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Magnetic properties of $Nd(Fe,Ti)_{12}$ and $Nd(Fe,Ti)_{12}N_x$ films of perpendicular texture

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$Nd(Fe,Ti)_{12}$ and $Nd(Fe,Ti)_{12}N_x$ films textured with c axis perpendicular to the film plane were synthesized by sputtering. For the $Nd(Fe,Ti)_{12}$ films a spin reorientation took place at about 170 K. For the $Nd(Fe,Ti)_{12}N_x$ films, the anisotropy field is about 85 kOe at room temperature and 130 kOe at 5 K, corresponding to a coercivity of 2.7 and 12 kOe, respectively. Initial magnetization curves show a behavior characteristic of nucleation type of magnetization reversal mechanism. Attempts were made to fit the experimental data with Kronmüller's magnetization reversal model. The results suggest that nucleation with extended sites is responsible for the reversal.

INTRODUCTION

Higano *et al.* were probably the first to report the astonishing effect of nitriding on some rare earth-transition metal(RE-TM) compounds in 1987. They concluded that[1]: "Nitriding resulted in a lattice expansion, increase in the saturation magnetization and change in Curie temperature." Coey and co-workers reported the first detailed study[2] of various aspects of these compounds, and their results have stimulated much of the succeeding work.[3]

The synthesis of $SmFe_{12}$ films by sputtering has been reported.[4,5] But $SmFe_{12}N_x$ or $Sm(Fe,Ti)_{12}N_x$ is not a good candidate for permanent magnets because its planar magnetic anisotropy leads to a low coercivity (H_c). $Nd(Fe,Ti)_{12}N_x$ is a promising candidate for permanent-magnet applications because of its high uniaxial anisotropy field (H_k), high Curie temperature and high saturation magnetization (M_s).[6]

Previously we reported the preliminary results on the $Nd(Fe,Ti)_{12}$ and $Nd(Fe,Ti)_{12}N_x$ films.[7] A report on similar films has been given also recently by Navarathna *et al.*[8] In this paper we will discuss the magnetic properties of these sputtered films. Low temperature measurements on the $Nd(Fe,Ti)_{12}N_x$ films will be emphasized. The magnetization reversal mechanism will also be discussed with available models.

EXPERIMENTAL METHODS

$Nd(Fe,Ti)_{12}$ films were prepared in a dc magnetron sputtering system on heated Ta substrates.[5] The base pressure was better than 5×10^{-7} Torr. After the deposition, nitrogen gas was introduced into the chamber with a pressure up to about 200 Torr at various sample temperatures for various lengths of time; for example, a 400 °C anneal for one hour was typically used. All films reported on in this paper had a nominal thickness of about 1 μm.

RESULTS AND DISCUSSION

Figure 1 shows x-ray diffraction (XRD) patterns for films deposited at different substrate temperatures under an argon pressure of 1.5 mTorr. The XRD patterns are fitted nicely to the (002), (202), (222), and (004) lines of the tetragonal $ThMn_{12}$ structure. The films have the c axis preferentially aligned perpendicularly to the film plane. For a substrate temperature higher than 430 °C, the small portion of α-Fe-like phase starts to increase and the (002) texture gets worse. For a substrate temperature much lower than 340 °C the films have only XRD peaks of the α-Fe-like phase. For all the measurement shown below, the films made at a substrate temperature of 390 °C were used. The XRD pattern for the nitride film was given in Ref. 7, which is similar as the

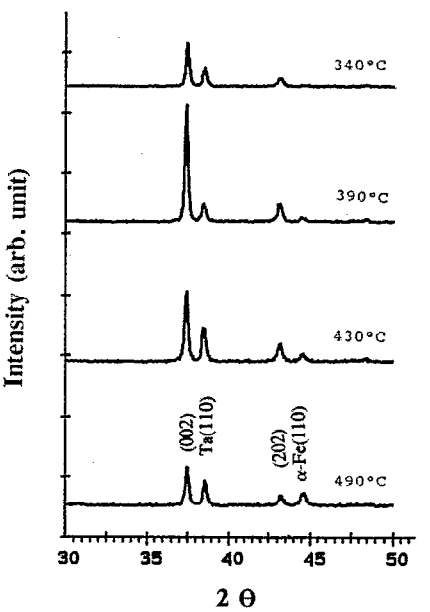

FIG. 1. XRD patterns of $Nd(Fe,Ti)_{12}$ films prepared on Ta under various substrate temperatures and at a sputtering gas pressure of 1.5 mTorr.

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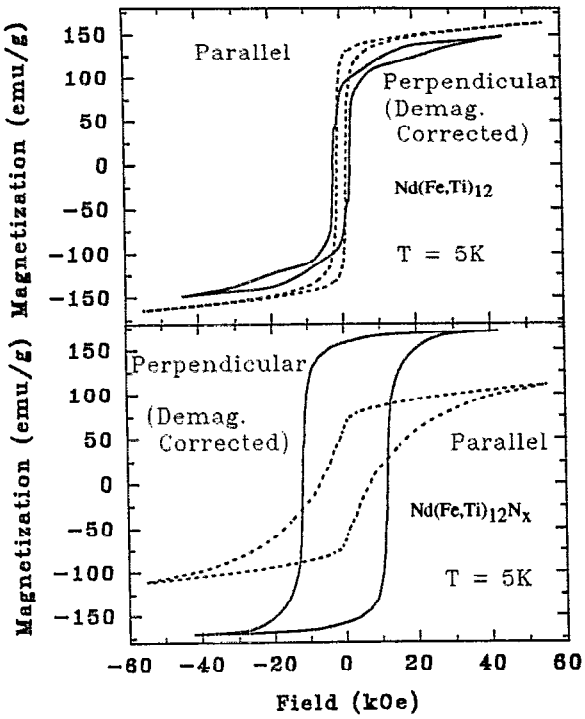

FIG. 2. Hysteresis loops of a $Nd(Fe,Ti)_{12}$ film and a $Nd(Fe,Ti)_{12}N_x$ film of about 1 μm thick measured at 5 K with a high applied field of 55 kOe.

390 °C pattern for the film before nitriding as shown in Fig. 1 with only positions shifted to lower angles.

When films are annealed *in-situ* at a temperature of 400 °C for about one hour in a nitrogen atmosphere, a unit-cell-volume expansion of 3.6% is observed. Based on the reported data[9] we estimated the nitrogen content to be about $x=0.9$. This accompanies a dramatic change in magnetic properties.

Figure 2 shows typical hysteresis loops for a $Nd(Fe,Ti)_{12}$ film and a $Nd(Fe,Ti)_{12}N_x$ film of about 1 μm thick measured at a low temperature of 5 K. For the perpendicular loops the applied field has been corrected for demagnetization. It is noted that for the $Nd(Fe,Ti)_{12}$ film the perpendicular magnetization is below the parallel magnetization, which is different from the case of room temperature measurement.[7]

Figure 3 shows the temperature dependence of the magnetization for films before and after nitriding. The procedure for this measurement was first to lower the temperature to 5 K and apply the maximum field of 55 kOe in the film plane, then decrease the field to 200 Oe, which is much lower than the coercivity, 2.7 kOe, and then measure the magnetization while increasing the temperature. A spin reorientation is observed at around 170 °C for the $Nd(Fe,Ti)_{12}$ film. For the $Nd(Fe,Ti)_{12}N_x$ film no such phenomenon is observed. This means that the spin reorientation is suppressed by nitriding.

Figure 4 shows the initial magnetization curves of a nitride film measured at 5 K from both a thermally demagnetized and a field-demagnetized state. The hysteresis loop in the first quadrant is also shown. The magnetization for the thermally demagnetized film rises rapidly to near saturation at a field well below the coercivity. For the field-

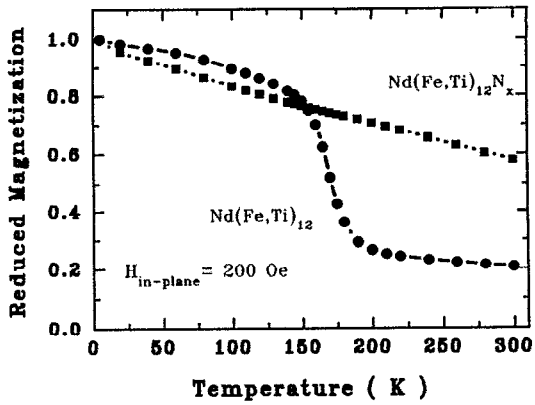

FIG. 3. Temperature dependence of magnetization for a $Nd(Fe,Ti)_{12}$ film and a $Nd(Fe,Ti)_{12}N_x$ film under a low field of 200 Oe.

demagnetized film the initial curve is much lower. These features are similar to those observed for NdFeB sintered magnets, which are believed to be strong indications of a nucleation type of magnetization-reversal mechanism.[3] Hysteresis loops were measured at different temperatures with a SQUID magnetometer. The temperature dependencies of M_s, H_k, H_c determined from the loops for a nitride film are plotted in Fig. 5. H_c decreases from 12 kOe at 5 K to 2 kOe at 350 K. At room temperature H_c is about 2.7 kOe. M_s decreases about 10% when the temperature increases from 5 K to room temperature. Kronmüller and co-workers[10] have derived a simplified formula for the coercivities due both to nucleation and pinning mechanisms by assuming that there are spatial fluctuations of the first-order anisotropy constant K_1 and the exchange constant A:

$$H_c(T)=\alpha(T)\,\frac{2K_1(T)}{M_s(T)}-N_{\text{eff}}M_s(T), \tag{1}$$

where for pinning

$$\alpha_K^{\text{pin}}(T)=a\,\frac{r_0}{\delta_B}\quad\text{when}\quad r_0\ll\delta_B, \tag{2}$$

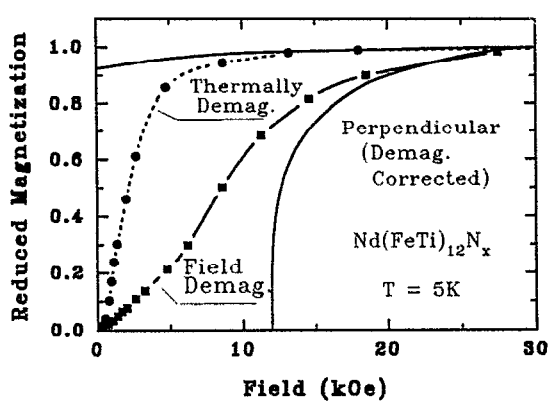

FIG. 4. Initial magnetization curve measured from a thermally demagnetized and field demagnetized state of a $Nd(Fe,Ti)_{12}N_x$ film at a low temperature of 5 K.

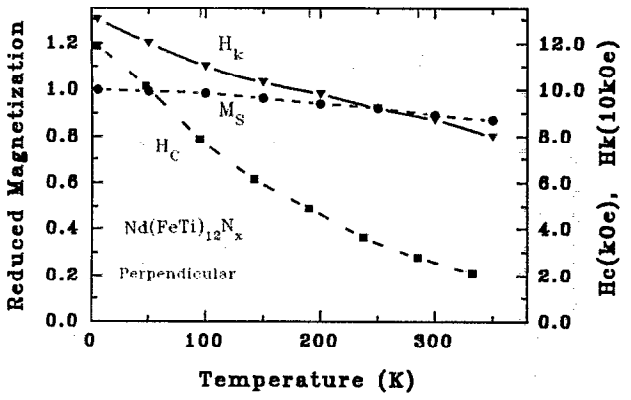

FIG. 5. Temperature dependence of the parameters M_s, H_k, H_c extrapolated from hysteresis loops for a $Nd(Fe,Ti)_{12}N_x$ film.

$$\alpha_K^{\text{pin}}(T) = b\,\frac{\delta_B}{r_0} \quad \text{when} \quad r_0 \gg \delta_B, \tag{3}$$

and where

$$\delta_B = \pi\sqrt{\frac{A}{K_1}} \tag{4}$$

is the domain-wall width for a uniaxial crystal and r_0 is the pinning-center size.

Equation (1) also applies to reversal by nucleation according to different sizes of the nucleation site (r_0):

$$\alpha_K^{\text{nuc}} = 1 - \frac{r_0^2}{\delta_B}, \quad \text{when} \quad r_0 \ll \delta_B, \tag{5}$$

$$\alpha_K^{\text{nuc}} = \frac{\delta_B}{\pi r_0}, \quad \text{when} \quad 2\pi r_0 \approx \delta_B, \tag{6}$$

$$\alpha_K^{\text{nuc}} = 1 - \frac{\Delta K}{K_1}, \quad \text{when} \quad 2\pi r_0 \gg \delta_B. \tag{7}$$

We assume that the exchange constant A for the main region[11] is 10^{-6} erg/cm and the domain-wall thickness δ_B is given by Eq. (4). After fitting the experimental results to the cases presented in Eqs. (2) to (7), the main results of this analysis are summarized as follows: (a) Assuming reversal by pinning with small pinning centers, the fitting according to Eq. (2) gives an r_0 value of 0.03 Å, which is unreasonably small though the H_c/M_s vs $2K/M_s^2/\delta_B$ curve shows a near linear relationship; (b) assuming reversal by pinning with large pinning centers according to Eq. (3), the experimental results are shown in Fig. 6(a). H_c/M_s does not vary linearly with $2K/M_s^2\delta_B$, so this case can be excluded; (c) for reversal by nucleation with small nucleation sites, the experimental results are fitted according to Eq. (5). The value of r_0 obtained is 150 Å, which is inconsistent with the small site assumption; (d) for reversal by nucleation with medium size nucleation sites, no linear relationship is expected since Eq. (6) and Eq. (3) have similar δ_B dependence; (e) for reversal by nucleation with extended nucleation sites, Fig. 6(b) gives

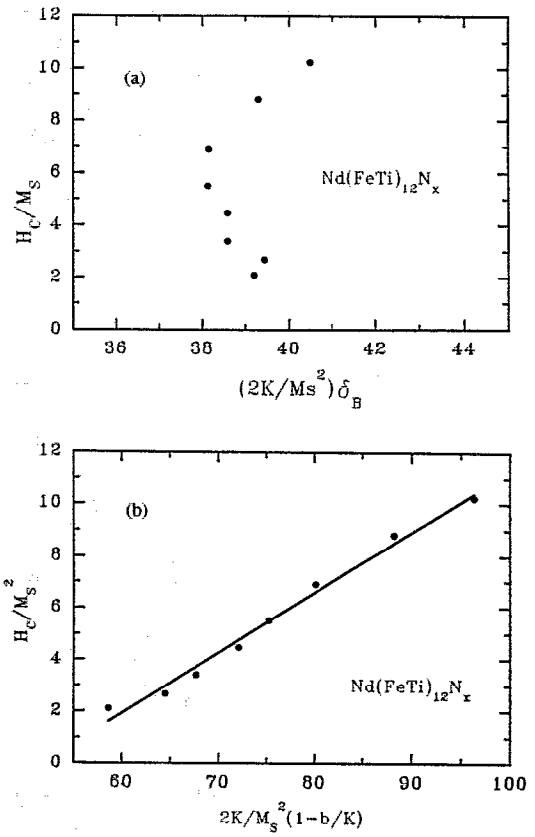

FIG. 6. (a) Experimental results for pinning with large pinning sites; (b) Experimental results and fitting results for nucleation with large sites for a $Nd(Fe,Ti)_{12}N_x$ film.

the experimental results and fitting results according to Eq. (7). There is a reasonably good linear relationship between H_c/M_s and $2K/M_s^2(1-b/K)$. The prefactor in Eq. (1) is about 0.23 and N_{eff} is about 3.8.

ACKNOWLEDGMENT

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