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Structural, magnetic and magnetostrictive studies of $Tb_{0.27}Dy_{0.73}(Fe_{1-x}Al_x)_2$

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Abstract

Measurements of magnetic properties, X-ray diffraction and magnetostriction were made on $Tb_{0.27}Dy_{0.73}(Fe_{1-x}Al_x)_2$ ($x=0.1$, 0.2, ..., 0.7) compounds. It was found that the system has the cubic $MgCu_2$ structure over almost the whole (Fe,Al) concentration range investigated, except for a narrow intermediate range ($x=0.4$–0.6) where the hexagonal $MgZn_2$ structure appears. With increasing Al content x, the lattice constant a increases linearly with x. The first replacement of Fe results in a marked decrease in the Curie temperature, which is followed by a slight decrease in T_C with x. A linear decrease in magnetostriction of $|\lambda_{\parallel}-\lambda_{\perp}|$ at room temperature with x was also observed from 1530×10^{-6} for $x=0$ to 36×10^{-6} for $x=0.3$. The saturation magnetization σ_s exhibits a complex concentration dependence in the $Tb_{0.27}Dy_{0.73}(Fe_{1-x}Al_x)_2$ system: in the range $x<0.5$, σ_s increases linearly with x and, for $x=0.5$–0.6, σ_s decreases and then increases again. An enhancement of the magnetic 'hardness' in this system was also observed at low temperature.

1. Introduction

Some of the cubic rare earth (R)-Fe Laves phases have unusually large anisotropy energies and extremely large room temperature magnetostrictions and have been under investigation for several decades [1, 2]. The huge magnetostriction was attributed to the large spin orbit interaction between the electron spins and the specially anisotropic 4f charge cloud, giving rise to strong coupling between the magnetic anisotropy, exchange and elastic energies. In efforts to decrease the anisotropy energy, many pseudobinary compounds with compensated anisotropy have been reported. Among these, $Tb_{0.27}Dy_{0.73}Fe_2$ is well known to be a developed as commercial material with compensated anisotropy and a large magnetostriction. In contrast, it was found that upon substitution of Al in $R(FeAl)_2$, a structural transition and a variety of field dependences of the magnetization occurred at low temperature [3–11]. It was assumed that these behaviours result from the high anisotropy of the $DyAl_2$ or $TbAl_2$ matrix, and local disorder of the (Fe,Al) sublattice. In this paper, the changes in the magnetic properties and magnetostriction upon substitution of Al for Fe in intermetallic compounds of $Tb_{0.27}Dy_{0.73}(Fe_{1-x}Al_x)_2$ will be reported.

2. Experimental details

The intermetallic compounds $Tb_{0.27}Dy_{0.73}(Fe_{1-x}Al_x)_2$ ($x=0.1$–0.7) were prepared by arc melting of a stoichiometric mixture of the raw materials Tb, Dy, Fe and Al with 99.9% purity in an Ar atmosphere. The ingots were annealed in a vacuum quartz tube at 1073 K for one week. Phase analysis was carried out by means of X-ray powder diffraction with Co Kα radiation. The magnetization measurements at 1.5 K were carried out using an extracting sample magnetometer in applied fields up to 5.2×10^3 kA m^{-1} ($\mu_0 H=6.5$ T) on the samples of ø4 mm in diameter and 6 mm thick. The temperature dependence of the magnetization was measured on a vibrating sample magnetometer. A strain gauge was used to measure the magnetostriction of the specimen (ø12 mm$\times$1.5 mm) in an applied field up to 1.6×10^3 kA m^{-1} at room temperature.

3. Results and discussion

From the results of X-ray diffraction, it was found that the system exhibits cubic $MgCu_2$ structure over almost the whole (Fe,Al) concentration range investigated, except for a narrow intermediate range ($x=0.4$–0.6), where the hexagonal $MgZn_2$ structure appears. In addition with increasing Al content x, the lattice constant a increases linearly with x from 7.345 Å for $x=0$ to 7.695 Å for $x=0.7$. This is shown in Fig. 1, where $a=(3^{1/2}a^2c)^{1/3}$ is the 'hypothetical C15 lattice constant for the C14 structure alloys'. Previously, the structure transition in the intermediate range of Al

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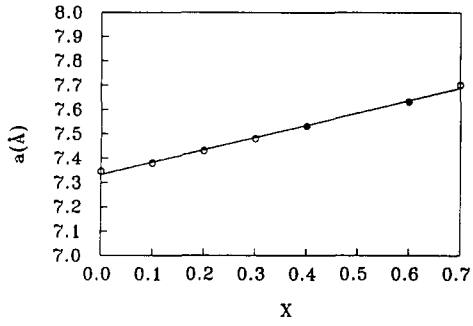

Fig. 1. The lattice constant a as a function of the Al content x in $Tb_{0.27}Dy_{0.73}(Fe_{1-x}Al_x)_2$ compounds: ○, cubic $MgCu_2$ structure; ●, hexagonal $MgZn_2$ structure.

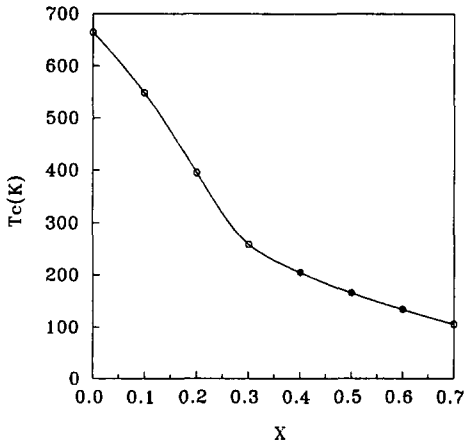

Fig. 2. The dependence of T_C on the Al content x in $Tb_{0.27}Dy_{0.73}(Fe_{1-x}Al_x)_2$ compounds: ○, cubic $MgCu_2$ structure; ●, hexagonal $MgZn_2$ structure.

content also was observed in other $R(Fe,Al)_2$ compounds [4]. It was suggested that the phase change in the Fe-rich range is driven by the increase in the electron concentration, whereas size-effects were regarded to be important in the Al-rich region. Also, previous investigations indicated that the local disorder of the (Fe,Al) sublattice in the $MgCu_2$-type $R(Fe, Al)_2$ system has a marked effect on the line width in X-ray diffraction and the magnetic behaviour.

From the temperature dependence of magnetization measured at an applied field $H=80$ kA m^{-1} ($\mu_0 H=0.1$ T), the Curie temperature T_C of each sample was obtained by plotting σ^2 *vs.* temperature. Figure 2 gives the T_C dependence on the Al content x. Compared with other $R(Fe, Al)_2$ systems [5, 8], the concentration dependence of the ordering temperature exhibits a similar trend: T_C decreases significantly with the first Al substitution up to $x=0.3$; with further increase in the Al content, T_C decreases slowly. According to the results of Sima *et al.* [8] and Hilscher *et al.* [10], it is evident that, for $x<0.22$, the 3d–3d exchange plays the most significant role in the magnetic ordering. For $x>0.22$, the 3d–3d exchange fails to establish effectively the magnetic ordering and the 3d–4f exchange increases in importance in the vicinity of the hexagonal region, while the 4f–4f exchange becomes predominant in the Al-rich range.

By fitting the magnetization curves of $\sigma(H)$ measured at 1.5 K to the law of approach to saturation, expressed as

$$\sigma=\sigma_s(1-a/H-2b/H^2)+\chi_p H$$

the saturation magnetization σ_s was derived for the various $Tb_{0.27}Dy_{0.73}(Fe,Al)_2$ samples. A linear increase in the saturation magnetization with increasing Al content was obtained for $x<0.5$. It seems that the concentration dependence of the saturation magnetization is more complex on the Al-rich side, as shown in Fig. 3. The initial linear increase in σ_s can be explained by a gradually decreasing concentration of the Fe moments in the (Fe,Al) sublattice of the ferrimagnetic $Tb_{0.27}Dy_{0.73}(Fe,Al)_2$ owing to the substitution of Al for Fe. According to the results of Sima *et al.* [8], the complex dependence of σ_s on x is attributed to a non-collinear magnetic structure in the $R(Fe,Al)_2$ system, where fluctuating orientations of the local easy axes of the Dy, Tb and Fe moments exist. It is proved that the magnetic disorder is caused mainly by local lattice distortion, in turn caused by differences in the ionic radii of the substituting elements.

In addition, the $Tb_{0.27}Dy_{0.73}(Fe_{1-x}Al_x)_2$ system, similarly to other $R(Fe,Al)_2$ systems [3, 8], displays a behaviour typical of 'hard' magnetic materials. This is shown by properties such as the following.

(1) Critical fields exist in the $\sigma(H)$ curve at low temperature. Typical jumps were found in the $\sigma(H)$ curves at 1.5 K for all the samples with Al. Figure 4 gives an example for $x=0.5$.

(2) There is a downturn at lower temperatures in the $\sigma(T)$ curves of $(Tb,Dy)(Fe,Al)_2$ for various fields.

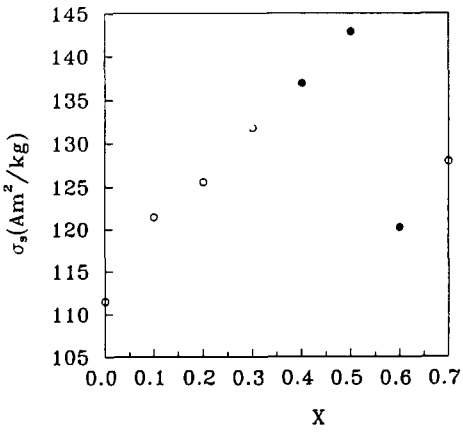

Fig. 3. The concentration dependence of the saturation magnetization σ_s in $Tb_{0.27}Dy_{0.73}(Fe_{1-x}Al_x)_2$ compounds: ○, cubic $MgCu_2$ structure; ●, hexagonal $MgZn_2$ structure.

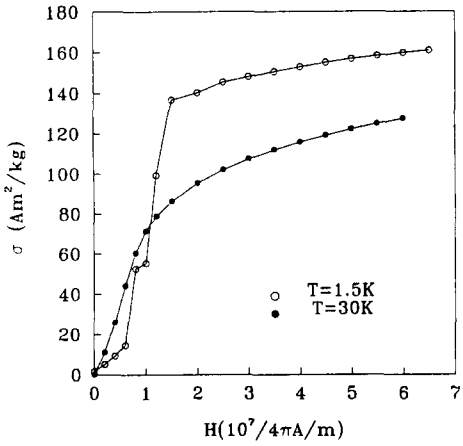

Fig. 4. $M(H)$ curves of $Tb_{0.27}Dy_{0.73}(Fe_{1-x}Al_x)_2$ at 1.5 and 30 K.

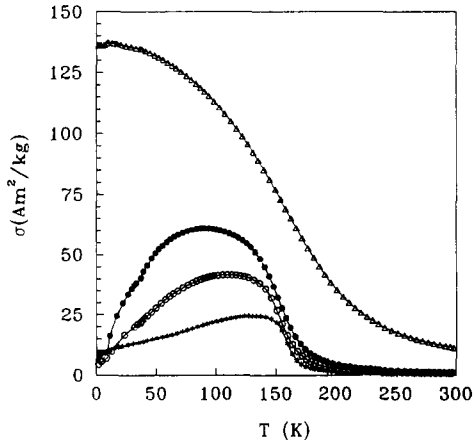

Fig. 5. $M(T)$ curves of $Tb_{0.27}Dy_{0.73}(Fe_{1-x}Al_x)_2$ in various fields after cooling in a zero field: *, $H=0.1\times10^7/4\pi$ A m^{-1}; ○, $H=0.25\times10^7/4\pi$ A m^{-1}; ●, $H=0.4\times10^7/4\pi$ A m^{-1}; △, $H=4.0\times10^7/4\pi$ A m^{-1}.

The results shown in Fig. 5 reflect the temperature dependence of the pinning field.

(3) There are remarkably high values of the coercivity H_c at low temperature: for instance, for $x=0.5$, we found $H_c=240$ kA m^{-1} at 30 K, while at 295 K we found 8 kA m^{-1} only. It is supposed that the fluctuations in the exchange as well as in the local crystal field are responsible for these pinning effects, which lead to the creation of energy barriers which affect the magnetization processes in low magnetic fields and at low field strength. With increasing temperature and field strength, the domain wall propagation should proceed more easily.

The magnetostriction of the $Tb_{0.27}Dy_{0.73}(Fe_{1-x}Al_x)_2$ compounds has been measured in directions parallel ($\lambda_{\parallel}$) and perpendicular ($\lambda_{\perp}$) to the applied magnetic field up to 1.6×10^3 kA m^{-1}. The magnetostriction of $|\lambda_{\parallel}-\lambda_{\perp}|$, measured at room temperature in a field of 1.6×10^3 kA m^{-1} is shown as a function of Al content x in Fig. 6. It was found that $|\lambda_{\parallel}-\lambda_{\perp}|$ drops from

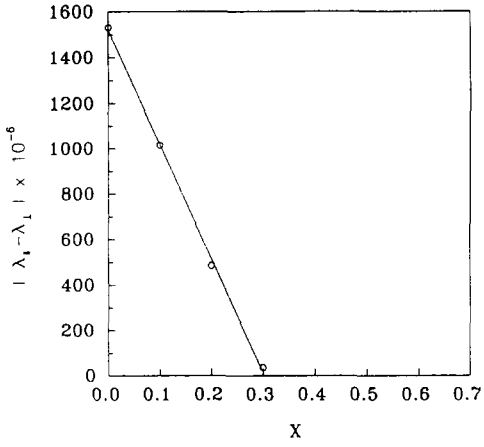

Fig. 6. The room temperature magnetostriction of $|\lambda_{\parallel}-\lambda_{\perp}|$ measured in a field of $2\times10^7/4\pi$ A m^{-1} as a function of the Al content x in $Tb_{0.27}Dy_{0.73}(Fe_{1-x}Al_x)_2$ compounds.

1530×10^{-6} for $x=0$ to 36×10^{-6} for $x=0.3$. It is well known that the very large magnetostriction in RFe_2 compounds is due to the interactions of the anisotropic cloud of the 4f electrons with the crystal field and Fe sublattice. According to this single-ion model, the magnetostriction varies with temperature as $\sigma_R{}^3(T)$. Assuming the R sublattice moments σ_R decrease with decreasing Curie temperature, the rapid decrease in T_C with increasing x value results in a particularly strong decrease in the room temperature value of $\sigma_R(T)$, which in turn leads to a marked reduction in $|\lambda_{\parallel}-\lambda_{\perp}|$ with Al content.

In conclusion, the appearance of structural transitions, a complex concentration dependence of σ_s and an increase in the magnetic hardness in the (Tb,Dy)-$(Fe,Al)_2$ system was attributed to two main factors: (1) high anisotropy of the rare earth sublattice; (2) local disorder of the (Fe,Al) sublattice [3, 8], leading to lattice distortions, fluctuating orientations of the local axes of easy magnetization and pinning effects of narrow domain walls at structural defects or clusters inhomogeneities. Small amounts of substituted Al in the (Tb, Dy)Fe_2 compounds cause strong reductions in T_C and consequently result in rapidly decreasing values of $|\lambda_{\parallel}-\lambda_{\perp}|$ with Al content at room temperature.

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