Comments and Addenda

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Spin-orientation diagram of the pseudobinary $Tb_{1-x}Dy_xFe_2$ Laves compounds

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The spin-orientation diagram of $Tb_{1-x}Dy_xFe_2$ is constructed by ^{57}Fe Mössbauer measurements and compared with single-ion calculations. The $4f$-crystal-field interaction dominates the magnetocrystalline anisotropy properties, though the presence of additional contributions to the magnetic anisotropy free energy is indicated.

Nonmajor cubic symmetry axes of easy magnetization have been established in some mixed rare-earth ternary $R^1_{1-x}R^2_xFe_2$,[1,2] as well as in binary $CeFe_2$,[3] $SmFe_2$,[4] and $HoFe_2$[5] cubic Laves compounds. These unusual axes of magnetization were found to occur over a relatively broad temperature interval, within which the easy axis of magnetization $\vec{n}$ rotates continuously over a wide range of directions, usually from one major cubic axis to another. It was believed[1,2] that the appearance of unusual, or transition regions, in the spin-orientation diagrams of the $R_{1-x}R_xFe_2$ compounds, is due either to coexisting domains of different major-axis spin alignments, or to additional, noncubic anisotropies, besides that which arises from the $4f$-crystal-field interaction. Recent calculations[6] have demonstrated that the single-ion model, applied to the rare-earth ion, accounts for unusual axes of easy magnetization within the framework of cubic symmetry. These calculations involved the diagonalization of the single-ion Hamiltonian—with the $4f$-crystal-field interaction the only source of magnetocrystalline anisotropy—for $\vec{n}$ parallel to the three major cubic axes, as well as for other directions of $\vec{n}$ which were confined to the $(1\bar{1}0)$ and (100) planes. Using the eigenvalues of the single-ion Hamiltonian, the magnetocrystalline anisotropy free energy per ion $F_R(\vec{n}, T)$ is calculated. For a ternary $R_{1-x}R_xFe_2$ compound

$$F(x, \vec{n}, T) = (1-x)F_{R^1}(\vec{n}, T) + xF_{R^2}(\vec{n}, T).$$

The easy direction of magnetization, of a given composition, at a given temperature, is that for which the free energy has its lowest value.

We report ^{57}Fe Mössbauer-effect measurements that were made in order to reexamine and supplement the previously reported[1] spin-orientation diagram of the $Tb_{1-x}Dy_xFe_2$ system. The analysis of the present Mössbauer results of $Tb_{1-x}Dy_xFe_2$ follows a least-squares-fitting procedure which is described elsewhere,[2,3] and enables determination of the direction of $\vec{n}$ relative to the cubic-cell axes. The present study was prompted by the single-ion calculations,[6] which revealed regions of unusual spin alignments in $Tb_{1-x}Dy_xFe_2$, at temperatures and compositions that have not been investigated before. Anomalous Mössbauer spectra were recorded then[1] at the [111]-[100] phase boundary above 70 K only, and were not ascribed to the $4f$-crystal-field interaction.

Figure 1 presents both the calculated and experimental spin-orientation results for the $Tb_{1-x}Dy_xFe_2$ system. The solid lines in the figure are the single-ion boundaries between [111], [100], and unusual regions (shaded area) as calculated in Ref. 6. The calculation employs crystal-field parameters $A_4 = 36a_0^{-4}$ K and $A_6/A_4 = -0.043a_0^{-2}$.[6] The exchange field, $\mu_B H_{ex} = -150$ K was taken to be temperature independent up to 300 K.[6] The calculated directions of easy magnetization within the shaded area of the spin-orientation diagram are listed in Table I. The dashed lines in Fig. 1 are the experimental boundaries between regions of different spin alignments in $Tb_{1-x}Dy_xFe_2$. These are in agreement with recent elasticity data on $Tb_{1-x}Dy_xFe_2$.[7] It is observed that unusual directions of $\vec{n}$ are obtained experimentally within only a part of the calculated unusual region (shaded area) of the

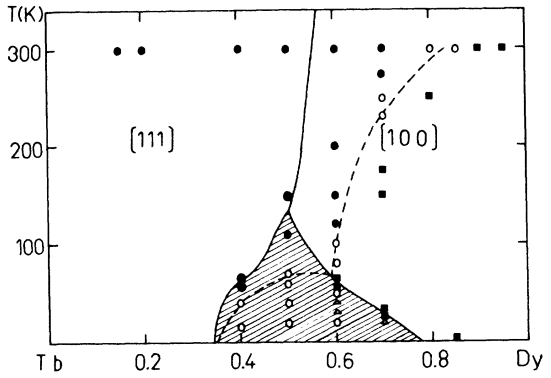

FIG. 1. Spin-orientation diagram of the $Tb_{1-x}Dy_xFe_2$ system. The solid and dashed lines are the calculated and experimental boundaries, respectively, between regions with [111], [100], and unusual (shaded area) directions of easy magnetization. Solid circles and squares correspond to [111]- and [100]-type Mössbauer spectra, respectively. Open circles and triangles correspond to $\langle uuw\rangle$- and $\langle uv0\rangle$-type spectra which, within the shaded area, were fitted using the calculated directions of easy magnetization (Table I).

spin-orientation diagram (Fig. 1). Within this part of the diagram, the Mössbauer results could be adequately fitted using the calculated spin alignments (Table I). The possibility that $\vec{n}$ in $Tb_{1-x}Dy_xFe_2$ is not confined to a cubic symmetry plane—so that four inequivalent iron sites are obtained—could not be verified using the present data. Evidently, a four-site fit, employing more adjustable parameters, should be at least as adequate as the [100] single-site, [111] two-site, and $\langle uuw\rangle$ or $\langle uv0\rangle$ three-site fits.

The single-ion calculations[6] fail to reproduce the displacement of the experimental [111]-[100] boundary towards a region of higher Dy content, nor could the narrow regime of unusual spin alignments above[1] 70 K be accounted for. Though extremely sensitive to the values of the crystal-field parameters employed,[1-6] the calculated spin-orientation diagram[6] could not be significantly improved by using different A_4 and A_6/A_4 over a wide range of values. The discrepancies between the calculated and experimental results may be attributed to sources of magnetocrystalline anisotropy other than the $4f$-crystal-field interaction. However, the reproduction of the general features of the experimental spin-orientation results justifies the present single-ion treatment.

TABLE I. Calculated directions of easy magnetization in $Tb_{1-x}Dy_xFe_2$, within the unusual region of the spin-orientation diagram (shaded area in Fig. 1). The angle θ is between $\vec{n}$, which is confined to the indicated cubic plane, and the [001] axis. Since the magnetic anisotropy Hamiltonian was calculated for a discrete set of 30 directions of $\vec{n}$, the indicated angle θ is correct to within 4°.

x	T (K)	θ (deg)	Plane	Axis
0.4	10–40	29	$(1\bar{1}0)$	$\langle uuw\rangle$
0.4	50–60	35	$(1\bar{1}0)$	$\langle uuw\rangle$
0.5	10–120	23	$(1\bar{1}0)$	$\langle uuw\rangle$
0.5	130	29	$(1\bar{1}0)$	$\langle uuw\rangle$
0.6	10–20	16	$(1\bar{1}0)$	$\langle uuw\rangle$
0.6	30–40	11	(100)	$\langle uv0\rangle$
0.6	50	8	$(1\bar{1}0)$	$\langle uuw\rangle$
0.7	10–20	6	(100)	$\langle uv0\rangle$

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[1]U. Atzmony, M. P. Dariel, E. R. Bauminger, D. Lebenbaum, I. Nowik, and S. Ofer, Phys. Rev. B 7, 4220 (1973).

[2]G. Dublon, U. Atzmony, M. P. Dariel, and H. Shaked, Phys. Rev. B 12, 4628 (1975).

[3]U. Atzmony and M. P. Dariel, Phys. Rev. B 10, 2060 (1974).

[4]A. M. van Diepen, H. W. de Wijn, and K. H. J. Buschow, Phys. Rev. B 8, 1125 (1973); U. Atzmony, M. P. Dariel, E. R. Bauminger, D. Lebenbaum, I. Nowik, and S. Ofer, in *Proceedings of the Tenth Rare Earth Conference, Carefree, Arizona, 1973*, edited by C. J. Kevane and T. Moeller (U. S. AEC, Oak Ridge, 1973), p. 605.

[5]U. Atzmony and M. P. Dariel, AIP Conf. Proc. 24, 662 (1974).

[6]U. Atzmony and M. P. Dariel, Phys. Rev. B 13, 4006 (1976).

[7]H. Klimker and M. Rosen (private communication).