

Giant Room-Temperature Magnetostrictions in TbFe₂ and DyFe₂[†]

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Huge magnetically induced strains (~0.2%) were observed in cubic TbFe₂ at room temperature. An anomalous "positive" temperature dependence for the magnetostriction is found for DyFe₂, yielding a peak in the magnetostriction exceeding 600 × 10⁻⁶ above room temperature. The source of the magnetostriction is the large strain-dependent anisotropy of the rare-earth ion situated at the cubic sites in the RFe₂ lattice. The magnetoelastic energy of the rare-earth ions in these compounds is estimated to be 2 to 5 times larger than that of the rare-earth elements themselves.

We wish to report the discovery of the largest room temperature magnetostrictions ever observed. These occur in the Laves-phase rare-earth-iron compounds TbFe₂ and DyFe₂. Previously all "giant" (i. e., >10⁻³) magnetostrictions were restricted to cryogenic temperatures. The rare-earth elements, for example, have the largest known magnetostrictions near 0°K,^{1,2} but are not magnetic at room temperature. The iron titanates have a similar behavior.³ Because of this huge magnetostriction, TbFe₂ and similar compounds may have considerable technological importance.

The origin of the magnetostriction is the large strain-dependent anisotropy of the rare-earth ions situated at cubic sites in the RFe₂ (R = rare earth) lattice. It remains large at room ambients because of the large rare-earth-iron exchange interaction which aligns the rare-earth spins even at elevated temperatures. The magnitude of this interaction is unique to the iron compounds.⁴ In the cobalt and nickel Laves-phase terbium compounds, the Curie temperatures are depressed to -35 and -227°C, respectively,^{5,6} whereas for the iron compound T_C ≅ 430°C.^{7,8}

We have measured the magnetostriction of arc-cast polycrystalline YFe₂, TbFe₂, and DyFe₂ from 78°K to room temperature. Standard strain-gage techniques were used.¹ The room-temperature magnetostrictions of TbFe₂ and DyFe₂ are shown in Fig. 1. The magnetostriction λ_s of YFe₂ is only ~2 × 10⁻⁶, indicating the negligible contribution of the iron ions to the magnetostriction. A comparison of these strains with those of typical polycrystalline metal and ferrites is shown in Table I. The magnetically induced strains in TbFe₂ are over 10 times the largest reported values and 100 times typical magnetostrictive strains. In the interpretation of Table I it is important to note that these are *polycrystalline* values. It is quite possible that higher values of magnetostriction will be found in single crystals of TbFe₂ and DyFe₂.

In some respects, the magnetostriction of TbFe₂ and DyFe₂ appears similar. However, on closer

examination it is seen that the Dy compound is far from saturation, indicating a substantially larger magnetic anisotropy. This leads to an unusual temperature dependence of the magnetostriction. In Fig. 2 we show that at room temperature the magnetostriction (λ_{||} - λ_⊥) of DyFe₂ is increasing with increasing temperature, in contrast to the usual negative (∂λ/∂T). In fact, both the magnetostriction (600 × 10⁻⁶) and its slope (+3.5 × 10⁻⁶/°K) are huge. As the temperature decreases, a reversal in sign occurs in the magnetostriction. At 25 kOe, this temperature is 112°K. On the other hand, at the same field, the magnetostriction of TbFe₂ is near saturation at all temperatures. A plot of Δl/l vs 1/H for H → ∞ at each temperature

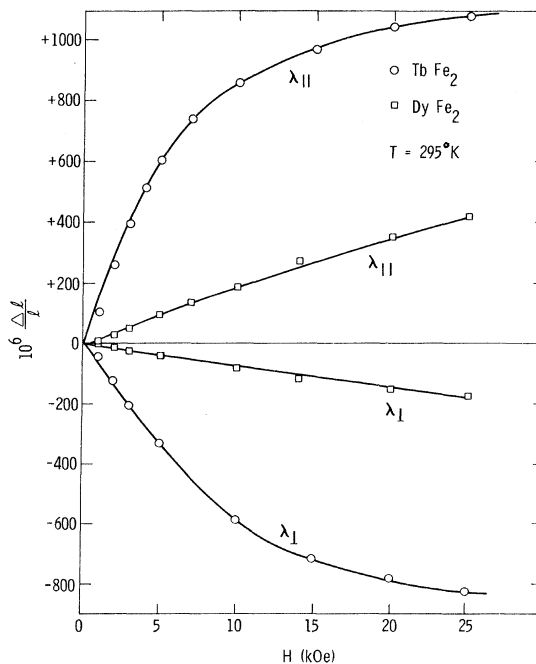


FIG. 1. Room-temperature magnetostriction of TbFe₂ and DyFe₂. λ_{||} and λ_⊥ denote magnetostrictions parallel and perpendicular to the applied field directions.

TABLE I. Typical saturation magnetostrictions of polycrystalline metals and ferrites at room temperature.^a

	$\lambda_s \times 10^6$		$\lambda_s \times 10^6$
Fe	-9 ^b	TbFe ₂	>1200
Ni	-35 ^b	Fe ₃ O ₄	40 ^c
Co	-62 ^b	CoFe ₂ O ₄	-110 ^c
Tb, Dy	~0 ^d	NiFe ₂ O ₄	-26 ^c
YFe ₂	2	Y ₃ Fe ₅ O ₁₂	-2 ^e

^aAt saturation: $(\Delta l/l) = \lambda_{||} - \lambda_{\perp} = \frac{2}{3}\lambda_s$.

^bSee R. M. Bozorth, *Ferromagnetism* (Van Nostrand, New York, 1951).

^cSee J. Smit and H. P. J. Wijn, *Ferrites* (Wiley, New York, 1959).

^dNonmagnetic at room temperature.

^eA. Clark, B. DeSavage, W. Coleman, E. Callen, and H. Callen, *J. Appl. Phys.* **34**, 1296 (1963).

is linear above 15 kOe. Saturation strains obtained in this way are indicated by the dotted line on Fig. 2.

Callen and Callen⁹ and Callen and Shtrikman¹⁰ have shown that the temperature dependence of the magnetostriction follows the well known $m^{l(l+1)/2}$ power law for magnetic anisotropy¹¹ at low temperatures, and the normalized hyperbolic Bessel function $\hat{I}_{l+1/2}(\mathcal{L}^{-1}(m))$ in the molecular-field approximation at all temperatures. Here m is the reduced magnetic moment, $\mathcal{L}^{-1}(m)$ is the inverse Langevin function, and l is the order of the spin operator in the magnetoelastic Hamiltonian (for lowest order,

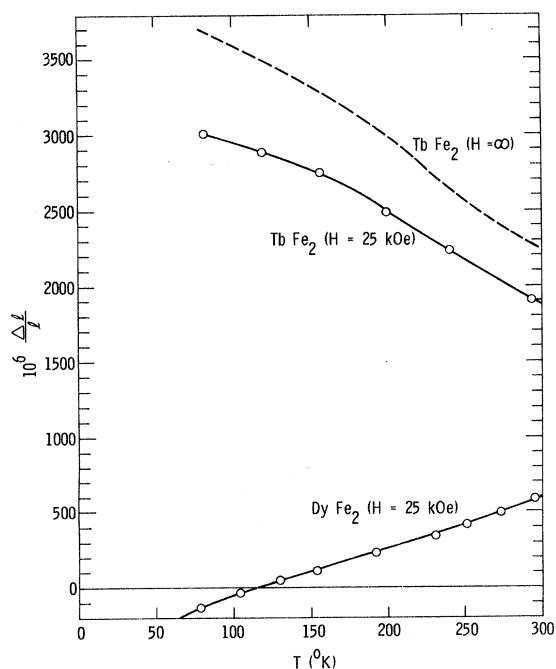


FIG. 2. Magnetostriction $\lambda_{||} - \lambda_{\perp}$ of TbFe₂ and DyFe₂. (At saturation $\Delta l/l = \frac{2}{3}\lambda_s$ for an isotropic polycrystal.)

$l=2$.) At low temperatures the hyperbolic Bessel function reduces to the power law. In Fig. 3, we plot the saturation values of the magnetostriction for TbFe₂ vs m_r^3 , where m_r is the reduced magnetization of the Tb sublattice calculated from the magnetization measurements of Burzo.¹² The solid line is the theoretical curve. Note the good agreement. The sole source of the magnetostriction is the rare-earth ions, and its extrapolated value at 0°K is 4080×10^{-6} . This is of the same magnitude as the magnetostriction of the polycrystalline rare-earth elements. The density of the rare-earth ions in Tb metal, however, is 60% larger than in TbFe₂. Thus, assuming equal magnetostrictions for elemental terbium and TbFe₂ at 0°K, the contribution to the magnetoelastic energy of the Tb ions in TbFe₂ is appreciably larger than that in terbium itself. (The presently known largest magnetoelastic energies are found in hexagonal terbium and dysprosium.) Because the rare-earth metals are also soft and have very low elastic moduli (whereas TbFe₂ is hard), we estimate that the magnetoelastic energy of the Tb ions on the cubic sites of TbFe₂ is 2 to 5 times larger than that of Tb ions on the hexagonal sites of the element.

The evaluation of the 0°K magnetostriction of TbFe₂ makes it possible to estimate the saturation magnetostriction in the other heavy rare-earth RFe₂ compounds. We utilize the theory of Tsuya

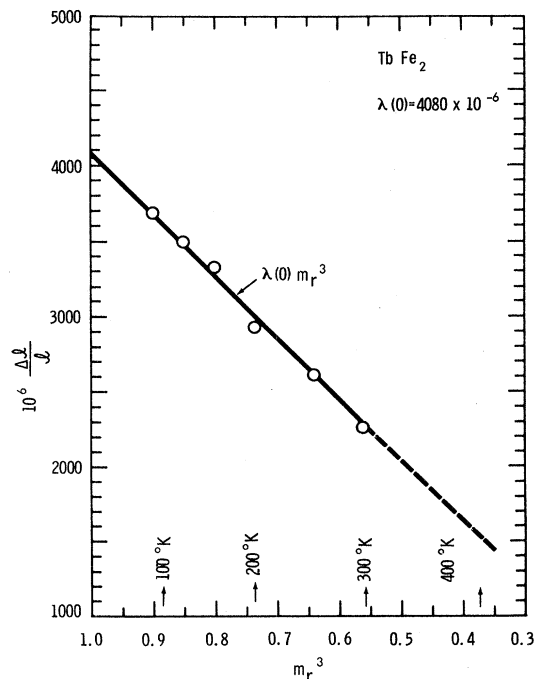


FIG. 3. Dependence of the magnetostriction of TbFe₂ on the cube of the reduced magnetization of the Tb sublattice. See text.

TABLE II. Theoretical saturation values of $\lambda_{\parallel} - \lambda_{\perp}$ for RFe₂ compounds.

T(°K)	$(\lambda_{\parallel} - \lambda_{\perp}) \times 10^6$				
	TbFe ₂	DyFe ₂	HoFe ₂	ErFe ₂	TmFe ₂
0	4080	3920	1490	-1450	-3450
300	2260	1610	520	-320	-520

*et al.*¹³ For simplicity, we assume that the angular momentum J of the rare-earth ion is a good quantum number and that the elastic moduli of the compounds are equal. Letting α denote the Stevens equivalent-operator coefficient,¹⁴ and $\langle r_f^2 \rangle$ the average square of the radius of the 4f electrons, the ratio of the magnetostriction coefficients for compounds i and j at $T = 0^\circ\text{K}$ is given by

$$\frac{\lambda_i(0)}{\lambda_j(0)} = \frac{\alpha_i J_i (J_i - \frac{1}{2}) \langle r_f^2 \rangle_i}{\alpha_j J_j (J_j - \frac{1}{2}) \langle r_f^2 \rangle_j}.$$

The calculated coefficients for the compounds DyFe₂ through TmFe₂ are compared to those of TbFe₂ at 0°K and 300°K in Table II. The values of $\langle r_f^2 \rangle$ were taken from Freeman and Watson.¹⁵ The magnetostriction at 300°K was determined from the temperature dependence of the magnetization,¹² assuming that $\lambda(T) = \lambda(0) \hat{I}_{5/2}(\mathcal{L}^{-1}(m_r))$ and that the iron-sublattice magnetization is temperature independent below

300°K . All coefficients exceed 10^{-4} , even at room temperature. The apparent discrepancy between the measured magnetostriction of 600×10^{-6} in DyFe₂ at 25 kOe and its calculated room temperature value of 1610×10^{-6} is explained by the fact that our measurements were taken below saturation. TbFe₂ has the largest positive magnetostriction coefficient at both absolute zero and room temperature. TmFe₂ has the largest negative coefficient. At 0°K , the magnitudes of the magnetostriction of TmFe₂ and TbFe₂ are almost equal. However, the reduced magnetization m_r of the heavy rare-earth compounds at room temperature decreases with increasing atomic number; thus the ratio of the 0°K magnetostriction to the room-temperature value is much larger for the heavier rare-earth compounds.

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Note added in proof. The huge magnetostrictions reported here were made on arc-cast polycrystalline samples. We find still larger values for these materials after annealing. For example, the highest room-temperature values for $\lambda_{\parallel} - \lambda_{\perp}$ of TbFe₂ are 2630×10^{-6} at 25 kOe and 2940×10^{-6} at saturation. The 0°K extrapolated value for annealed TbFe₂ is 5100×10^{-6} , which is to be compared to the value of 4080×10^{-6} that appears in Table II for the unannealed sample.

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