Magnetization of alloys of \((\text{Tb}_{1-x}\text{Y}_x)_{0.8}\text{Sm}_{0.2}\text{Fe}_2\) system in stationary and pulsed magnetic fields

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Abstract. The paper presents the results of studying the alloy magnetization of the multicomponent system \((\text{Tb}_{1-x}\text{Y}_x)_{0.8}\text{Sm}_{0.2}\text{Fe}_2\). The substitution parameter in this system takes the following values: \(x = 0, 0.2, 0.4, 0.6, 0.8, 1\). In the system, the atoms of magnetic terbium were replaced by atoms of weakly magnetic samarium and nonmagnetic yttrium. X-ray diffraction analysis determined the structure and lattice constant for all compositions. Phase analysis was carried out. It is known that by varying the concentration of components, temperature, and external fields, one can successfully influence the magnitude of exchange interactions leading to one or another type of magnetic ordering in the rare-earth intermetallic metals. Therefore, the main goal in this work was to investigate the possibility of observing the magnetic phase transition – ferrimagnetism – ferromagnetism – in the alloys of the system from the curves of the field dependence of magnetization in strong fields up to 60 T. The studies were carried out at the liquid helium boiling point. The main magnetic characteristics of the alloys of this system were determined, such as saturation magnetization \(\sigma_s\), magnetic moment \(\mu\) and magnetic moment \(\mu_{\text{Fe}}\) on iron atoms. Their dependences on the concentration of yttrium were established. The possible values of the magnetic moment per formula unit in the case of ferromagnetic ordering of these alloys were calculated theoretically. It was concluded that these fields are insufficient to observe the ferrimagnetism – ferromagnetism phase transition.

1. Introduction

Intermetallic compounds based on the rare earth metals are widely known in science and technology due to their unique magnetic properties. Among the numerous rare earth intermetallic compounds, a special place is given to \(\text{RE}\text{Fe}_2\), where \(\text{RE}\) is a rare earth element. Compounds of this stoichiometry belong to Laves phases and are presented in two structural types: cubic structure of \(\text{MgCu}_2\) (structural type C15) and hexagonal structure of type \(\text{MgZn}_2\) (structural type C14) [1]. They combine a relatively simple crystal structure and important magnetic properties such as high Curie temperatures, giant magnetostriction, and a large magnetocaloric effect [2, 3].

Most of the rare earth intermetallic compounds are in a magnetically ordered state within a certain temperature range [4]. The exchange interactions are one of the main reasons for magnetic ordering appearance in rare earth metal (REM) intermetallic compounds. As is known, the exchange
interactions can have different signs, they can be both positive and negative. Depending on the sign of the integral of the exchange interaction in magnets, a ferro- or ferrimagnetic order of arrangement of the magnetic moments of the atoms that make up one or another magnet is implemented. Most often, they are presented in the form of two-sublattice magnets, where one sublattice consists of rare-earth R-ions, and the other consists of 3d-transition ions. Most RFe2 compounds have collinear magnetic structures where the magnetic moments of R and 3d sublattices are parallel to each other. Moreover, for light rare earth elements, the total magnetic moments \( M_R \) and \( M_{3d} \) of the rare earth and 3d transition sublattices are forwarded to the same direction; therefore, the total magnetic moment is equal to \( M_{RFe2} = M_R + 2M_{3d} \) and we are dealing with ferromagnetism.

For heavy rare-earth elements, the total magnetic moments \( M_R \) and \( M_{3d} \) of the rare-earth and 3d sublattices are antiparallel; therefore, the total magnetic moment is equal to the ratio \( M_{RFe2} = M_R - 2M_{3d} \) and, therefore, these are ferrimagnets. For some compounds, it is possible to observe a magnetic phase transition from a ferrimagnetic state to a ferromagnetic state in appropriate fields. This effect is created by applying a strong magnetic field, when all magnetic moments tend to line up parallel to the applied field, as shown in Fig. 1.

The study of the magnetism nature, the mechanisms of magnetization and the determination of the main magnetic characteristics is a rather urgent problem in the physics of magnetic phenomena. Therefore, the purpose of this work was to use strong magnetic fields, statistical and pulsed, to observe the processes of rotation of the magnetic moments of individual sublattices, as well as to observe the phenomenon of a ferromagnetic state induced by an external magnetic field.

![Figure 1. Magnetic structure of ferrimagnet without external field application (a) and in a strong magnetic field inducing a ferromagnetic state (b)](image)

At the moment, the maximum value of the magnetic field strength available to scientists is about 100–120 T [5]. Such experiments are one-time experiments. Besides, they are expensive due to their technical complexity. Experiments in the fields up to 60 T are relatively accessible and are being implemented in several laboratories around the world [6, 7]. However, papers describing such experiments are not often found in the literature. The importance of this kind of research stems from the fact that REM intermetallic compounds are widely used in various branches of electrical engineering, optoelectronics, computer technology, micro- and nanoelectronics, and also as small displacement sensors. The most important thing is that there is an acute problem of obtaining magnetic materials of a new type with controlled set of physical and ochemical parameters.

2. Methods and Materials
The objects of research in this work are intermetallic alloys of rare earth metals with iron of the type \( (Tb_{1-X}Y_X)_{0.8}Sm_{0.2}Fe_2 \) synthesized on the basis of TbFe2. The system studied during this investigation is
multicomponent. As mentioned above, magnetically active terbium atoms are replaced or substituted by nonmagnetic samarium atoms in a stationary concentration of 20 % and then terbium atoms in the rare-earth sublattice are substituted by nonmagnetic yttrium atoms. The substitution parameter in this system is \( x = 0, 0.2, 0.4, 0.6, 0.8, 1.0 \).

The synthesis of alloys was carried out on the basis of high-purity rare-earth metals and iron (99.95 %) in an arc furnace with a non-consumable tungsten electrode on a water-cooled copper hearth of a special design in an atmosphere of purified inert gas (argon) at normal pressure. Then the samples were subjected to homogenizing annealing for two weeks.

X-ray spectra were recorded at room temperature using Panalytical Emryren diffractometer with copper anode (operating modes \( I = 40 \) mA, \( U = 40 \) kV), in the Bragg-Brentano geometry at a pitch of 0.026°, in the angle range from 5° to 140° using two-dimensional detector Pixel3D, a system of variable slits, a nickel filter on a diffracted beam. The diffraction patterns contained Cu – K\(_{a1}\) and Cu – K\(_{a2}\) emission lines. The unit cell parameters were determined from the reflections in the angular range \( 2\theta = 15–105° \). According to our data, with an increase in the concentration of yttrium in the compounds \( \text{Tb}_{1–x}\text{Y}_x\text{Sm}_{0.2}\text{Fe}_2 \), the cubic lattice parameter increased slightly linearly.

The phase composition of the samples was investigated using the Rietveld analysis in the Powred Cell 2.4 program. The analysis showed that only the extreme in this system alloys \( \text{Tb}_{0.8}\text{Sm}_{0.2}\text{Fe}_2 \), \( \text{Y}_{0.8}\text{Sm}_{0.2}\text{Fe}_2 \), for which the substitution parameter \( x = 0 \) and 1.0, respectively, are single-phase and have a cubic structure of the Laves C15 phase. This structure belongs to the space group Fd\(_{3m}\)–Oh.

In more complex compositions \( \text{Tb}_{0.8}\text{Y}_{0.2}\text{Sm}_{0.2}\text{Fe}_2 \), \( \text{Tb}_{0.6}\text{Y}_{0.4}\text{Sm}_{0.2}\text{Fe}_2 \), \( \text{Tb}_{0.4}\text{Y}_{0.6}\text{Sm}_{0.2}\text{Fe}_2 \) (the value of the substitution parameter for which is \( x = 0.2, 0.4, 0.6 \), and 0.8, respectively), a small content of the second phase with a crystal structure of the PuNi\(_3\) type, belonging to the space group R-3m, was observed, the content of which varied from 8 to 10 %.

The investigations of the magnetization of compounds were carried out using two methods. In the first method, the magnetization of compounds was measured in static magnetic fields on using standard PPMS-14 magnetometer (Quantum Design, USA) in the fields up to 14 T.

In the second method, the magnetization was measured in high pulsed fields up to 60 T using special equipment in a laboratory of high magnetic fields (EMFL, Dresden, Germany). A magnetic field pulse was generated in the solenoid by a discharge of electricity accumulated in a bank of capacitors with a total capacity of 1.44 MJ. The rise time of the magnetic field in the sample area is 7 ms, the total pulse time is about 25 ms. Magnetization was measured by integrating the voltage generated in a precisely compensated coil system surrounding the sample.

All measurements of magnetization in this work were carried out at a boiling point of liquid helium of 4.2 K.

3. Experimental Results

The alloys of this system are based on the \( \text{TbFe}_2 \) compound. It is interesting that it has a cubic crystal structure with rhombohedral distortions. It also has the highest Curie temperature among the known rare earth intermetallic compounds of stoichiometry \( \text{RFe}_2 \). Curie temperatures are the temperature of the magnetic phase transition from an ordered ferromagnetic state to a disordered, paramagnetic state. Based on the literature data [4], the Curie temperature of the \( \text{TbFe}_2 \) compound is \( T_c = 711 \) K, the magnetic moment per formula unit is \( \mu = 1.74 \mu_\text{B} \).

In the \( \text{Tb}_{1–x}\text{Y}_x\text{Sm}_{0.2}\text{Fe}_2 \) system, the yttrium and samarium are introduced into the rare-earth terbium sublattice, which affect the properties and the main magnetic characteristics of the \( \text{TbFe}_2 \) compound depending on the substitution parameter \( x \). Earlier, in stationary magnetic fields up to 14 kOe, there were discovered a number of phenomena, such as the phenomenon of spin reorientation, the phenomenon of magnetic compensation, and the inversion of the sign of the magnetostriction constants [8-10] depending on the parameter \( x \). The main magnetic characteristics of alloys in the indicated fields are given in Table 1.
Table 1. Main magnetic characteristics of alloys (\(\text{Tb}_{1-x}\text{Y}_x\text{Sm}_{0.2}\text{Fe}_2\))

| \(x\) | Alloys/Properties | \(G_s\), emu/q | \(M,M_B\) | \(T_c\), K |
|---|---|---|---|---|
| 0 | \(\text{Tb}_{0.8}\text{Sm}_{0.2}\text{Fe}_2\) | 74 | 3.56 | 672 |
| 0.2 | \(\text{Tb}_{0.64}\text{Y}_{0.16}\text{Sm}_{0.2}\text{Fe}_2\) | 48 | 2.20 | 613 |
| 0.4 | \(\text{Tb}_{0.48}\text{Y}_{0.32}\text{Sm}_{0.2}\text{Fe}_2\) | 33 | 1.47 | 601 |
| 0.6 | \(\text{Tb}_{0.32}\text{Y}_{0.48}\text{Sm}_{0.2}\text{Fe}_2\) | 11 | 0.47 | 576 |
| 0.8 | \(\text{Tb}_{0.16}\text{Y}_{0.64}\text{Sm}_{0.2}\text{Fe}_2\) | 50 | 2.02 | 568 |
| 1 | \(\text{Y}_{0.8}\text{Sm}_{0.2}\text{Fe}_2\) | 64 | 2.45 | 539 |

The investigation of alloy magnetization (\(\text{Tb}_{1-x}\text{Y}_x\text{Sm}_{0.2}\text{Fe}_2\)) in static fields up to 14 T was also carried out at a temperature of 4.2 K. Fig. 2 shows field dependences of the magnetization \(M(H)\). These dependences for all alloys of the system show that the \(M(H)\) curves reach saturation rather quickly. The saturation magnetization values were calculated for these compositions by extrapolating the \(M(1/H)\) dependence to the region of high fields [11].

The magnetic structure of (\(\text{Tb}_{1-x}\text{Y}_x\text{Sm}_{0.2}\text{Fe}_2\)) compounds can be considered in the model of three sublattices of terbium, samarium, and iron, the magnetic moments of which are collinearly oriented. The magnetic moment on the atoms of the heavy rare earth metal Tb is \(9 \mu_B\) and is ordered antiparallel to the magnetic moment of iron, while the magnetic moment on the atoms of the light rare earth metal of samarium is \(0.7 \mu_B\) [11] and is ordered in the same direction as the magnetic moment of iron. By diluting the rare-earth sublattice with non-magnetic rare-earth metals with yttrium, one can observe the phenomenon of magnetic compensation and find the compensation composition.

To determine the concentration value of yttrium in the compensation composition, theoretical calculations were carried out. Since the magnetic moments of the sublattices (Tb, Sm, Fe) can be considered collinear, the value of the total magnetic moment is determined by the following formula:

\[
\mu_{\text{calc}} = 2\mu_{\text{Fe}} + \mu_{\text{Sm}} - \mu_{\text{Tb}} \tag{1}
\]

where \(\mu_{\text{Sm}} = 0.2 \cdot 0.7 \mu_B = 0.14 \mu_B\) is the magnetic moment of the sublattice Sm [15];

\(\mu_{\text{Tb}} = 0.8 \cdot (1-x) \cdot 9 \mu_B\) is the magnetic moment of the sublattice Tb depending on the substitution parameter \(x\);

\(\mu_{\text{Fe}} = 1.45 \mu_B\) is the magnetic moment of the sublattice Fe defined on the basis of magnetization compound YFe\(_2\) [12, 13].

Figure 2. Field dependences of magnetization of (\(\text{Tb}_{1-x}\text{Y}_x\text{Sm}_{0.2}\text{Fe}_2\)) compounds at a temperature \(T = 4.2\) K in stationary fields up to 14T
Applying the values of magnetic moments of the sublattices to the formula (1), a linear dependence of total magnetic moment on the concentration of yttrium is obtained

\[ \mu_{\text{calc}} = |7.2x - 4.16| \tag{2} \]

Fig. 3 shows the concentration dependences of the experimental values of the saturation magnetization for compositions with \( x = 0, 0.2, 0.4, 0.6, 0.8, \) and \( 1 \) and the dependence of the total magnetic moment calculated by the formula (2). From the expression (2), the theoretical value of the yttrium concentration in the compensation composition was \( x_{\text{comp}} \approx 0.58 \) at \( \mu_{\text{calc}} = 0 \).

**Figure 3.** Calculated dependence of total magnetic moment (solid blue line) on the yttrium concentration \( x \) and experimental values of saturation magnetization (red dots) at a temperature of 4.2 K for \((Tb_{1-x}Y_x)_{0.8}Sm_{0.2}Fe_2\)

In the region \( x < x_{\text{comp}} \), the contribution to the total magnetization from the terbium sublattice dominates, and in the region \( x > x_{\text{comp}} \), the main contribution to the magnetization comes from the iron sublattice (and, to a lesser extent, from the samarium sublattice). Therefore, the concentration dependence shows a minimum at \( x = x_{\text{comp}} \). Fig. 3 shows that the experimental values of saturation magnetization are in good agreement with theoretical calculations. It can also be seen from this figure that the composition with the yttrium concentration \( x = 0.6 \) is the closest to the compensation composition. A similar result was previously discovered by us experimentally in stationary fields of 2 kOe in the temperature range from 90 to 300 K (fig. 4) [8].
Figure 4. Dependence of magnetization of alloys \((\text{Tb}_{1-x}\text{Y}_x)_{0.8}\text{Sm}_{0.2}\text{Fe}_2\) on the substitution parameter \(x\) in the temperature range from 90 to 300 K in a field of 2 kOe.

Further studies of the magnetization of alloys of the \((\text{Tb}_{1-x}\text{Y}_x)_{0.8}\text{Sm}_{0.2}\text{Fe}_2\) system were carried out in pulsed magnetic fields up to 60 T. Fig. 5 shows the field dependences of the magnetization of the \((\text{Tb}_{1-x}\text{Y}_x)_{0.8}\text{Sm}_{0.2}\text{Fe}_2\) system at a temperature of 4.2 K. It is seen that the magnetization curves, as in static fields, quickly reach saturation. A slight increase in magnetization is observed for the compounds with the substitution parameter \(x = 0.4\) and \(x = 0.6\). These are the compositions near the compensation composition \(x = 0.58\).

Figure 5. Field dependences of magnetization \((\text{Tb}_{1-x}\text{Y}_x)_{0.8}\text{Sm}_{0.2}\text{Fe}_2\) at temperature \(T = 4.2\) K in pulsed fields up to 60 T.

As mentioned earlier, RFe\(_2\)-type compounds with heavy rare-earth metals have ferrimagnetic ordering. When a strong magnetic field is applied, it is presumably possible to achieve a ferromagnetic state, since the magnetic moments of each sublattice will tend to line up parallel to the applied field.

Assuming that the collinear structure of the magnetic moments of the sublattices is preserved, it is possible to calculate the magnetic moment of the \((\text{Tb}_{1-x}\text{Y}_x)_{0.8}\text{Sm}_{0.2}\text{Fe}_2\) compounds for the ferri- and
ferromagnetic states. Only for the ferromagnetic state, the sign of the terbium magnetic moment $\mu_{\text{Tb}}$ in relation (1) will be positive.

Table 2 shows the results of calculating total magnetic moments for the ferri- and ferromagnetic states of the compounds $(\text{Tb}_{1-x}\text{Y}_x)_{0.8}\text{Sm}_{0.2}\text{Fe}_2$.

| $x$ | $\mu_{\text{Sm}}$ ($\mu_B$) | $\mu_{\text{Fe}}$ ($\mu_B$) | $\mu_{\text{Tb}}$ ($\mu_B$) | $\mu_{\text{calc}}^{\text{ferri}}$ ($\mu_B$) | $\mu_{\text{calc}}^{\text{ferro}}$ ($\mu_B$) |
|-----|-----------------|-----------------|-----------------|-----------------|-----------------|
| 0   | 7.20            | 4.16            | 10.24           | 6.00            | 3.16            |
| 0.2 | 5.76            | 2.72            | 8.80            | 5.20            | 3.60            |
| 0.4 | 4.32            | 1.28            | 5.60            | 4.40            | 2.20            |
| 0.6 | 2.88            | 0.16            | 3.04            | 2.36            | 0.84            |
| 0.8 | 1.44            | 1.60            | 3.04            | 2.00            | 0.96            |
| 1   | 0               | 3.04            | 3.04            | 1.52            | 1.52            |

The magnetization dependences on the concentration $Y$ for the $(\text{Tb}_{1-x}\text{Y}_x)_{0.8}\text{Sm}_{0.2}\text{Fe}_2$ compounds at 4.2 K in a static field of 16 T and a pulsed field of 60 T are shown in Fig. 6. It was shown above that the smallest spontaneous magnetization is observed at $x_{\text{comp}} = 0.6$. In high fields, the magnetization at $x = 0.6$ also exhibits a minimum. It should be noted that for extreme concentrations, the magnetization in the range from 16 to 60 T practically does not change, which indicates a high value of the exchange field for the compositions $\text{Tb}_{0.8}\text{Sm}_{0.2}\text{Fe}_2$ and $\text{Y}_{0.8}\text{Sm}_{0.2}\text{Fe}_2$. Near the compensation composition, the magnetization value, even in high pulsed fields up to 60 T, remains rather low and amounts to 0.7 $\mu_B$/f.u. in a 60T field with $x = 0.6$. Nevertheless, this value of magnetization is 4 times higher than the calculated magnetic moment for the ferrimagnetic state (see Table 2). A similar behavior is observed for the composition with $x = 0.4$. This fact indicates that in these two compositions (near the compensation) a magnetic field of 60 T makes it possible to destroy the collinear magnetic structure.

The comparison of theoretical calculations of the total magnetic moments of the compounds and the corresponding magnetization curves obtained from the experiment showed that the ferromagnetic state in the fields up to 60 T in compounds $(\text{Tb}_{1-x}\text{Y}_x)_{0.8}\text{Sm}_{0.2}\text{Fe}_2$ is not implemented (Fig. 4). The values of the total magnetic moments for the ferromagnetic state (Table 2) are much greater than the value of the saturation magnetization obtained from the experiment. Thus, in compositions with $x = 0$, 0.2, and 0.8, the effect of an external magnetic field is 60 T less than the exchange field required to destroy the anticollinear ferrimagnetic structure. In the compositions with $x = 0.4$ and 0.6 in the field range from 10 to 60 T, a linear increase in magnetization is observed (Fig. 5), which can be explained by the development of noncollinear magnetic structure in a strong magnetic field.

4. Conclusion

In the intermetallic compounds of the C15 type, three types of exchange interactions are distinguished:

- Between transitional 3d-ions and (3d–3d–interaction).
- Between rare-earth ions and transitional 3d–ions and (R–3d interaction).
- Between rare-earth ions (R–R–interaction).

In the RFe$_2$ compound, the strongest are the 3d – 3d exchange interactions, and the weakest are the R– exchange interactions [14, 15]. It is the 3d–3d–interactions that determine the Curie temperature of the rare-earth intermetallic compounds. The exchange R-3d–interaction is intermediate in magnitude between 3d–3d and R–R–interactions and determines the type of magnetic ordering, i.e. ferro- or ferrimagnetic.

The alloys studied in this work can be represented in the model of three magnetic sublattices – terbium, samarium, and iron. The magnetic moment on the atoms of the heavy rare earth metal Tb is ordered antiparallel to the magnetic moment of iron, and the magnetic moment on the atoms of the light rare earth metal Sm is ordered in the same direction as the magnetic moment of iron. Consequently, the exchange interaction between the Fe and Sm sublattices has a positive sign, while
the exchange interaction between the Fe and Tb sublattices has a negative sign. Thus, in these alloys, the exchange interactions of Tb-Fe and Sm-Fe are competing.

When the rare-earth sublattice of terbium is diluted with a nonmagnetic element yttrium in the (Tb$_{1-x}$Y$_x$)$_{0.8}$Sm$_{0.2}$Fe$_2$ system, only the effects of dilution of the magnetic rare-earth sublattice and an increase in the distance between the terbium and samarium ions are observed. This leads to a change in the magnitude of the Tb-Fe and Sm-Fe exchange interactions competing with each other, and therefore, one could assume that it is possible to obtain a state in which the magnetic field energy would be sufficient to overcome the value of the negative exchange interaction Tb -Fe and carry out a magnetic phase transition from a ferrimagnetic state to a ferromagnetic one. However, in all compounds, with the exception of the extreme alloy Y$_{0.8}$Sm$_{0.2}$Fe$_2$, which was originally a ferromagnet, the ferrimagnetic state is retained in the magnetic fields up to 60 T.

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