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Magnetostriction of $\langle 1\ 1\ 0 \rangle$ oriented crystals in Tb_{0.36}Dy_{0.64}(Fe_{1−x}Co_x)₂ ($x = 0$ –0.30) alloys

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Abstract

Oriented polycrystalline rods of Tb_{0.36}Dy_{0.64}(Fe_{1-x}Co_x)₂ (where $x = 0.00, 0.10, 0.15, 0.20, 0.30$) have been successfully prepared by zone melting unidirectional solidification with high growth velocity of 240 mm/h and 900 mm/h. The measurement results show that (110) preferred orientation can be formed in the quaternary alloys with dendritic solidified morphologies. The magnetostriction was measured as a function of applied magnetic field up to 4 kOe and compressive stresses of 5 MPa and 10 MPa over a wide temperature range from −80 to 100 °C. The magnetostriction "jump" effect was obvious in all the Tb_{0.36}Dy_{0.64}(Fe_{1-x}Co_x)₂ (1 1 0) oriented crystals at room temperature and elevated temperature (100 ◦C), but not so obvious at cryogenic temperature (−80 ◦C). A satisfactory magnetostrictive property was obtained in the $\langle 110 \rangle$ oriented Tb_{0.36}Dy_{0.64}(Fe_{0.85}Co_{0.15})₂ crystal with the value of 1810×10^{-6} under 14 MPa pre-stress and 5 kOe magnetic field at room temperature.

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1. Introduction

Giant magnetostrictive materials based on (Tb,Dy)Fe₂ compound can be used in acoustic transducers, sensors and actuators because of their huge magnetostriction [\[1–3\].](#page-6-0) Considerable work has been done to improve the magnetostriction and extend the operating temperature range by partly substituting other rare earth elements for Tb or Dy, or other transition metals for Fe [\[4–6\].](#page-6-0) It is known that the (Tb,Dy)Fe2 compounds exhibit different magnetostriction properties which depend on their easy magnetization direction (EMD). The EMD of (Tb,Dy)Fe₂ compounds is $\langle 1 1 1 \rangle$ for $T > T_r$ and $\langle 100 \rangle$ for $T < T_r$ [\[7\],](#page-6-0) where T_r is the spin reorientation temperature of the compounds. Therefore, the large magnetostriction at room temperature can be kept to cryogenic temperature if T_r is low enough. Clark et al. have

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predicted that it is possible to tailor $Tb_xDy_{1-x}(Fe_{1-y}Co_y)_2$ to obtain optimum magnetostrictive properties over a wide temperature range, because Co substitution for Fe increases the Curie temperature T_c of Terfenol-D. However, the increase in Tb/Dy ratio decreases the spin reorientation temperature *T*^r [\[6\].](#page-6-0)

Due to the strongly magnetostrictive anisotropy, $\langle 1 1 1 \rangle$ oriented rods should exhibit excellent magnetostriction in the alloys. However, it was proved to be difficult to prepare oriented $(Tb, Dy)Fe₂$ crystals with the $\langle 1 1 1 \rangle$ preferred growth direction even by seed technique [\[8,9\].](#page-6-0) Good magnetostrictive property can be obtained from $\langle 1\,1\,2 \rangle$ oriented crystals [\[1\], a](#page-6-0)nd in some cases, the magnetostriction of $\langle110\rangle$ oriented crystals is not lower than that of $\langle 1\ 1\ 2 \rangle$ oriented crystals [\[10\].](#page-6-0) Our group has prepared $\langle 1 1 0 \rangle$ oriented (Tb,Dy)Fe₂ crystals by zone melting unidirectional solidification with a large ve-locity range [\[11\]. S](#page-6-0)hi et al. reported that $\langle 1\ 1\ 0 \rangle$ oriented crystals of the quaternary alloy $Tb_{0.5}Dy_{0.5}(Fe_{0.95}Mn_{0.05})_2$ can be grown by directional solidification [\[12\].](#page-6-0)

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In the present paper, we have prepared grain-aligned Tb_{0.36}Dy_{0.64}(Fe_{1−x}Co_x)₂ materials with high growth velocity of 240 mm/h and 900 mm/h and measured their magnetostrictive properties in the temperature range from −80 to 100 ◦C. Distinct dendritic morphologies were observed and the grains were regularly aligned along the solidification direction (the axial direction of the rod). A clear magnetostriction jump effect can be observed in all the crystals at room temperature and elevated temperature, but not at cryogenic temperature.

2. Experimental

The purity of the constituents was as follows: Tb, Dy and Fe 99.9%, Co 99.8%. Samples of Tb_{0.36}Dy_{0.64}(Fe_{1-x}Co_x)₂ alloys were prepared by arc melting of the appropriate constituent metals in a high-purity argon atmosphere then arc cast into rods with a diameter of 6.8 mm. Oriented crystals were fabricated by a self-made crystal growth device. It has a super high temperature gradient around 400 ◦C/cm and the rare earth melting loss can be minimized by control of temperature and growth velocity. The oriented rods with a diameter of 7.2 mm were prepared with a velocity of 240 mm/h and 900 mm/h. An optical microscope was used to observe the solidified morphology of the longitudinal and transverse sections. A Regaku D/max 2200 pc X-ray diffractometer with Cu K α radiation was used to check the preferred orientations on the transverse section. The magnetostrictive strains were measured by standard resistant strain gauge on samples with a length of 25 mm in the temperature range from −80 to 100 ◦C. A gas pressure cell was employed to produce 0–15 MPa axial pre-stress. Elevated and cryogenic temperatures were reached in a homemade heating device with the error less than ± 1 °C.

3. Results and discussion

3.1. 110- *preferred orientation formation*

Transverse sections of the rods prepared with growth velocities of 240 mm/h and 900 mm/h were employed to determine the crystal orientations by X-ray diffraction, as shown in Fig. 1. The ratio of the intensity of the (2 2 0) peak and $(1 1 3)$ peak $I_{(220)}/I_{(113)}$ is 4.07 for $x = 0.00$, 1.05 for $x = 0.10$, 3.06 for *x* = 0.15, 1.83 for *x* = 0.20 and 1.70 for *x* = 0.30, which can be calculated from the X-ray patterns. It demonstrates that the (2 2 0) peak is remarkably the strongest one among all the diffraction peaks for each specimen, since it is known that the (1 1 3) diffraction peak is the strongest one of the $(Tb, Dy)Fe₂$ Laves phase for polycrystalline samples, this indicated that all the oriented crystal possess $\langle 1\ 1\ 0 \rangle$ preferred orientation. Oxide and RFe₃ phases cannot be seen in the X-ray patterns. It demonstrates that the $\langle 1\,1\,0 \rangle$ preferred orientation of Tb_{0.36}Dy_{0.64}(Fe_{1−x}Co_x)₂ alloys can be suc-

Fig. 1. X-ray diffraction patterns from the transverse sections of the oriented Tb_{0.36}Dy_{0.64}(Fe_{1-x}Co_x)₂ samples with growth velocities of 240 mm/h or 900 mm/h.

cessfully obtained with high growth velocity by the present unidirectional solidification method.

The solidification morphologies of the longitudinal sections of each oriented sample obtained with growth velocities of 240 mm/h and 900 mm/h were examined and are shown in [Fig. 2. I](#page-2-0)t is clear that all the $\langle 1\ 1\ 0 \rangle$ oriented crystals show dendritic morphologies, and the phases were arranged uniformly and regularly. The rare-earth rich phase was distributed at the boundaries of the matrix Laves phase. No 1:3 phases can be seen in the microstructures. The solid–liquid interface remains planar when the growth velocity is low enough, and becomes unstable with the increase of the growth velocity, where the dendritic morphologies developed in the oriented Tb_{0.36}Dy_{0.64}(Fe_{1−x}Co_x)₂ crystals when grown with velocities of 240 mm/h and 900 mm/h. In fact, during the quick unidirectional solidification, crystals with different orientations compete with each other, and the preferred growth of the crystals will dominate other growth directions and gradually govern the whole transverse section. $\langle 1\ 1\ 0 \rangle$ oriented TbDyFe crystals grow with two kinds of $\{111\}$ -twin growth mech-anisms as described by Jiang et al. [\[13\].](#page-6-0) A two- $\{1\ 1\}$ -twin growth mechanism was found for low velocity growth, and a single-{111}-twin growth mechanism for high velocity growth. The $\langle 1\ 1\ 0 \rangle$ oriented Tb_{0.36}Dy_{0.64}(Fe_{1−x}Co_x)₂ crystals obtained with velocities of 240 mm/h or 900 mm/h were formed by the single- $\{111\}$ -twin growth mechanism.

Typical optical morphologies from the transverse sections of $Tb_{0.36}Dy_{0.64}Fe_2$ and $Tb_{0.36}Dy_{0.64}(Fe_{0.85}Co_{0.15})_2$ are shown in [Fig. 3.](#page-4-0) It also shows that the crystals grown with 240 mm/h or 900 mm/h have dendritic morphologies. It is clear that the crystals were formed by many thin flats, as reported by Wu et al. [\[9\].](#page-6-0) The quantity of the rare-earth rich phase of $Tb_{0.36}Dy_{0.64}Fe_2$ as shown in [Figs. 2a and 3a](#page-2-0) was larger than that of $Tb_{0.36}Dy_{0.64}$ (Fe_{0.85}Co_{0.15})₂ as shown in [Figs. 2c and 3b.](#page-2-0) This indicates that the addition of Co decreases the amount of rare-earth rich phase. In another

Fig. 2. Solidified morphologies of the longitudinal sections of the oriented Tb_{0.36}Dy_{0.64}(Fe_{1-x}Co_x)₂ samples. (a) $x = 0.0$; (b) $x = 0.10$; (c) $x = 0.15$; (d) $x = 0.15$ 0.20; (e) $x = 0.30$.

Fig. 2. (*Continued*).

word, the Co addition is beneficial for the formation of the Laves phase, similar as in the Pr_{0.4}D_{V0.6}(Fe_{1−x}Co_x)₂ [\[14\]](#page-6-0) and Tb_{0.2}Pr_{0.8}(Fe_{1−x}Co_x)₂ systems [\[15\].](#page-6-0)

3.2. Magnetostriction

[Fig. 4](#page-4-0) shows the magnetostrictive properties of oriented crystals with compositions of $Tb_{0.36}Dy_{0.64}Fe_2$ and $Tb_{0.36}Dy_{0.64}$ (Fe_{0.9}Co_{0.1})₂ at room temperature. As shown in [Fig. 4a,](#page-4-0) for an oriented $Tb_{0.36}Dy_{0.64}Fe_2$ crystal, the magnetostriction in an applied field of 4 kOe is 794 \times 10⁻⁶ under free conditions, and increases to 997 \times 10⁻⁶ and 1186 \times 10⁻⁶ when the pre-stress is 5 MPa and 10 MPa, respectively. The increase ratio of the magnetostriction is 25.6% and 49.4%. As shown in [Fig. 4b,](#page-4-0) for an oriented $Tb_{0.36}Dy_{0.64}$ (Fe_{0.90}Co_{0.10})₂ crystal, the magnetostriction in 4 kOe is 810 \times 10⁻⁶, 1027 \times 10⁻⁶ and 1296 \times 10⁻⁶ under free conditions, or prestress of 5 MPa and 10 MPa, respectively. The increase ratio of the magnetostriction is 26.8% and 60.0%. The conclusion can be drawn that an obvious magnetostrictive jump

effect was obtained in the oriented $Tb_{0.36}Dy_{0.64}Fe_2$ and Tb_0 36Dy_{0.64}(Fe_{0.90}Co_{0.10})₂ crystals when pre-stress was applied at room temperature. Under free conditions, the magnetostriction in $4kOe$ of Tb_{0.36}Dy_{0.64}Fe₂ is smaller than that of $Tb_{0.36}Dy_{0.64}$ (Fe_{0.90}Co_{0.10})₂. Clark et al. considered that Co addition decreases the saturated magnetostriction λ_s in Tb_{0.3}Dy_{0.7}(Fe_{1-x}Co_x)_{1.9} polycrystalline materials [\[16\].](#page-6-0) The different magnetostriction between $Tb_{0.36}Dy_{0.64}Fe_2$ and $Tb_{0.36}Dy_{0.64}$ (Fe_{0.90}Co_{0.10})₂ oriented crystals must be related with the different preferred orientation degree, besides the composition difference.

[Fig. 5](#page-5-0) shows the magnetostrictive properties of oriented $Tb_{0.36}Dy_{0.64}$ (Fe_{0.85}Co_{0.15})₂, $Tb_{0.36}Dy_{0.64}$ (Fe_{0.80}Co_{0.20})₂ and $Tb_{0.36}Dy_{0.64}$ (Fe_{0.70}Co_{0.30})₂ crystals at room temperature (20 \degree C), elevated temperature (100 \degree C), and cryogenic temperature $(-80 °C)$. As shown in [Fig. 5a](#page-5-0), under free conditions, the magnetostriction in 4 kOe of the oriented Tb_{0.36}Dy_{0.64}(Fe_{0.85}Co_{0.15})₂ crystal is 1507×10^{-6} , 1224 \times 10⁻⁶ and 1024 \times 10⁻⁶ at −80 °C, 20 °C and 100 °C, respectively. The magnetostriction in 4 kOe decreases

Fig. 3. Typical optical morphologies from the transverse sections of oriented samples. (a) $Tb_{0.36}Dy_{0.64}Fe_2$; (b) $Tb_{0.36}Dy_{0.64}(Fe_{0.85}Co_{0.15})_2$.

Fig. 4. Magnetostriction of oriented samples at room temperature (20 °C). (a) $Tb_{0.36}Dy_{0.64}Fe_2$; (b) $Tb_{0.36}Dy_{0.64}(Fe_{0.9}Co_{0.1})_2$.

Fig. 5. Magnetostriction of oriented samples at room temperature (20 ◦C), elevated temperature (100 ◦C), and cryogenic temperature (−80 °C). (a) Tb_{0.36}Dy_{0.64}(Fe_{0.85}Co_{0.15})₂; (b) Tb_{0.36}Dy_{0.64}(Fe_{0.8}Co_{0.2})₂; (c) $Tb_{0.36}Dy_{0.64}$ (Fe_{0.7}Co_{0.3})₂.

with increasing the temperature. When 10 MPa pre-stress was applied, the magnetostriction increases to $1627 \times$ 10^{-6} , 1677×10^{-6} and 1368×10^{-6} , respectively, the corresponding ratio is 7.9%, 37% and 33.6%. It suggests that the magnetostrictive jump effect in the oriented $Tb_{0.36}Dy_{0.64}$ (Fe_{0.85}Co_{0.15})₂ crystal is very obvious at room temperature (20 \degree C) and elevated temperature (100 \degree C), but almost does not occur at cryogenic temperature (−80 ◦C).

In the $Tb_{0.36}Dy_{0.64}$ (Fe_{0.80}Co_{0.20})₂ and $Tb_{0.36}Dy_{0.64}$ $(Fe_{0.70}Co_{0.30})_2$ oriented crystals, their magnetostrictive properties are similar to $Tb_{0.36}Dy_{0.64}(Fe_{0.85}Co_{0.15})_2$, as shown in Fig. 5b and c.

As seen from [Figs. 4 and 5,](#page-4-0) an obvious magnetostrictive jump effect can be observed in all the $\langle 1\,1\,0 \rangle$ oriented crystals at room temperature and elevated temperature. According to our previous work [\[11\],](#page-6-0) λ^{P} , the magnetostriction measured under a pre-stress can be calculated from the following equation:

$$
\lambda^{P} = 3\lambda_{111}(\alpha_{1}\alpha_{2}\beta_{1}\beta_{2} + \alpha_{2}\alpha_{3}\beta_{2}\beta_{3} + \alpha_{3}\alpha_{1}\beta_{3}\beta_{1})
$$

+
$$
\frac{3}{2}\lambda_{100}(\alpha_{1}^{2}\beta_{1}^{2} + \alpha_{2}^{2}\beta_{2}^{2} + \alpha_{3}^{2}\beta_{3}^{2} - \frac{1}{3})
$$
 (1)

where α_i and β_i are the direction cosines of the magnetization and measurement direction, respectively, with respect to the cubic axis. Because $\lambda_{111} \gg \lambda_{100}$ [\[1\],](#page-6-0) the value of λ_{100} was ignored here. Following the single-{111}-twin growth mechanism, the magnetostriction λ^P of both the $\langle 110 \rangle$ and $\langle 112 \rangle$ oriented Tb_{0.36}Dy_{0.64}(Fe_{1−x}Co_x)₂ samples should be equal to 1.25 λ_{111} . Substituting $\lambda_{111} = 1640 \times$ 10^{-6} into Eq. (1), the theoretical value of Tb_{0.36}Dy_{0.64}Fe₂ should be 2050 \times 10⁻⁶. Co addition decreases the value of λ_{111} in Tb_{0.3}Dy_{0.7}(Fe_{1-x}Co_x)_{1.9} alloys [\[16\],](#page-6-0) due to the negative contribution to the magnetostriction from the Co sublattice [\[17\].](#page-6-0) It can be estimated that the Co addition also decreases the value of λ_{111} in the Tb_{0.36}Dy_{0.64}(Fe_{1−x}Co_x)₂ alloys. As shown in Fig. 5a, the magnetostriction of $Tb_{0.36}Dy_{0.64}$ (Fe_{0.85}Co_{0.15})₂ $\langle 1 1 0 \rangle$ oriented crystal in 5 kOe under 14 MPa pre-stress at room temperature is 1810×10^{-6} , which is approaching the theoretical value. It is reasonable to assume that the $\langle 110 \rangle$ oriented Tb_{0.36}Dy_{0.64}(Fe_{1-x}Co_x)₂ crystals obtained with high growth velocities follow the single-{111}-twin growth mechanism.

4. Conclusions

By zone melting unidirectional solidification, $\langle 110 \rangle$ oriented crystals of Tb_{0.36}Dy_{0.64}(Fe_{1−x}Co_x)₂ ($x = 0.00, 0.10,$ 0.15, 0.20, 0.30) alloys can be successfully prepared with high growth velocities of 240 mm/h and 900 mm/h. Developed dendrites are observed in both the longitudinal and transverse sections of the $\langle 110 \rangle$ oriented crystals. The Co addition is beneficial for Laves phase formation in Tb_{0.36}Dy_{0.64}(Fe_{1−x}Co_x)₂ alloys. A clear magnetostrictive jump effect is observed at room temperature $(20\degree C)$ and elevated temperature (100 \degree C), but it is less clear at cryogenic temperature (-80° C). A rough calculation was made based on the single- $\{111\}$ -twin growth mechanism, and the experimental magnetostriction of the $\langle 110 \rangle$ oriented $Tb_{0.36}Dy_{0.64}$ (Fe_{0.85}Co_{0.15})₂ crystal is approaching the theoretical value. It can be considered that the $\langle 1\,1\,0 \rangle$ oriented Tb_{0.36}Dy_{0.64}(Fe_{1−x}Co_x)₂ crystals obtained with high growth velocities were formed by the single- $\{111\}$ -twin growth mechanism.

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