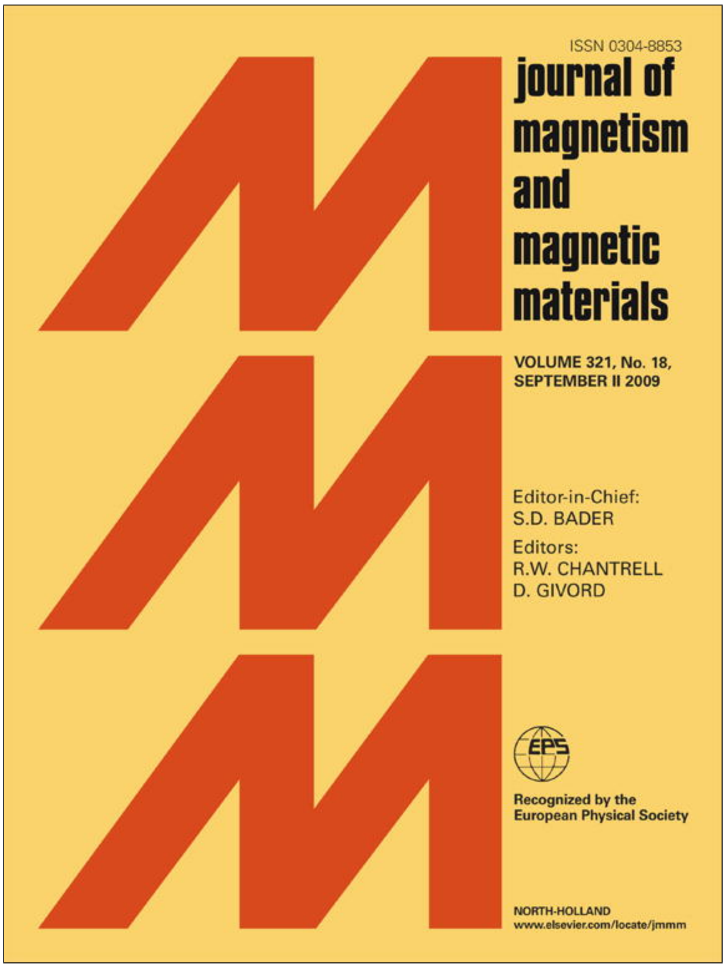
ISSN 0304-8853
journal of magnetism and magnetic materials
VOLUME 321, No. 18, SEPTEMBER II 2009
Editor-in-Chief: S.D. BADER
Editors: R.W. CHANTRELL D. GIVORD
EPS
Recognized by the European Physical Society
NORTH-HOLLAND
www.elsevier.com/locate/jmmm

Journal of Magnetism and Magnetic Materials 321 (2009) 2792–2794

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Growth temperature dependence of the hysteretic behavior of $Ni_{0.5}Zn_{0.5}Fe_2O_4$ thin films

J. Prado [a], M.E. Gómez [a], P. Prieto [a,*], A. Mendoza [b]

[a] Thin Film Group, Department of Physics, Center of Excellence for Novel Materials—CENM, Universidad del Valle, A.A. 25360 Cali, Colombia
[b] Magnetic Materials and Nanostructures Group, Department of Physics, Universidad Nacional de Colombia, carrera 45 No 26-85, Bogotá, Colombia

ARTICLE INFO

Article history:
Received 11 December 2008
Received in revised form
30 March 2009
Available online 17 April 2009

PACS:
75.50.Gg
75.60.−d
75.60.Ch

Keywords:
Ferimagnetics
Domain effect
Magnetization curve
Hysteresis
Domain wall

ABSTRACT

Herein, a discussion of the effect of deposition temperature on the magnetic behavior of $Ni_{0.5}Zn_{0.5}Fe_2O_4$ thin films. The thin films were grown by r.f. sputtering technique on (1 0 0) MgO single-crystal substrates at deposition temperatures ranging between 400 and 800 °C. The grain boundary microstructure was analyzed via atomic force microscopy (AFM). AFM images show that grain size (ϕ~70–112 nm) increases with increasing deposition temperature, according to a diffusion growth model. From magneto-optical Kerr effect (MOKE) measurements at room temperature, coercive fields, H_c, between 37and 131 Oe were measured. The coercive field, H_c, as a function of grain size, reaches a maximum value of 131 Oe for ϕ ~93 nm, while the relative saturation magnetization exhibits a minimum value at this grain size. The behaviors observed were interpreted as the existence of a critical size for the transition from single- to multi-domain regime. The saturation magnetization (21 emu/g$<M_s<$60 emu/g) was employed to quantify the critical magnetic intergranular correlation length ($L_c\approx$166 nm), where a single-grain to coupled-grain behavior transition occurs. Experimental hysteresis loops were fitted by the Jiles–Atherton model (JAM). The value of the k-parameter of the JAM fitted by means of this model (k/μ_o~50 A m^2) was correlated to the domain size from the behavior of k, we observed a maximum in the density of defects for the sample with ϕ~93 nm.

1. Introduction

Growth of magnetically soft ferrite thin films is motivated by their potential applications [1,2]; particularly, the study of NiZn ferrites with spinel structure is of current theoretical and technological interest. The magnetic properties of soft magnetic materials can be understood by adequate theoretical models [3,4]. Given their low conductivity, NiZn ferrites are used for high-frequency applications. For optimum performance at these high frequencies, their magnetic hysteresis should also be as narrow as possible; said property is related to domain wall motion, and several studies have been conducted on the domain structure of poly-crystalline NiZn ferrites, along with their relationship to average grain size [5,6]. Generally, the magnetic quality of ferrite films is also mainly dependent on the extrinsic properties, i.e., morphological properties like single- and multi-domain structures, grain size distribution, grain boundary, and defects. The properties mentioned have direct influence on some parameters of the hysteresis behavior; especially, saturation magnetization and coercive field are highly sensitive to said properties. The existence of multi-domains can be interpreted as coupled grains. It has been shown that this coupling appears at a magnetic-correlation length that depends on the grain sizes [3]. In this work, we estimated the value of the correlation length for the transition from single-grain to coupled-grain behavior. Extrinsic properties and magnetic properties were correlated in this work through the Jiles–Atherton model (JAM) [4,7]; mean k-pinning parameter, which depends on the morphological properties mentioned above.

2. Experimental details

$Ni_{0.5}Zn_{0.5}Fe_2O_4$ films were produced via a high oxygen pressure r.f. sputtering process and deposited onto 10 mm × 10 mm (1 0 0)-MgO single-crystal substrates. A NiZn ferrite target with a $Ni_{0.5}Zn_{0.5}Fe_2O_4$ composition was prepared by solid-state reaction of NiO, ZnO, and Fe_2O_3 powders by using a sintering temperature of 1200 °C. Oxygen pressure during deposition was 2.1×10^{-1} mbar. Deposition temperatures were 600, 650, 700, 750, and 800 °C. During the whole deposition process, temperature remained stable to within less than 1 °C. Deposition rate was ~1.3 nm/min. The thickness of the samples analyzed was around 97 ± 3 nm. X-ray diffraction (XRD) was conducted with a Philips

---

[*] Corresponding author. Tel.: +57 2 3394610; fax: +57 2 3393237.
E-mail address: pprieto@calima.univalle.edu.co (P. Prieto).

0304-8853/$ - see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.jmmm.2009.04.015

X-ray diffraction unit (Model PW1710) using Cu K_α ($\lambda = 1.5405$) radiation. The atomic force microscopy (AFM) was used for surface morphological studies. The hysteresis curve was measured at room temperature by using a vibrating spectro-magnetometer (VSM) from Quantum Design^{TM} Physical Property Measuring System (PPMS). The *k*-parameter was determined by modeling the magnetization curves by using the JAM [7] model. The hysteretic curves were obtained by using a magneto-optical Kerr effect (MOKE) System. A diode laser with a 532 nm wavelength was used as the light source for MOKE measurements. The longitudinal Kerr signal was obtained by monitoring the changes of the reflective intensity, proportional to the magnetization of the sample. The signals produced at the detector were relayed to the computer through optical fiber. All MOKE measurements were performed at room temperature.

3. Results and discussion

The crystal structure of the samples was analyzed by using XRD diffraction. The diffraction peaks indicate a highly textured film, corresponding to single-phase cubic spinel oxides [8], where the lattice parameter of 0.838 ± 0.001 nm was obtained through a refinement method. Grain size, as a function of deposition temperature, is shown in Fig. 1. The inset shows AFM images of NiZn ferrite thin film surface (1 μm × 1 μm) grown at: (a) 600 °C, (b) 650 °C, (c) 750 °C, and (d) 800 °C. It can be deduced that grain size ($\phi \sim 70$–112 nm) increases with increasing deposition temperature to a saturation value. Average ϕ values and standard geometric deviation, as extracted from the inset figure for different samples, are listed in Table 1. The change in grain size is associated to growth processes. The nucleation process followed by a diffusion mechanism induces grain size increase.

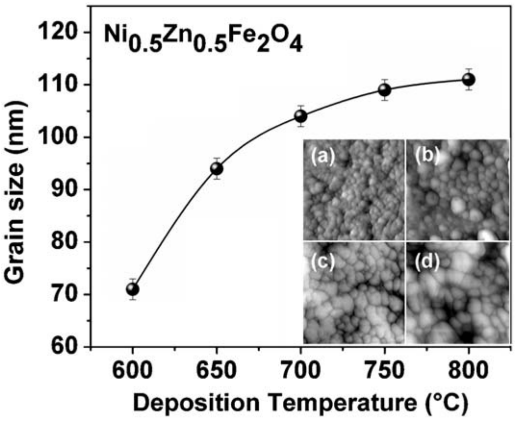

Fig. 1. Grain size as a function of deposition temperature. Solid lines are a guide to the eye. Inset shows AFM images of a NiZn ferrite thin film surface (1 μm × 1 μm) grown at: (a) 600 °C, (b) 650 °C, (c) 750 °C, and (d) 800 °C.

Table 1
Grain size ϕ (nm); H_c values; *k*-pinning parameter; and saturation magnetization obtained for samples grown at different temperatures.

Growth temperature (°C)	ϕ (nm)	H_c (Oe) ± 10%	k/μ_0 (Am^2)	M_s (emu/g)
600	71 ± 3	95	62 ± 1	15
650	93 ± 3	131	98 ± 2	11
700	104 ± 3	114	73 ± 2	12
750	109 ± 4	73	46 ± 1	47
800	111 ± 4	37	44 ± 2	70

The VSM *M*(*H*) curve at room temperature is shown in Fig. 2 for a sample grown at a deposition temperature of 750 °C. All samples were measured with both techniques, VSM and MOKE. From these measurements, the coercive field, H_c, and saturation magnetization, M_s, were extracted, with values measured between 37 and 131 Oe, 21 and 60 emu/g for H_c and M_s, respectively (Fig. 3). Above $\phi = 70$ nm, the increase in saturation magnetization as a function of the grain size – as expected – is observed (Table 1). The increase in saturation magnetization as a function of the growth temperature (above 650 °C) could be explained on the basis of grain growth. As grain size increases, magnetization also increases. The lower value of magnetization as compared to the bulk (~600 emu/g) could be due to a large grain boundary volume present in nano-crystalline thin films. The saturation magnetization at the growth temperature of 600 °C is slightly higher than at a growth temperature of 650 °C, possibly due to the increase in the intergranular magnetic correlations length.

The coercive field, H_c, reaches a maximum value of 131 Oe for the sample with $\phi = 93 \pm 3$ nm (e.g., for the sample grown at 650 °C); while the relative saturation magnetization exhibits a minimum value for this grain size. Such behavior can be interpreted as the existence of a critical grain size for the transition from single- to multi-domain regime, as claimed by Desai [5]. This behavior has been discussed by Löfler et al. [3], where they introduce the concept of a transition from single-grain to coupled-grain through a magnetic correlation length.

The maximum in the coercivity, H_c, has been related to the correlation length, *L*, indicating a coupling between adjacent grains for a critical grain size ϕ_c [3], where the change in magnetic properties and the energy wall domains start to play an important

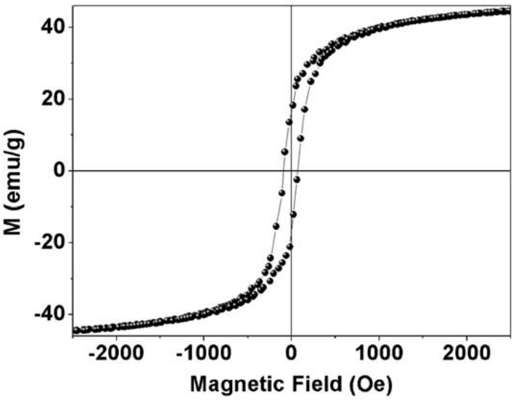

Fig. 2. Hysteresis loop for a $Ni_{0.5}Zn_{0.5}Fe_2O_4$ thin film grown at 750 °C.

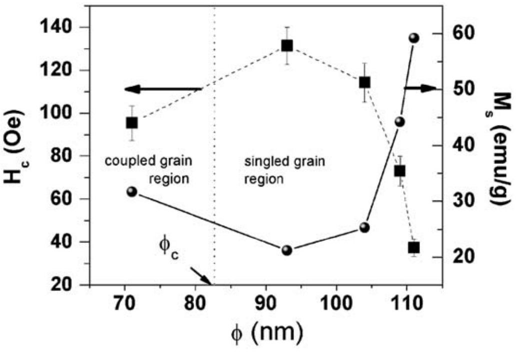

Fig. 3. Grain size dependence of the coercivity (dashed line) and relative saturation magnetization (solid line) of $Ni_{0.5}Zn_{0.5}Fe_2O_4$ thin films. Dotted line represents the theoretical boundary between the coupled-grain and the single-grain regions; critical grain size is also indicated.

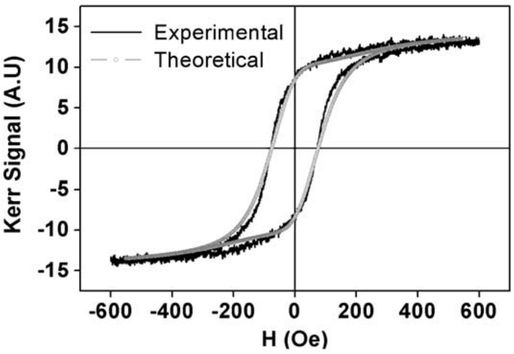

Fig. 4. Theoretical fitting of the MOKE experimental hysteresis loop (800 °C deposition temperature) by means of the Jiles–Atherton Model.

role in the magnetic behavior. In the limit where $L > \phi_c$, it is possible to consider

$$L \approx 2\phi \tag{1}$$

while, for $L < \phi$ a minimization with respect to L results in

$$L = (16/9)\delta_{eff}^4/\phi^3 \tag{2}$$

where δ_{eff} is the domain wall width, which depends on the exchange constant. To estimate a first approximation of the L value, we calculated the wall width, δ_{eff}, from the Bloch model [9] by using: the anisotropy constant, $K_a = 0.3 \times 10^5\,\mathrm{erg/cm^3}$; the exchange constant, $C = 2.5 \times 10^{-6}\,\mathrm{erg/cm}$; the unit cell parameter, $a = 8.4 \times 10^{-8}\,\mathrm{cm}$; the film thickness, $d \approx 100\,\mathrm{nm}$; and saturation magnetization M_s in the 21–60 emu/g range. The δ_{eff} obtained was ~83 nm. At the transition point from single-grain to coupled-grain behavior, the δ_{eff} corresponds approximately to the critical grain size, ϕ_c, value: $\delta_{eff} \approx \phi_c$. In the vicinity of this value, a change in the magnetic properties is also observed as a result of the intergranular magnetic correlations. The value of $\phi_c \sim 83\,\mathrm{nm}$ corresponds quite well to the experimental value of 93 nm, roughly observed in Fig. 3. Through Eq. (1), the estimated values for the correlation length were $L(\phi = 93\,\mathrm{nm}) \approx 186\,\mathrm{nm}$, and $L_c(\phi_c = 83\,\mathrm{nm}) \approx 166\,\mathrm{nm}$. The latter corresponds to the minimum value expected in an L vs. ϕ curve. The limit between the coupled-grain and the single-grain regions is indicated by the dashed line in Fig. 3.

We used the Jiles–Atherton Model [7,10] to fit the experimental hysteresis loops obtained by MOKE. The Jiles–Atherton Model is based on domain wall motion of an irreversible component due to wall displacement and a reversible component due to domain wall bending. The k-pinning parameter of the model, $k = \mathrm{d}E_{lost}/\mathrm{d}M$, is the change in the energy that appears due to domain wall motion when magnetization reversal proceeds in the magnetization $M(H)$ measurements. The k-parameter is summarized in Table 1. This parameter represents energy loss in magnetization processes and it is usually attributed to defects, stress, distortion in the cell, and grain boundaries [4]. Fig. 4 shows a Kerr signal curve for the sample grown at 800 °C, fitted by the Jiles–Atherton model. The k-value, as a function of grain size, is displayed in Fig. 5. In the uncoupled-grain region ($\phi > \phi_c$), the reduction in the value of k with the increase of ϕ could be attributed to the reduction in the coupling of the grain boundary; this is the boundary of the multi-domain region, consistent with an uncoupled-grain region. Given that, we do not expect variations in the density of other defects like second phases and crystallinity, we can attribute the coupling of the grain boundary as one of the predominant factors in the hysteretic behavior of the films.

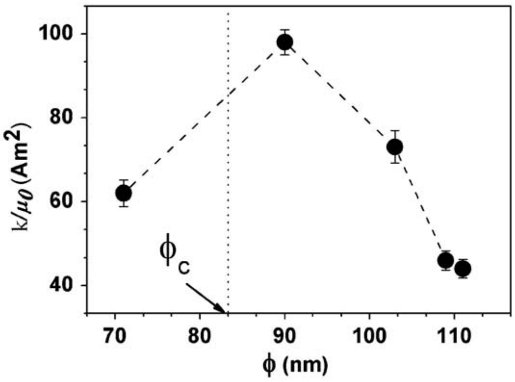

Fig. 5. The k-pinning parameter as a function of grain size. Solid lines are a guide to the eye. Dashed line represents the theoretical boundary between the coupled-grain and the single-grain regions; critical grain size is also indicated.

In summary, grain size distribution in the NiZn ferrite films shows average values in the range from 70 to 111 nm. Experimental coercive field, H_c, takes maximal values at a grain size of $\phi = 93 \pm 3\,\mathrm{nm}$. From a magnetic-correlation model in nanostructured ferromagnets, a critical grain size was estimated at $\phi_c \sim 83\,\mathrm{nm}$. At this value, the corresponding correlation length would be $L_c = 166\,\mathrm{nm}$. The k-pinning parameter was fitted by using the Jiles–Atherton Model with values in the range 40–100 A m^2. The k-pinning parameter is mainly dependent on morphological properties like grain size. From the k behavior, a multidomain regime is expected for $\phi > 93\,\mathrm{nm}$, consistent with the magnetic-correlation length model. From the results obtained, we consider that the films grown at low temperatures ($T \approx 600\,°\mathrm{C}$) have the highest potential for applications, e.g., in microwave applications, since these fulfill two requirements: low-coercivity and high-resistivity as a result of their small grain sizes.

Acknowledgment

This work was partially supported by Center of Excellence for Novel Materials—CENM, under Colciencias Contract no. 043-2005.

References

[1] R.G. Welch, J. Neamtu, M.S. Rogalski, S.B. Palmer, Mater. Lett. 19 (1996) 199.
[2] A. Goldman, Handbook of Ferromagnetic Materials, Kluwer, 1999, p 333.
[3] J.F. Löffler, H.B. Braun, W. Wagner, Phys. Rev. Lett. 85 (2000) 1990–1993.
[4] D.C. Jiles, J.B. Thoelke, M.K. Devime, IEEE Trans. Magn. 28 (1992) 27.
[5] M. Desai, S. Prasad, N. Venkataramani, I. Samjdar, A.K. Nigman, R. Krishnan, J. Appl. Phys. 91 (2002) 2220.
[6] Q. Yan, R.J. Gambino, S. Sampath, L.H. Lewis, L. Li, E. Baumberger, A. Vaifya, H. Xion, Acta Mater. 52 (2004) 3347.
[7] K. Chwastek, J. Szczyglowski, Math. Comput. Simul. 71 (2006) 206–211.
[8] M. Sorescu, L. Diamandescu, R. Swaminathan, M.E. McHenry, M. Feder, J. Appl. Phys. 97 (2005) 10G105.
[9] A. Aharoni, Introduction to the Theory of Ferromagnetism, Second ed., Oxford University Press, Oxford, 2000, pp. 157–162.
[10] V.H. Calle, F. Cuéllar, C. Calle, O. Marín, J. Roa-Rojas, D. Arias, O. Guzmán, W. Lopera, P. Prieto, A. Mendoza, Phys. Status Solidi C 4 (2007) 4197.