X-ray studies of multicomponent Sm$_{0.2}$(Tb,Y)$_{0.8}$Fe$_2$ alloys

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Abstract. The multicomponent Sm$_{0.2}$(Tb$_{1-x}$Y$_x$)$_{0.8}$Fe$_2$ system was obtained by the arc melting method, in which atoms with a high magnetic moment of terbium are replaced by yttrium atoms that do not carry a noticeable magnetic moment. In this system, varying not only the composition, but also external factors (temperature, magnetic field, etc.), it is possible to influence competing exchange interactions and observe a number of unique phenomena, such as, for example, the phenomenon of magnetic compensation. Using the method of high-temperature and low-temperature X-ray diffraction, the phase composition and atomic-crystalline structure of the Sm$_{0.2}$(Tb$_{1-x}$Y$_x$)$_{0.8}$Fe$_2$ alloys ($x = 0, 0.2, 0.4, 0.6, 0.8, 1$) were studied.

1. Introduction
The system of quasi-binary compounds (Sm,Tb)Fe$_2$ with the Laves structure of phases is an important and interesting object of research, both from the fundamental and practical viewpoints [1, 2]. Giant and, at the same time, different sign magnetostrictive deformations in TbFe$_2$ and SmFe$_2$, caused by the large anisotropy of the Tb$^{3+}$ and Sm$^{3+}$ ions, the difference in the ordering of the magnetic moments of Tb and Sm, relative to the Fe sublattice, are the cause of the complex behavior of both structural and magnetic properties of Sm$_{1-x}$Tb$_x$Fe$_2$ compounds with varying their composition (0 ≤ $x$ ≤ 1) and external factors (temperature, magnitude of the applied magnetic field, etc.) [3-6].

To elucidate the role that magnetoactive ions play in the processes of magnetic ordering and various structural phase transitions associated with them, systems of alloys with isomorphic substitution of these magnetoactive ions by nonmagnetic analogs, for example, yttrium, are often used. Thus, in the Tb$_{1-x}$Y$_x$Fe$_2$ system [7], isomorphic substitution of yttrium atoms in the rare-earth sublattice of terbium atoms made it possible not only to experimentally determine the role of terbium in the appearance of features of the thermal expansion of TbFe$_2$, but also to trace how the magnetic "dilution" affects spontaneous magnetostriction. Studies have shown that, at room temperature, rhombohedral structural distortions are observed in Tb$_{1-x}$Y$_x$Fe$_2$ alloys only in the concentration range 0 ≤ $x$ ≤ 0.55. The magnitude of these distortions increases with decreasing temperature, while with increasing concentration of yttrium, on the contrary, decreases in absolute value. Tb$_{1-x}$Y$_x$Fe$_2$ alloys...
with a high yttrium content have a C15 cubic structure at room temperature. With decreasing temperature, a magnetic-structural phase transition occurs in such alloys and the structure of the alloys becomes rhombohedral.

In a multicomponent system (Sm,Tb,Y)Fe2, the isomorphic substitution of yttrium atoms in the rare-earth sublattice of samarium and terbium atoms makes it possible to influence the competing exchange interactions. It was shown in [8, 9] that a continuous series of substitutional solid solutions is formed in the Sm0.2(Tb1-xYx)0.8Fe2 system. We studied the features of the field and temperature dependences of the magnetostriction of multicomponent alloys Sm0.2(Tb1-xYx)0.8Fe2, as the properties of the most sensitive to changes in exchange interactions, and also revealed the main mechanisms responsible for the formation of features in the field and temperature dependences of magnetostriction.

The aim of this work is a detailed study of the structure of the multicomponent system Sm0.2(Tb1-xYx)0.8Fe2 in a wide temperature range, including the region of magnetic and structural phase transitions, and construction of a magnetic phase diagram. The main research method will be X-ray diffraction analysis. However, along with the main method, for comparison, we also plan to use the strain-gauge method.

2. Experimental details

The synthesis of Sm0.2(Tb1-xYx)0.8Fe2 (x = 0, 0.2, 0.4, 0.6, 0.8, 1) was carried out in an electric arc furnace from Leybold-Heraeus (ingot weight 20 g) with a non-consumable tungsten electrode on a water-cooled copper hearth of a special structure in an argon atmosphere at normal pressure. Metals of high purity (99.978 wt %) were taken as the starting components. The samples were subjected to subsequent homogenizing annealing in sealed evacuated quartz ampoules at a temperature of 850-900°C for 2 weeks.

The certification of alloys carried out by the method of X-ray phase analysis on a DRON-2 diffractometer (CuKα radiation) is described in [9]. In this work, a study of the crystal structure of alloys at room temperature was carried out on a Rigaku SmartLab3 high-resolution diffractometer (CuKα radiation). Investigations of the crystal structure in the temperature range 80-700 K were carried out by X-ray powder diffraction on a SuperNova diffractometer (Agilent). The survey was performed at the wavelength Kα of molybdenum radiation $\lambda = 0.71073$ Å using a Cobra temperature attachment. The obtained diffraction patterns were processed by the full-profile Rietveld analysis in the Fullprof software environment.

The strain-gauge method was employed for measuring thermal expansion in the temperature range 80-320 K. This method has already been described in detail in Ref. [10].

3. Results and discussion

Phase analysis of the samples, carried out using a high-resolution diffractometer, confirmed the data previously obtained at Refs. [9]. In the quasi-binary alloys Sm0.2Y0.8Fe2, Sm0.2Tb0.8Fe2, the amount of the main cubic Laves phase with a structure of the MgCu2 type reaches 96%. Figure 1 shows the experimentally obtained diffraction spectra of the studied alloys at room temperature. As an example, for the Sm0.2(Tb0.8Y0.2)0.8Fe2 composition, the calculated spectrum of the main phase of the MgCu2 type is shown, and the positions of the Bragg peaks of the main phase are indicated by blue dashes. As can be seen, in more complex investigated alloys (Sm,Tb,Y)Fe2, additional peaks appear from impurity phases, the main of which is the RFe3 phase (crystal structure of the PuNi3 type). Our calculations showed that the content of the main phase in the substituted compositions (Sm,Tb,Y)Fe2 varies within 90-95%.
It is known [11] that strong anisotropic magnetoelastic interactions can lead to various distortions of the lattice, determined by the easy axis of magnetization of the crystal. However, according to studies [8], alloys of the Sm$_{0.2}$(Tb$_{1-x}$Y$_x$)$_{0.8}$Fe$_2$ system have magnetostrictive deformations at room temperature of $\sim 10^{-4}$ in magnetic fields up to 1.2 T. These distortions are insignificant and, therefore, the crystal structure of Sm$_{0.2}$(Tb$_{1-x}$Y$_x$)$_{0.8}$Fe$_2$ can be considered as pseudocubic (like MgCu$_2$). The values of the parameters and volume of the unit pseudocubic cell of the studied compositions Sm$_{0.2}$(Tb$_{1-x}$Y$_x$)$_{0.8}$Fe$_2$ at room temperature are shown in Table 1. Data analysis shows that the dependence of the parameters on the concentration of yttrium is rather complex.

For a more detailed study of changes in the lattice parameters, X-ray studies were carried out in a wide temperature range. The temperature dependences of the unit cell parameters were obtained for all investigated compositions. Figure 2a shows the temperature dependence of the pseudocubic cell parameter of Sm$_{0.2}$Tb$_{0.8}$Fe$_2$ as an example. A monotonic increase in the cell parameter is observed, and in the region of the transition temperature to the magnetically disordered phase (at Curie temperature) equal to $T_C = 673$ K [5], there is a change in the slope of the $a(T)$ curve. It should be especially noted that the sample gradually amorphizes upon heating and, in the temperature range above 600 K, the intensity of reflections noticeably decreases (inset in Figure 2a), which significantly reduces the accuracy of determining the cell parameter. It is seen that in the temperature region of 690 K all peaks practically disappear and merge with the background. Nevertheless, the obtained values of the Curie temperatures of all the studied compositions of the Sm$_{0.2}$(Tb$_{1-x}$Y$_x$)$_{0.8}$Fe$_2$ system correlate well with the data of magnetic measurements [9] and are given in Table 1.

| Y content | a, Å   | V, Å$^3$ | $T_{S_{R2}}$, K | $T_{S_{R1}}$, K | $T_C$, K |
|-----------|--------|---------|---------------|---------------|---------|
| $x = 0$   | 7.3592(2) | 398.56  | 100           | 130           | 670     |
| $x = 0.2$ | 7.3615(2) | 398.93  | 95            | 136           | 665     |
| $x = 0.4$ | 7.3625(2) | 399.09  | 88            | 150           | 640     |
| $x = 0.6$ | 7.3639(2) | 399.32  | 95            | 215           | 610     |
| $x = 0.8$ | 7.3666(2) | 399.76  | 97            | 215           | 580     |
| $x = 1$   | 7.3658(2) | 399.63  | 98            | 179           | 540     |
Figure 2b shows the concentration dependences of the pseudocubic cell of the compounds of the Sm$_{0.2}$(Tb$_{1-x}$Y$_x$)$_{0.8}$Fe$_2$ system, obtained at different temperatures. It is seen that at low temperatures (T = 100 and 200 K), with an increase in the concentration of yttrium, a monotonic, close to linear, increase in the unit cell parameter is observed. At temperatures above room temperature (T = 400, 460, 525 K), the $a(x)$ dependence is not monotonic; a minimum is observed at yttrium concentrations $x = 0.6$. Earlier in Ref. [9] for Sm$_{0.2}$(Tb$_{1-x}$Y$_x$)$_{0.8}$Fe$_2$ with Y concentration $x = 0.6$, authors observe a magnetic compensation, i.e. mutual cancellation of magnetic moments of the sublattices (Tb, Sm and Fe), which is fully consistent with the model of a three-sublattice collinear ferrimagnet.

![Figure 2](image)

**Figure 2.** (a) Temperature dependence of the pseudocubic cell parameter of Sm$_{0.2}$Tb$_{0.8}$Fe$_2$. Inset: temperature behavior of the most intense spectral peaks in the temperature range 523 - 738 K. (b) Dependence of the unit cell parameter of the compositions of the Sm$_{0.2}$(Tb$_{1-x}$Y$_x$)$_{0.8}$Fe$_2$ system as a function of the Y content at different temperatures.

![Figure 3](image)

**Figure 3.** Temperature dependences of the relative volume change of Sm$_{0.2}$(Tb$_{0.6}$Y$_{0.4}$)$_{0.8}$Fe$_2$ compound obtained by different methods (a) and the magnetic phase diagram of the Sm$_{0.2}$(Tb$_{1-x}$Y$_x$)$_{0.8}$Fe$_2$ system (b).

The study of the values of the lattice parameter $a$ and the volume of the unit cell $V$ was carried out for all investigated compositions in a wide temperature range by both diffractometric and tensometric methods [12]. Figure 3a shows the temperature variation of the relative volume ($V - V_0)/V_0$ of the sample with yttrium content $x = 0.4$ in the range from 80 to 300 K. The diffractometric method showed a monotonic increase in the unit cell volume with increasing temperature. Measurements by
the strain-gauge method, carried out with a step of 1 K, made it possible to reveal features in the temperature dependence of the relative change in volume. Thus, at temperatures $T_{SR1} = 150$ K and at $T_{SR2} = 88$ K, kinks are observed in the temperature dependence, indicating a magnetic-structural (spin-reorientation) phase transition. Similar features are observed for all compositions of the Sm$_{0.2}$Tb$_{1-x}$Y$_x$Fe$_2$ system.

Summing up our and literature data on the structure and phase transitions in the studied system of compounds, we constructed a magnetic phase diagram (Figure 3b). In the Sm$_{0.2}$Y$_{0.8}$Fe$_2$ compound, as shown in [13], there are two phase transitions (similar to those in the Y-free SmFe$_2$ compound [14-18]). Recall that in SmFe$_2$ at low temperatures the crystal structure is tetragonal and YFe$_2$ or LuFe$_2$ do not have phase transitions [19]. In Sm$_{0.2}$Y$_{0.8}$Fe$_2$, the low-temperature transition temperature and tetragonal distortions of the cubic structure are retained. In other words, as the temperature decreases in the Sm$_{0.2}$Y$_{0.8}$Fe$_2$ compound, a transition from the cubic phase to the tetragonal phase occurs through the intermediate phase (shown in Figure 3b by the shaded region). With a decrease in the yttrium content (and an increase in the terbium content, respectively), the crystal lattice at room temperature exhibits ever larger rhombohedral distortions, which is associated with a large anisotropy of terbium ions [20-26]. The presence of two spin-reorientation phase transitions is retained for all compositions of the system. The data obtained is consistent with data for other systems (see, for example Refs. [27-29]). In this case, the temperature of the low-temperature transition ($T_{SR2}$) practically does not change with a change in $x$, while the temperature of the high-temperature transition ($T_{SR1}$) changes and the dependence $T_{SR1}(x)$ has a maximum in the concentration range $x = 0.6$ - 0.8. As follows from early studies [8, 9], at $x = 0.6$, both the phenomenon of compensation of the total magnetization and compensation of magnetostriction are observed. In addition, the value of thermal expansion is also minimal for the composition with $x = 0.6$. Specific features in the temperature and field dependences of the thermal expansion and magnetostriction of multicomponent alloys can be quite attractive for their use in practice [30-32].

4. Conclusions
X-ray measurements carried out in the wide temperature region, including the Curie temperature. The determined $T_C$ values are in good agreement with the data obtained using magnetometric measurements. For the compounds of Sm$_{0.2}$Tb$_{1-x}$Y$_x$Fe$_2$ system under study, it decreases monotonically with an increase in the yttrium content in the range from 670 to 540 K. It was found that when the samples are heated above 600 K, their partial amorphization occurs. Along with the X-ray research method in the temperature range 80 - 320 K, the strain-gauge method was used. It was found that at low temperatures all the compounds under study demonstrate two spin-reorientation phase transitions. Based on the research results, a magnetic phase diagram was constructed.

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