



Research articles

Magnetostriptive properties of Tb_{0.2}Gd_{0.8} single crystal in high magnetic fieldsS.A. Nikitin^a, T.I. Ivanova^a, A.I. Zvonov^a, Yu.S. Koshkid'ko^{b,*}, J. Ćwik^b, K. Rogacki^b^a Lomonosov Moscow State University, Faculty of Physics, 119991 Moscow, Russia^b Institute of Low Temperature and Structure Research, PAS, 50-950 Wrocław, Poland

ARTICLE INFO

Keywords:

Magnetic materials
Magnetostriktion
Single crystal
Rare-earth alloys
Magnetocrystalline anisotropy
High magnetic fields

ABSTRACT

Temperature and field dependencies of the magnetostriction of the Tb_{0.2}Gd_{0.8} single crystal were measured in strong magnetic fields up to 14 T in a temperature range of 4.2–300 K. The temperature dependencies of five magnetostriction constants $\lambda_1^{a,0}$, $\lambda_2^{a,0}$, $\lambda_1^{a,2}$, $\lambda_2^{a,2}$ and $\lambda_{\gamma,2}$ were determined, using Clark's theory for rare-earth hexagonal close-packed single crystals. Giant values of linear and volume magnetostriction due to forced magnetostriction were found in the Tb_{0.2}Gd_{0.8} single crystal ($\lambda_{pc} = 1.6 \times 10^{-3}$) in the region of the Curie temperature (near room temperatures). It is shown that giant values of forced magnetostriction are caused by the sharp dependence of exchange interactions on interatomic distances along the hexagonal the *c* axis.

1. Introduction

Magnetostriction phenomena is defined as the mechanical deformation of a magnetic material caused by the changes in magnetization degree or magnetization direction. J. Joule first identified this effect in 1842. The change in volume due to the sample magnetization is called volume magnetostriction. The interest in new magnetostrictive materials was steadily growing in recent years. The giant anisotropic magnetostriction in rare-earth metals (REM) was discovered by physicists of Moscow State University in 1960 [1,2]. The magnetostriction of Tb and Dy single crystals was investigated by Legvold et al. [3,4], the study of the magnetostriction of Sm and Tm single crystals were presented in [5,6]. Giant volume magnetostriction in the region of room temperatures was discovered in Fe – REM compounds [7]. REM and rare-earth-based alloys and compounds form an important class of magnetostrictive materials with giant magnetostriction [8–10]. It is important to note that the large volume magnetostriction was found recently in the Y₂Fe₁₇ compound in the area of room temperatures [11].

There are two known types of magnetostriction mechanism: isotropic magnetostriction (caused by pair exchange interactions) and anisotropic magnetostriction (caused by single-ion interactions) [4–7,12]. The theory of magnetostriction was created and formulated by Callens [12] and Clark et al. [13,14]. Clark proposed the mathematical expression of magnetostriction for hexagonal close-packed (HCP) single crystals with magnetostriction constants depending on temperature and magnetic field [14]. The theory of magnetostriction

for 3d-4f intermetallic compounds was developed by Kuzmin [15]. The magnetostrictive materials are widely used in modern technologies, in particular, in the production of precision transducers and actuators [8,9].

The aim of the present study was to investigate the magnetostriction both in the low temperature region and in the area of the Curie temperature on the Tb_{0.2}Gd_{0.8} single crystal in strong magnetic fields up to 14 T and in temperature range 4.2 K–300 K; to determine magnetostrictive constants and contributions to magnetostriction caused by exchange and magnetocrystalline interactions.

Tb_xGd_{1-x} alloys form a continuous series of solid solutions with the HCP structure (P6₃/mmc space group). The nature and behavior of the magnetocaloric effect and magnetization in high magnetic fields were analyzed in the Tb_{0.2}Gd_{0.8} single crystal [16]. Tb_xGd_{1-x} alloys with *x* > 0.06 are highly magnetically anisotropic, with the hard magnetization axis matching the *c* crystallographic axis [17]. A relatively high Curie temperature is observed in Tb_{0.2}Gd_{0.8} alloy due to high Gd concentration and giant values of magnetostriction caused by Tb content (which has high values of single-ion magnetostriction). As Tb_xGd_{1-x} alloys possess both strong exchange interactions and giant magnetostriction values, they become promising objects in terms of theoretical research in field of REM magnetism and in practical applications.

2. Experimental

The complete research of the magnetic and magnetostriction

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Received 30 May 2019; Received in revised form 25 March 2020; Accepted 3 May 2020

Available online 04 May 2020

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properties demands a large single crystal with perfect crystal structure and homogeneous distribution of alloy components throughout the crystal volume. The most suitable for this purpose is the Czochralski method (pulling crystals from the melt).

The experimental samples were produced by Czochralski method in «Giredmet» Institute (Moscow, Russia). For the growth of the $\text{Tb}_{0.2}\text{Gd}_{0.8}$ single crystal Tb and Gd of 99.99 at.% purity were used. Tb and Gd are known to form a continuous series of substitutional solid solutions when alloying. The numerous homogenization remeltings provide distribution of alloy components uniform throughout the sample [17].

Single crystal pulling was carried out at a speed of 0.3 mm per minute and the rotation rate of 20 rpm in a vacuum of 10^{-6} Torr. The single crystal obtained has the form of a cylindrical rod with a diameter of 6–10 mm and a length of 40–60 mm.

The experimental specimens were cut from a single crystal in the form of a parallelepiped (with sizes $6 \times 4 \times 4$ mm) in such a way that each side was perpendicular to one of the main crystallographic axes. Orientation of the samples was carried out by X-ray diffraction method. Then the samples were etched by 30% HNO_3 ethanol solution for depth of approximately 0.2–0.5 mm on each cut side and annealed for 10 h at a temperature of 800 degrees Celsius in a pure argon atmosphere, to remove surface-deformed layer.

The measurement of magnetostriction was carried out on the PPMS equipment using the SK-06-030 TY-350 type strain gauge. This type of strain gauge allows measuring the magnetostrictive deformation of the sample simultaneously in two mutually perpendicular directions. The strain gauge was glued to the sample surface containing the main crystallographic axes c and b .

3. Results and discussion

The experimental measurements of magnetostrictions were performed on the $\text{Tb}_{0.2}\text{Gd}_{0.8}$ single crystal both along the easy magnetization b axis ($\lambda_{cb}, \lambda_{bb}$) and along the hard magnetization c axis ($\lambda_{cc}, \lambda_{bc}$). The first index denotes the magnetic field direction, the second one denotes crystallographic direction of magnetostriction measurement.

The magnetic moments of the $\text{Tb}_{0.2}\text{Gd}_{0.8}$ single crystal rotate within a basal plane in external magnetic field $H \perp c$. In a field $H \parallel c$ the magnetic moments rotate to c axis and the angle between basal plane and magnetic moments appears. In a field $H \parallel c$ exceeding the anisotropy field the magnetic moments orient along c axis.

3.1. Linear magnetostriction

Measurements of the λ_{bb} magnetostriction show that in magnetic field $H \parallel b$ the sample's deformation along b axis initially increases sharply at relatively low magnetic fields (not exceeding 1 T) and reaches giant values. At $T = 4.2$ K the λ_{bb} magnetostriction reaches value of 0.8×10^{-3} in 1 T magnetic field (see Fig. 1). In magnetic fields higher than 1 T the λ_{bb} magnetostriction converges to saturation. However, with the sample's temperature rising and approaching the Curie temperature $T_C \sim 280$ K, the λ_{bb} magnetostriction value increases nearly linearly with the magnetic field.

The λ_{bc} magnetostriction at low temperatures possesses very low values $\sim 10^{-5}$, however it reaches a narrow maximum in the area of the Curie temperature (Fig. 2) with giant values 1.3×10^{-3} in 10 T magnetic field. As it can be seen the temperature dependencies of the λ_{bb} and λ_{bc} magnetostrictions show a maximum in the Curie temperature region (Fig. 2).

To understand the real mechanisms responsible for the appearance of a giant Curie anisotropy and giant values of magnetostriction, it is important to study the temperature dependencies for all types of magnetostriction measured in this work: λ_{cb} , λ_{bb} , λ_{cc} and λ_{bc} . Fig. 3 depicts the $\lambda(T)$ curves in magnetic field $\mu_0 H$ equal to 8 T. As it can be seen, the behavior and nature of these dependencies are noticeably different. The λ_{bb} magnetostriction is determined mainly by rotation of

the magnetic moments in the basal plane and domain walls shifting, which are damped close to the Curie temperature. It follows from a smooth decrease of λ_{bb} magnetostriction (see Fig. 2) with temperature increase from $T = 4.2$ K ($\lambda_{bb} = 1.2 \times 10^{-3}$) to the Curie temperature $T_C \sim 280$ K ($\lambda_{bb} = 1 \times 10^{-4}$). In magnetic field H oriented along the b axis (easy magnetization axis) the magnetization vector retains its direction and therefore there are no rotation processes. But domain walls shifting and forced magnetization (which is most intense near the Curie temperature) remain.

The magnetostriction λ_{bc} which values are minimal in the range of low temperatures and reach sharp maximum in the area of T_C , is determined mainly by forced magnetization process. This process is completely determined by intense orientation of magnetic moments in external magnetic field through thermal fluctuations near the Curie temperature.

For λ_{cc} magnetostriction the similar behavior of temperature and field dependencies is observed. In the region of low temperatures in magnetic field $H \parallel c$ the rotation of the magnetization vector from the easy magnetization plane (basal plane) to the direction of hexagonal c axis occurs. A strong external magnetic field overcomes the anisotropy field and magnetization vector rotation approaching the Curie temperature concludes. Further growth of λ_{cc} magnetostriction occurs due to the forced magnetization, thus the maximum of λ_{cc} magnetostriction is reached at T_C (see Figs. 2 and 3). The maximum of value of λ_{cc} exceeds 10^{-3} in a field $\mu_0 H = 8$ T. This fact indicates a strong dependence of exchange interactions on interatomic distances along the c axis. The λ_{cb} magnetostriction has relatively low values $\sim 0.3 \times 10^{-3}$ at low temperatures in a field $\mu_0 H = 8$ T and moreover around T_C (see Figs. 2 and 3). The low values of λ_{cb} magnetostriction are caused by weak dependence of exchange interactions on interatomic distances in basal plane.

3.2. Volume magnetostriction

The change in volume due to the magnetization of the sample is called volume magnetostriction (denoted by ω). The expressions for determining the volume magnetostriction were obtained from the equations derived by Callens [12]:

$$\omega_c = \lambda_{ca} + \lambda_{cb} + \lambda_{cc} \quad (1)$$

$$\omega_b = \lambda_{ba} + \lambda_{bb} + \lambda_{bc} \quad (2)$$

where ω_c is the volume magnetostriction in magnetic field applied along the c axis, ω_b is the volume magnetostriction in magnetic field applied along the b axis. λ_{ba} values were taken from [17]. In the case of $\text{Tb}_{0.2}\text{Gd}_{0.8}$ single crystal with the easy magnetization plane $\lambda_{cb} = \lambda_{ca}$, (see Eq. (3), (7) and (8)). The temperature dependencies $\omega_b(T)$ are shown in Fig. 4 in various magnetic fields from 2 to 10 T. In the area of low temperatures ω_b reaches giant values ($\omega_b \sim 0.8 \times 10^{-3}$). Then ω_b gradually decreases with increasing temperature up to T_C . However, at T_C the $\omega_b(T)$ curve has a maximum, $\omega_b \sim 1.6 \times 10^{-3}$ in a field $\mu_0 H = 10$ T. The $\omega_c(T)$ curve also demonstrates a maximum at T_C ($\omega_c \sim 1.15 \times 10^{-3}$). At low temperatures (see Fig. 5) the ω_c value is almost ten times less than the ω_b value, $\omega_c = 0.35 \times 10^{-3}$. The close values of ω_c and ω_b at T_C point out that these magnetostrictions are mainly caused by the forced magnetization, because external magnetic field $\mu_0 H$ exceeds the magnetic anisotropy field $\mu_0 H_a$. The measurements of magnetic anisotropy constants of the $\text{Tb}_{0.2}\text{Gd}_{0.8}$ single crystal, made in our previous work [17], showed that the value of magnetic anisotropy field ($\mu_0 H_a = K_2/2M_s$) does not exceed $\mu_0 H_a = 8$ T at temperatures $T > 160$ K. The magnetic anisotropy field steadily decreases with temperature increase.

4. Theoretical description of magnetostriction

The theoretical description of the magnetostriction phenomenon

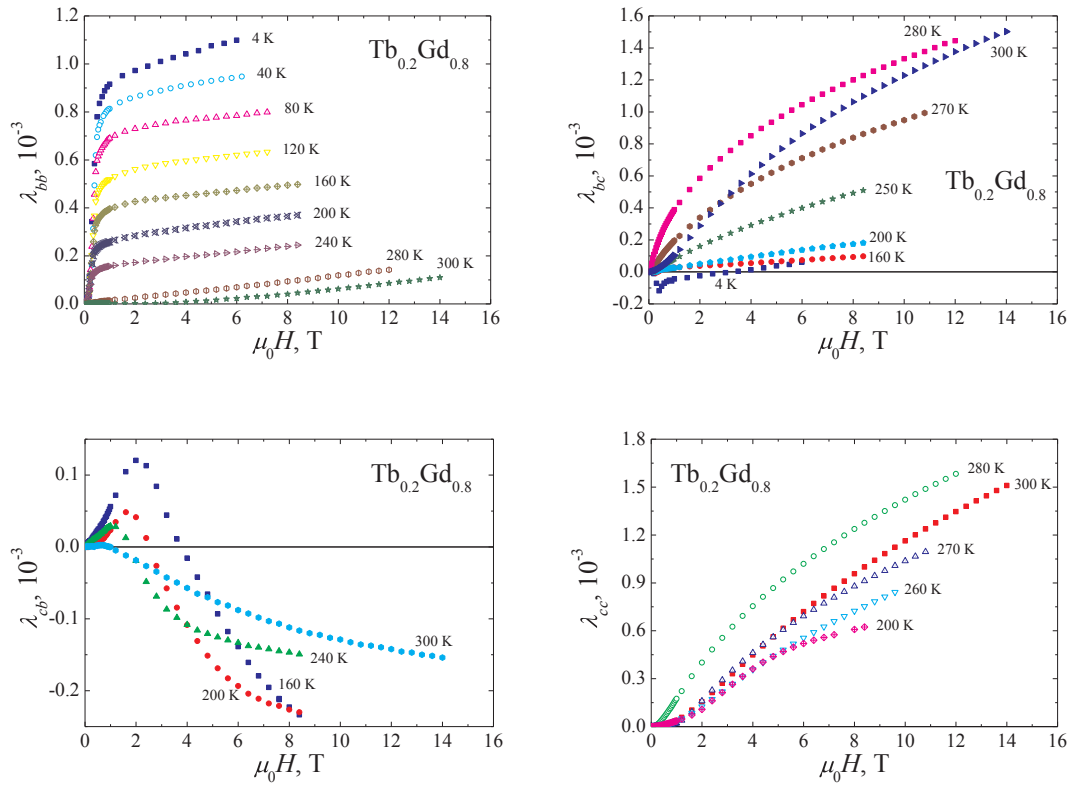


Fig. 1. Magnetic field dependencies of the λ_{bb} , λ_{bc} , λ_{cb} , λ_{cc} magnetostrictions at various temperatures.

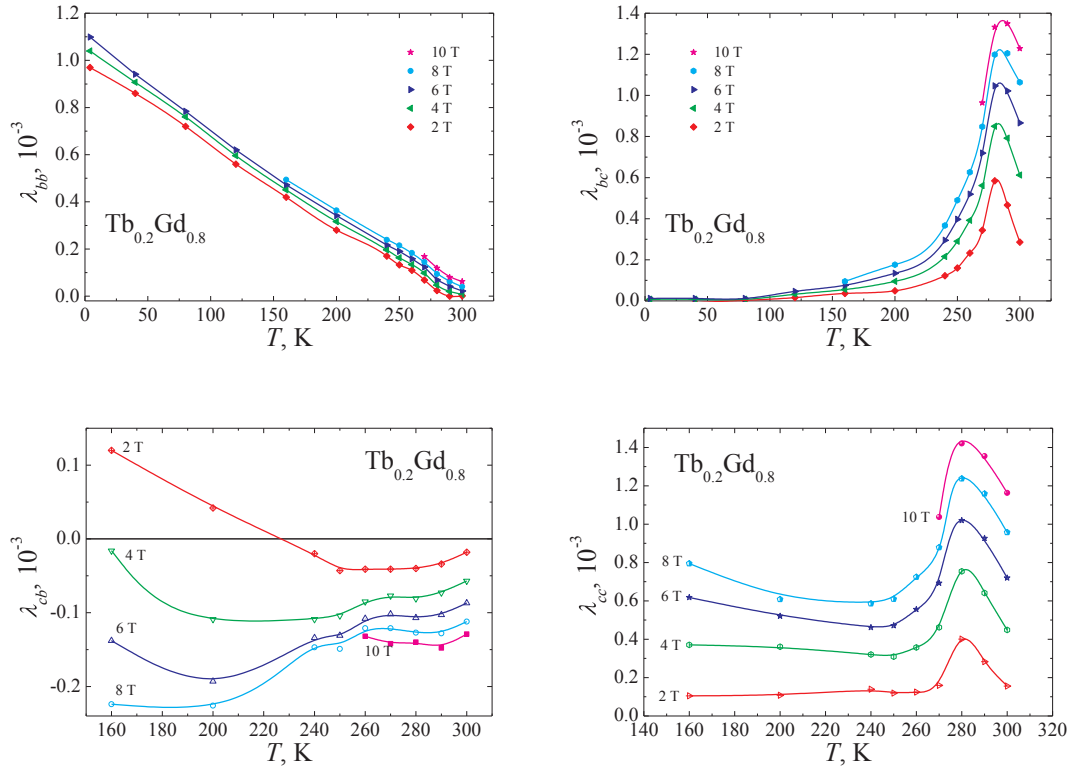


Fig. 2. Temperature dependencies of the λ_{bb} , λ_{bc} , λ_{cb} , λ_{cc} magnetostrictions in various magnetic fields.

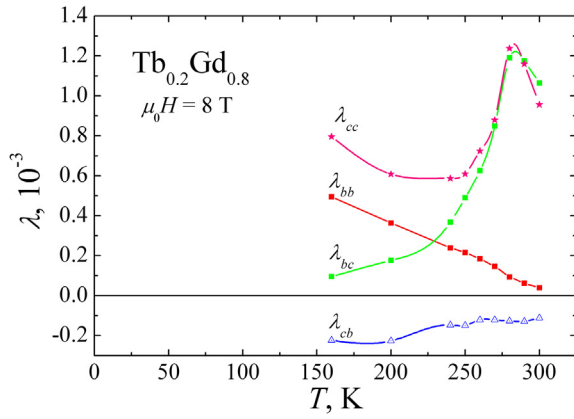


Fig. 3. Temperature dependencies of the λ_{cb} , λ_{bb} , λ_{cc} , λ_{bc} magnetostrictions in magnetic field $\mu_0 H = 8$ T.

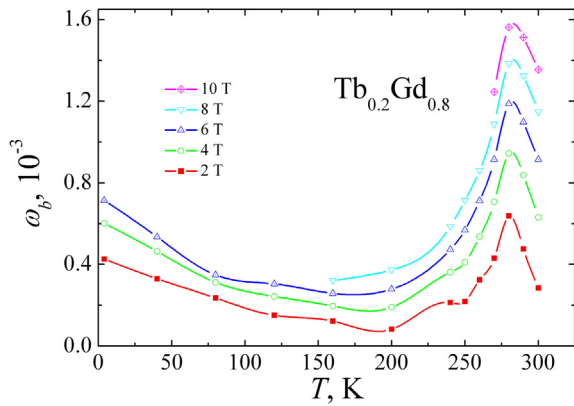


Fig. 4. Temperature dependencies of the ω_b volume magnetostriction in various magnetic fields.

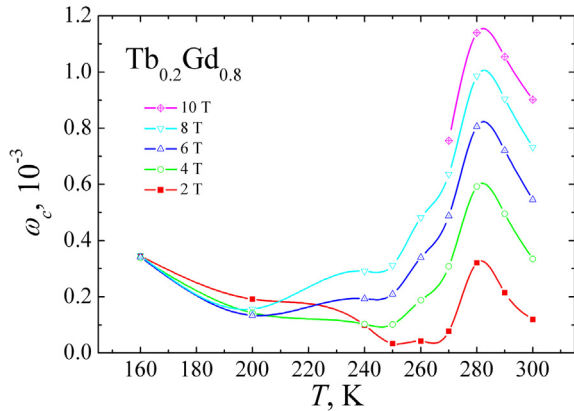


Fig. 5. Temperature dependencies of the ω_c volume magnetostriction in various magnetic fields.

was given by A. Clark et al. [13,14] and by Callens [12]. A. Clark [14] described the magnetostriction in rare-earth hexagonal crystals by following equation:

$$\lambda = \lambda_1^{\alpha,0} (\beta_x^2 + \beta_y^2) + \lambda_2^{\alpha,0} \beta_z^2 + \lambda_1^{\alpha,2} (\beta_x^2 + \beta_y^2) \left(\alpha_z^2 - \frac{1}{3} \right) + \lambda_2^{\alpha,2} \beta_z^2 \left(\alpha_z^2 - \frac{1}{3} \right) + \lambda^{\gamma,2} \left[\frac{1}{2} (\beta_x^2 - \beta_y^2) (\alpha_x^2 - \alpha_y^2) + 2\beta_x \beta_y \alpha_x \alpha_y \right] \quad (3)$$

where α_x , α_y , α_z are the direction cosines of the magnetization and β_x , β_y , β_z are those of the measurement direction. The $\lambda_1^{\alpha,0}$ and $\lambda_2^{\alpha,0}$ are isotropic magnetostriction constants, arising from the two-ion

exchange interaction term in the Hamiltonian, with values depending only on the absolute value of spontaneous magnetization M_s of the crystal (independent of M_s direction). Moreover, $\lambda_1^{\alpha,0}$ determines the strain in the basal plane of the crystal, $\lambda_2^{\alpha,0}$ determines the strain along the hexagonal c axis. The $\lambda_1^{\alpha,2}$ and $\lambda_2^{\alpha,2}$ are the constants of the anisotropic magnetostriction (arising from a sum of both single- and two-ion interactions), which describe the expansion and elongation of sample with preserving the symmetry of the crystal. The $\lambda^{\gamma,2}$ constant describes the distortion of circular symmetry in the basal plane, i.e. orthorhombic distortion. They depend on both the magnitude of the magnetization and its direction.

If the external magnetic field exceeds the magnetic anisotropy field (the maximum value of $\mu_0 H_a = K_2/2M_s$), then the direction of the field and the direction of magnetization vector coincide. Therefore, according to Eq. (3) λ_{bb} , λ_{bc} , λ_{cc} , λ_{cb} , λ_{ca} and λ_{ba} magnetostrictions can be presented as follows:

$$\lambda_{bb} = \lambda_1^{\alpha,0} - \frac{1}{3} \lambda_1^{\alpha,2} + \frac{1}{2} \lambda^{\gamma,2} \quad (4)$$

$$\lambda_{bc} = \lambda_2^{\alpha,0} - \frac{1}{3} \lambda_2^{\alpha,2} \quad (5)$$

$$\lambda_{cc} = \lambda_2^{\alpha,0} + \frac{2}{3} \lambda_2^{\alpha,2} \quad (6)$$

$$\lambda_{cb} = \lambda_1^{\alpha,0} + \frac{2}{3} \lambda_1^{\alpha,2} \quad (7)$$

$$\lambda_{ca} = \lambda_1^{\alpha,0} + \frac{2}{3} \lambda_1^{\alpha,2} \quad (8)$$

$$\lambda_{ba} = \lambda_1^{\alpha,0} - \frac{1}{3} \lambda_1^{\alpha,2} - \frac{1}{2} \lambda^{\gamma,2} \quad (9)$$

Then the magnetostriction constants $\lambda_1^{\alpha,2}$, $\lambda_2^{\alpha,2}$ and $\lambda^{\gamma,2}$ can be determined from Eqs. (4)–(9) using experimental data on λ_{bb} , λ_{bc} , λ_{cc} , λ_{cb} , λ_{ba} :

$$\lambda_1^{\alpha,2} = \lambda_{cb} - \lambda_{bb} + \frac{1}{2} \lambda^{\gamma,2} \quad (10)$$

$$\lambda_2^{\alpha,2} = \lambda_{cc} - \lambda_{bc} \quad (11)$$

$$\lambda^{\gamma,2} = \lambda_{bb} - \lambda_{ba} \quad (12)$$

Temperature dependencies of the magnetostriction constants are presented in Fig. 9 in magnetic field $\mu_0 H = 8$ T, which exceeds the magnetic anisotropy field ($H \parallel c$) and demagnetizing field ($H \parallel b$) (Fig. 6).

The constants $\lambda_1^{\alpha,0}$ and $\lambda_2^{\alpha,0}$ can be found only from thermal expansion measurement data by using it to determine spontaneous magnetostriction. Results of the thermal expansion measurement in the

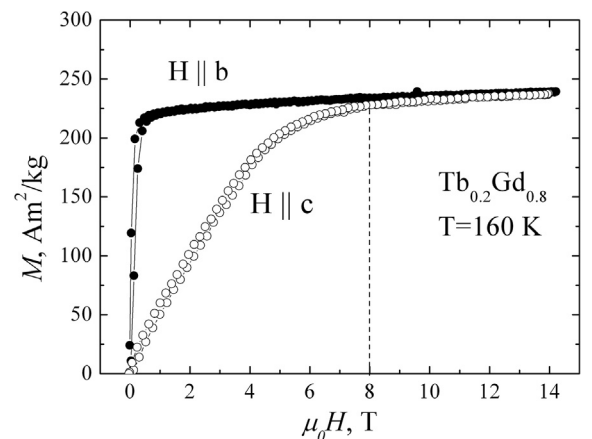


Fig. 6. Magnetic field dependencies of the magnetization along the c -axis and the b -axis at 160 K.

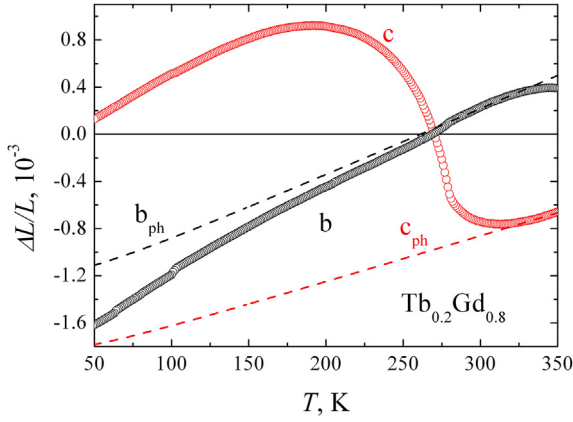


Fig. 7. Temperature dependencies of thermal expansion along *b* and *c* axes in zero magnetic field. The phonon contribution to thermal expansion is shown by dashed lines.

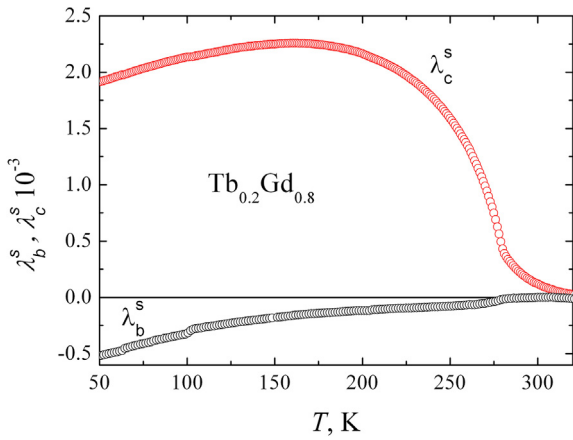


Fig. 8. Temperature dependencies of spontaneous magnetostrictions along *b* and *c* axes in zero magnetic field.

absence of a magnetic field are shown in Fig. 7. The spontaneous magnetostriction temperature dependencies obtained by subtracting the phonon contribution to the thermal expansion of the Tb_{0.2}Gd_{0.8} single crystal are presented in Fig. 8. Using Eq. (3) it is possible to obtain equations that determine spontaneous magnetostriction in the basal plane and along the *c* axis in the absence of a magnetic field:

$$\lambda_{bb}^s = \lambda_1^{\alpha,0} - \frac{1}{3}\lambda_1^{\alpha,2} + \frac{1}{2}\lambda^{\gamma,2} \quad (13)$$

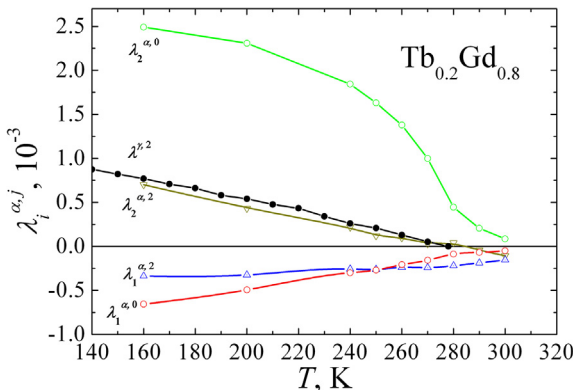


Fig. 9. Temperature dependencies of magnetostriction constants in magnetic field $\mu_0 H = 8$ T.

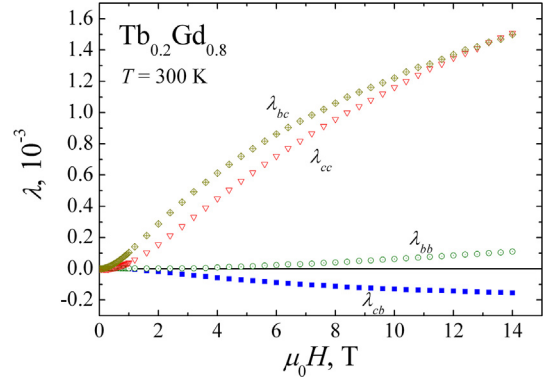


Fig. 10. Magnetic field dependencies of the λ_{cb} , λ_{bb} , λ_{cc} , λ_{bc} magnetostrictions at $T = 300$ K (above the Curie temperature).

$$\lambda_{bc}^s = \lambda_2^{\alpha,0} - \frac{1}{3}\lambda_2^{\alpha,2} \quad (14)$$

The temperature dependence of magnetostriction constants was determined using Eqs. (10)–(14) and experimental data Figs. 1, 2, 8. The temperature dependence of $\lambda^{\gamma,2}$ constant presented in Fig. 9 was determined using the equation $\lambda^{\gamma,2}(T) = \lambda^{\gamma,2}(4.2 \text{ K})(M_s(T)/M_s(4.2 \text{ K}))^3$ and $\lambda^{\gamma,2}(4.2 \text{ K}) = 1.5 \times 10^{-3}$ value obtained by Nikitin et al. [17].

The temperature dependencies of anisotropic magnetostriction constants $\lambda_1^{\alpha,2}$ and $\lambda_2^{\alpha,2}$ demonstrate maximum absolute values at low temperatures (see Fig. 9). With further increase in temperature up to T_C there is almost a linear decrease in the absolute values of $\lambda_1^{\alpha,2}$ and $\lambda_2^{\alpha,2}$. Magnetocrystalline interaction can be estimated using the values of magnetic anisotropy constants that sharply decrease approaching T_C (according to our previous results [16]). Thus, the contribution to magnetostriction from the single-ion interaction decreases when approaching the Curie temperature, and near T_C the contribution from the two-ion exchange interaction becomes prevailing. The competition of two contributions: a single-ion magnetocrystalline interaction and a two-ion exchange interaction, leads to complex temperature dependencies of the linear and volume magnetostrictions in the Tb_{0.2}Gd_{0.8} single crystal.

As mentioned above the applied magnetic field considerably exceeds the magnetic anisotropy field near T_C . The magnetostriction of the forced magnetization was observed in the field up to 14 T (Fig. 10). The λ_{cc} and λ_{bc} magnetostrictions at $T = 300$ K nearly coincide and reach the giant value of 1.4×10^{-3} , while λ_{cb} and λ_{bb} reach the values one order of magnitude lower. It was found that the λ_{cc} and λ_{bb} magnetostriction increases proportionally to the square of magnetization in the region of the Curie temperature (see Fig. 11) according to the equation:

$$\lambda_{cc} = \beta M^2 - \lambda_c^s \quad (15)$$

$$\lambda_{bb} = \beta M^2 - \lambda_b^s \quad (16)$$

where λ_c^s and λ_b^s is the spontaneous magnetostriction, and β is the magnetoelastic coefficient. It should be noticed that the values of spontaneous magnetization, defined from curves $\lambda_{cc}(M^2)$ and $\lambda_{bb}(M^2)$ close to the values found by thermal expansion data (Fig. 11). Therefore λ_{cc} is due to the forced magnetostriction near the Curie temperature.

We can conclude that the forced magnetostriction around T_C is caused by a strong anisotropy of exchange interaction and the experimental data obtained suggests the strong correlation between the exchange interaction and interatomic distances along the *c* axis.

5. Conclusion

The giant linear and volume magnetostriction was found at room (near T_C) and low temperatures in the Tb_{0.2}Gd_{0.8} single crystal. The

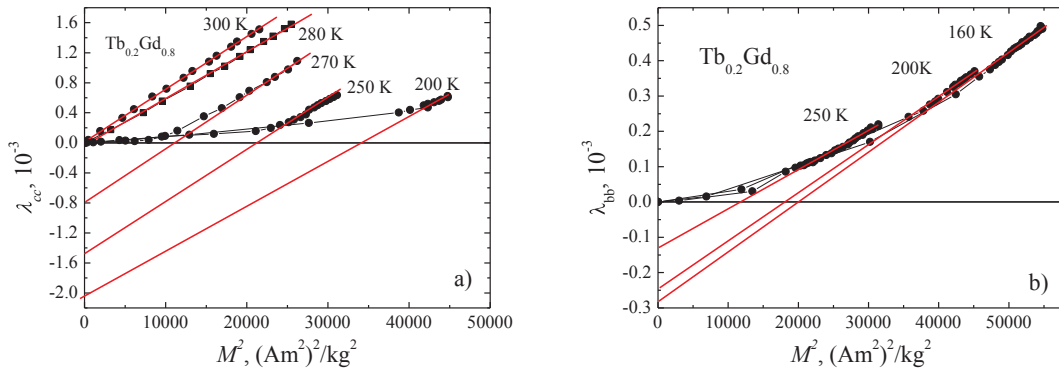


Fig. 11. λ_{cc} and λ_{bb} as function of M^2 . Lines intercept the λ -axis at the value of respective spontaneous magnetostrictions λ_c^s and λ_b^s .

temperature and field dependencies of the magnetostrictions along various crystallographic directions were measured. The temperature dependencies of magnetostriction constants $\lambda_1^{\alpha,0}$, $\lambda_2^{\alpha,0}$, $\lambda_1^{\alpha,2}$, $\lambda_2^{\alpha,2}$ and $\lambda^{\gamma,2}$ were determined. It was found that the λ_{cc} magnetostriction is a linear function of square of magnetization (M^2) and λ_{bb} grows nonlinearly with M^2 . The magnitude of spontaneous magnetostriction λ_c^s was determined from both the experimental curves of the thermal expansion and by using the dependencies $\lambda_{cc}(M^2)$ and $\lambda_{bb}(M^2)$. Therefore λ_{cc} behavior was effected by the forced magnetization near the Curie temperature.

It is shown that in the region of low temperatures the magnetostriction is caused by both magnetocrystalline anisotropy and exchange interactions. It is established that the two-ion exchange interaction is responsible for the giant magnetostriction arising along the c axis near T_C since in this direction the exchange interactions strongly depend on the interatomic distances. The results obtained are crucial for solving fundamental problems in the theory of magnetostriction for rare-earth based alloys. These results can also provide a constructive guide for the further development of new magnetostrictive materials, which can be used in precise transducers and actuators that are operated by magnetic field.

6. Author statement

All co-authors contributed equally to the article. Work on the article was led by prof. Nikitin.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This research was funded from the budget of the Institute of Low Temperature and Structure Research, and supported by RFBR (Russia) Grant № 16-02-00472 A.

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