant c_{44} is taken to be 4.87×10^{11} dynes/cm² for all compounds. From the measured values of K_1^{int} , the intrinsic anisotropy constants and $\Delta K_1/K_1^{\text{int}}$ are calculated. According to single-ion theories, where J is large and the magnetically induced levels are assumed to be nearly evenly spaced, the expression for the temperature dependence of the anisotropy is given by:

$$K_1^{\text{int}}(T) = K_1^{\text{int}}(0) \hat{I}_{9/2}[\chi^{-1}(m_R(T))]$$

and

$$\Delta K_1(T) = \Delta K_1(0) \hat{I}_{5/2}^2[\chi^{-1}(m_R(T))].$$

Here the small temperature dependence of c_{44} is neglected. $\hat{I}_{\ell+1/2}$ is the hyperbolic Bessel function of order $\ell+1/2$ normalized to one at $T = 0$ K; χ^{-1} is the inverse Langevin function and m_R is the reduced rare earth sublattice magnetization. In Fig. (5) we show by the solid line the calculated anisotropy based upon $K_1^{\text{int}}(0)$ of 4.7 and -3.8 ($\times 10^8$ erg/cm³) for DyFe_2 and TmFe_2 , respectively. The values of ΔK_1 of -2.4 and -2.7 ($\times 10^8$ erg/cm³) for DyFe_2 and TmFe_2 were taken from magnetostriction data. We find that with one adjustable constant per compound, $K_1^{\text{int}}(0)$, the fit to the data is within 20% over the temperature range of the experiments. For DyFe_2 , the Dy sublattice magnetization was taken from Mössbauer spectra by Bowden et al.¹² For TmFe_2 , the sublattice magnetization was calculated from the total magnetization reported in this paper less that of the iron sublattice inferred from Bowden et al.¹²

TABLE II. MAGNETOELASTIC AND INTRINSIC CONTRIBUTION TO THE MAGNETIC ANISOTROPY (ergs/cm³).

RFe_2	$\Delta K_1(300)^*$ $\times 10^{-4}$	$K_1^{\text{int}}(300)$ $\times 10^{-4}$	$\frac{\Delta K_1(300)}{K_1^{\text{int}}(300)}$
SmFe_2	-970	---	---
TbFe_2	-1330	-6300	.21
DyFe_2	-350	2450	-.14
ErFe_2	-20	-310	.06
TmFe_2	-9.7	-43	.22

* Calculated from λ_{111} values taken from A. E. Clark, J. R. Cullen, O. D. McMasters and E. R. Callen, *AIP Conf. Proc.* **29**, 192 (1976) and R. Abbundi and A. E. Clark, *J. Appl. Phys.* **49**, 1969 (1978).

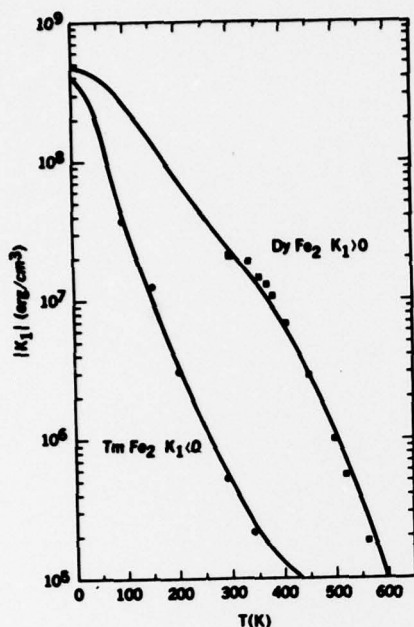


Fig. 5 Anisotropy constant K_1 for single crystals DyFe_2 and TmFe_2 as a function of temperature. The solid curve represents the anisotropy calculated from single-ion theory.

The anisotropy constant K_1 of $\text{Tb}_{0.27}\text{Dy}_{0.73}\text{Fe}_2$ equals zero near room temperature. K_2 remains negative. The anisotropy is DyFe_2 -like ([100] easy) at low temperatures and TbFe_2 -like ([111] easy) at high temperatures. The fields required for magnetic saturation along the principal directions are shown in Fig. (6). $H_{111} = H_{100}$ at 283 K where $H_{110} = 1.5$ kOe. The anisotropy fields drop rather sharply from very high values at low temperatures. On the high temperature side, the slopes are not so steep, with the anisotropy constant K_1 reaching a peak value of -1.5×10^6 erg/cm³ at 380 K. The rapid drop of the anisotropy constants of both binary TbFe_2 and DyFe_2 over this temperature range accounts for this rather low peak value.

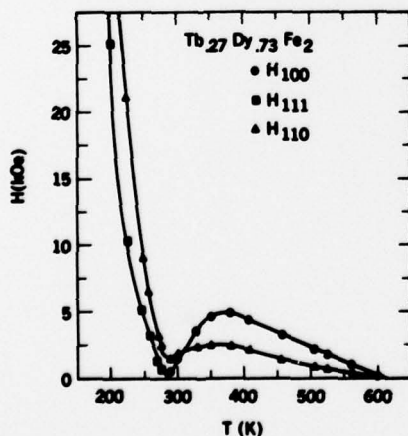


Fig. 6 Fields required for magnetic saturation along the principal crystallographic directions in single crystal $\text{Tb}_{0.27}\text{Dy}_{0.73}\text{Fe}_2$ as a function of temperature.

Magnetization rotation and domain wall motion can be effected at low applied magnetic fields when the magnetic anisotropy is low. The maximum energy transformed from the magnetic to the elastic systems is $\frac{1}{2} c_{44} \lambda_{111}^2$. Defining $f = \frac{1}{2} c_{44} \lambda_{111}^2 / M H_{110}$ as a figure of merit of magnetostrictive transduction, we calculate for TbFe_2 , $f = 0.016$; and for DyFe_2 , $f = 0.015$. For the pseudo-binary $\text{Tb}_{0.27}\text{Dy}_{0.73}\text{Fe}_2$ compound, $f = 0.5$ with a peak near 283 K. In Fig. (7), we compare $\frac{1}{2} c_{44} \lambda_{111}^2 / M H_{110}$ to the relative magnetomechanical coupling factor k for temperatures near anisotropy compensation (maximum $k = 0.6$). Note that the slightly different pseudobinary composition ($\text{Tb}_{0.26}\text{Dy}_{0.74}\text{Fe}_2$) used for the magnetomechanical coupling measurements shifts the compensating temperature to a little higher temperature. On the low temperature side of the anisotropy compensation, there is a rapid roll-off in both k and $\frac{1}{2} c_{44} \lambda_{111}^2 / M H_{110}$. At high temperature, the roll-off is not as severe, and in fact for k , rather small. The major source of the magnetostrains and the high coupling probably arises from domain wall motion rather than magnetization rotation. Above the anisotropy compensation temperature, the anisotropy remains low enough to prevent domain wall pinning at defect sites.

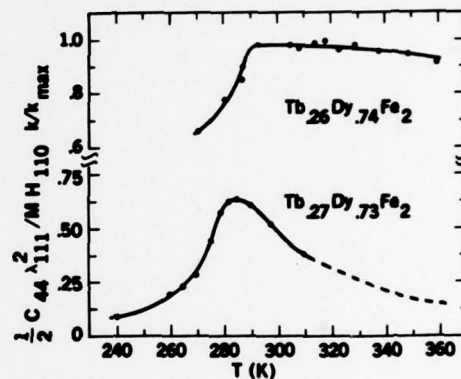


Fig. 7 Comparison of the relative magnetomechanical coupling factor for $\text{Tb}_{0.26}\text{Dy}_{0.74}\text{Fe}_2$ with $\frac{1}{2} c_{44} \lambda_{111}^2 / M H_{110}$ (figure of merit for magnetostrictive transduction) for single crystal $\text{Tb}_{0.27}\text{Dy}_{0.73}\text{Fe}_2$ as a function of temperature.

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