MAGNETIZATION AND MAGNETIC ANISOTROPY OF TbFe₂, DyFe₂, Tb₀.₂₇Dy₀.₇₃Fe₂ and TmFe₂

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

ABSTRACT

Saturation magnetizations were measured on single crystals of TbFe₂, DyFe₂, Tb₀.₂₇Dy₀.₇₃Fe₂, and TmFe₂. 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, K₁(0)'s, as determined from magnetic fields required for saturation, are huge (~5 x 10⁶ erg/cm³). For these highly magnetostrictive compounds, the magnetoelastic contribution to the anisotropy K₂ ≈ 4x₁₀⁶ erg/cm³ 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 DyFe₂ and ErFe₂ exceed 5 x 10⁶ erg/cm³; at room temperature, anisotropies of TbFe₂ and DyFe₂ exceed 2 x 10⁶ erg/cm³. The RFe₂ compounds also possess huge magnetostrictions, which also extend to room temperature for TbFe₂ and SmFe₂. In this paper we report magnetization measurements on single crystals of TbFe₂, DyFe₂, and TmFe₂. 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 K₁, a sample allowed to freely strain possessing an additional contribution ΔK₁ arising from the magnetoelastic energy. Even though a large number of materials possess relatively large magnetostrictions, only in a few materials, e.g. TbFe₂, DyFe₂, and TmFe₂, has the proper rare earth ratio Fe₂/R₂ compensate the total magnetic anisotropy constant (K₁ + ΔK₁) near room temperature while at the same time maintaining a large magnetostriction. Hence near room temperature, ΔK₁ and the intrinsic contribution K₁tot individually are large, but their sum K₁tot = K₁ + ΔK₁ = 0.

MAGNETIZATION AND MAGNETIC ANISOTROPY

The magnetizations of polycrystalline TbFe₂, DyFe₂, and TmFe₂ have been known for some time. Measurements on single crystal DyFe₂ at low temperatures were reported earlier. 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 TbFe₂ through TmFe₂. Single crystals of TbFe₂, DyFe₂, and TmFe₂ were grown by O. D. McMeekers by horizontal zoning and Czochralski techniques. The magnetic moments of these crystals were measured from 4 K to above their Curie temperatures (except for TbFe₂) utilizing a vibrating sample magnetometer. In Fig. 1 the magnetic moments measured along their respective easy axes are compared with the earlier values of Burzo for polycrystals. In all cases, the moments of the single crystals are substantially higher. In the heavy RFe₂ series, the rare earth-iron exchange energy decreases with increasing rare earth atomic number approximately according to [(g-1)(J+1)]¹. Thus for TmFe₂, 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 TbFe₂ and DyFe₂, 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 νᵣ = gJν₀.
For DyFe₂, TbFe₂, and Tb₀.₂₇Dy₀.₇₃Fe₂, the magnetic anisotropy constant ₖ₁ was calculated from the fields required for magnetic saturation along the [100], [110] and [111] directions. The anisotropy constants ₖ₁ for DyFe₂ and TbFe₂ are plotted in Fig. (5). Except at the very low temperatures, the anisotropy of DyFe₂ is much larger than that of TbFe₂. The single-ion theory has been successful in fitting the temperature dependence of the magnetostriction of the RFe₂ compounds as well as the anisotropy of ErFe₂. Here we apply that theory to DyFe₂ and TbFe₂. It is important to realize that even in the non-compensating RFe₂ compounds, because of the high magnetostrictions, ₄₆₅₉ is not negligible.

Taking the magnetoelastic contribution to be ₄₆₅₉ = ₇₉₄₄ λ₁₁₁₂ and utilizing the single crystal magnetostriction data published earlier, we arrive at the values shown in Table II. The elastic constant ₄₆₅₉ is taken to be 4.87 x 10⁻¹³ dynes/cm² for all compounds. From the measured values of ₖ₁, the intrinsic anisotropy constants and ₄₆₅₉ are calculated. According to single-ion theories, where ₗ 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:

ₖ₁(₉₄₃₅) = ₖ₁(₉₄₃₅₉) [₄₆₅₉⁻¹(₉₄₅₉₉(T))] and

₄₆₅₉(₉₄₃₅) = ₄₆₅₉(₉₄₃₅₉) [₄₆₅₉⁻¹(₉₄₅₉₉(T))].

Here the small temperature dependence of ₄₆₅₉ is neglected. ₄₆₅₉ is the hyperbolic Bessel function of order ₄₆₅₉ normalized to one at ₉₄₅₉₉ = 0 K; ₄₆₅₉ is the inverse Langevin function and ₉₄₅₉₉ is the reduced rare earth sublattice magnetization. In Fig. (5) we show by the solid line the calculated anisotropy based upon ₄₆₅₉(₉₄₃₅₉) of 4.7 and -3.8 (x 10⁻¹³ ergs/cm²) for DyFe₂ and TbFe₂, respectively. The values of ₄₆₅₉ of -2.4 and -2.7 (x 10⁻¹³ ergs/cm²) for DyFe₂ and TbFe₂ were taken from magnetostriction data. We find that with one adjustable constant per compound, ₄₆₅₉(₉₄₃₅₉), the fit to the data is within 20% over the temperature range of the experiments. For DyFe₂, the sublattice magnetization was taken from Mössbauer spectra by Bowden et al. For TbFe₂, the sublattice magnetization was calculated from the total magnetostriction reported in this paper less that of the iron sublattice inferred from Bowden et al.

### Table II. Magnetoelastic and Intrinsic Contribution to the Magnetic Anisotropy (ergs/cm²).

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For TbFe₂, TmFe₂ and Tb₀.₂₇Dy₀.₇₃Fe₂, the magnetic anisotropy constant ₖ₁ was calculated from the fields required for magnetic saturation along the [100], [110] and [111] directions. The anisotropy constants ₖ₁ for TbFe₂ and TmFe₂ are plotted in Fig. (5). Except at the very low temperatures, the anisotropy of DyFe₂ is much larger than that of TbFe₂. The single-ion theory has been successful in fitting the temperature dependence of the magnetostriction of the RFe₂ compounds as well as the anisotropy of ErFe₂. Here we apply that theory to DyFe₂ and TbFe₂. It is important to realize that even in the non-compensating RFe₂ compounds, because of the high magnetostrictions, ₄₆₅₉ is not negligible.

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The anisotropy constant $K_1$ of Tb$_{0.27}$Dy$_{0.73}$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). $H_{111} = H_{110}$ 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 $K_2$ reaching a peak value of $-1.5 \times 10^6$ erg/cm$^3$ 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.

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 $\frac{1}{2} c_{44} a^2 H_{111}^2$. Defining $f = \frac{\frac{1}{2} c_{44} a^2 H_{111}^2}{\langle H_{111} \rangle}$, 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 Tb$_{0.26}$Dy$_{0.74}$Fe$_2$ compound, $f = 0.5$ with a peak near 283 K. In Fig. (7), we compare $\frac{1}{2} c_{44} a^2 H_{111}^2$ to the relative magnetomechanical coupling factor $k$ for temperatures near anisotropy compensation (maximum $k = 0.6$). Note that the slightly different pseudo-binary composition (Tb$_{0.27}$Dy$_{0.73}$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 $\frac{1}{2} c_{44} a^2 H_{111}^2$. 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.

REFERENCES

7. O. D. McMasters, Iowa State University, Ames, Iowa.
9. See for example C. Kittel, Rev. Mod. Phys. 21, 541 (1949). Note $\langle H_{111} \rangle < \langle H_{110} \rangle$.