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Low-temperature magnetostriction and distortions in the rare-earth Laves phases

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Abstract. The effects of partial substitution of dysprosium in $Tb_{0.2}Dy_{0.8}Co_2$ by terbium and gadolinium on the structure and magnetic properties have been studied. Two compositions, $Tb_{0.3}Dy_{0.7}Co_2$ and $Tb_{0.2}Dy_{0.7}Gd_{0.1}Co_2$ have been synthesized. Their crystal structure, in contrast to the structure of the original compound, has both tetragonal distortions and rhombohedral distortions at lower temperatures. Anomalies of magnetostriction and magnetocaloric effect near the observed magnetic phase transitions have been studied. The sign-alternating temperature dependences of the longitudinal and transverse magnetostrictions associated with various types of crystal structure distortions of the alloys are revealed.

1. Introduction

The rare earth Laves phases RCo_2 with cubic $MgCu_2$-type structure (C15) exhibit a number of interesting effects such as large magnetovolume effect, significant anisotropic magnetostriction, and large magnetocaloric effect [1-4]. The special symmetry of the cubic Laves phase leads to a significant rhombohedral distortion in the (111) direction in the magnetic state. This behavior was observed in $TbCo_2$. However, the metamagnetic transitions in $DyCo_2$ and $GdCo_2$ cause a crystal symmetry reduction from the cubic (paramagnetic) to a tetragonal (ferrimagnetic) structure [5,6]. Thus, in the system of binary compounds based on $(Tb_xDy_{1-x})Co_2$, at $x \leq 0.2$, the distortions are tetragonal, and for $x > 0.2$, rhombohedral [7]. In the system of binary compounds based on $(Tb_xGd_{1-x})Co_2$, for $x \leq 0.9$, the distortions are tetragonal, and for $x > 0.9$, the distortions are rhombohedral [8].

In this work, three compounds are considered: $Tb_{0.2}Dy_{0.8}Co_2$, $Tb_{0.3}Dy_{0.7}Co_2$, $Tb_{0.2}Dy_{0.7}Gd_{0.1}Co_2$, the first of which should have a tetragonal distortion when going into a magnetically ordered state, and the other two are rhombohedral [7-10]. Also, according to the analysis of literature data, in the compounds $Tb_{0.3}Dy_{0.7}Co_2$, $Tb_{0.2}Dy_{0.7}Gd_{0.1}Co_2$ there should be spin-reorientational phase transitions. These transitions are observed in these compounds at low (80–200 K) temperatures. The magnetostriction of such compounds is usually studied in the region of room temperature; a detailed analysis of the temperature dependence of magnetostriction in the region of magnetic phase transitions is often absent. The aim of the work was to study the magnetic, magnetocaloric and magnetostriction properties near magnetic phase transitions of the studied compounds and to identify the dependence of these properties on the type of crystal structure distortion in a magnetically ordered state.

2. Materials synthesis and experimental details

The alloys were prepared by arc melting of stoichiometric amounts of constituent elements under a high purity argon atmosphere. The purity of rare-earth metals and Co was 99.9 and 99.95 %, respectively. The alloys were remelted three times and annealed at 900 C for one week to obtain homogeneous samples. Their crystal structure was determined by powder X-ray diffraction (XRD) at room temperature. The XRD patterns were recorded at a 2q scanning step of 0.02 (at the 2-s exposition) on a Rigaku Ultima IV (Japan) powder diffractometer with CuKα radiation. The qualitative and quantitative phase analysis was performed using a program PDXL integrated with the international database ICDD. To observe the lattice distortion with temperature and determine the crystal symmetry, we employed X-ray diffractometer Supernova (Agilent) with MoKα radiation The temperature of the sample was controlled by the Oxford Cobra Cryosystem, which can provide a temperature range of 80 K to 300 K.

Magnetization and magnetocaloric effect were investigated in the 80 - 350 K temperature range in fields of up to 1.8 T with the use of the MagEq MMS 901 setup (Russia) by the induction and direct (measurement of adiabatic temperature change) method, respectively.

The magnetostriction was studied by the strain-gauge method on the polycrystalline samples in the temperature range 80–350 K in external magnetic fields up to 1.2 T. The field applied both along and perpendicularly the direction of the strain-gauge measurement, that allows to recording both longitudinal ($\lambda_{||}$) and transverse ($\lambda_{\perp}$) magnetostriction, respectively.

3. Results and discussion

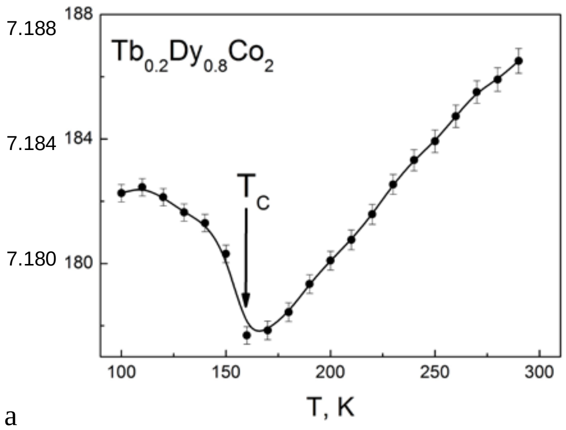
Rietveld refinement of the powder XRD patterns has shown that the all compounds crystallize in the C15-type cubic Laves phase structure ($MgCu_2$, space group Fd3m) at room temperature. All studied materials are single-phase. The parameters of the crystal cell are shown in table 1.

It was determined that the value of the lattice parameter decreases on cooling from room temperature to the certain temperature and grows upon cooling below it (figure 1a). Such temperature behavior of the unit cell parameter is observed in all investigated compounds and may indicate a structural distortion in the vicinity of Curie temperature (T_C). According to the $TbCo_2$-$DyCo_2$ phase diagram (figure 1b), for $Tb_{0.2}Dy_{0.8}Co_2$ below the transition temperature to a magnetically ordered state, the cubic lattice is distorted tetragonally, and for $Tb_{0.3}Dy_{0.7}Co_2$ is distorted rhombohedrally. In the $Tb_{0.3}Dy_{0.7}Co_2$, as the temperature decreases, a spin-reorientation (SR) phase transition was observed, during which the crystal lattice acquires tetragonal distortions (figure 1b). A similar situation was expected in the $Tb_{0.2}Dy_{0.7}Gd_{0.1}Co_2$. To confirm reliably the type of distortion in X-ray diffraction studies, it is necessary to identify the splitting of the corresponding diffraction peaks.

Table 1. Lattice parameters, Curie temperatures and the types of magnetic phase transition in investigated compounds.

Compound	a, Å	V, Å^3	T_C, K	Transition type (at T_C)	T_{SR}, K
$Tb_{0.2}Dy_{0.8}Co_2$	7.191	371.85	161	I	-
$Tb_{0.3}Dy_{0.7}Co_2$	7.190	371.69	169	I	~110
$Tb_{0.2}Dy_{0.7}Gd_{0.1}Co_2$	7.201	373.40	184	II	~150

However, in our experiment, the use of a copper anode did not allow us to see the splitting due to the application of X-ray fluorescence radiation, and the diffraction lines obtained on the molybdenum anode are grouped close to each other, and this cannot be corrected for splitting in such diffraction patterns. The use of an anode of iron [11] apparently, can make it possible to obtain spectra with good splitting (we plan in the future). In this work, we turned to magnetic research.



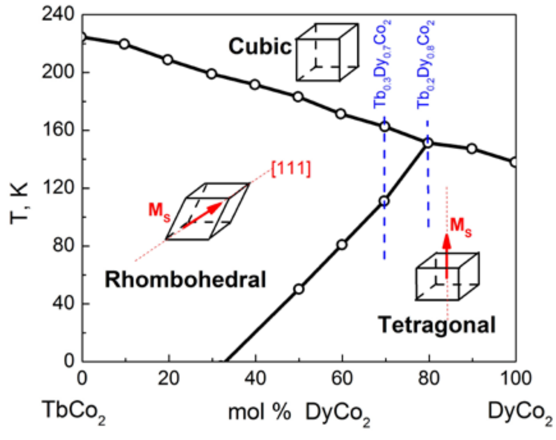


Figure 1. Temperature dependence of $Tb_{0.2}Dy_{0.8}Co_2$ lattice parameters and phase diagram of $TbCo_2$-$DyCo_2$[7].

Figure 2 shows the temperature dependences of the magnetization of compounds in a 0.1 T magnetic field. From the the peak in the curve corresponding to the first derivative (see inset), it is easy to determine the Curie temperature. As expected, an increase in the Curie temperature is observed upon partial substitution of dysprosium. The obtained values are given in Table 1, the values of $(Tb,Dy)Co_2$ are in good agreement with the literature values [12-14]. To determine the type of phase transition, as well as to refine the Curie temperature, the Belov – Arrott curves were constructed for the compounds $Tb_{0.2}Dy_{0.8}Co_2$ and $Tb_{0.2}Dy_{0.7}Gd_{0.1}Co_2$ (figure 3). The magnetization curves for $Tb_{0.3}Dy_{0.7}Co_2$, as well as a detailed analysis of the type of magnetic transition (T_C), were given earlier [15–17]. It was found that in the compounds $Tb_{0.2}Dy_{0.8}Co_2$ and $Tb_{0.3}Dy_{0.7}Co_2$, the transition is of first order, while in the compound with gadolinium the type of transition changes to the second. There are no significant anomalies in the low-temperature region, reliably confirming the presence of SR phase transition.

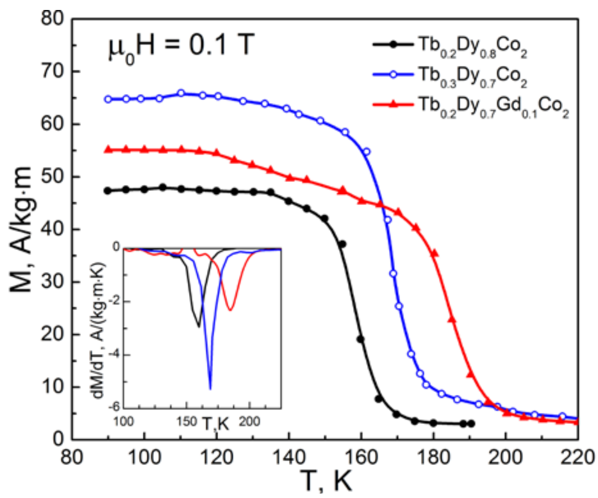


Figure 2. Temperature dependence of the magnetization of the investigated compounds. Inset: first derivative of the magnetization dM/dT.

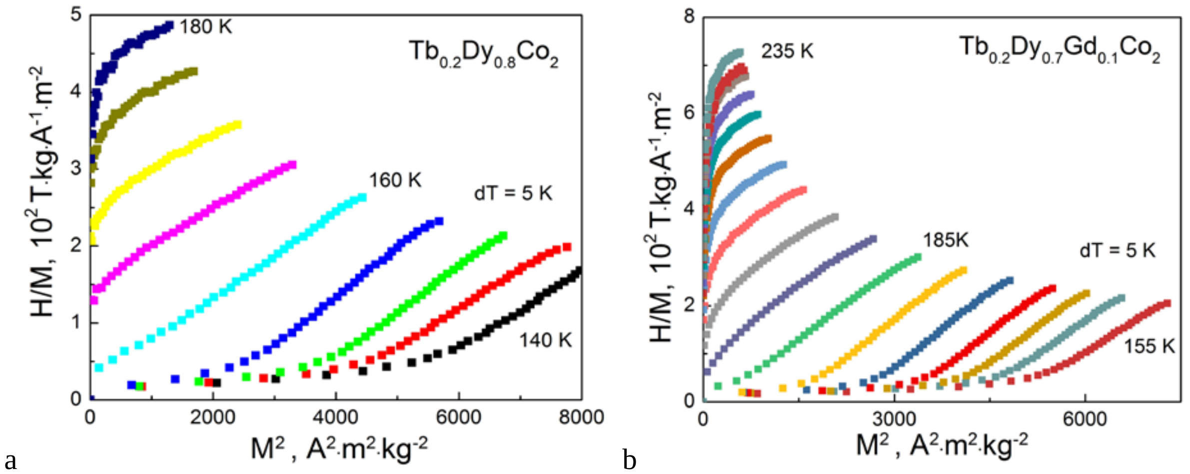


Figure 3. Belov-Arrott plots for $Tb_{0.2}Dy_{0.8}Co_2$ (a) and $Tb_{0.2}Dy_{0.7}Gd_{0.1}Co_2$ (b) compounds.

For the studied compounds, the magnetocaloric effect (MCE) was measured by the direct method. Figure 4 shows the dependence of the adiabatic temperature change on the temperature change in the external magnetic field 1.8 T. In the T_C region, MCE maxima are observed. The determined transition temperatures are in good agreement with the temperature at which a maximum of the MCE is observed. The value of the MCE is significantly reduced on partial substitution of dysprosium and for the compound $Tb_{0.2}Dy_{0.7}Gd_{0.1}Co_2$ it is 1 K/T. No significant anomalies in the low temperature region were found.

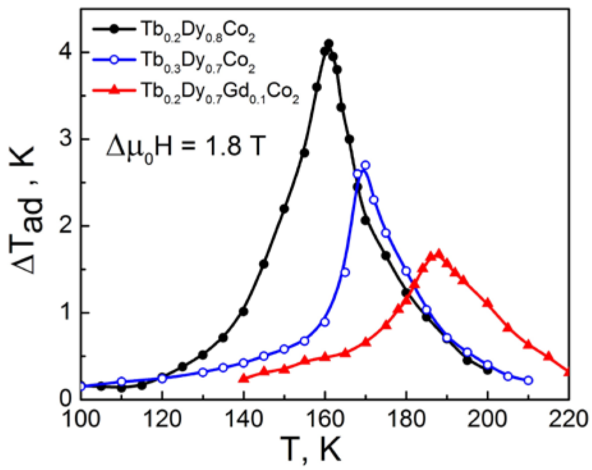


Figure 4. Temperature dependence of adiabatic temperature change obtained by direct method at $\Delta\mu_0 H = 1.8$ T.

As our previous studies have shown, magnetostriction deformations [9,18] are the most sensitive to magnetostructural transitions. Indeed, the temperature dependences of the longitudinal and transverse magnetostrictions of the studied compounds are of the greatest interest (figure 5). For the $Tb_{0.2}Dy_{0.8}Co_2$ parent compound at liquid nitrogen temperature, the longitudinal magnetostriction is negative, while the transverse magnetostriction is positive. This means, the cubic lattice shrinks, which is similar to the behavior of $SmFe_2$ [19]. As the temperature increases, the absolute values of magnetostriction decreases monotonically. Near the Curie temperature, both longitudinal and transverse magnetostrictions have a positive sign. The maximum value of magnetostriction corresponds to the Curie temperature and does not depend on the magnitude of the applied magnetic field.

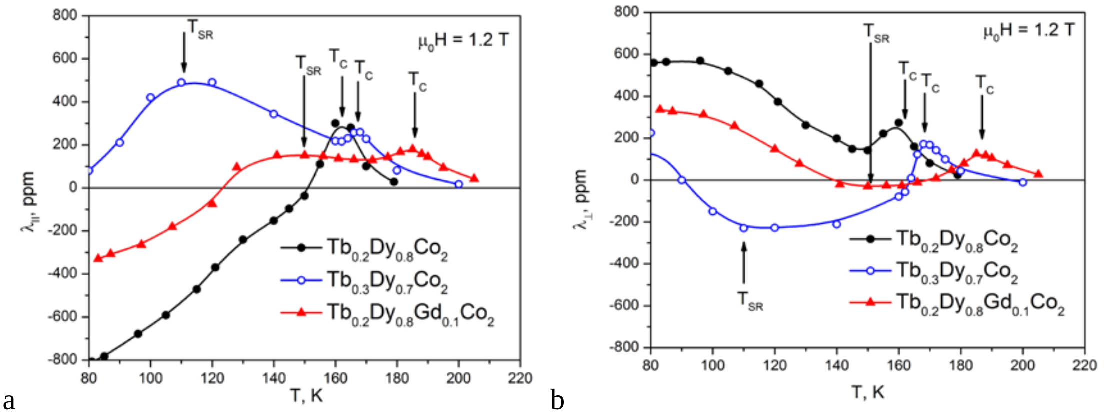


Figure 5. Temperature dependence of the longitudinal (a) and transverse (b) magnetostriction of the investigated compounds.

For the $Tb_{0.3}Dy_{0.7}Co_2$ compound, below the Curie temperature, the longitudinal magnetostriction is positive and the transverse magnetostriction is negative. This fact clearly confirms a different type of distortion of the cubic lattice than in the $Tb_{0.2}Dy_{0.8}Co_2$ compound. In this case, the cubic lattice is stretched, which is similar to the stretching along the <111> direction of the cubic lattice of $TbFe_2$ compound in a magnetically ordered state. With a further decrease in temperature, the longitudinal and transverse magnetostrictions experience an extremum and decrease in absolute value. The temperature of this extremum (T~110 K) corresponds to the temperature of spin reorientation in the compound and agrees well with the transition temperature in the phase diagram (figure 1b) for this compound. This temperature, as well as a change in the sign of magnetostriction, as shown by our studies, depend on the magnitude of the applied magnetic field. At temperatures below 80 K, it appears that the signs of the longitudinal and transverse magnetostriction for the compounds $Tb_{0.3}Dy_{0.7}Co_2$ and $Tb_{0.2}Dy_{0.8}Co_2$ are the same, which indicates the same type of distortion of the cubic lattice, in this case tetragonal.

For the $Tb_{0.2}Dy_{0.7}Gd_{0.1}Co_2$ compound, the temperature behavior of the longitudinal and transverse magnetostriction is similar to that for $Tb_{0.3}Dy_{0.7}Co_2$. However, in comparison with it, the extremes in the region of spin reorientation are more diffuse and the absolute values of magnetostriction are lower. Nevertheless, it can be argued that when going from a magnetically disordered to a magnetically ordered state in the $Tb_{0.2}Dy_{0.7}Gd_{0.1}Co_2$ compound, the cubic lattice is distorted rhombohedrally (pulled along the <111> axis), and at a temperature close to 150 K, the spin-oriented phase transition occurs and the type of distortion changes to tetragonal.

4. Conclusions

The magnetic, magnetocaloric, and magnetostrictive properties of the compound $Tb_{0.2}Dy_{0.8}Co_2$ and compounds with partial dysprosium substitution with terbium and gadolinium ($Tb_{0.3}Dy_{0.7}Co_2$, $Tb_{0.2}Dy_{0.7}Gd_{0.1}Co_2$) were investigated at temperature range 80 - 220 K in magnetic fields up to 1.8 T. The parameters of the crystal cell at room temperature (in the paramagnetic state) and the temperature value (T_C) and the type of transition to the magnetically ordered state are determined. With a partial dysprosium substitution with both terbium and gadolinium, the Curie temperature increases, and the maximum value of the magnetocaloric effect (ΔT_{ad}) decreases markedly. The temperature dependences of the longitudinal and transverse magnetostrictions of the $Tb_{0.3}Dy_{0.7}Co_2$ and $Tb_{0.2}Dy_{0.7}Gd_{0.1}Co_2$ compounds revealed anomalies indicating spin-reorientation phase transitions. The relationship between the magnitude and sign of longitudinal and transverse magnetostriction and the type of distortion of the cubic lattice of the studied Laves phases is revealed.

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