Influence of the giant magnetostriction on the antiferromagnetic–ferromagnetic phase transition in terbium–yttrium alloys

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An experimental study is made of the phase transition from spiral antiferromagnetism to ferromagnetism in single crystals of terbium–yttrium alloys. Measurements are made of the critical magnetic field $H_{\rm cr}$ at which the transition occurs, the jump in the magnetization and magnetostriction at the transition, the thermal expansion, and the magnetic anisotropy in the basal plane. It is established on the basis of a thermodynamic analysis that the height of the energy barrier separating the ferromagnetic and antiferromagnetic phases in terbium–yttrium alloys decreases upon cooling of the sample and upon introduction of yttrium into terbium mainly on account of the giant spontaneous exchange magnetostriction along the spiral axis (the axis of the helical antiferromagnetic structure). It is shown that the transition from antiferromagnetism to ferromagnetism in terbium–yttrium alloys is due to the giant spontaneous magnetostrictive strains.

One of the most characteristic features of the magnetic ordering in heavy rare earth metals and their alloys is that upon cooling below a certain temperature Θ_2 the samples, as a result of a paramagnetic–antiferromagnetic phase transition, develop a periodic antiferromagnetic structure of the spiral or static-longitudinal-spin-wave type. This structure is stable upon cooling to a temperature Θ_1, where it breaks up as a result of an antiferromagnetic–ferromagnetic (AFM–FM) phase transition. While the formation of the periodic magnetic structures in rare earth metals has found theoretical explanation,[1] there is divided opinion concerning the nature of the AFM–FM transition. The breakup of the antiferromagnetic structure at $T = \Theta_1$ has been variously attributed to the magnetic anisotropy[2] and its magnetoelastic part,[3] the magnetoelastic exchange energy,[4–7] the splitting of the energy bands at "superzone" boundaries,[8,9] and a temperature dependence of the exchange parameters.[1] An extremely promising approach to the investigation of the physical mechanisms responsible for these transitions is the integrated study of the magnetic and other properties of rare-earth-metal alloys, in which it is possible to purposefully alter the basic interactions governing the stability of the magnetic structures.

In the present paper we report an experimental study of the AFM–FM phase transition in alloys of terbium with yttrium for the purpose of elucidating how this transition is influenced by the magnetic anisotropy, the magnetostriction, and the exchange interaction. To answer these questions, we measured the magnetization, magnetic anisotropy, magnetostriction, and thermal expansion for single-crystal samples of these alloys. The technology for growing the single crystals and the techniques for measuring the magnetic and other properties have been described elsewhere.[10,11]

Terbium–yttrium alloys are known to form continuous solid solutions with the hexagonal crystal lattice.[12,13] According to the neutron-diffraction data,[14] at temperatures between Θ_1 and Θ_2 in these alloys the magnetic moments of atoms belonging to the same basal plane of the lattice are parallel, while the magnetic moments of adjacent basal planes are rotated with respect to one another by a certain angle α. As a result, a spiral antiferromagnetic structure is formed, its axis coinciding with the hexagonal axis c of the crystal.

From our measurements of the temperature and field dependence of the magnetization, magnetostriction, and thermal expansion in terbium–yttrium alloys $\mathrm{Tb}_x\mathrm{Y}_{1-x}$ we determined the magnetic and magnetostriction characteristics needed for finding the various energy contributions to the thermodynamic potential.

We studied in greatest detail the magnetic phase transitions for $\mathrm{Tb}_x\mathrm{Y}_{1-x}$ alloys with terbium contents $x = 0.91$ and $x = 0.835$. For these alloys we found the magnetic transition temperatures $\Theta_1 = 170$ K, $\Theta_2 = 217$ K for $x = 0.91$ and $\Theta_1 = 120$ K, $\Theta_2 = 205$ K for $x = 0.835$.

In the temperature interval Θ_1–Θ_2 a magnetic field applied in the basal plane (perpendicular to the spiral axis) in terbium–yttrium alloys induces a transition from a spiral antiferromagnetic (SAFM) to a ferromagnetic phase when the field exceeds a certain critical value $H_{\rm cr}$. As can be seen in Fig. 1 (curve 1), $H_{\rm cr}$ increases as the temperature is reduced below Θ_2, reaches a maximum, and then decreases linearly upon further cooling. At $T = \Theta_1$, where $H_{\rm cr} = 0$, a spontaneous SAFM–FM transition occurs.

The magnetization jump ΔI observed at $H = H_{\rm cr}$ on the magnetization isotherms $I(H)$ increases monotonically upon cooling below the temperature Θ_2 (curve 2 in Fig. 1).

The magnetostriction constant $\lambda^{\gamma,2} = \lambda_\parallel - \lambda_\perp$, which is equal to the difference between the longitudinal and transverse magnetostrictions $\lambda_\parallel$ and $\lambda_\perp$, respectivity, as measured in the basal plane, increases rapidly upon cooling below Θ_2 (curve 3 in Fig. 1), leading to a strong spontaneous magnetostrictive distortion of the crystal lattice along the crystallographic directions in the basal plane.

In the temperature interval Θ_1–Θ_2 in a magnetic field $H > H_{\rm cr}$ along the spiral axis (the c axis), the samples exhibit a "spiral" magnetostriction λ_c (curve 4 in Fig. 1) which is manifested in a discontinuous increase in the lattice parameter c at the SAFM–FM transition. This λ_c increases upon

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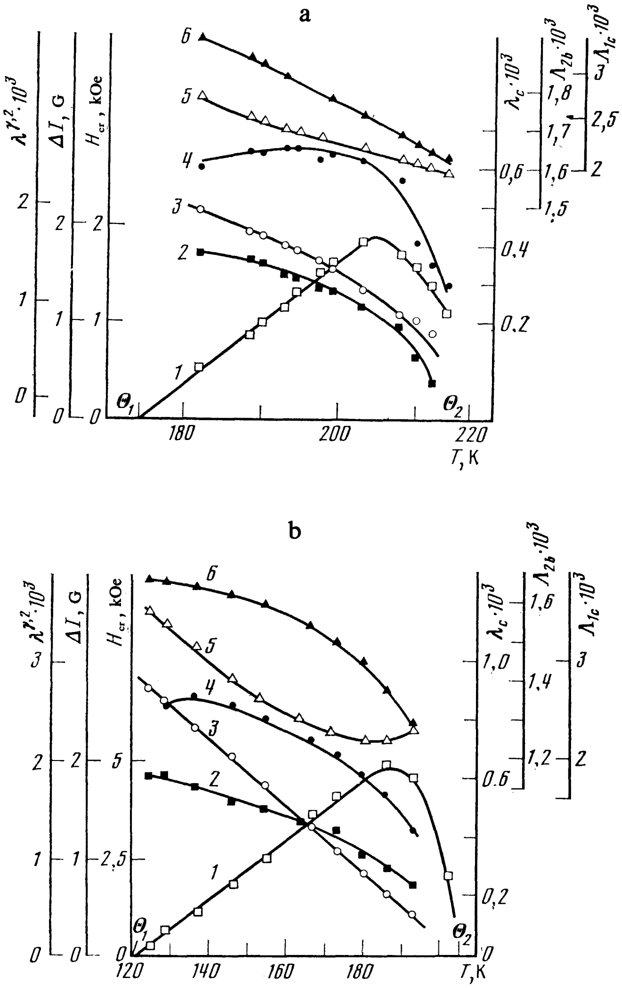

FIG. 1. Temperature dependence of the magnetic and magnetostriction parameters for the alloys $Tb_{0.91}Y_{0.09}$ (a) and $Tb_{0.835}Y_{0.165}$ (b): 1) the critical magnetic field H_{cr}, 2) the magnetization jump ΔI at the SAFM–FM transition, 3) the magnetostriction constant $\lambda^{\gamma,2}$, 4) the "spiral" magnetostriction (the relative change in the dimensions along the c axis at $H = H_{cr}$), 5) the spontaneous magnetostriction in the ferromagnetic phase along the b axis, 6) the spontaneous magnetostriction in the antiferromagnetic phase along the c axis.

cooling at temperatures $T < \Theta_2$. At temperatures $T < \Theta_1$ the magnetostriction along the c axis is two orders of magnitude smaller than at the SAFM–FM transition.

To determine the magnetoelastic energy in terbium–yttrium alloys we measured the thermal expansion of single crystals along the principal crystallographic directions. Measurements at $H = 0$ permitted determination of the relative strain of the crystal in the AFM phase. Measurements in a field $H = 50$ kOe applied along the axis of easy magnetization (the b axis) in the basal plane yielded the relative strain in the FM phase, since the 50-kOe field exceeded the maximum value of H_{cr} over the entire temperature range.

From the experimental data on the thermal expansion of the single crystals along the principal crystallographic directions, we determined the relative strains in the FM and AFM phases:

$$\Lambda_{2c}=\frac{c_2(T)-c(0)}{c(0)},\quad \Lambda_{1c}=\frac{c_1(T)-c(0)}{c(0)},$$
$$\Lambda_{2b}=\frac{b_2(T)-b(0)}{b(0)},\quad \Lambda_{1b}=\frac{b_1(T)-b(0)}{b(0)}. \tag{1}$$

Here $c_i(T)$ and $b_i(T)$ are the dimensions of the crystal along the crystallographic directions c and b, with the subscript $i = 1$ corresponding to the AFM phase and $i = 2$ to the FM phase, and $c(0)$ and $b(0)$ are the dimensions of the crystal along the crystallographic directions c and b in the absence of the exchange interaction and spontaneous magnetostriction. In this paper we have evaluated $c(0)$ and $b(0)$ using available data on the thermal expansion of lutecium,[15] which is analogous to other heavy rare earth metals in its crystal and electronic structure but which does not have a spontaneous magnetic moment. The thermal expansion of lutecium can to sufficient accuracy be taken as the phonon part of the thermal expansion of terbium and its alloys with yttrium if a small correction is introduced for the difference in the Debye temperature of lutecium and the alloys by the method described in an earlier paper.[16]

Let us consider the change in the energy contributions at the SAFM–FM phase transition for the case in which the magnetic field H is directed along the easy axis b in the basal plane: $\mathbf{H}\|\mathbf{b}$. As $H\rightarrow 0$ this transition goes over to the spontaneous transition at the temperature Θ_1. For $H \neq 0$ the temperature $\Theta_1(H)$ depends on the field.

For further description of this transition it is important that in both the FM and AFM phases the magnetic moments of the atoms lying in the same basal plane form ferromagnetic layers within which the magnetic moments of all the atoms are oriented parallel.

By virtue of the huge uniaxial anisotropy,[11] the magnetic moments at the SAFM–FM transition in a field $\mathbf{H}\|\mathbf{b}$ do not come out of the basal plane. Therefore, the uniaxial-anisotropy energy remains constant at the transition,and, consequently, the only change in the magnetic anisotropy energy is in the basal-plane component: ΔE_a^b.

The energy of the exchange interaction between atoms in the basal plane at the transition remains constant, whereas the energy of the exchange interaction between atoms lying in different basal planes changes markedly at $H = H_{cr}$, since the angle between the magnetic moments of the layers changes discontinuously from a value α to zero.[14] The interlayer turn angle α is the order parameter for the spiral ordering. The discontinuous changes occurring in the angle α, the magnetization, magnetostriction, and magnetocaloric effect at $H = H_{cr}$ indicate that the SAFM–FM transition is a first-order phase transition[17] (here we are not considering temperatures in the vicinity of Θ_2).

Thus, for describing the SAFM–FM transition, the following energy contributions should be included in the thermodynamic potentials Φ_2 and Φ_1 of the FM and AFM phases, respectively:

$$\Phi_2=F_{2\,\mathrm{exch}}+E_{2\,\mathrm{me}}+E_{2a}{}^b-HI_2, \tag{2}$$

$$\Phi_1=F_{1\,\mathrm{exch}}+E_{1\,\mathrm{me}}+E_{1a}{}^b-HI_1, \tag{3}$$

where $F_{2\mathrm{exch}}$ and $F_{1\mathrm{exch}}$ are the free energies of the exchange interaction between the magnetic layers, $E_{2\mathrm{me}}$ and $E_{1\mathrm{me}}$ are

TABLE I. Change in the components of the thermodynamic potential per atom at $H = H_{cr}$ for the alloy $Tb_{0.835}Y_{0.165}$.

T, K	$-\Delta E^{c}_{me}$, K	$-\Delta E^{b}_{a}$, K	$-(\Delta E^{c}_{me}+\Delta E^{b}_{a})$, K	$-\Delta E_{me}$, K	$-E_{1me}$, K	$-\Delta E^{b}_{a}$, K	$H_{cr}\Delta I$, K	ΔF_{exch}, K
125	0,65	0.21	0,86	0,83	1,38	0,032	0.125	0,987
137	0,64	0.15	0.79	0.60	1,42	0,025	0,429	1,05
155	0.56	0.088	0,65	0,57	1,42	0,011	0,844	1,43
173,5	0,41	0,036	0,45	0,39	1.30	0,0045	1.16	1,55
186,5	0,27	0.011	0,28	0,30	1.10	0,0016	0,95	1,25

the magnetoelastic energies, and I_2 and I_1 are the magnetizations; the subscripts 1 and 2 refer to the AFM and FM phases, respectively. The thermodynamic potentials of the phases are equal at the transition point in the case of a first-order phase transition at $H = H_{cr}$, and from the condition $\Phi_2 = \Phi_1$ we thus have

$$H_{cr}\Delta I=\Delta F_{exch}+\Delta E_{me}+\Delta E_{a}^{\text{б}}. \qquad (4)$$

Here ΔI is the jump in the magnetization at $H - H_{cr}$, $\Delta F_{exch} = F_{2exch} - F_{1exch}$ is the jump in the free energy of the exchange interaction between layers, ΔE^{b}_{a} is the jump in the energy of magnetic anisotropy in the basal plane, and ΔE_{me} is the jump in the magnetoelastic energy.

Since the magnetic moments are oriented along the axis of easy magnetization in the ferromagnetic phase and at various angles to the principal crystallographic directions in the basal plane in the spiral magnetic structure, the jump in the magnetic anisotropy energy at $H = H_{cr}$ is given by

$$\Delta E_{a}^{\text{б}}=-K_6. \qquad (5)$$

Our measurements of the magnetic anisotropy constant K_6 showed that this constant, which describes the anisotropy upon rotation of the magnetization I_s in the basal plane, falls rapidly as the temperature is raised from 4.2 K to the point Θ_2. Therefore, in the temperature interval Θ_1–Θ_2 the jump in the magnetic anisotropy energy ΔE^{b}_{a} at $H = H_c$ is small compared to the jump in the magnetoelastic energy ΔE_{me} (see Table I and Fig. 2).

The magnetoelastic energy, which is due to the magnetostrictive distortion of the crystal lattice in the basal plane, changes at $H = H_{cr}$ by the amount[3]

$$\Delta E_{me}^{\text{б}}=-{}^1/_4(C_{11}-C_{12})(\lambda^{\gamma,2})^2, \qquad (6)$$

where C_{11} and C_{12} are the elastic moduli. We evaluated ΔE^{b}_{em} by substituting into formula (6) the values obtained in this study for $\lambda^{\gamma,2}$ and the known values[18,19] of C_{11} and C_{12} for terbium. It is seen from Fig. 2 and Table 1 that the condition $E^{b}_{me} > \Delta E^{b}_{a}$ holds everywhere in the temperature range Θ_1–Θ_2. The total change in the magnetoelastic energy at the SAFM–FM transition is given by[4]

$$\Delta E_{me}=-{}^1/_2C_{11}^{\alpha}\Delta(\varepsilon^{\alpha,1})^2-{}^1/_2C_{22}^{\alpha}\Delta(\varepsilon^{\alpha,2})^2+\Delta E_{me}^{\text{б}}, \qquad (7)$$

where $\varepsilon^{\mu j}$ are the reversible deformations (strains) with the symmetry of the hexagonal close-packed structure, and C^{μ}_{jk} are the symmetric elastic constants. The strains $\varepsilon^{\alpha,1}$ and $\varepsilon^{\alpha,2}$ can be evaluated by the following formulas[4]:

$$\varepsilon^{\alpha,1}=\Lambda_{2c}+2(\Lambda_{2b}-{}^1/_2\lambda^{\gamma,2}), \qquad \varepsilon^{\alpha,2}={}^1/_3[\Lambda_{2c}-(\Lambda_{2b}-{}^1/_2\lambda^{\gamma,2})] \qquad (8)$$

for the ferromagnetic phase and

$$\varepsilon^{\alpha,1}=\Lambda_{1c}+2\Lambda_{1b}, \qquad \varepsilon^{\alpha,2}=3^{-1/2}(\Lambda_{1c}-\Lambda_{1b}) \qquad (9)$$

for the antiferromagnetic phase, where Λ_{ic} and Λ_{ib} are given by formulas (1).

The values of $\varepsilon^{\alpha,1}$ and $\varepsilon^{\alpha,2}$ evaluated from the experimentally determined spontaneous magnetostriction for single crystals of the alloy $Tb_{0.835}Y_{0.165}$ are given in Table II.

These values were used in formula (7) together with published values of the elastic constants of terbium[18,19] to evaluate the changes in the magnetoelastic energy ΔE_{me} at the SAFM–FM transition.

It should be noted that the elastic constants C_{jk} differ only slightly in the series of heavy rare earth metals and in yttrium, and their temperature dependence is such[18,19] that they vary no more than 10–15% over the temperature interval 4.2–300 K; the temperature dependence of these constants is therefore not important for estimating the energy contributions. The values found for ΔE_{me} in Tb_xY_{1-x} alloys are given in Table 1 and Fig. 2. It should be noted that

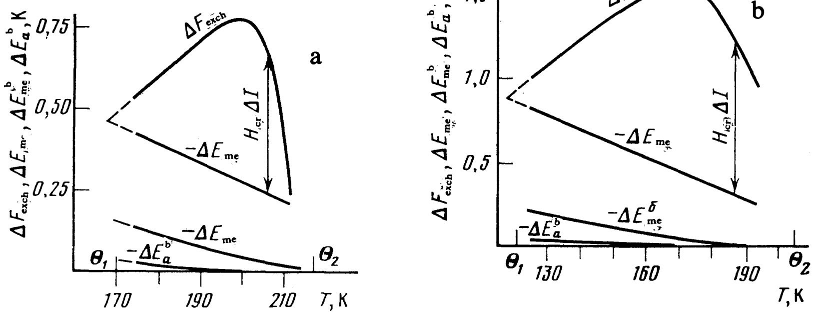

FIG. 2. Change in the components of the thermodynamic potenial per atom at $H = H_{cr}$ in the alloys $Tb_{0.91}Y_{0.09}$ (a) and $Tb_{0.835}Y_{0.165}$ (b).

TABLE II. Strains $\varepsilon_i^{\alpha,1}$ and $\varepsilon_i^{\alpha,2}$ in the ferromagnetic ($i=2$) and antiferromagnetic ($i=1$) phases for the alloy $\mathrm{Tb}_{0.835}\mathrm{Y}_{0.165}$.

T, K	$\varepsilon_2^{\alpha,1}\cdot 10^3$	$\varepsilon_2^{\alpha,2}\cdot 10^3$	$\varepsilon_1^{\alpha,1}\cdot 10^3$	$\varepsilon_1^{\alpha,2}\cdot 10^3$
125	5,16	2,61	4,45	2,05
129	5,22	2,58	4,53	2,01
137	5,32	2,50	4,67	1,94
146,5	5,33	2,40	4,78	1,83
155	5,35	2,28	4,88	1,71

relation (7) for the change in the magnetoelastic energy at $H > H_{\mathrm{cr}}$ includes the change due to the strain along the hexagonal axis c as well as the change due to the strain in the basal plane.

The change ΔF_{exch} in the exchange interaction energy at $H = H_{\mathrm{cr}}$ was evaluated from thermodynamic equation (4) by substituting in the calculated values of ΔE_a^b and ΔE_{me} and the experimental values of H_{cr} and ΔI. It is seen in Fig. 2 that ΔF_{exch} initially increases as the sample is cooled below Θ_2. This can be attributed to growth of the spontaneous magnetization of the magnetic layers. Upon further cooling ΔF_{exch}, after reaching a maximum, begins to decrease, apparently because of a decrease in the interlayer turn angle.[14]

Near the temperature Θ_1 of the SAFM–FM transition, ΔE_{me} is comparable in absolute value with ΔF_{exch} (see Fig. 2). This means that the increase ΔF_{exch} in the exchange energy at the transition is compensated by a decrease ΔE_{me} in the magnetoelastic energy. As a result, there is a transition to a stabler ferromagnetic phase which has a lower thermodynamic potential at temperatures $T = <\Theta_1$. The magnetic anisotropy energy ΔE_a^b, as is seen from Table I and Fig. 2, has an insignificant influence on the transition, inasmuch as $\Delta E_a^b \ll \Delta E_{\mathrm{me}}$.

The governing role in the magnetoelastic contribution ΔE_{me} is played by that part of the magnetostrictive strain which leads to a change in the volume and is due to the strain along the c axis (the spiral axis). This part of the magnetoelastic energy can be evaluated from the relation

$$\Delta E_{\mathrm{me}}^{c} = -{}^1/_2 C_{33}(\Lambda_{2c}^2 - \Lambda_{1c}^2), \tag{10}$$

where C_{33} is an elastic constant. The quantities appearing in formula (10) were evaluated from the experimental data and formula (1) by the method described above.

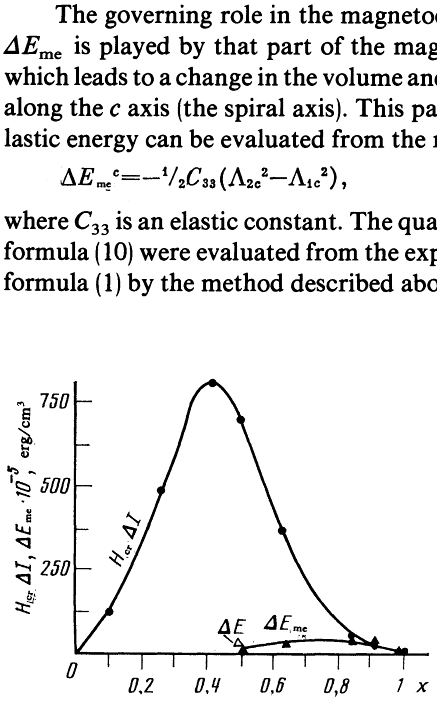

FIG. 3. Concentration dependence of the maximum energy barrier $H_{\mathrm{cr}}\Delta I$ in $\mathrm{Tb}_x\mathrm{Y}_{1-x}$ alloys.

It is seen from Table 1 that the value ΔE_{me}^c differs only slightly from the net change in the magnetoelastic energy ΔE_{me}. The author has previously shown[20] that the magnetostriction along the c axis is due to the dependence of the exchange energy on the lattice parameter c. This means that at $H = H_{\mathrm{cr}}$ the leading contribution to the change ΔE_{me} in the magnetoelastic energy is that of the magnetostriction due to the dependence of the exchange integrals on the lattice parameter c, which determines the distance between the magnetic layers in the FM and AFM phases. The magnetic anisotropy energy ΔE_a^b and the magnetoelastic energy ΔE_{me}^b in the basal plane (see Fig. 2 and Table I) have a much smaller influence on the SAFM–FM transition.

The data obtained in this study on the concentration dependence of the energy barrier separating the FM and AFM phases in $\mathrm{Tb}_x\mathrm{Y}_{1-x}$ alloys confirm the conclusion that the magnetostriction exerts the controlling influence on the SAFM–FM transition. It is seen in Fig. 3 that the maximum height of this barrier, i.e., the maximum value of the product $H_{\mathrm{cr}}\Delta I$ of the critical field H_{cr} and the magnetization jump ΔI, at first grows rapidly as the terbium is diluted with yttrium, reaches a maximum at $x \approx 0.4$, and then decreases on account of the decrease of both H_{cr} and ΔI. The concentration dependence of $(H_{\mathrm{cr}}\Delta I)_{\max}$ can be explained by a change in the energy spectrum of the conduction electrons, a splitting of the energy bands by the superzone boundaries, and by a deformation of the Fermi surface as a result of the spiral magnetic ordering and the magnetic dilution. The magnetic data[21] imply that the spontaneous SAFM–FM transition at temperature Θ_1 is observed in $\mathrm{Tb}_x\mathrm{Y}_{1-x}$ only at sufficiently high terbium concentrations $x > 0.835$. For $x > 0.8$, as can be seen in Fig. 3, $\Delta E_{1\mathrm{me}}$ becomes approximately equal to the energy barrier between the FM and AFM phases. In dilute alloys with $x < 0.63$, ΔE_{me} is smaller than this energy barrier by one or two orders of magnitude. For this reason the magnetoelastic interaction in dilute $\mathrm{Tb}_x\mathrm{Y}_{1-x}$ alloys does not lead to a spontaneous transition, and the SAFM–FM transition is observed in dilute alloys only in the presence of a field $H > H_{\mathrm{cr}}$. The interaction responsible for the magnetic anisotropy energy is also incapable of causing a spontaneous transition, since $\Delta E = \Delta E_{\mathrm{me}} + \Delta E_a^b$ is small compared to $H_{\mathrm{cr}}\Delta I$ (see Fig. 3).

Thus, the thermodynamic estimates and experimental data imply that the giant spontaneous exchange magnetostriction in terbium–yttrium alloys causes the decrease of the thermodynamic potential upon cooling to be stronger in the FM phase than in the AFM phase. As a result, during cooling in the absence of magnetic field there occurs a spontaneous transition at Θ_1 from the AFM to the FM phase, which at $T < \Theta_1$ has a lower thermodynamic potential.

By considering the results of the present study in conjunction with other data[4–7] which indicate that the magnetostriction plays a governing role in the SAFM–FM transition in dysprosium and holmium, we can conclude that the AFM–FM transition in heavy rare earth metals and their alloys is caused by the giant spontaneous magnetostrictive strains which arise upon magnetic ordering.

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