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# Magnetic and Magnetostrictive Properties of Rare Earth Intermetallic Compounds

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***Abstract*—This work presents an overview of research results in magnetic and magnetostrictive properties of rare earths-based intermetallic compounds conducted by Russian and foreign researchers. A special attention is paid to multicomponent alloys, as variation in component concentration, temperature and external fields allows for successful influence over the exchange interaction, thus leading to unique phenomena in the area of phase transformations.**

***Keywords—intermetallic compounds, Laves phases, crystalline structure, magnetostriction***

## I. INTRODUCTION

Currently, development of modern areas of electronics, magnetic data recording, computing, micro and nanoelectronics, as well as other areas of technology and physics that use magnetic sensors, strongly demand production of a new type of magnetic materials. Currently, there is a pressing challenge in creating such magnetostrictive material that have high values of magnetostriction in a given range of temperatures and magnetic fields.

Production of new compositions requires a detailed analysis of many properties of the metals used in creating alloys with a predefined set of physical and chemical properties.

It is known, that intermetallic compounds may be satisfactory described as a system of two of more metals with a well-defined and simple stoichiometry.

Starting from the second half of the last century, much attention is given to so-called Laves phases, named after Fritz Laves, a German crystal chemist. These compounds are formed by atoms of two types, A and B, at that, the A-type atoms have a larger atomic radius than the B-type atoms. An ideal ratio of component radii is $R_A:R_B = 1.225$. Starting from 1960s, intermetallic compounds of rare earths with iron-group elements (Mn, Fe, Co and Ni). They combine a relatively simple structure with unique magnetic properties, such as giant magnetostriction [1] and large magnetocaloric effect [2]. First of all, it is $RT_2$, compounds, where Ris a rare earth element, T is a transition metal Fe, Co, Ni or Mn). As a rule, the $RT_2$ intermetallides lack a homogeneity area. Deviations form stoichiometry usually lead to appearance of a two-phase state, and in some cases to changes in the structural type. The structural phase transitions are usually accompanied with distortions of the atomic crystal structure. They are mainly caused by a spontaneous magnetostriction that in these intermetallides may reach giant values on the order of $10^{-3}$.

There are such intermetallic compounds among the $RT_2$ Laves phases, which together with a large value of spontaneous magnetostriction also demonstrate a giant magnetostriction under an external magnetic field. Such compounds find practical applications in underwater echo sounding, optronics, hydraulics and automation as ultrasound transducers, sensors and displacement sensors.

## II. METHODS AND MATERIALS

$RT_2$ type of stoichiometry is seen in two structural types [3]: Laves phase C15 with cubic symmetry and Laves phase C14 with hexagonal symmetry (Figure 1).

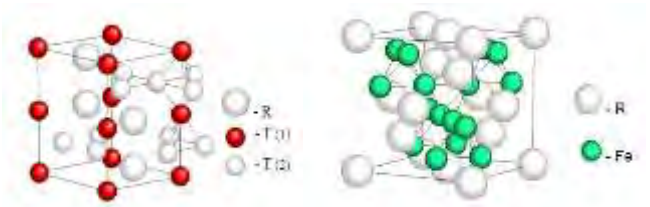


Fig. 1. Crystalline structure of Laves phases with a $RT_2$ stoichiometry: (a) hexagonal structure of C14, (b) cubic structure of C15.

The structure of intermetallic compounds considered in this work was studied with the methods of X-ray diffractometry. In particular, in [4], X-ray diffraction spectra obtained at various temperatures were processed with a Rietveld full-profile analysis in the Fullprof programming environment.

Magnetostriction measurements are conducted either with a dilatometer [4], whose operation principle is based upon measuring electrical capacitance changing with the size of the specimen, or by a strain gage (tensometric) method. The scope of an inquiry in all the studies was limited to 300 K.

As the authors of the works [4-14] state, all the studied alloys were single phase after annealing and had a structural type of Laves phase C15.

## III. Results

In [4], the authors studied spontaneous magnetostriction and that induced by an external magnetic field for two multicomponent alloys $Tb_{0.3}Dy_{0.35}Ho_{0.35}Co_2$ and $Tb_{0.8}Dy_{0,1}Gd_{0.1}Co_2$. These alloys demonstrate a high level of magnetostrictive deformations in the area of weak magnetic fields, mainly due to compensation of magnetic anisotropy in their rare earth sublattice [5]. To reveal phase transitions of the order-chaos type in alloys $Tb_{0.3}Dy_{0.35}Ho_{0.35}Co_2$ and $Tb_{0.8}Dy_{0,1}Gd_{0.1}Co_2$ , a thermal range of 110 – 250 K was selected.

In the work, it has been established that with decreased temperature in both of the studied alloys ($Tb_{0.8}Dy_{0.1}Gd_{0.1}Co_2$ and $Tb_{0.3}Dy_{0.35}Ho_{0.35}Co_2$) there is a transition from cubic to rhombohedric phase. Table 1 shows the structural parameters and Curie temperature values of the studied alloys.

TABLE 1. Curie temperature and parameters of unit cell of the studied intermetallides and $TbCo_2$

| Alloy | Tc, K | T> Tc | T < Tc | |
|---|---|---|---|---|
| | | $a_0$, A | a, A | c, A |
| $TbCo_2$ | 240 | 7.209 | 5.09 | 12.514 |
| $Tb_{0.8}Dy_{0.1}Gd_{0.1}Co_2$ | 243 | 7.208 | 5.107 | 12.459 |
| $Tb_{0.3}Dy_{0.35}Ho_{0.35}Co_2$ | 148.5 | 7.189 | 5.091 | 12.437 |

It is evident from the table that the parameters obtained correlate well with the data known from literature for compounds of similar composition.

The values of spontaneous volumetric magnetostriction that appears at transition from magnetically unordered to magnetically ordered state in the vicinity of the Curie temperature were evaluated by the authors with the following expression [6]:

$$\omega_S = (V_m(T) - V_p(T))/V_P(T),$$

where $V_m$ is the volume of the unit cell at a given temperature, $V_p$ is the volume of the unit cell in the paramagnetic area.

It has been established that in the transition area the value of spontaneous magnetostriction of the $Tb_{0.3}Dy_{0.35}Ho_{0.35}Co_2$ alloy comprises about $\omega_S \approx 5 \cdot 10^{-3}$, which is larger than that of the $Tb_{0.8}Dy_{0.1}Gd_{0.1}Co_2$ alloy ($\omega_S \approx 2 \cdot 10^{-3}$). For a more precise determination of the Curie temperature in the studied compounds, a thermomagnetic analysis was conducted in the temperature range of 4.2–300 K in a constant magnetic field of 0.04 T. The results are given in Figure. 3.

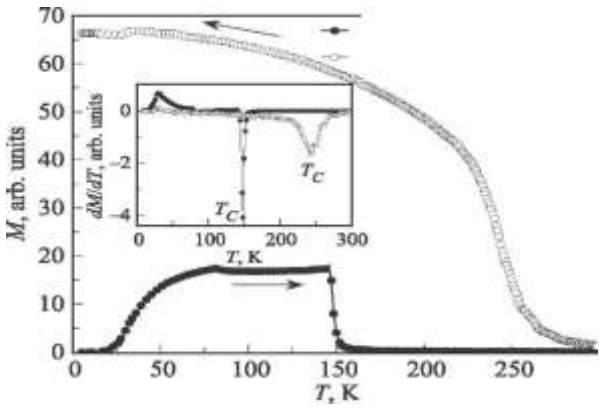


Fig. 3. Thermomagnetic analysis of the $Tb_{0.3}Dy_{0.35}Ho_{0.35}Co_2$ and $Tb_{0.8}Dy_{0.1}Gd_{0.1}Co_2$ alloys. In the insert, there is a thermal dependence of magnetization derivative dM/dT.

As it is evident from the figure, with increased temperature magnetization increases practically to zero and reaches its maximum value that is then kept up to the $T_C$, where there is an abrupt drop in magnetization observed due to a transition to a paramagnetic state.

When measuring during cooling, the external magnetic field impedes fixation of the domain walls, leading to an increase in magnetization with reduction of temperature. Besides, transition from magnetically-ordered state to unordered one is intermittent in $Tb_{0.3}Dy_{0.35}Ho_{0.35}Co_2$ and gradual in $Tb_{0.8}Dy_{0.1}Gd_{0.1}Co_2$, which is an evidence of different types of magnetic phase transition.

Professor A.S. Ilyushin et al in this work used a curve differentiation method for *M(T)* to determine the values of the Curie temperatures. These data are also shown in Table 1. The work has revealed that in the low temperature range (50-80 K) there are thermal anomalies on the thermal dependence curves *M(T)* and *dM/dT(T)*, which most probably witness the spin flipping in these compounds in the stated temperature range, that is, there are additional order-disorder transitions.

Work [4] also studied changes in the value and sign of magnetostriction of $Tb_{0.3}Dy_{0.35}Ho_{0.35}Co_2$ and $Tb_{0.8}Dy_{0.1}Gd_{0.1}Co_2$ specimens depending on temperature and application of external magnetic field. All the stated dependences demonstrate their maximum values at the Curie temperature, including voluminous magnetostriction. The volumetric magnetostriction attains its maximum value at $T_c$

equal to $1.2 \cdot 10^{-3}$ in the field of 8 T. An exception is anisotropic magnetostriction $\lambda_{anis}$ (T), which attains its maximum value not in the vicinity of the Curie temperature, but in the range of temperatures between 50 and 80 K, which is related to a spin reorientation taking place in this range (Fig. 4).

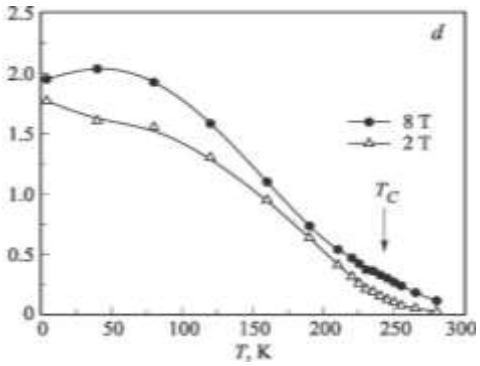


Fig. 4. Thermal dependences of magnetostriction of the $Tb_{0.8}Dy_{0.1}Gd_{0.1}Co_2$ alloy under external magnetic fields of 2 and 8 T.

Giant anisotropic magnetostriction is observed in $RCo_2$ compounds under low temperatures not only due to localization of 4*f*-magnetic moment of the rare earth atoms, but also, as it is shown in [6], due to cobalt atoms having a localized portion of spin density of 3*d*-electrons. Due to the above, a comparison between the anisotropic and volumetric magnetostriction in the low temperature range is especially interesting, as well as that in the range of transition from the magnetically-ordered to the magnetically-disordered state. For the studied $Tb_{0.8}Dy_{0.1}Gd_{0.1}Co_2$ alloy, field dependences of longitudinal and transversal magnetostriction under various temperatures from 4.2 to 275 K in the magnetic field of up to 8 T and the values of $\lambda_\omega(H)$ and $\lambda_{anis}(H)$ were calculated. From the Figure 5, it is evident, that in a wide range of magnetic fields from 2 to 8 T at a $T$ = 50 K, the anisotropic magnetostriction exceeds the volumetric magnetostriction by a factor of 4. In the vicinity of the Curie temperature, the ratio between the volumetric and anisotropic magnetostriction is 3:1.

For the $Tb_{0.3}Dy_{0.35}Ho_{0.35}Co_2$ compound this ratio was equal to 10:1, which is an evidence of significant fluctuation processes in the vicinity of Tc.

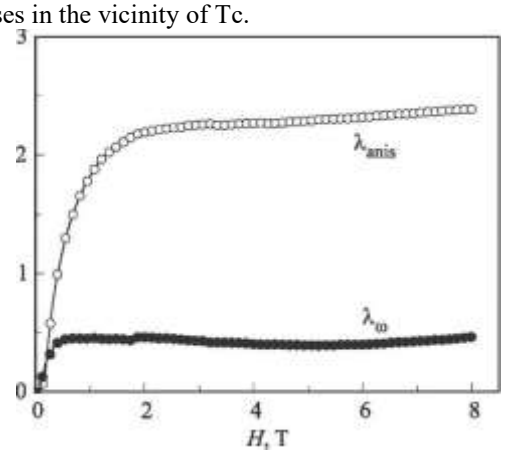


Fig. 5. Field dependences of volumetric $\lambda_\omega$ and anisotropic $\lambda_{anis}$ magnetostriction of the $Tb_{0.8}Dy_{0.1}Gd_{0.1}Co_2$ alloy at a temperature of 50 K.

An important place among the intermetallic compounds of rare earths with the iron-group metals is taken up by heat-resisting multicomponent nickel-based alloys. Nickel, that has high strength and corrosion-related characteristics, has another advantage in comparison with other metals, which is that its alloying with significant amounts of other elements does not cause formation of phases that degrade the mechanical characteristics of alloys. Thus, research into physical properties of nickel-based alloys, their structure and multitude of structural phase transitions is a priority for condensed-matter physics in general and for aerospace industry in particular.

N.V. Mushnikov et al [7] studied the structural and magnetic features of rare earth intermetallides $TbNi_2Mn_x$ in a concentration range $0 < x < 1.5$. In such alloys, the range of cubic phase with vacancies in $Tb_{1-x}Ni_2$ compounds is $0<x<0.05$.

Recently, it has been discovered [8] that $RNi_2Mn$ alloys with $R$ = Tb, Dy, Ho and Er crystallize into a $MgCu_2$-like structure, as $RNi_2$ and $RMn_2$. The Mn atoms variably take both rare earth positions and those of Ni, a phenomenon that has never been registered before for rare earth Laves phases. The $RNi_2Mn$ alloys are ferromagnetics. The highest value of $T_C$ = 131 K was observed for $TbNi_2Mn$. Spontaneous magnetic moment of $TbNi_2Mn$, determined from magnetization measurements amounted to 4.8 $\mu_B$ per formula unit [8], which is significantly less than the values $gJ\mu_B = 9\mu_B$ for the $Tb^{3+}$ ion. Observed low value of magnetic moment of $RNi_2Mn$ was interpreted in this work in an assumption that the Ni atoms are practically devoid of the magnetic moment, while the Mn atoms have a large magnetic moment directed in the opposite direction to the moment of rare earth atoms.

However, the research results given in [9] showed that Ni in these compounds has a magnetic moment of about 0.3 $\mu_B$/atom. Presence of magnetic moment at the nickel atoms is atypical of R-Ni compounds with high concentration of R-element, where usually the valence electrons of the R-atoms completely fill up the nickel's d-zone. Studies of the $Gd_XY_{1-X}Ni_5$ system have shown that nickel is non-magnetic in $YNi_5$, while on introduction of gadolinium there appears an induced magnetic moment of up to 0.16 $\mu_B$/atom of Ni [10].

As manganese partially substitute both nickel and R-atoms in the cubic structure of $RNi_2$, then, changing the manganese concentration may be expected to lead to production of new compounds with different magnetic properties, including a higher Curie temperature. The most important for $TbNi_2Mn_X$ -like compounds are their magnetoelastic properties as a function of manganese concentration.

One of the main practical applications of rare earth intermetallides with the structure conforming to Laves phases is in magnetostrictive materials. The $TbNi_2$ compound has a giant magnetostriction, just as $TbFe_2$, but only at low temperatures. Thus, it is of interest to study the influence of manganese concentration onto magnetostriction of TbNi2Mnx compounds. In Figure 6 there are loops of longitudital $\lambda\|$ and transversal $\lambda\perp$ magntostriction of $TbNi_2$ and $TbNi2Mn_{0.5}$ specimens as measured at 4.2 K. For a specimen with x = 0, the

value of the anisotropic magnetostrictive deformation is ($\lambda\| - \lambda\perp$) = $2.4 \cdot 10^{-3}$, which supports the findings of [11].

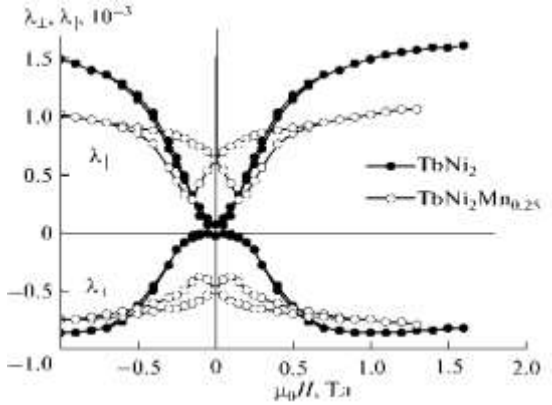


Fig. 6. Loops of longitudinal and transversal magnetostriction of $TbNi_2$ and $TbNi_2Mn_{0.25}$ at 4.2 K.

The value of the transversal magnetostriction is about half of that of the longitudinal magnetostriction. This is typical for isotropic polycrystalline cubically-symmetric specimens.

The loops of magnetostriction of the TbNi2Mn0.5 sample show a significant difference in form from those of the initial TbNi2 compound. For them the ratio $\lambda\| = 2\lambda\perp$ no longer holds. The magnetic hysteresis is clearly seen in the magnetostriction loops. It should be noted, that in the weak field are the curves of $\lambda\|$ and $\lambda\perp$ do not close. Thus, the magnetic state (type of domain structure) depends on magnetic history. The value of anisotropic magnetostrictive deformation ($\lambda\| - \lambda\perp$) does not depend on the magnetic texture of the specimens. However, when manganese is added, the magnetostriction value decreases monotonically. As giant magnetostriction of rare earth intermetallides is determined by interaction of anisotropic *f*-electron cloud of a rare earth atom with the crystalline field of the lattice, one of the causes for reduced magnetostriction is a gradual reduction of Tb concentration in the samples. However, when moving from $TbNi_2$ to $TbNi_2Mn$, the terbium concentration changes only from 33 to 25 at%, while the magnetostriction changes more than threefold.

Magnetic anisotropy may be spontaneous as well. Intermetallic compounds of rare earths with iron $RFe_2$, that have a cubic Laves phase structure similar to $MgCu_2$ have a large spontaneous anisotropic magnetostriction at room temperature. Spontaneous anisotropy leads to distortions of the crystalline lattice so noticeable at a temperature below the magnetic ordering temperature, that they may be registered with an X-ray method. Spontaneous magnetoelastic lattice distortions in compounds with R atoms that have a non-zero orbital moment are caused by form anisotropy of the 4*f*-electron shell of the rare earth ions. When an external magnetic field is applied, change in orientation of the 4*f* shell with respect to surrounding ions creating the crystalline field causes giant magnetostrictive deformation of lattice that are used in practice in various magnetostrictive sensors.

Due to a single-ion nature of interactions between the 4*f* ions and crystalline field of the lattice, in quasibinary systems of the type $(R^I_{1-x}R_x^{II})Fe_2$, where $R^I$ and $R^{II}$ are rare earth metals with magnetostriction constants of the opposite sign, e.g., Er and Tb, when passing through a critical concentration, there is a change of sign of the magnetostriction constant $\lambda_{111}$ without a change in the type of anisotropy.

L.A. Stashkova et al [12] presented the results of X-ray research into structure of $Er_{1-x}Tb_xFe_2$ ($0 \le x \le 0.6$) compounds and their hydrides. Ordering of hydrogen, absorbed by $RFe_2$ compounds may cause a crystalline lattice deformation that is significantly larger than the magnetoelastic deformation.

Figure 8 shows experimental and design X-ray diffraction pattern of the $Er_{0.8}Tb_{0.2}Fe_2$ alloy. Diffraction patterns of other compounds have qualitatively similar look.

The studied compounds have a ferrimagnetic structure below their Curie temperature, which is 590 K for $ErFe_2$ and 698 K for $TbFe_2$. Both compounds demonstrate giant magnetostriction. For the $Er_{1-x}Tb_xFe_2$ system at low temperatures, one shall expect a linear change in magnetostriction depending on concentration *x*. At the room temperature, the linearity may be somewhat disturbed due to a difference in Curie temperature between $ErFe_2$ and $TbFe_2$. In linear approximation, the $Er_{0,89}Tb_{0,11}Fe_2$ compound shall correspond to the magnetostriction compensation point. Except for this compound, the crystalline lattice of all the compounds is distorted at a room temperature due to magnetoelastic interactions and is a pseudocubic lattice. Indeed, the detailed analysis of X-ray line profiles shows that there are rhombohedric distortions of structure.

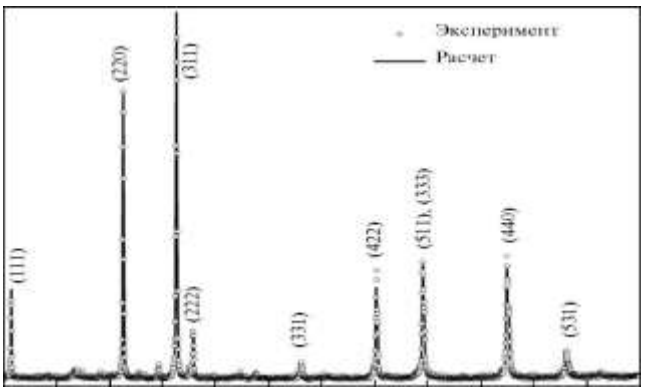


Fig. 8. Powder pattern of the $Er_{0,8}Tb_{0,2}Fe_2$ alloy: experiment (points) and calculations (solid line).

Figure 9 shows a concentration dependence of the magnetostriction constant $\lambda_{111}$ of the $Er_{1-x}Tb_xFe_2$ alloys plotted from the experimental values of magnetostriction for boundary compounds (dark squares) with considerations for the single-ion nature of changes in the magnetostriction value depending on concentration (dash line) and from the results of [12] (light squares).

From the $\lambda_{111}$ (*x*) dependence it is evident, that magnetostriction compensation is in the vicinity of the

$Er_{0.89}Tb_{0.11}Fe_2$ compound, that is, in the area of terbium concentration $x$ = 0.11, the magnetostriction constant $\lambda_{111}$ changes its sign from negative to positive.

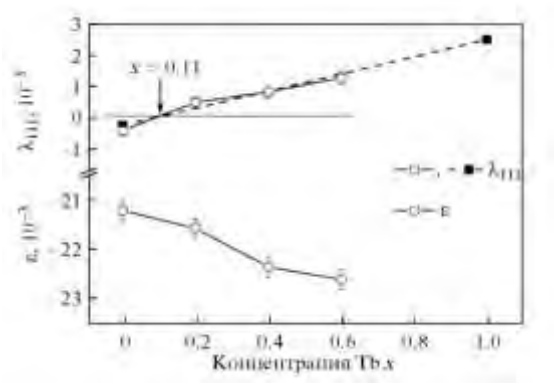


Fig. 9. Dependence of the magnetostriction constant $\lambda_{111}$ and the value of rhombohedric distortions ε in the system of $Er_{1-x}Tb_xFe_2$ alloys depending on terbium concentration.

Together with the change of sign of the magnetostriction constant as seen in the latter work, a significant interest is recently paid to the phenomenon of spin reorientation. This phenomenon was studied by professor S.A. Nikitin [13] and his disciples on a $DyFe_{11}Ti$ monocrystal by measuring angular and field dependences of rotating mechanical momentum acting on a specimen put in a magnetic field. It has been shown, that in this compound there are two spin-reoriented phase transitions (SRT): a second order transition at $T_1$ = 250 K and a first order transition at $T_2$ = 122 K.

New data on the nature of these transitions may be obtained by studies of magnetoelastic effects. With this aim, the authors of [13] conducted a study of magnetostriction in the SRT area on a $DyFe_{11}Ti$ monocrystal using a strain gage method.

Figure 10 shows longitudinal ($\lambda_{\parallel}>0$) and transversal ($\lambda_{\perp}<0$) magnetostriction as a function of temperature. The strain sensor was glued to the surface of the disk in parallel to the *c* axis. The magnetic field was applied in parallel ($\lambda_{\parallel}$) and perpendicularly ($\lambda_{\perp}$) to the **c axis.** It is evident, that during the heating the specimen in the range of 78-100 K, there is a sharp increase of $\lambda_{\parallel}(T)$ observed. The maximum value of magnetostriction is reached in the temperature range of 100-140 K. Spin reorientation first order phase transition is also observed in the same range.

From this figure, it is evident, that the longitudinal magnetostriction in the field **H**||**c** is maximal in the vicinity of $T_2$. Here, the efficient field of magnetic anisotropy is small and the external magnetic magnetic field applied along the **c axis** causes a rotation of magnetic moments out of the basic plane ($\theta = 90°$) in a direction of the magnetic field, which leads to a strong increase in magnetostriction. When the field is strengthened, the maximum at the $\lambda_{\parallel}$ (T) curve moves in the direction of lower temperatures, as the transition temperature $T_2$, apparently lowers with the field increase.

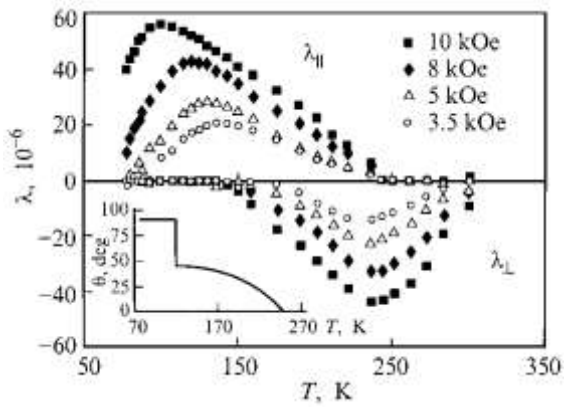


Fig. 10. Longitudinal $\lambda_{\parallel}$ and perpendicular $\lambda_{\perp}$ magnetostriction of $DyFe_{11}Ti$ as a function of temperature in various magnetic fields.

Field induced magnetostriction is also very important, its isotherms are shown in Fig. 11. It is evident, that at the temperature T = 78 and 86 K there is no saturation on the magnetostriction isotherms $\lambda_{\parallel}(H)$, pointing to a higher value of the magnetic anisotropy.

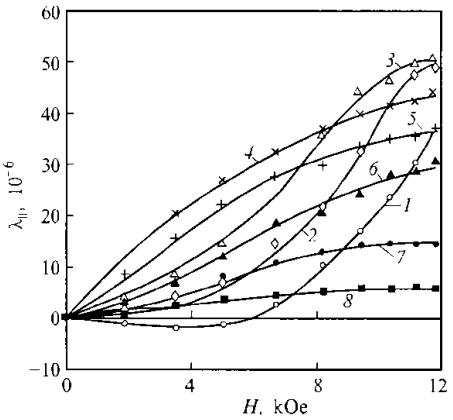


Fig. 11. Isotherms of longitudinal magnetostriction $\lambda_{\parallel}$ in a $DyFe_{11}Ti$ monocrystal. T, K: *1* — 78, 2 — 86, *3* — 100, *4* — 143, *5* — 158, *6* — 190, 7 — 221, *8* — 236.

Spontaneous magnetostriction along the **c** axis of the crystal with tetragonal crystalline lattice was calculated with the formula:

$$\lambda_c=\lambda_2^{\alpha,0}+\lambda_2^{\alpha,2}(\cos^2\theta-1/3), \quad (1)$$

where $\lambda_2^{\alpha,0}$and $\lambda_2^{\alpha,2}$ are magnetostriction constants, and θ is an angle between the magnetic moment $\mathbf{M_s}$ and the tetragonal **c** axis.

Experimental research of the authors of [13] and their theoretic calculations allow for a conclusion that magnetostriction in $DyFe_{11}Ti$ in the SRT range is due to processes of magnetic moment rotation of a rare earth metal sublattice under the influence of the field. The main microscopic mechanism of magnetostriction in this monocrystal is, apparently, the single-ion magnetostriction

determined by interaction between the anisotropic electron shell of the Dy3+ magnetic ion and crystalline field of the lattice.

In connection with spin reorientation phase transitions, studies of a $Sm_xTb_{1-x}Fe_2$ quasibinary system appear as especially interesting for the following compositions x = 0; 0.1; 0.2; 0.25; 0.3; 0.4; 0.5; 0.75; 1.0 [14]. It is known, that the initial compound in this system, $SmFe_2$ is a ferromagnetic with a Tc = 700 K, while $TbFe_2$ is a ferrimagnetic with a Tc = 711 K. Their atomic crystalline structure is isotypic with the cubic Laves phase C15. However, analysis of the results of X-ray examination has shown that the cubic structure of all the alloys underwent rhombohedric distortions already at a room temperature. At that, in the concentration range from x = 0 to x = 0.4, the values of the rhombohedric distortions $\varepsilon_{rh}$ have the negative sign, that is, $\varepsilon_{rh}<0$. In alloys whose composition is within the range from $x = 0.5$ to $x = 1$, the cubic cell C15, unlike in the previous case, is elongated along the [111] direction, so that $\varepsilon_{rh}>0$. It was also established that in the range of terbium concentration $x = 0.45$-0.5, there is a structural phase transition from the rhombohedric phase to the purely cubic one, that is, there is a distortive structural phase transition. At that, the stated concentration of $x \approx 0.45$ is the point of magnetic phase transition between ferromagnetism and ferrimagnetism. It is the area of these concentrations, where one may produce a compound with an almost compensated magnetocrystalline anisotropy, and thus, the giant magnetostriction may be expected. The study has shown that there is a clear relation between the type f atomic crystalline lattice distortion and the type of magnetic order. The thermal studies of alloys pertaining to this system have shown that in the temperature range from 5 to 300 K in the concentration range of $x < 0.45$, two spin reorientation phase transitions take place in the system (Fig.12) from the state with the direction of the axis of easy magnetisation along the crystallographic direction [111] to the direction [110].

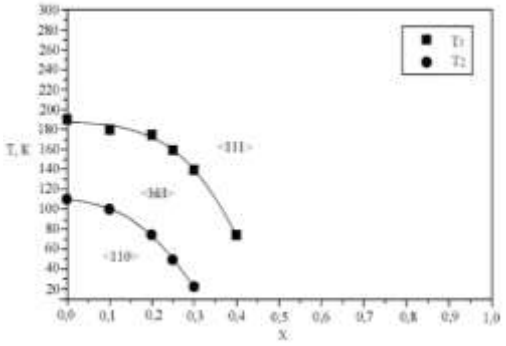


Fig. 12. Magnetic spin reorientation phase diagram of the $Sm_{1-x}Tb_xFe_2$ system.

Spin reorientation in the alloys of this system is also accompanied with appearance of angular magnetic phases (hkl), leading to complex distortions of their atomic crystalline structure and reduction in crystal symmetry [15].

## IV. Conclusion

The studies of magnetic and magnetostrictive properties of rare earth intermetallides presented in the review allow considering a fundamental problem of the modern condensed stated physics, namely, finding relations between the composition, structure and physical properties of a compound.

Research into relationships between the crystalline structure properties of Laves phases with their magnetic properties and their correlation in the area of magnetic phase transitions for multicomponent systems with the Laves phase-like structure allows for a targeted search of a new type of magnetic materials with optimal physicochemical properties.

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