Magnetoelastic lattice distortion in the spin-reorientation region of Tb$_2$Dy$_{1-x}$Fe$_2$

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The spin-reorientation transition from [111] to [100] easy axes in Tb$_2$Dy$_{1-x}$Fe$_2$ for $0.29<x<0.395$ has been studied by X-ray diffraction and a.c. susceptibility measurements. It was determined from the type of the magnetoelastic lattice distortion that the spin-reorientation occurs through an angular $<uvw>$-type phase in annealed samples, whereas as-cast samples show the transition through a mixture of $<100>$ and $<111>$ phases. After annealing the transition temperature lowered by 10-20 degrees.

Key words: intermetallic compounds, spin-reorientation, susceptibility, X-ray diffraction, internal stress, spontaneous magnetostriction

1. Introduction

Rare-earth quasi-binary intermetallic Tb$_2$Dy$_{1-x}$Fe$_2$ compounds crystallize in the cubic MgCu$_2$-type structure (Laves phase C15). Due to the giant anisotropic magnetostriction, they exhibit a large spontaneous magnetoelastic lattice distortion in the magnetically ordered state below 700 K. The symmetry of the distortion is determined by the orientation of the easy axis. When Tb atoms are partially substituted by Dy in TbFe$_2$, the magneto-crystalline anisotropy decreases. At a definite temperature for the concentration range $0.25<x<0.45$ the spin-reorientation transition (SRT) occurs. The easy axis changes its orientation from [111] direction to [100] when the temperature decreases. The SRT comes through an intermediate phase, which is stable within a narrow temperature range. The available data on the easy axis orientation in the intermediate phase are contradictory. According to$^{1,2)}$, the SRT occurs through a gradual rotation of the magnetization vector from [111] toward [100] within (110) plane. Indeed, authors of$^3$ reported a mixture of $<100>$ and $<111>$-type phases within the temperature range of SRT. Besides, the reported transition temperatures are different.

SRT are usually studied by the Mössbauer effect, a.c. susceptibility, magnetization and sound velocity measurements. Due to the giant spontaneous magnetoelastic lattice distortion in high-magnetostrictive compounds, X-ray diffraction technique can be used for the SRT study.$^4$ Present paper reported the study of the spin-reorienation in Tb$_2$Dy$_{1-x}$Fe$_2$ for $0.29<x<0.395$ by X-ray diffraction and a.c. susceptibility measurements.

2. Experimental details

The alloys were prepared by an induction melting under argon atmosphere. As-melted samples contain a small amount (~3 %) of (Tb$_2$Dy)$_2$Fe$_3$ phase together with a pure (Tb$_2$Dy) phase. After annealing at 1100°C for 24 hours the samples were confirmed to be single phase with MgCu$_2$ structure.

A.c. susceptibility ($\chi$) was measured in a field 100 A/m of the frequency 74 Hz in the temperature range 77-300 K. X-ray diffraction studies were performed with a monochromatized Cr K$_\alpha$ radiation in the temperature range 77-300 K using a low-temperature vacuumized chamber. Powdered samples were attached to a copper sample holder by a grease having high thermal conductivity. The experimental shape of X-ray diffraction lines was compared with the calculated ones. The details of the fitting procedure were described elsewhere.$^5$

3. Results and discussion

Figure 1 shows temperature dependences of the susceptibility measured for three samples of different composition. The minimum of the magneto-crystalline anisotropy energy near SRT is responsible for a maximum observed on $\chi$(T) dependences. The maximum broadened with decreasing $x$ and shifted to the high temperature region. X-ray diffraction studies showed that the temperature of the maximum of $\chi$(T) dependence (T$_m$)

![Fig.1. Temperature dependence of a.c. susceptibility for Tb$_2$Dy$_{1-x}$Fe$_2$ with different x.](image-url)
Fig. 2. Concentration dependence of the spin-reorientation temperature for TbDy₁₋ₓFe₂.<br>

corresponds to the center of the SRT, where a maximum rate of the structural changes is observed. Figure 2 shows the concentration dependence of the susceptibility maximum of the studied samples.<br>

The temperature of the SRT was found to be different for as-cast and annealed samples. The annealing reduces the SRT temperature by 10 - 20 degrees. Figure 3 shows the difference between χ(T) dependences for as-cast and annealed samples of two compositions: x=0.35 and 0.395. It should be noted that the width of χ(T) maximum for Tb₀.₃₉₅Dy₀.₆₀₅Fe₂ remains unchanged, indicating the homogeneous distribution of substituted elements over the sample even before annealing. The shift of the SRT after annealing can be attributed to the possible change of Tb/Dy ratio. Such explanation assumes that available in the samples before annealing phases (Tb,Dy)Fe₃ and pure (Tb,Dy) enriched by Tb. According to our estimations, appearance of just 2.5 % of TbFe₃ or less than 1 % of pure Tb can be responsible for the observed Tₘ shift.<br>

Not only the temperature but also the type of SRT was found to depend on the sample annealing. Figures 4 and 5 showed the shape of (440) diffraction line in the temperature range of SRT for as-cast and annealed Tb₀.₃₉₅Dy₀.₆₀₅Fe₂ sample, respectively. The experimental lines were compared with the calculated one in the assumption of both gradual rotation of the magnetization vector and the mixture of <100> and <111> phases. As-cast sample shows the spin-reorientation transition through the mixture of phases, whereas in the annealed sample the magnetization vector gradually rotates from [100] to [111] direction within (110) plane, forming [uvw]-type angular phase.<br>

Observation of the SRT of different type in the same sample allows us to understand the above mentioned discrepancy of available data.

Fig. 3. Temperature dependences of a.c. susceptibility for Tb₀.₃₉₅Dy₀.₆₀₅Fe₂: as-cast (dashed line) and annealed at 1100°C for 24 h (solid line).

Fig. 4. The shape of the (440) diffraction line for as-cast Tb₀.₃₉₅Dy₀.₆₀₅Fe₂ at different temperatures. Dashed lines show the calculated positions and intensities of the splitted Kₐ₁ reflex.
Fig. 5. The shape of (440) diffraction line for annealed \( \text{Tb}_{0.395}\text{Dy}_{0.605}\text{Fe}_2 \) at different temperatures. Dashed lines show the calculated positions and intensities of the split K\(_a1\) reflex.

By minimizing the difference between the intensities of experimental and calculated (440) diffraction lines using the standard least-square fitting, we obtained the temperature dependence of the angle between [100] axis and the spontaneous magnetization vector (Fig. 6). The rotation was assumed to be within (110) plane.

The change in the type of SRT can be attributed to the influence of internal stresses caused by the structural defects due to relatively fast cooling of the alloy. Since the magnetostriction \( \lambda \) is strongly anisotropic for these compounds (\( \lambda_{111} \gg \lambda_{100} \)), the internal stress produces an additional uniaxial anisotropy oriented along <111>-type direction closest to the axis of the stress. The coexistence of the parent cubic and the additional uniaxial anisotropies suppresses the gradual rotation of the magnetic moment and leads to the first-order magnetization process.

Analysis of the shape of X-ray diffraction lines allows us to assume a crystallographic alignment of the studied powdered samples. The alignment was especially significant for the alloys with \( x = 0.3 \) and 0.31, having a small magnetocrystalline anisotropy near the room temperature. Easy axes were oriented preferably in the plane of the sample holder.

4. Conclusion

In order to clarify the origin of the spin-reorientation transition in giant-magnetostrictive \( \text{Tb}_{0.395}\text{Dy}_{0.605}\text{Fe}_2 \) quasi-binary compounds, several alloys for \( 0.29 < x < 0.395 \) were prepared and studied by a.c. susceptibility and X-ray diffraction measurements. It was found that as-cast alloys exhibit the SRT through a mixture of <111> and <100> magnetic phases, whereas annealed alloys show a gradual easy axis rotation through <uvw>-type angular phase. Additionally, the temperature the SRT lowered by 10 - 20 degrees after annealing.

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References