Investigation of magnetic properties of Tb(Co$_{1-x}$Ni$_x$)$_5$ compounds

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Neutron-diffraction and magnetic methods are used to investigate the crystalline and magnetic structure of the compounds Tb(Co$_{1-x}$Ni$_x$)$_5$. It is shown that the Ni ions occupy predominately the 2*c* positions and Co the 3*g* positions of a crystal cell with a CaCu$_5$ structure. The magnetic phase diagram is constructed. It is concluded from an analysis of the concentration dependences of the magnetizations of the 2*c* and 3*g* sublattices that the magnetic moments at the Co ions are constant. It is shown that the Ni atoms have in the investigated compounds a magnetic moment that depends on the concentration *x*. Estimates based on the assumption that the R-R and R-Co exchange interaction energies are constant lead to the conclusion that an important contribution is made by the Co-Co interactions to the Curie temperature of compounds with small CO contents ($x \geqslant 0.8$). It is established that the character of the temperature dependence of the 2*c*-sublattice magnetization is connected with the direction of the easy magnetization axis of the crystal, and this leads to a jump of the magnetization in the easy plane–easy axis transition. It is concluded on this basis that the dominant contribution to the magnetic anisotropy of the Co sublattice of compounds of the RCo type is made by Co atoms located in 2*c* positions.

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A tremendous magnetocrystalline anisotropy energy, comparable with the energy of the exchange interactions, is realized in rare-earth intermetallic compounds of the type R (M$_{1-x}$M'$_x$)$_5$. As shown in Refs. 1–6, this circumstance leads to substantial peculiarities in their magnetic properites (e.g., anisotropy is observed in the magnetizations of the rare earth (RE) and 3*d* sublattices). The study of the ensuing questions is in fact only in the initial stage. An investigation of these systems is promising from the point of view of obtaining the information needed to solve some general problems concerning the nature of the magnetism of rare-earth and 3*d*-transition metals and their alloys.

So far, the most investigated is the group of compounds of the type R(Co$_{1-x}$Ni$_x$)$_5$. As a rule, the average magnetic moments were determined by magnetic measurements, which yield no information on the individual atomic properties. As a result, even in the simplest case when *R* is not magnetic (R = La, Y), the analysis of the results is difficult and at times even impossible, and the interpretations offered are therefore ambiguous. Thus, for example, the anomalous behavior of the concentration dependence of the average magnetic monet $\bar{m}(x)$ and of the Curie point $T_C(x)$ in the nickel-rich region are differently treated in different papers for the same alloys. The difficulties are increased manyfold when it comes to interpret the value of $\bar{m}(x)$ of compounds containing a magnetoactive rare-earth element. Therefore, notwithstanding the relatively large accumulated material, many aspects of the magnetic nature and structure of these compounds remain unexplained for lack of neutron-diffraction data.

The purpose of the present paper is a comprehensive study of the structure and of the magnetic state of Tb(Co$_{1-x}$Ni$_x$)$_5$ alloys, using magnetic measurements and neutron diffraction. The chosen system is preferable to others for a number of reasons. It is known that Tb has a small cross section for neutron absorption ($\sigma_{\rm abs}^{\rm Tb} = 26\times10^{-28}$ m^2, $\sigma_{\rm abs}^{\rm Sm} = 6000\times10^{-28}$ m^2, $\sigma_{\rm abs}^{\rm Dy} = 400\times10^{-28}$ m^2) and a large magnetic moment. This ensures an increased sensitivity of the diffraction pattern to changes of the magnetic state. Another extremely important fact is that one of the limiting alloys of the system has a temperature spin-orientational transition. It is of interest to study the influence of alloying with nickel on the characteristics of this transition and to use its value to study the influence of the magnetic anisotropy on the magnetic moments and on the temperature dependences of the sublattice magnetizations. In particular, replacement of Co with nickel in TbCo$_5$ should lower T_C and lead, in accord with Ref. 7, to a more pronounced manifestitation of the influence of the anisotropy on the previously observed[8] magnetization of the Co sublattice.

SAMPLES AND EXPERIMENTAL TECHNIQUE

The Tb(Co$_{1-x}$Ni$_x$)$_5$ (x = 0, 0.2, 0.4, 0.5,[1)] 0.6, 0.8, 1.0) were made by melting terbium (purity 99 wt.%), Co, and Nb (both 99.99% pure) in an induction furnace, in alundum crucibles and in an argon atmosphere. Homogenizing annealing at 1400 K was carried out in an argon atmosphere for 24 hours. Spherical single-crystal samples of 2 mm diam were cut from individual grains of the ingots. They were used to study the temperature dependence of the spontaneous magnetization with the aid of a low-frequency vibromagnetometer. The measurements were performed in a 9 kOe field parallel to the easy magnetization axis (on a free-standing sample).

Powders with particle sizes $d < 10^{-2}$ cm were prepared for the x-ray diffraction and neutron diffraction investigation. The x-ray diffraction was investigated at room temperture with a DRON-2 diffractometer. The neutron-diffrac-

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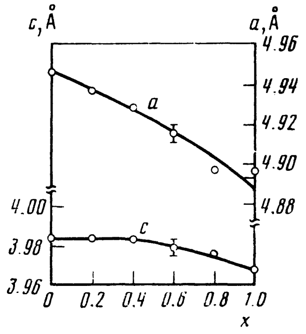

FIG. 1. Concentration dependences of the parameters a and c of the crystal cell of the compounds $Tb(Co_{1-x}Ni_x)_5$.

tion patterns were obtained with apparatus mounted on one of the horizontal channels of an IVV-2 reactor. The neutron wavelengths were 1.29×10^{-8} and 1.70×10^{-8} cm.

We investigated the angle interval $0.0409 < \sin\theta / \lambda < 0.3$. In the analysis of the neutron diffraction patterns it was assumed that $b_{Tb} = 0.76 \times 10^{-12}$ cm and $b_{Co} = 0.28 \times 10^{-12}$ cm. The form factors of Tb^{3+}, Co, and Ni were taken from Refs. 9 and 10. The temperature in the interval 4.2–300 K was determined with a Cu-FeCu thermocouple, and in the 300–900 interval with a chromel-alumel thermocouple. The respective measurement accuracies were ± 1.0 K and ± 2 K.

ATOMIC STRUCTURE

The $Tb(Co_{1-x}Ni_x)_5$ compounds were synthesized for the first time ever. This made in necessary to carry out a thorough analysis of their atomic structure. A study of the x-ray diffraction pattern shows that all the $Tb(Co_{1-x}Ni_x)_5$ compounds have a hexagonal structure of the $CaCu_5$ type (space group P6/mmm). The Tb^{3+} ions occupy the position $1a$ (0, 0, 0), while Co and Ni occupy the positions $2c$ (1/3, 2/3, 0) and $3g$ (1/2, 0, 1/2). From an analysis of the intensities of the peaks on the neutron diffraction patterns it follows that the cobalt and nickel are not uniformly distributed amount the $2c$ and $3g$ sites: the Co ion occupies predominantly the $3g$ sites and Ni the $2c$ sites. This is indicated also, in accord with Ref. 11, by the nonlinear concentration dependence of the lattice parameters a and c in the system considered (Fig. 1). A similar distribution of the atoms over nonequivalent sites of the d-sublattice was observed in the alloys of the systems $Y(Co_{1-x}Ni_x)_5$ and $La(Co_{1-x}Ni_x)_5$ (Ref. 12). In contrast to Ref. 12, in our case we used neutron diffraction patterns obtained at $T > T_C$ and containing no magnetic contributions. The probabilities P of filling the $2c$ sites with Ni ions in the various compositions are listed in Table I. It can be seen that the predominance of substitution of Ni in the $2c$ sites and of Co in the $3g$ sites is most strongly pronounced at $x \approx 0.5$. The experimental $P(x)$ dependence is well described by the third-degree polynomial

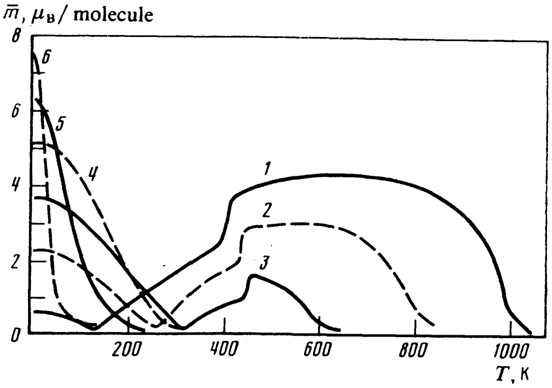

FIG. 2. Temperature dependences of the average magnetic moment $\bar{m}$ of the compounds $Tb(Co_{1-x}Ni_x)_5$. The numbers correspond to the following values of x: 1 — 0; 2 — 0.2; 3 — 0.4; 4 — 0.6; 5 — 0.8; 6 — 10.

$$P(x) = 1.557x - 0.349x^2 - 0.208x^3. \tag{1}$$

Measurements performed on $TbCo_2Ni_3$ and $TbCoNi_4$ in the interval 300–950 K have shown that within the limits of the sensitivity of the method (± 0.02) the distribution of the $3d$ ions over the $2c$ and $3g$ sites can be regarded as independent of temperature. This has made it possible to use the P values obtained at $T > T_C$ to analyze the magnetic structure and to determine the sublattice magnetic moments m_{Tb}, m_c and m_g in the entire temperature interval (m_{Tb}) is the magnetic moment of the rare-earth sublattice, while m_c and m_g are the average magnetic moments of the sublattices consisting of Co and Ni atoms in the $2c$ and $3g$ position, respectively). The values of P obtained by us for different x are close to those given in (12) for $Th(Co_{1-x}Ni_x)_5$, $Y(Co_{1-x}Ni_x)_5$ and $La(Co_{1-x}Ni_x)_5$. This confirms once more the concusion drawn in Ref. 12, that the Ni and Co ion distribution is independent of the type of rare-earth element in the compounds considered.

MAGNETIC MEASUREMENTS

Figure 2 shows the results of the magnetic measurements of the alloys $Tb(Co_{1-x}Ni_x)_5$ with $x = 0$, 0.2, 0.4, 0.6, 0.8, and 1.0. It can be seen that with increasing x the alloy magnetization $\bar{m}$ at 4.2 increases almost linearly. The dependence of the Curie temperature $T_C(x)$ on the concentration is more complicated; it can be easily divided into two sections,

TABLE I. Experimental results (the values of m_{Tb}, m_c, m_g, and $\bar{m}$ correspond to $T = 4.2$ K).

x	P	m_{Tb}, μ_B	m_c, μ_B	m_g, μ_B	$\bar{m}$, μ_B/ /molecule	T_c, K
0	0	8.5±0.4	1.60±0.15	1.72±0.15	0.5	980±20
0.2	0.31	8.5±0.4	1.29±0.15	1.51±0.15	2.2	790±20
0.4	0.53	8.7±0.4	0.92±0.15	1.25±0.15	3.6	590±10
0.6	0.76	8.5±0.4	0.63±0.15	0.93±0.15	5.2	330±10
0.8	0.93	7.9±0.4	0.33±0.20	0.4±0.20	6.3	120±10
1.0	1.0	7.3±0.3	0.0±0.2	0.0±0.2	7.6	28±2

linear in x from 0 to 0.4, and with some deviation from linearity at $0.4 < x < 1.0$. The maximum $T_C - 28$ K corresponds to $x = 1$.

A compensation point T_{comp} is found on the $\bar{m}(T)$ plots for $x = 0$, 0.2, and 0.4. As can be seen from Fig. 2, in this set of comounds T_{comp} increases from 130 to 300 K with changing x. The $\bar{m}(T)$ plots of these three alloys show above T_{comp} an anomaly wherein the rate of change of the alloy magnetization increases sharply in a narrow temperature interval. According to Ref. 1, in compounds in which R has the larger magnetic moment this is determined mainly by the change of the magnetization of the rare-earth sublattice in the case of a spin-reorientation transition (SRT). With increasing x, the SRT interval shifts somewhat towards higher temperatures and becomes slightly narrower. The $m(T)$ curves for the alloys with $x = 0.6$, 0.8, and 1.0 are relatively simple. It can be seen from their shape that the magnetization of the rare-earth sublattice magnetization in these alloys is larger than that of the $3d$ lattice, and the easy magnetization axis lies in the basal plane in the entire investigated temperature interval.

MAGNETIC PHASE DIAGRAM

Figure 3 shows a neutron diffraction pattern of $TbCo_2Ni_3$ at 4.2 K. The neutron diffraction pattern of the other compounds of the system differ from it, at all temperatures, only in the reflection intensities. The magnetic-structure wave vector $\mathbf{K} = 0$ (the magnetic and atomic unit cells coincide). All the magnetic structures with $\mathbf{K} = 0$ that are symmetry-allowed for RCo_5-type crystals were established in Ref. 13 by a group-theoretical method. In the analysis of the neutron diffraction patterns obtained for all these structures in the entire temperature interval, the R factor is minimized with respect to the moduli of the magnetic moments:

$$R = \sum_i |J^i_{\text{obs}} - J^i_{\text{calc}}| \;/\; \sum_i J^i_{\text{obs}}, \tag{2}$$

where J^i_{obs} is the observedmagnetic component of the i-th reflection; J^i_{calc} is the calculated intensity of the i-th magnetic reflection; $i = (100)$, (001), (110), (200), (111), (002), (102). Good agreement between the calculated and observed intensities is obtained when the magnetic moments $\mathbf{m}_c$ and $\mathbf{m}_g$ of the d-ions in the $2c$ and $3g$ positions are parallel to each other and are antiparallel to the Tb moments ($\mathbf{m}_{\text{Tb}}$). Figure 4 shows the magnetic phase diagram plotted from magnetic and neutron-diffraction measurement data. The Curie points in the diagram were determined from the temperatures at which the magnetic reflections on the neutron-diffraction pattern vanished. The values of T_C obtained from magnetic measurements are somewhat higher than those indicated in Fig. 4, a fact known to be due to a some influence of the applied magnetic field (9 kOe). In the region under the curve EDC (Fig. 4), the magnetic moments lie in the basal plane. In the upper part of the diagram, bounded by the lines AD and DB, they are parallel to the c axis. In the narrow section between the lines BD and DE are realized the so-called canted structures, that can be regarded as the result of superposition of the two considered magnetic structures.

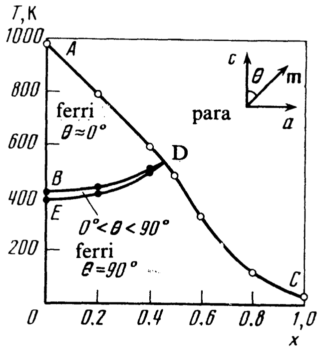

FIG. 4. Magnetic phase diagram of the compound $Tb(Co_{1-x}Ni_x)_5$: ○—Curie temperature, ●—lower and upper SRT temperatures.

MAGNETIC MOMENTS

1. The form factors of the cobalt and nickel atoms are practically identical, so that the reflection intensities together with the rare-earth magnetic moment m_{Tb} can yield only the average magnetic moments m_c and m_g. They are connected with the individual magnetic moments by the relations

$$m_c = (1-P)m_c^{\text{Co}} \pm P m_c^{\text{Ni}}, \tag{3}$$

$$m_g = \frac{3-5x+2P}{3} m_g{}^{\text{Co}} \pm \frac{5x-2P}{3} m_g{}^{\text{Ni}}, \tag{4}$$

where m_c^{Ni}, m_c^{Co}, m_g^{Ni}, and m_g^{Co} are the magnetic moments of the nickel and cobalt atoms in the positions $2c$ and $3g$, respectively. The coefficients preceding these moments are the probabilities of substitution of the corresponding sites by

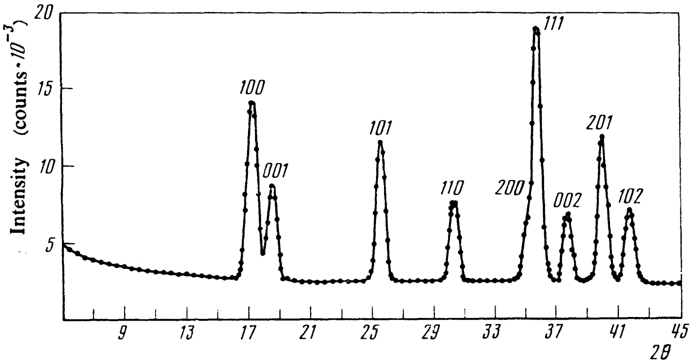

FIG. 3. Neutron diffraction pattern of the compound $TbCo_2Ni_3$ at $T = 4.2$ K.

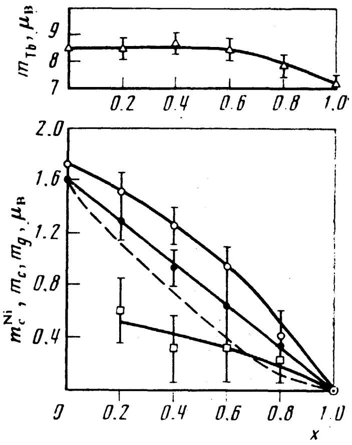

FIG. 5. Concentration dependences of m_{Tb}, m_g, m_c, m_c^{Ni} at 4.2 *K*: Δ — m_{Tb}; ● — m_g; ○ — m_c; □ — m_c^{Ni}; dashed curve—calculated plot of $m_c(x)$ under the conditions $m_c^{Ni} = 0$ and $m_c^{Co} = 1.6\,\mu_B$.

cobalt and nickel atoms. The (—) signs in (3) and (4) correspond to the case when the magnetic moments of the cobalt and nickel atoms in the *d*-sublattice are oppositely oriented. This possibility was pointed out in Ref. 14.

Replacing *P* in (3) and (4) by (1) we obtain the functions $m_c(x)$ and $m_g(x)$ in analytic form

$$m_c(x) = (1 - 1.557x + 0.349x^2 + 0.208x^3)m_c^{Co} \pm (1.557x - 0.349x^2 - 0.208x^3)m_c^{Ni}, \quad (5)$$

$$m_g(x) = (1 - 1.629x - 0.233x^2 - 0.139x^3)m_g^{Co} \pm (0.629x + 0.233x^2 + 0.139x^3)m_g^{Ni}. \quad (6)$$

Table I and Fig. 5 show the values of m_{Tb}, m_c, and m_g obtained by us for all the alloys. Analysis of the experimental $m_c(x)$ and $m_g(x)$ dependences with the aid of relations (5) and (6) leads to the conclusion that the nickel atoms in the intermediate-region alloys have an appreciable magnetic moment. Indeed, to reconcile the experiment with the values calculated from (5) it is necessary to ascribe to the Co atoms extremely high magnetic moments. For example, in the alloys with $x = 0.8$ the value $m_c^{Ni} = 0$ corresponds to $m_c^{Co} = 5.4\ \mu_B$, a physically unrealistic value. There are grounds for assuming that the magnetic moment at the Co atoms in either position, at least in most of the concentration interval, does not change at all and remains constant. Favoring this conclusion, for example, is the linear dependence of the Curie temperature on the concentration (see Fig. 4) in the region $0 < x < 0.4$, where the mixing rule is well satisfied. Assuming in this connection that, just as in $TbCo_5$, we have $m_c^{Co} = 1.60 \pm 0.15\mu_B$ and $m_g^{Co} = 1.72 \pm 0.15\mu_B$ at all *x*, we obtain good fit of the calculated curves (5) and (6) to the experiment if m_g^{Ni} for all *x* is equal to $(0 \pm 0.15)\,\mu_B$ and m_c^{Ni} decreases smoothly when *x* changes from 0 to 1, from a value $(0.6 \pm 0.15)\,\mu_B$ to $(0 \pm 0.15)\,\mu_B$. It can thus be regarded as established by our results that an appreciable magnetic moment is present at the nickel atoms in the $Tb(Co_{1-x}Ni_x)_5$ alloys and that it depends on the Co concentration.

The absence of a magnetic moment at the nickel atoms in compounds with $x = 1$ is usually attributed to the filling of the 3*d* band of nickel by the valence electrons of the rare-earth metal. The appearance of a magnetic moment at the nickel in the mixed $R\,(Co_{1-x}Ni_x)_5$ compounds can be due to a change in the band structure when cobalt is added. This may be the consequence of the drawing away of the electron density towards the cobalt atoms.

The absence of any magnetic moment whatever for nickel in $R\,(Co_{1-x}Ni_x)_5$ with nonmagnetic R (Ref. 14) indicates that an important role in the formation of this moment in $Tb(Co_{1-x}Ni_x)_5$ is played also by the magnetic moment of the rare-earth ion (more accurately, by its spin). In this sense the compound considered here is no exception: analysis of the data for certain compounds with magnetic R, such as Sm, Gd, or Dy (Refs. 15–17) does not exclude the possibility of explaining their magnetic properties by assuming a magnetic moment for the nickel atoms.

2. As seen from Fig. 5, we can regard m_{Tb} as constant in the region $0 \leqslant x \leqslant 0.4$. It decreases then and reaches 7.3 μ_B at $x = 1$.

The moments of Tb and Ni$(7.3 \pm 0.3)\,\mu_B$ $(0.00 \pm 0.2)\,\mu_B$ obtained in $TbNi_3$ agree with those given in Ref. 18. Unfortunatly, the measurement accuracy attained is insufficient ($\pm 0.2\,\mu_B$) and the question of the presence of a magnetic moment at the Ni atoms in $TbNi_5$ remains open. The published opinions concerning this matter differ. For example, it is assumed in Ref. 17 that m_{Ni} in this compound is zero. It was concluded in Ref. 19 on the basis of measurements of the magnetization and of the paramagnetic susceptibility of $TbNi_5$ that the *d* band of nickel has an orientational polarization equal to 1 μ_B per cell (corresponding to a magnetic moment $\sim 0.2\,\mu_B$ for the Ni atom).

The complicated dependence of the magnetic moment of the rare earth ion on the concentration, established in this paper, is apparently a common property of the compounds of the type considered. It is impossible to predict with any degree of accuracy the character of this dependence, which must be determined by neutron diffraction for each compound. It is possible that such investigations will necessitate changes in the conclusions based on the assumption that the magnetic moment of the RE ion is constant in the compounds $Sm(Co_{1-x}Ni_x)_5$ (Ref. 15), $Gd(Co_{1-x}Ni_x)_5$ (Ref. 16), and $Dy(Co_{1-x}Ni_x)_5$ (Ref. 17).

MAGNETIZATION AND CURIE TEMPERATURE

In contrast to the compounds $La(Co_{1-x}Ni_x)_5$ and $Y(Co_{1-x}Ni_x)_5$ with nonmagnetic R (Refs. 14 and 19), the $m_d(x)$ dependence obtained by us for $Tb(Co_{1-x}Ni_x)_5$, from neutron-diffraction measurements is monotonic (Fig. 6). It is described in the entire concentration region by a single expression constituting some modification of the displacement law:

$$m_d(x) = 2m_c(x) + 3m_g(x), \quad (7)$$

$m_c(x)$ and $m_g(x)$ in (7) are calculated from (3) and (4). This means that, in contrast to alloys with alloys with nomagnetic *R*, the contribution of the cobalt atoms to the *d*-sublattice magnetization is proportional in the entire concentration region to their numbers in the 2*c* and 3*g* sites, i.e., the cobalt atoms play the same role in the formation of the *d*-sublattice

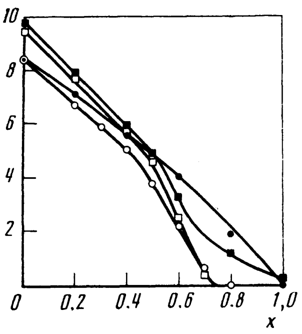

FIG. 6. Concentration dependences of the Curie temperatures T_C, of the magnetic moment m_d of the d sublattices of the compounds $Y(Co_{1-x}Ni_x)_5$ and $Tb(Co_{1-x}Ni_x)_5$; ○, □ — m_d T_C and of TC of the compounds $Y(Co_{1-x}Ni_x)_5$ [14]; ● ■ — m_d $Tb(Co_{1-x}Ni_x)_5$.

magnetization at all x. Thus, complete substitution of nonmagnetic R with magnetic R in $R(Co_{1-x}Ni_x)_5$ and addition of supplementary R-Co and R-R exchange interactions to the existing Co-Co exchange interactions alters radically the state of the alloys in the nickel-rich region, viz., the paramagnetic compounds become ferrimagnetic at $x > 0.78$, the magnetic moments at the Co atoms remain constant at all x, and an induced magnetic moment appears at the nickel atoms. Near the $TbNi_5$ composition, the number of Co atoms is small and the bulk of them has in the nearest surroundings only nonmagnetic or weakly magnetic Ni atoms. A ferrimagnetic (relative to the magnetic moment of the rare-earth atoms) of the magnetic moments of Co is produced here in fact only on account of the R-Co exchange interaction. The compounds considered constitute each in this concentration region a magnetic matrix of exchange-interacting R ions, in which are placed Co ions that are for the most part non-interacting (92% of the Co atoms are solitary at $x = 0.99$). For such Co atoms the R-Co interaction serves as an external field that is practically the only cause of the appearance of the spontaneous magnetization of the d sublattice.

With decreasing x, the number of solitary Co atoms decreases rapidly (at $x = 0.85$ only 15% of the Co atoms have no other atoms in their immediate vicinity). With increasing number of nearest Co-Co groups, conditions are produced in the alloys for the onset of exchange Co-Co bonds and for their participation in the magnetic ordering of the atoms.

The Curie temperatures in the $Tb(Co_{1-x}Ni_x)_5$ alloys, obtained by neutron diffraction, are given in Table I. Figure 6 shows the concentration dependences of $T_C(x)$ for $Tb(Co_{1-x}Ni_x)_5$ and $Y(Co_{1-x}Ni_x)_5$ (the latter taken from Ref. 14). It can be seen that in contrast to $T_C(x)$ for systems with nonmagnetic R, which have in the nickel-rich region $(0.78 < x < 1.0)$ a section with zero T_C, the Curie point on the $T_C(x)$ plot for $Tb(Co_{1-x}Ni_x)_5$ always differs from zero. We can estimate roughly the role of each of the interactions R-Co R-R, and Co-Co in the formation of T_C of alloys in the nickel-rich region. We assume that the R-R contribution to the overall T_C remains constant over the entire concentration interval and is equal to the value of T_C for the $TbNi_5$ alloy (28 K). If it is assumed that the exchange parameter $I_{R\text{-}Co}$ is the same in the entire concentration interval and that the contribution of the R-Co interactions to the resultant T_C of the alloy is determined by the mixing rule, i.e., it depends linearly on the number of atoms in the alloy, its value for the alloy with $x = 0.85$ should be 29 K (in fact, in accordance with Ref. 3, the total energy of the exchange bond of one Tb atom with five Co atoms is $I_{R\text{-}Co} = 145$ K, meaning 29 K per bond with one Co atom). The combined contribution to T_C from the R-R and R-Co interactions in this alloy is 57 K. If only the aforementioned three types of interaction are significant in the alloys, the difference 63 K between the measured T_C of the $TbCoNi_4$ alloy and the calculated R-R and R-Co contributions can be due only to the Co-Co interactions. These rough estimates suggest thus that in the nickel-rich region the Co-Co interactions in the $Tb(Co_{1-x}Ni_x)_5$ alloys make a substantial contribution to the overall exchange energy.

TEMPERATURE DEPENDENCES OF THE MAGNETIZATIONS OF THE RARE EARTH AND *d* SUBLATTICES

1. As shown in Refs. 3 and 8, the temperature dependence of the Co-sublattice magnetization in $TbCo_5$ depends on the orientation of the magnetization relative to the crystal axes. The fact that the magnetic moment of the Co sublattice depends on the magnetization direction (along or across the c axis) was established for the alloys YCo_2Ni_3 and YCo_3Ni_2 (Ref. 2) at 4.2 and 100 K and for YCo_5 (Ref. 5). That the modulus of the magnetic moment of cobalt depends on its orientation follows also from an analysis of the concentration dependence of the spontaneous magnetic moment in the $Tb_xY_{1-x}Co_5$ system at 4.2 K (Ref. 2).

The foregoing experiments indicate that the magnetic crystalline anisotropy of the sublattice plays an important role in the formation of the magnetization of this sublattice in compounds of the type RCo_5.

It was assumed in the cited references, in essence, that all the atoms of the Co sublattices are equally responsible for the observed effects. However, the Co atoms in the positions $2c$ and $3g$ are under different condtions (local symmetry), so that we can assume that the behavior and the properties of these atoms, as well as the role they play in the formation of the properties of the entire crystal, can differ substantially. There are published data that offer evidence in favor of these assumption. Analysis of the results of measurements on $Y(Co_{1-x}Fe_x)_5$, $Th(Co_{1-x}Fe_x)_5$ and YCo_5 has made it possible in Refs. 20–23 to conclude that it is just the Co atoms in the 2c positions which are responsible for the anisotropy of the entire d sublattice. Our present data lead to the same conclusion. Indeed, the magnetization temperature dependences $m_c(T)$ and $m_g(T)$ in $Tb(Co_{1-x}Ni_x)_5$ with $x = 0, 0.1$, and 0.4 with spin-reorientation transition differ greatly. Figure 7 shows for the alloy $TbCo_4Ni$, typical curves on which this effect is particularly pronounced. It can be seen that at temperature below the spin-orientational transition in this alloy the magnetization of the $2c$ sublattice varies with temperature much more rapidly than at temperatures above this

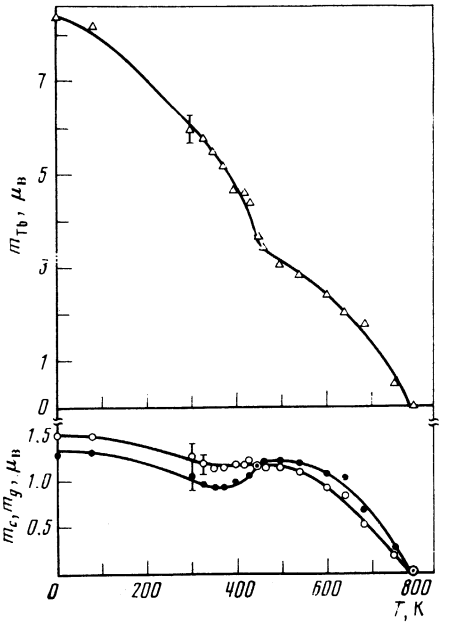

FIG. 7. Temperature dependences of the magnetizations of the R sublattice and of the two d sublattices in $TbCo_4Ni$: Δ — m_{Tb}; ○ — m_g; ● — m_c

transition, and the character of $m_c(T)$ as a whole can be understood only by assuming that the value of m_c in the case when the easy-magnetization axis of the crystal is parallel to the c axis $[m_c^{\parallel}(T)]$ is somewhat larger than the corresponding value $(m_c^{\perp}(T))$ when the easy magnetization axis is perpendicular to the axis, so that the $m_c^{\parallel}(T)$ plot lies higher than the $m_c^{\perp}(T)$ plot at all temperatures. The positive magnetization jump observed in the reorientation region corresponds to a transition from the $m_c^{\perp}(T)$ curve to $m_c^{\parallel}(T)$. The magnitude of the "jump" is $\Delta\ m_c \approx 0.2\ \mu_B$ per Co atom in the $2c$ sublattice, meaning $\sim 15\%$ of the maximum magnetization of the $2c$ sublattices ($\sim 1.3\ \mu_B$), which is reached at a temperature ~ 500 K. As shown in Ref. 24 with Ni as the example, the magnetization anisotropy in d metals is due to spin-orbit interaction. Comparison of Δm mesured for Ni and Fe in Ref. 25 with Δm obtained for the compounds YCo_2Ni_3 and YCo_3Ni_2, in which the anisotropy energy is larger by three orders of magnitude, has enabled the authors of Ref. 2 to conclude that Δm is determined by the ratio of the anisotropy and exchange energies (E_{an}/E_{exch}). According to Ref. 5, the Δm of the entire cobalt sublattice in YCo_3 is 3% of its magnetization. For only two atoms of the $2c$ position we obtain $\Delta m_c = 0.15\ \mu_B$, in good agreement with our resutls.

The plot of $m_g(T)$ is practically monotonic. The observed difference between the $m_c(T)$ and $m_g(T)$ curves is evidence that the magnetocrystalline anisotropy of the Co atoms in different positions differs greatly. In $2c$ positions it is apparently tremendously large, and in $3g$ sites it is much smaller. It can thus be assumed that practically the entire magnetization anisotropy of the d sublattice is due to the Co $2c$ atoms, which make also the dominant contribution to the magnetocrystalline anisotropy of this sublattice.

Figure 7 shows an experimental plot of $m_{Tb}(T)$ for $TbCo_4Ni$, with a form typical of compounds with $x = 0, 0.2$, and 0.4 (compounds with spin-reorientation transition). It can be seen from the figure that the experimental plots of m_{Tb} differ greatly on the right and left sides of the transition, thus pointing to a dependence of the rare-earth sublattice magnetization on the orientation of the vector m_{Tb} relative to the crystallographic axes. The m_{Tb} plot for alloys with $x \geqslant 0.6$, in which the easy-magnetization axis maintains the same direction in the entire region of existence of the magnetic order, contain no singularities.

A negative jump of the rare-earth sublattice magnetization is observed for the alloys with $x = 0$, 0.2, and 0.4 in the spin-reorientation region.

An expression for the rare-earth sublattice magnetization $m_R(T)$ as a functions of temperature in RCo_5 compounds is given in Ref. 1. An attempt to fit it to the experimental $m_{Tb}(T)$ curve in mixed alloys of the system $Tb(Co_{1-x}Ni_x)_5$ was not succesful. At all values of $I_{R\text{-}Co}$ and of K_R,[2] the calculated curve does not fit the experimental curves. This was to be expected, since the mentioned expression for $m_R(T)$ is valid only for systems similar to RCo_5, in which all the rare-earth atoms have identical suroundings. In $Tb(Co_{1-x}Ni_x)_5$ alloys the surroundings of the rare-earth element (number of Co atoms in the nearest surrounding of the Tb atoms), and consequently also its exchange binding to the Co sublattice, vary from atom to atom, a circumstance that must be allowed for when generalizing the expression obtained in Ref. 1 for $m_R(T)$.

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[1] Only the Curie temperature was determined for the alloy with $x = 0.5$.
[2] K_R is the anisotropy constant of the R sublattice.

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