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Spin configuration and magnetostriction in $Nd_{0.9}Tb_{0.1}(Fe_{1-x}Co_x)_{1.9}$ $(0 \le x \le 0.55)$ alloys

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ABSTRACT

Polycrystalline magnetostrictive alloys $Nd_{0.9}Tb_{0.1}(Fe_{1-x}Co_x)_{1.9}$ $(0 \le x \le 0.55)$ were synthesized by high-pressure annealing. Measurements of crystal structure, Curie temperature, magnetization, spin-reorientation temperature and magnetostriction were made on the alloys. X-ray diffraction results show that all the samples exhibit cubic Laves phase with $MgCu_2$-type structure. The variations of Curie temperature and saturation magnetization are interpretable due to 3d band filling. The spin configuration diagram of $Nd_{0.9}Tb_{0.1}(Fe_{1-x}Co_x)_{1.9}$ was constructed. The spin-reorientation temperatures of the samples first increase and then decrease with the substitution of Co for Fe. The anisotropy minimum and low-filed large magnetostriction is realized around $x = 0.05$.

1. Introduction

The C15 cubic Laves phase compounds $REFe_2$ (RE = rare earth) are well known for their giant magnetostriction at room temperature [1]. But they are unsuitable as practical materials because of their high magnetic anisotropy. An excellent magnetostrictive material, showing an effective efficiency in converting electrical energy to mechanical one, should have the characters of large low-field magnetostriction and low magnetic anisotropy. Thus, much effort has been paid to $(RE_1, RE_2)Fe_2$ pseudobinary anisotropy compensation system, such as $Tb_xDy_{1-x}Fe_2$ and $Tb_xHo_{1-x}Fe_2$ [2–7]. However, little attention be paid to light rare-earth Nd-based magnetostrictive alloys because $NdFe_2$ cubic Laves phase cannot be obtained under normal atmosphere [8]. Recently, we reported that $Nd_xTb_{1-x}Fe_{1.9}$ $(0 \le x \le 1)$ cubic Laves alloys could be synthesized by a method of high-pressure annealing and the anisotropy compensation is realized in $Nd_{0.9}Tb_{0.1}Fe_{1.9}$ [9]. As we know, the magnetostrictive properties of the alloys are very sensitive to the compositions or the spin-reorientation temperature (T_{sr}). In general, the T_{sr} of the alloy needs to be approach to the room temperature as close as possible to get the low anisotropy and large low-field magnetostriction. Many works were focused on regulating the proportion of the rare earth in $(RE_1, RE_2)Fe_2$ alloys to make the T_{sr} to be near the room temperature [10,11]. Meanwhile, previous investigations have shown that the T_{sr}, the anisotropy and magnetostriction of the $REFe_2$ could be significantly changed with the substitution of Fe by other transition metals [12–16]. In this paper, the composition of the compound is chosen at $Nd_{0.9}Tb_{0.1}Fe_{1.9}$, which is the anisotropy compensation point in $Nd_xTb_{1-x}Fe_{1.9}$ system. The aims of the present work is to investigate the effects of Co substitution for Fe on the magnetic properties of the pesudobinary $Nd_{0.9}Tb_{0.1}Fe_{1.9}$ alloys.

2. Experimental procedures

Ingots with $Nd_{0.9}Tb_{0.1}(Fe_{1-x}Co_x)_{1.9}$ $(0 \le x \le 0.55)$ stoichiometry were prepared by melting the constituent metals in a magneto-controlled arc furnace in a high-purity argon atmosphere. The as-cast ingots were then annealed at 900 °C under a high pressure of 6 GPa for 30 min. After that, all samples were annealed at 400 °C for 48 h in vacuum quartz capsules. Conventional X-ray diffraction (XRD) analysis was carried out using Cu Kα radiation with a Rigaku D/Max-gA diffractometer. The Curie temperature was detected by a thermal gravitation analyzer with a vertical gradient magnetic field under the samples. Magnetization curves at room temperature were measured by a superconducting quantum interference device (SQUID) magnetometer at fields up to 53 kOe. The temperature dependence of the ac initial susceptibility was recorded by the SQUID magnetometer to detect the T_{sr}. The linear magnetostriction was measured using standard strain-gauge technique in directions parallel ($\lambda_{//}$) or perpendicular ($\lambda_{\perp}$) to applied magnetic fields at room temperature.

3. Results and discussion

The XRD patterns for $Nd_{0.9}Tb_{0.1}(Fe_{1-x}Co_x)_{1.9}$ with different Co contents are presented in Fig. 1. It is found that all the alloys mainly

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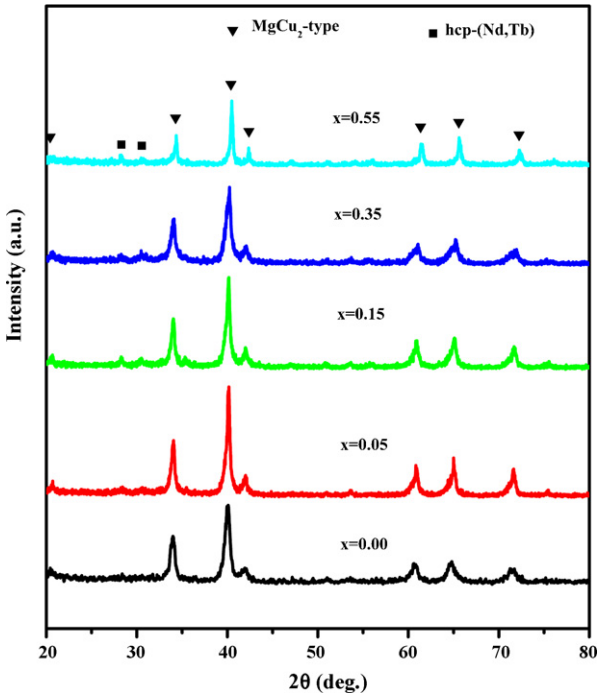

Fig. 1. XRD patterns of $Nd_{0.9}Tb_{0.1}(Fe_{1-x}Co_x)_{1.9}$ alloys.

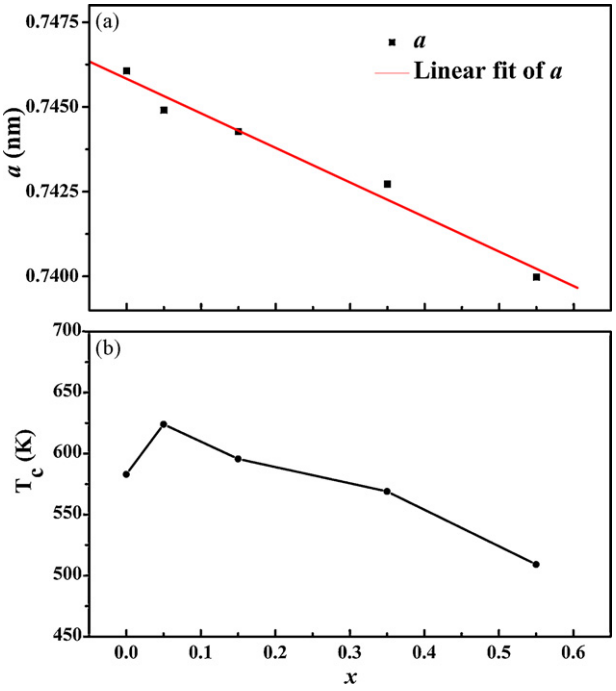

Fig. 2. (a) The lattice parameter a and (b) the Curie temperatures T_c of $Nd_{0.9}Tb_{0.1}(Fe_{1-x}Co_x)_{1.9}$ with different concentration of Co.

consist of C15 cubic Laves phase with $MgCu_2$-type structure, coexisting with minor impurities, i.e. hcp-(Nd, Tb) phases. No $REFe_3$ phases or RE_2Fe_{17} phases can be found in the alloys. However, the cubic Laves phase could not be obtained by a traditional high-temperature annealing method with Co content less than 60% [17]. It is generally believed that the atom radius plays an important role in the formation of $REFe_2$ C15 cubic Laves alloys. The ideal ratio of RE and Fe is 1.225, however, the ratio of Nd and Fe is larger than the ideal one. Therefore, in our present work, the formation of the cubic Laves phase should be ascribed to the effect of the high pressure. In consistence with the previous report, high-pressure synthesis is an effective method to prepare high-Nd content compounds [8,9].

The lattice parameters a of the Laves phase in $Nd_{0.9}Tb_{0.1}(Fe_{1-x}Co_x)_{1.9}$ compounds derived from the XRD data are shown in Fig. 2(a). The lattice parameter decreases from 0.746 nm for $Nd_{0.9}Tb_{0.1}Fe_{1.9}$ to 0.739 nm for $Nd_{0.9}Tb_{0.1}(Fe_{0.45}Co_{0.55})_{1.9}$, which suggests that Co with smaller radius takes up the site of Fe in cubic lattice.

The Curie temperature T_c as a function of Co contents is shown in Fig. 2(b). The Curie temperature increases with increasing Co content and reach a maximum value at $x = 0.05$. After that, further increasing Co decrease T_c. According to the Stoner–Wohlfarth description, the Curie temperature T_c is shown below [18]:

$$T_c^2 = T_F^2(\bar{I} - 1)$$

where T_F represents the degeneracy temperature, and $\bar{I} = IN(E_F)$, I is the effective Coulomb repulsion between the 3d electrons, and $N(E_F)$ is the 3d-electron density of states at the Fermi level, respectively. It can be assumed that the spin-up and spin-down bands of the 3d electrons are unsaturated in $Nd_{0.9}Tb_{0.1}Fe_{1.9}$. The Fermi level of the spin-down band resides in a region of a minimum in the density of states [19]. With a small amount of Co substitution, the electron concentration will increase and the spin-up band will move up and, as a result, the E_F increased. In this situation, an increase in E_F will lead to an increase of T_F^2 or T_c. With further increasing of Co content, it will lead to a decrease of T_F^2 and the decrease of T_c. The similar situation could also be found in $Sm_{0.88}Dy_{0.12}(Fe_{1-x}Co_x)_2$ [14] and $Y(Fe_{1-x}Co_x)_2$ [20] alloys.

Magnetic field dependence of magnetization at room temperature for the $Nd_{0.9}Tb_{0.1}(Fe_{1-x}Co_x)_{1.9}$ compounds is shown in Fig. 3. The magnetizations of the alloys are close to saturation in the applied field of 53 kOe. The saturation magnetization (M_s) derived from M–H curves is shown in the inset of Fig. 3. The composition dependence of M_s exhibits a similar trend with the variations of T_c:M_s increases to a maximum around $x = 0.05$ and decreases with further increasing Co content. This can be understood on the basis of the rigid-band model [20]. As mentioned above, the spin-up and spin-down bands of the 3d electrons are unsaturated in $Nd_{0.9}Tb_{0.1}Fe_{1.9}$. The electrons are firstly added to the spin-up band, and, the net 3d magnetic moment will increase. With further increasing the content of Co, the spin-up band

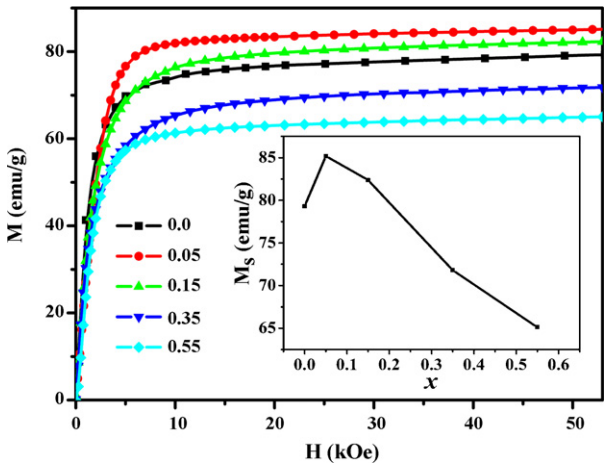

Fig. 3. Magnetization curves for $Nd_{0.9}Tb_{0.1}(Fe_{1-x}Co_x)_{1.9}$. The inset shows the saturation magnetization (M_s) with different concentration of Co.