Anomalies of magnetic and electric properties on change of topology of the Fermi surface in terbium-yttrium alloys

K. P. Belov, S. A. Nikitin, N. A. Sheludko, V. P. Posyado, and G. E. Chuprikov

Moscow State University

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On the basis of experimental data on the concentration dependence of the magnetic, magnetostrictive, and electrical properties of terbium-yttrium alloys, the conclusion is drawn that upon change of the concentration of the components in these alloys, there occurs a phase transition of order 2 1/2, caused by a substantial reconstruction of the Fermi surface when terbium is alloyed with yttrium.

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Alloys of the terbium-yttrium system are very suitable objects for study of the influence upon the exchange and magnetocrystalline interactions of changes of the mean interatomic distances between magnetically active atoms. The crystal lattice parameters of terbium and yttrium differ by only ~0.5%, and the yttrium atoms have no magnetic moment; therefore the yttrium in these alloys is an almost ideal magnetic solvent.[1] Over the whole concentration range, terbium and yttrium form solid solutions.[2] Studies have already been made of their magnetic[3] and magnetostrictive[4] properties and of their crystal structure.[6] Neutron-diffraction investigations[7] have shown that on cooling below a point Θ_2, these alloys undergo a transition from the paramagnetic to a magnetically ordered state with an antiferromagnetic helical magnetic structure, whose axis coincides with the hexagonal c axis. But the measurements so far have been made on polycrystalline specimens, and consequently important anomalies due to the anisotropy of the properties have escaped the attention of the investigators.

In the present communication, we present data on the concentration dependence of the anisotropy of the magnetic, magnetostrictive, and electrical properties of single crystals of terbium-yttrium alloys; and we also show that the anomalies in the behavior of these properties can be understood if we take into account the change of topology of the Fermi surface upon dilution of terbium by yttrium. The single crystals of $Tb_{1-x}Y_x$ alloys were grown by the Czochralski method at the State Rare Metals Research Institute. The technique for preparing single crystals of Tb-Y alloys, their structure, and an analysis of the concentration of gaseous and metallic impurities were presented earlier.[8,9] For measurement of the magnetostrictive properties and thermal expansion, disks were used, of diameter 0.7–0.9 cm and thickness 0.5–0.15 cm, cut in two planes containing the crystallographic directions $\langle 1\bar{2}10\rangle$, $\langle 10\bar{1}0\rangle$, or $\langle 10\bar{1}0\rangle$, $\langle 0001\rangle$. The specimens for measurement of the electrical resistance had a rectangular cross section $\sim 10^{-2}$ cm² and length 0.8–1.0 cm along crystallographic directions $\langle 10\bar{1}0\rangle$, $\langle 1\bar{2}10\rangle$, $\langle 0001\rangle$. Orientation along the indicated direction to within error 1° was accomplished by the Laue method. The electrical resistance was measured by a standard four-contact method with a potentiometric circuit. The thermal expansion and magnetostriction were investigated with strain gauges made of wire with a small galvanomagnetic effect at low temperatures. The measurements were made in magnetic fields up to 55 kOe, over the temperature interval 4.2–400 K. The magnetic susceptibility and magnetization were measured on a vibration magnetometer, over the temperature interval 78–500 K.

The paramagnetic susceptibility of $Tb_{1-x}Y_x$ alloys can be described by the Curie-Weiss law, both for a direction along the hexagonal axis and in the basal plane (Fig. 1). The effective magnetic moments per terbium atom, determined from the slope of the χ^{-1} vs T curves, increase monotonically from the value $\mu_{eff}=9.6\ \mu_B$ found for pure terbium (Fig. 2). By extrapolation of the μ_{eff} vs x curve to $x=1$, one finds that the effective magnetic moment per terbium atom in this case, when the concentration of terbium in yttrium is negligibly small, is $\mu_{eff}=11\ \mu_B$, which is smaller by $\sim 1.3\ \mu_B$ than the value $\mu_{eff}=9.72\ \mu_B$ for a free trivalent terbium ion. The increase of μ_{eff} on dilution of terbium by yttrium is apparently explained by polarization of conduction electrons as a result of exchange interaction.

The paramagnetic Curie point for the basal plane, $\Theta_p^{\perp}$, decreases monotonically with increase of the yttrium concentration, approaching zero as $x\rightarrow 1$. The paramagnetic Curie point for the hexagonal axis, $\Theta_p^{\parallel}$, reveals a different behavior: there is a hump on the $\Theta_p^{\parallel}$ vs x curve, and a negative value of $\Theta_p^{\parallel}$ for alloys containing more than 80% yttrium. The paramagnetic Curie point for a polycrystalline specimen, which we calculated by

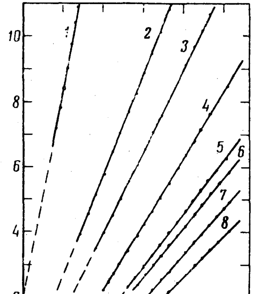

FIG. 1. Temperature variation of the inverse susceptibility of $Tb_{1-x}Y_x$ alloys. Yttrium content, at.%: Curve 1, 90; 2, 73; 3, 58.5; 4, 50; 5, 37.6; 6, 16.5; 7, 9; 8, ? [line omitted in original].

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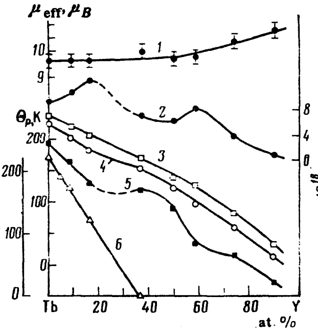

FIG. 2. Magnetic properties of $Tb_{1-x}Y_x$ alloys as functions of the yttrium concentration. 1, effective magnetic moment per terbium atom. 2, value of η. 3, paramagnetic Curie point for the basal plane. 4, paramagnetic Curie point of a polycrystal. 5, paramagnetic Curie point for the hexagonal axis. 6, temperature of the phase transition between ferromagnetism and helical antiferromagnetism.

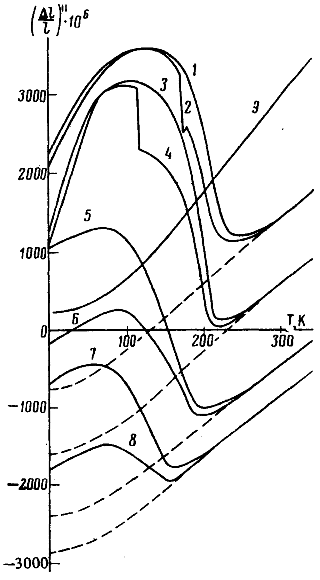

FIG. 4. Thermal expansion along the hexagonal axis of single crystals of $Tb_{1-x}Y_x$ alloys. Curves 1 and 2, Tb-9 at.% Y; 3 and 4, Tb-16.5 at.% Y; 5 and 6, Tb-37 at.% Y; 7 and 8, Tb-50 at.% Y; 9, Tb-90 at.% Y. Curves 2, 4, 6, and 8, $H = 0$; Curves 1, 3, 5, 7, and 9, $H > H_{cr}$. The dotted lines are the phonon contributions to the thermal expansion.

the formula $\Theta_p^{poly} = \frac{2}{3}\Theta_p^{\perp} + \frac{1}{3}\Theta_p^{\|}$, shows no departure from a monotonic decrease with increase of the yttrium content.

For layered antiferromagnets (in Tb-Y alloys the magnetic moments of atoms lying in the same basal plane are parallel and form a layer), according to the theory of second-order phase transitions,[10] the ratio $\Delta\Theta_p/\Theta_p$ ($\Delta\Theta_p = \Theta_p^{\perp} - \Theta_p^{\|}$) is equal to the ratio of the uniaxial anisotropy energy to the exchange-interaction energy. The relative values of the various contributions to the anisotropy have so far not been explained.[11-14] As was shown earlier,[14] if the anisotropy is caused by exchange interaction, the value of $\eta = \Delta\Theta_p\Theta_p a^3/(1.633 - c/a)$ is a quadratic function of the concentration of the components of the alloy. If η varies linearly with the concentration of the components, the anisotropy is of single-ion type.

According to the results of our measurements, the η vs x curve in $Tb_{1-x}Y_x$ alloys departs noticeably from the linear concentration dependence that was found earlier for gadolinium-disprosium rare-earth alloys.[14] In order to explain the reasons that lead to the anomalies of the concentration dependence of the paramagnetic Curie point $\Theta_p^{\|}$ and of the value of η, we shall consider the experimental results of measurement of the magnetostrictive and electrical properties and of the thermal expansion.

Figures 3 and 4 show data on the thermal expansion along the axis of easy magnetization b, located in the basal plane, and along the hexagonal axis c. The coefficients of thermal expansion measured in the paramagnetic range change smoothly with increase of the yttrium concentration, as is characteristic of solid solutions and correlates with the slight change of crystal structure in the paramagnetic state.[6] The anisotropy $\alpha_p^{\|} - \alpha_p^{\perp}$ of the coefficients of thermal expansion measured along the hexagonal axis ($\alpha_p^{\|}$) and in the basal plane ($\alpha_p^{\perp}$) increases at compositions near yttrium (Fig. 5).

On transition from the paramagnetic state to the magnetically ordered state below the point Θ_2, $Tb_{1-x}Y_x$ alloys undergo anisotropic spontaneous magnetostrictive deformations that are superposed on the phonon part of

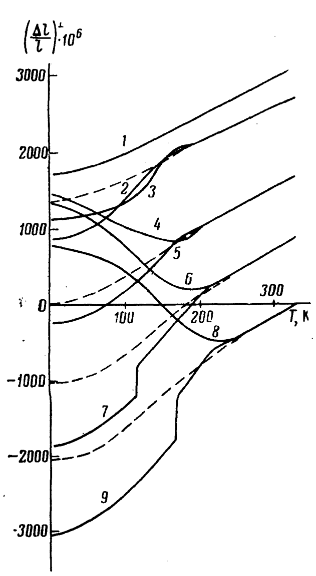

FIG. 3. Thermal expansion along the axis of easy magnetization of single crystals of $Tb_{1-x}Y_x$ alloys. 1, Tb-90 at.% Y. 2 and 3, Tb-50 at.% Y. 4 and 5, Tb-37 at.% Y. 6 and 7, Tb-16.5 at.% Y. 8 and 9, Tb-9 at.% Y. 2, 5, 7, and 9, $H = 0$; 1, 3, 4, 6, and 8, $H > H_{cr}$. The dotted lines are the phonon contributions to the thermal expansion.

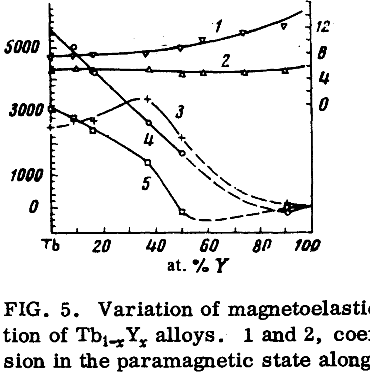

FIG. 5. Variation of magnetoelastic properties with composition of $Tb_{1-x}Y_x$ alloys. 1 and 2, coefficients of thermal expansion in the paramagnetic state along the hexagonal axis and in the basal plane. 3 and 5, spontaneous magnetostrictions λ_{0c} and λ_{0b}. 4, magnetostriction constant $\lambda^{\gamma,2}$.

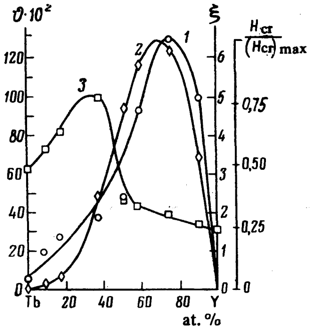

FIG. 6. Concentration dependence of ϑ, the relative magnitude of the hump on the ρ_m^c vs T curves (Curve 1); the maximum value of the critical field H_{cr} (Curve 2); and the ratio $\xi \equiv (d\rho_{ph}^b/dT)(d\rho_{ph}^c/dT)^{-1}$ (Curve 3) in $Tb_{1-x}Y_x$ alloys.

the thermal expansion. It is seen from Figs. 3 and 4 that at $T<\Theta_2$ there is a large increase of the relative elongation $(\Delta l/l)^{\parallel}$ of the specimen along the hexagonal axis, and a decrease of the relative elongation $(\Delta l/l)^{\perp}$ of the specimen in the basal plane along the axis b, in comparison with the phonon part $(\Delta l/l)_{phon}$ of the thermal expansion (the dotted curves in Figs. 3 and 4). The values of $(\Delta l/l)_{phon}$ were found from the thermal-expansion curves of lutecium, which has no magnetic contribution to the thermal expansion, with a correction for the difference in Debye temperatures.

For alloys containing less than 37 at.% yttrium, on cooling to points Θ_1 there occurs a transition from the helical antiferromagnetic state to a ferromagnetic; this shows up on the thermal-expansion curves as a discontinuous increase of the parameter c and decrease of the parameter b. Application of a magnetic field $H > H_{cr}$ along the axis b suppresses the jumps of the thermal expansion at the point Θ_1 and leads to a change of sign of the coefficient of thermal expansion along the axis b; this is caused by destruction of the helical magnetic structure in the interval Θ_1 to Θ_2 in a magnetic field exceeding a certain critical value H_{cr}, and orientation of the magnetic moments of the domains along the direction of the magnetic field.

The spontaneous magnetostrictive deformations along the c axis (λ_{0c}) and along the b axis (λ_{0b}) were determined at 4.2 K by subtracting from the experimental values of $(\Delta l/l)^{\parallel}$ and $(\Delta l/l)^{\perp}$, measured in a magnetic field $H>H_{cr}$, the phonon part of the thermal expansion; they reveal a noticeable difference. The values of λ_{0c} and λ_{0b} decrease sharply for $x>0.4$–0.5, and at $x=0.5$ the value of λ_{0b} changes sign. The magnetostriction constant $\lambda^{\gamma 2}$, equal to the difference between the longitudinal and transverse magnetostrictions in the basal plane for $H>H_{cr}$, also changes sign at $x=0.8$ (Fig. 5); this is unusual from the point of view of single-ion theory,[13,14] which predicts a linear variation of $\lambda^{\gamma 2}$ with concentration.

Thus although the thermal expansion in the paramagnetic range has no anomaly in its concentration dependence, the thermal expansion for $T<\Theta_2$, the spontaneous magnetostrictions λ_{0b} and λ_{0c}, and the field-induced magnetostriction, which is described by the constant $\lambda^{\gamma 2}$, are greatly different for compositions close to terbium and for compositions with a large content of yttrium. The thermal expansion in the paramagnetic range ($T>\Theta_2$) is determined primarily by the phonon spectrum of the crystal lattice, which changes little in heavy rare-earth metals as compared with yttrium. But the electronic contribution to the thermal expansion is negligibly small over almost the whole temperature range (including the range $T>\Theta_2$) except the low-temperature region ($T\ll\Theta_D$).[15]

The magnetostrictive deformations, according to calculations carried out for rare-earth metals,[16] depend significantly on the electronic structure, because of the fact that the magnitude of the s–f exchange interaction is determined by the properties of the conduction electrons and by the topology of the Fermi surface.[17] Anomalies in the concentration dependence of the magnetostrictive deformations can be explained by change of the electronic structure on dilution of terbium by yttrium; this shows up more clearly in analysis of the electrical properties of $Tb_{1-x}Y_x$ alloys.[9]

As follows from the results of measurements of the temperature variation of the electrical resistivity of these alloys,[9] at $T\lesssim\Theta_2$ there is a hump on the ρ^c vs T curves; this is explained by the appearance of energy gaps in the spectrum of the conduction electrons as a result of the formation of superzone boundaries when a helical magnetic structure is formed below the Θ_2 point. If we use the calculations carried out by Elliott and Wedgwood,[18] then it is easily shown that the magnitude of the hump is related to the exchange-interaction parameter and to the characteristics of the electronic structure by the formula

$$\vartheta=\frac{(\rho_M{}^c)_{\max}-(\rho_M{}^c)_{T>\Theta_2}}{(\rho_M{}^c)_{T>\Theta_2}}\sim\frac{3\pi\Gamma S m_{\max}}{4E_F k_F}\sum l_i, \tag{1}$$

where Γ is the integral of the s–f exchange interaction between the conduction electrons and the $4f$ electronic shell of the rare-earth ion; S is the spin of the ion; m_{max} is the relative magnetization at the temperature corresponding to the maximum $(\rho_M^c)_{max}$ on the ρ_M^c vs T curve; E_F is the Fermi energy; k_F is the Fermi wave vector; and l_i is the wave vector of the superzone boundary. The sum extends over all the superzone boundaries that intersect the Fermi surface perpendicularly to the axis of the helical structure.

The relative magnitude ϑ of the hump increases sharply with increase of the yttrium concentration in the alloys, reaching a maximum value at $x\sim 0.7$ (see Curve 1 in Fig. 6). This indicates a significant change, in the magnetically ordered state, of the area and position of the superzone boundaries with increase of the angle of the helicoid[7] as a result of dilution of terbium by yttrium.

Formula (1) was derived on the assumption of a quadratic dispersion law for the conduction electrons,[18] whereas the Fermi surface in terbium-yttrium alloys differs appreciably from a sphere.[9] For this reason, formula (1) can be used only for qualitative interpretation of the experimental results. One is struck by the fact that (see Fig. 6) there is a clear correlation between the concentration dependences of the quantity ϑ, which according to formula (1) and the work of Elliott and Wedgwood[18] describes the effect on the electrical conductivity of a gap in the energy spectrum of the conduction electrons, and the magnetic parameter $(H_{cr})_{max}$. The quantity H_{cr} is the field at which the helical magnetic ordering is destroyed, and $(H_{cr})_{max}$ is its maximum value for a given alloy. The experimentally observed interconnection of the magnetic and electric properties can be explained on the basis of Dzyaloshinskiĭ's theory,[19] according to which the nature of the ordering is determined to a significant degree by the energy spectrum of the conduction electrons and the topology of the Fermi surface. In order to obtain more complete information about the change of topology of the Fermi surface in $Tb_{1-x}Y_x$ alloys, we shall carry out an analysis of data[9] on the electrical properties of crystals of this system on the basis of theoretical calculations of the Fermi surface of rare-earth metals,[20] and by use of general theorems on the relation of the electrical conductivity to the projection of the Fermi surface on a plane perpendicular to the direction of the current.

In the relaxation-time approximation, which is widely used in the study of transport phenomena,[21] and in the approximation of validity of Matthiessen's rule for rare-earth metals,[20,22,23] one may write for the electrical resistivity along a given crystallographic direction

$$\rho=\left(\frac{4\pi^3\hbar}{e^2}\right)\left[1\Big/\int_{E_F}\tau_0 v_i\,dS_j+1\Big/\int_{E_F}\tau_{ph}v_i\,dS_j+1\Big/\int_{E_F}\tau_m v_i\,dS_j\right], \qquad (2)$$

where τ_0, τ_{ph}, and τ_m are the relaxation times for impurities, phonons, and magnons, v_i is the component of the electron velocity in the i-th direction, and dS_j is the projection of an element of the Fermi surface on the j-th direction. The experimental data are interpreted[20–23] by replacing v_i by its mean value over the Fermi surface and assuming that in a first approximation the relaxation times are isotropic. Then the relation (2) can be reduced to the simpler form

$$\rho^c=A/\tau_0\int_{E_F}dS_c+A/\tau_{ph}\int_{E_F}dS_c+A/\tau_m\int_{E_F}dS_c, \qquad (3)$$

where $\int dS_c$ is the projection of the Fermi surface on a plane perpendicular to the c axis. Analogous relations can be written for the b and a axes. The expression (3) gives a possibility of making certain estimates of the change of topology of the Fermi surface on the basis of results of measurements of electrical resistivity.

According to calculations,[20] the Fermi surface of heavy rare-earth metals departs markedly from the spherical surface for free electrons, and its projections on a plane normal to the hexagonal axis and on planes normal to the basal plane differ greatly in value. From experimental data[9] it follows that the phonon part of the electrical resistivity in the basal plane in the paramagnetic range increases rapidly with yttrium concentration for $x>0.4$. This indicates, according to formula (3), a decrease of the projection of the Fermi surface on planes normal to the a and b axes, and consequently a decrease of the "trunk" of the Fermi surface (see Ref. 20). A still more rapid decrease is observed for $x>0.4$ for the phonon part of the electrical resistivity along the c axis[9]; this may be attributed to a significant change of the projection of the Fermi surface, also, on the plane perpendicular to the c axis. The magnitude of $(d\rho^b_{ph}/dT)\,(d\rho^c_{ph}/dT)^{-1}$ changes rapidly near the same concentration $x\approx 0.4$–0.5 (Curve 3 in Fig. 6). This is evidence that the deformation of the Fermi surface occurs without conservation of its shape. Calculations[24] show that the Fermi surfaces of heavy rare-earth metals are quite similar, but there are certain distinctive features in terbium as compared with yttrium[20,24,25]: in terbium there is an M arm, whereas in yttrium this element of the Fermi surface is absent. Furthermore, the distance between plane sections of the "webbing" anomaly is appreciably smaller for terbium than for yttrium. From the results of our measurements it follows that the transition from a Fermi surface of one type to the other occurs in terbium-yttrium alloys at concentration 40–50 at.% yttrium. Here anomalies are observed in the concentration dependence of the anisotropy of the paramagnetic Curie point, the critical field, the magnetostriction, and the electrical resistivity.

According to our data (Curve 6 in Fig. 2), ferromagnetism in terbium-yttrium alloys exists only when the yttrium content is less than 40 at.%; in alloys with a larger yttrium content, there is observed on cooling only a transition from paramagnetism to helical antiferromagnetism. The experimental results presented above enable us to conclude that when the concentration of the components is changed in terbium-yttrium alloys, there occurs near the concentration 40–50 at.% yttrium a phase change of order $2\frac{1}{2}$, caused by a substantial change of topology of the Fermi surface and known in the literature as the I. M. Lifshitz transition.[26]

With such an interpretation of the experimental results, one thing in particular that becomes intelligible is the anomaly in the variation of the anisotropy of the paramagnetic Curie points with composition in terbium-yttrium alloys. Indirect exchange interaction in rare-earth metals and alloys occurs via the conduction electrons[17]; therefore upon change of the energy spectrum of the conduction electrons and of the Fermi surface, one may expect changes also in the contribution to the anisotropy from anisotropic exchange interaction.[11] Since the departure from the relation predicted on the basis of the single-ion anisotropy model amounts to ~30%, one may conclude that although, as in a number of other rare-earth metals and alloys, single-ion anisotropy is dominant, nevertheless there is a contribution of smaller amount from anisotropic exchange interaction. It must be emphasized that the anomalous composition dependence of the anisotropy of the paramagnetic Curie points is caused by the anomaly in the concentration dependence of the paramagnetic Curie point along the hexagonal axis, $\Theta_p^{\parallel}$, whereas the temperatures $\Theta_p^{\perp}$, Θ_p^p, and Θ_2, which characterize the exchange-interaction energy, vary monotonically with yttrium concentration (see Fig. 2), just as in other alloys of rare-earth metals with yttrium.[1]

Another reason for the anomalies that we observed in the concentration dependence of physical properties might be a rapid change of the exchange interaction with unsubstantial change of the electronic spectrum. But this assumption is less probable, since the anomalies are also observed for the phonon part of the electrical resistivity, measured at $T>\Theta_2$, where exchange interaction has no effect on the temperature coefficient of the electrical resistivity[9]; this, according to the relation (3), points unambiguously to a substantial reconstruction of the Fermi surface. This is also indicated by the change of sign that we observed in the galvanomagnetic effect (the change of electrical resistivity in a magnetic field) upon destruction of the helical magnetic structure, in the concentration range 40–50 at.% yttrium.

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