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MAGNETIC INTERACTIONS IN ΔM PLOTS FOR NANOCRYSTALLINE Sm-Fe-Ti ALLOYS

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Abstract: Nanocrystalline Sm–Fe–Ti alloys were obtained via mechanical alloying involving 5 h of milling. A characterization of nanocrystalline Sm-Fe-Ti alloys via transmission electron microscopy (TEM) shows the nanocrystalline characteristics of the alloys. Exchange and dipolar interactions were investigated by measuring isothermal remanent magnetization and normalized direct current demagnetization curves. The effects of the interaction field in analyzing the magnetic interactions in nanocrystalline Sm–Fe–Ti alloys were modelled using the ΔM plots.
Keywords: Magnetic interactions, ΔM plots, Henkel Plots

INTRODUCTION

Dipolar interactions, although difficult to study, can be investigated using isothermal remanent magnetization (m_r) and direct current demagnetization (m_d) curves. Although the exchange and dipolar interactions exist, magnetic interactions are even more difficult to study. Conversely, a decrease in the coercivity and squareness has been seen in CoNiFe nanowires because of the increase in dipolar interactions. A transition from single domain (SD) to pseudo single domain (PSD) and multi domain (MD) structures can be attributed to an increase in magnetostatic interactions [17], while that for CoFe nanowires has been proposed to be owing to a decrease in the magnetic moment and large distance between which can reduce magnetostatic interactions [21]. Formulation of a mathematical model for wires with dipolar interactions using the remanence curves is difficult. Nevertheless, is more difficult to formulate a mathematical model for nanocrystalline alloys exhibiting exchange and dipolar interactions. For the study of magnetic interactions researchers occasionally dilute the nanoparticles in a polymer to control average particle distance [12], which is vital to study a system with exchange and dipolar interactions. Additionally, the study of materials with the existence of the exchange and dipolar interactions is of actual interest because these effects are sometimes owing to the existence of a mean field or structural disorder, which can be screened [11]. Magnetic interactions can be studied using the measured m_r and the m_d curves [14]. Additionally, the existence of a mean field and the aleatory interactions increase the diversity of forms of seen in the Henkel [16]. In the nanocrystalline Sm-Fe-Ti alloys obtained by mechanical alloying study the exchange and dipolar interactions is actual due a that one of the phases that crystallize in this system is the phase of $SmFe_2$ that usually present magnetostrictive effects. Since occasionally the microscopic origin of magnetostrictive effects is ascribed to magnetic dipolar energy due to interatomic spacing. Where actual use the methods of the isothermal remanent magnetization (IRM), and the curve direct current demagnetization (DCD) due to that the method yields evidence about the microscopic level magnetic interactions in materials with more one phase [5]. The first section of this work presents the structural characterization of nanocrystalline Sm-Fe-Ti alloys via transmission electron microscopy (TEM) and shows the nanocrystalline characteristics of the alloys. The second section shows the measured m_r and m_d curves for nanocrystalline Sm–Fe–Ti alloys and the mathematical model derived using m_r and m_d curves. The third section supplies modeled and quantified magnetic interactions using the ΔM plot. The final section includes analysis of the Henkel for Sm–Fe–Ti alloys. The purpose of this work is to show the effects of the interaction field in analyzing the magnetic interactions in nanocrystalline Sm–Fe–Ti alloys using the ΔM plots.

MATERIAL AND METHODS

Nanocrystalline alloys of Sm–Fe–Ti was prepared via mechanical alloying in a high energy mill (Spex 8000) comprising a cylindrical container and a steel ball. In adding, Sm (99.9% Sigma Aldrich), Fe (99.9% Sigma Aldrich), and Ti (99.7% Sigma Aldrich) powders were used, the milling was carried out under atmosphere of Argon. Nanocrystalline Sm–Fe–Ti alloys were obtained after 5 h of milling and, the alloys were covered in Tantalum foil and annealing in high vacuum at temperature of 840 °C through 30 min in closed vycor ampules. The annealing was realized out under atmosphere of Argon subsequent quenching in air [18]. The structural characterization was accomplished diffractometer Rigaku model D-MAX 2200 with Cu-K radiation (λ = 1.5406 Å). X-ray diffraction patterns were measured with angular interval $30^o < 2\theta < 60^o$. The phases identified in the analysis of Xray diffraction was Fe, $Sm_{20}Fe_{70}Ti_{10}$, $Sm(Fe,Ti)_2$ and Sm_2O_3. The phases was indexed with the unit cells (96-901-3476) for Fe, (42-1201) for $Sm_{20}Fe_{70}Ti_{10,}$ (42-1049) for $Sm(Fe,Ti)_2$ and (15-0813) for $Sm_2O_{3.}$ The analysis of Xray diffraction confirm the analysis realized by area electron diffraction pattern presented in the Fig. 1(c). For measuring the m_r curves of the nanocrystalline Sm–Fe–Ti alloys, the alloy is in the demagnetized state. The m_r curves characterizes the remanent magnetization after the application and removal of a magnetic field. For measuring the m_d curves, a system of Sm–Fe–Ti nanoparticles was started in a sample that was already magnetized using a magnetic field, and a negative magnetic field was applied as well as removed to reach this state, that characterized the normalized DCD curve described by $m_{d.}$

Interval	$0<m_o<0.33$	$0.33<m_o<0.50$	$0.60<m_o<1$
Probe value	0.20	0.34	0.60
δm	$\delta m = -0.13 < 0$	$\delta m(0.34) = 0.014 > 0$	$\delta m(0.60) = 0.27 > 0$
Conclusions	Only dipolar interactions occur	The value of δm is positive due to appearance of the exchange interaction.	Only exchange interactions occur as previously shown [12]

Table 1.- Test of intersection of remanence curves confirming the mathematical model.

For a detailed characterization of the nanocrystalline Sm–Fe–Ti alloys, a right model was supplied using the $\frac{1}{2}(1-m_d)$ and m_r curves. An experimental model for the nanocrystalline Sm–Fe–Ti alloys considering the afore mentioned magnetic interactions is given using.

GENERAL CONSIDERATIONS

For the study the magnetic interactions in the materials customarily the IRM and DCD remanence curves, m_r and m_d. Henkel Plots and Plots ΔM versus Magnetic Field was applied.

IRM AND DCD REMANENCE CURVES, m_r AND m_d

For the IRM and DCD remanence curves, m_r is the normalized remanent magnetization after applying a magnetic field to the sample in a demagnetized state and m_d is the normalized remanent magnetization after application of a reverse magnetic field in a sample previously magnetized with a magnetic field [10]. These curves are plotted to understand the types of magnetic interactions. When a magnetic interaction occurs, the separation between the curves is more prominent. When such interactions are not present, the separations between the curves are less prominent, and the curves are equal one to the another.

HENKEL PLOTS

In the Henkel Plots the presence of demagnetization interactions related with the apparitions of a dominant local disorder is used in the Henkel plots for description of this comportment. This includes plotting vs. versus where is given using:

$$m_r=\int_0^{\alpha m_{IRM}} g(h)dh \quad (1)$$

where the result of the Integral is:

$$m_r= [\alpha m_{IRM}]_0^h \quad (2)$$

then if we substitute the superior and inferior limit the result is:

$$m_r= h \quad (3)$$

Then if defined the value of the m_d as the following integral:

$$m_d=1-2\int_0^{\alpha m_{IRM}} g(h)dh \quad (4)$$

Then m_d is equal to:

$$m_d= 1\text{-}2[\alpha m_{IRM}]_0^h \quad (5)$$

then if we substitute the superior and inferior limit the result is:

$$m_d= 1\text{-}2[h] \quad (6)$$

then combining the eq. 3 with 6 we found the relation of Wohlfarth

$$m_d= 1\text{-}2m_r \quad (7)$$

where the Wohlfarth model is considered, which has a straight line with slope equal to −2 [6]. Now, Henkel plots are significant for study the mean-field effects with no random interactions, and for the study of mean-field effects and random interaction.

PLOT ΔM VERSUS MAGNETIC FIELD.

The goal of the ΔM plots is to study magnetic interactions between the nanoparticles. ΔM plots is used for a non-interacting system of single-domain uniaxial particles. In ΔM plots can be written as:

$$m_r=\int_0^{h+H_{in}} g(h)dh \quad (8)$$

where H_{in} represent the interaction field and the result of the Integral is:

$$m_r= [h]_0^{h+H_{in}} \quad (9)$$

then if we substitute the superior and inferior limit the result is:

$$m_r= h + H_{in} \quad (10)$$

where $H_{in}= \alpha M + \beta(1\text{-}M^2)$ and m_d can be calculated as follows:

$$m_d=1\text{-}2\int_0^{h+H_{in}} g(h)dh \quad (11)$$

where the result of the Integral is:

$$m_d=1\text{-}2[h]_0^{h+H_{in}} \quad (12)$$

then if we substitute the superior and inferior limit the result is:

$$m_d=1\text{-}2\ (h + H_{in}) \quad (13)$$

In order to preserve field information we additional a futher term to the equation 13, and combining the equations (10) and (13) we defined the ΔM plots as:

$$m_d=1\text{-}2\ m_r + \Delta M \quad (14)$$

Using ΔM plots, it can be concluded that when there is no magnetic interaction, the ΔM plot would be a horizontal line (ΔM = 0). If the values of ΔM are positive in these curves, it is a sign that the dominant interaction between the nanoparticles is the exchange interactions, while when the values of ΔM are negative, it is concluded that the dominant magnetic interactions were the dipolar interactions [1,8,20]. Consequently, we can understand that physical meaning in the ΔM plots is that m_r represent the fraction of the total magnetic moment of the system switched at a definite field, while that m_d represent the fraction

switched in the DCD curve. The ΔM plots can be modeled by the following equation: $H_{in}=\alpha M + \beta(1-M^2)$ which represents the effects of the interaction field (symbolized by αM, where α represents the interaction field coefficient) and the second order term represents the fluctuation of the interaction field. Different values of the interaction field coefficient and a value of β = 0 can be used.

RESULTS

Fig. 1 shows the TEM analysis of the nanocrystalline Sm–Fe–Ti alloys obtained by mechanical alloying after 5 h of milling. Figs. 1 (a) and (b) show micrographs obtained in the bright and dark field régimes using the transmitted and diffracted beams respectively. Fig. 1 (c) shows the selected area electron diffraction pattern for the nanocrystalline Sm–Fe–Ti alloys.

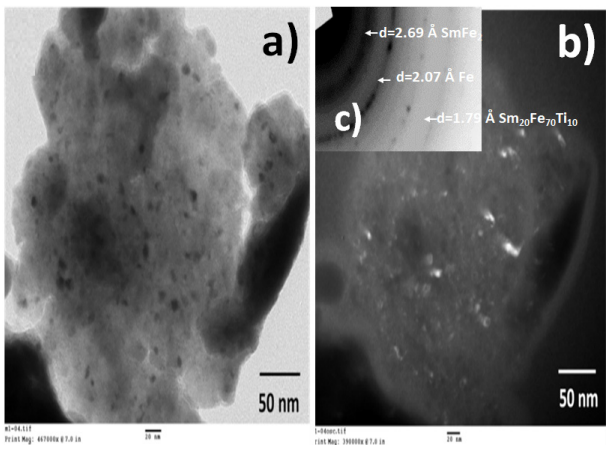


Fig.1. (a) Bright field and (b) dark field TEM images and (c) selected area electron diffraction pattern for nanocrystalline Sm–Fe–Ti alloy after 5 h of milling.

Fig. 1 (a) shows small dark fragments in the micrograph obtained and brighter region indicating the presence of more of one phase. Fig. 1 (b) shows particles with an average size of 8 nm. Fig. 1 (c) shows the indexed selected area electron diffraction pattern, which confirmed the presence of the $Sm_{20}Fe_{70}Ti_{10}$, $SmFe_2$, Fe, and Sm_2O_3 phases that is consistent with the phases found in the ternary phases diagram for the alloy of Sm-Fe-Ti annealing at 800 °C with a 20% at of Sm.

Fig. 2 shows the initial curve magnetization and a minor hysteresis loop for nanocrystalline Sm–Fe–Ti alloy annealing to a temperature of 840 °C and quenching in air; the hysteresis loops of the powders were measured using an alternating gradient magnetometer. In Ref.12 a coercivity of 349 and 402 Oe was observed for nanocrystalline Sm–Fe–Ti alloy, while that here a coercivity of 424 Oe was observed for nanocrystalline Sm–Fe–Ti alloy. To compare the values of the magnetization with other materials the magnetization was normalized.

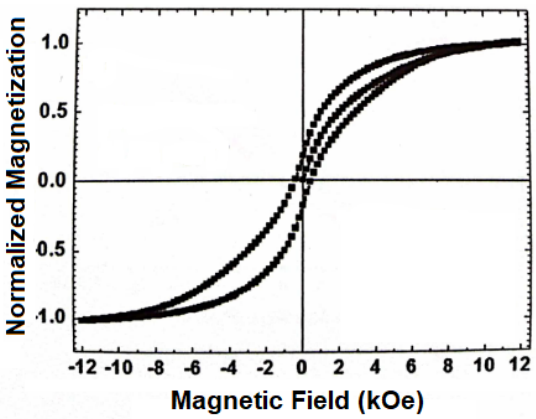


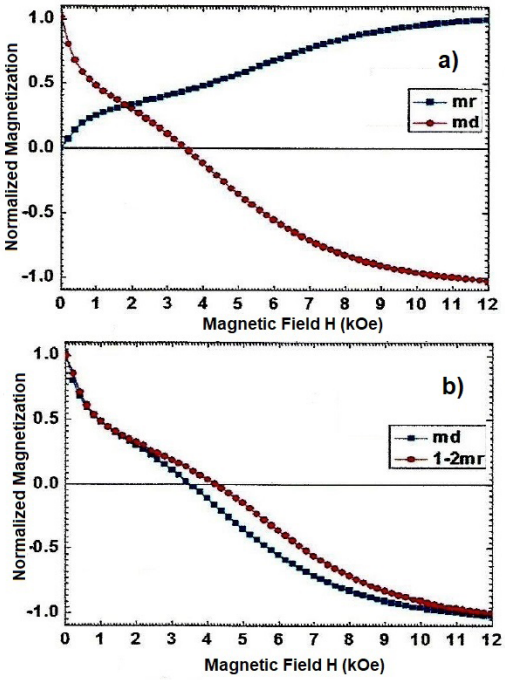
Fig. 2. Hysteresis loop for nanocrystalline Sm–Fe–Ti alloy annealing to a temperature of 840 °C and quenching in air.

Fig. 3 (a) shows the remanence curve for the nanocrystalline Sm–Fe–Ti alloys. Here, m_r is the normalized remanent magnetization after applying a magnetic field to the sample in a demagnetized state and m_d is the normalized remanent magnetization after application of a reverse magnetic field in a sample previously magnetized with a magnetic field of 12 kOe [10,15]. Fig. 3(b) and (c) shows that the separation between the remanence curve for the nanocrystalline Sm–Fe–Ti alloys is more prominent, and it can be concluded that the interactions occur.

For the nanocrystalline Sm–Fe–Ti alloys, the type of interactions with the following values of normalized magnetization, m_0, can be inferred through the intersection of the remanence curves using:

$$\delta m = m_0 - \frac{1}{3} \quad (15)$$

The following conclusions are derived from the intersection of remanence curves by confirming and testing the mathematical model as previously [2]. The Table 1 shows the test of intersection of remanence curves confirming the mathematical model. If m_0 = 1/3 and the δm= 0, there are no magnetic interactions. For the nanocrystalline Sm–Fe–Ti alloys the intersection of the remanence curves is in 0.34 related with arrival of the exchange interaction as previously shown [11]. The conclusions derived for the range $0.33 < m_0 < 0.50$ can be confirmed via quantification of the magnetic interactions using the ΔM plot. Figs. 3 (b) and 3 (c) shows normalized m_r and m_d curves, respectively, for the nanocrystalline Sm–Fe-Ti alloys.



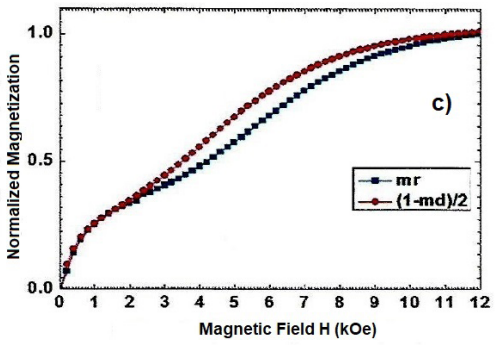


Fig. 3. (a) IRM and DCD remanence curves, m_r and m_d measured for Sm–Fe–Ti alloys. (b) Modified IRM (1-2m_r) and m_d remanence curves as a function of magnetic field and (c) Modified DCD (1-m_d)/2 and m_r remanence curves as a function of magnetic field.

$$\alpha = \frac{1}{9}H \quad (16)$$

obtained from the remanence curves, where α is the interaction field coefficient calculated from the m_r and m_d curves. When the magnetic field is $H = H_r$ and $H = H_d$, the values of H_r and H_d indicate the values of the field at which the normalized IRM curve is 0.5, and the field at which the DCD curve is zero., respectively. The value of ΔH can be obtained using:

$$\Delta H = H_r - H_d \quad (17)$$

Combining equations (16) and (17), the total interaction field can be calculated as follows:

$$\alpha_T = \frac{1}{9}\Delta H \quad (18)$$

Here, ΔH is significant. This value is used in the determination of the total field (H_{Tot}), which includes the contribution of the applied magnetic field (H_{app}) and the interaction field (H_{in}). When exchange and dipolar interactions exist in the materials and the dipolar interactions is dominant, find a representation for the magnetic interactions is crucial. Here an interaction field coefficient (α_T) is used

to represent these magnetic interactions for nanocrystalline Sm–Fe–Ti alloys that is equal to 1/9 ΔH. When the exchange and dipolar interactions exist in the material and the dipolar interactions are dominant, this interaction field coefficient value is different [15]. Conversely, if H in Eq. (16) is assumed to be equal to the dipolar interaction field (H_{dip}), which is equal to NM_sP, where N is the demagnetization factor, M_s is the saturation magnetization and P is the packing fraction, then the interaction field can be written as:

$$\alpha = \frac{1}{9} NM_sP \quad (19)$$

The relationship between interaction field (H_{in}) and magnetization (m) can be given using the following Eq. $H_{in} = \alpha m$. Using Eq. (19), the following equation for the interaction field was obtained:

$$H_{in} = \frac{1}{9} H_{dip} m \quad (20)$$

Subsequently, using Eq. (20), an equation for the total dipolar field (H^D_T) can be obtained as follows:

$$(H^D_T) = H_d + H_{in} \quad (21)$$

where Eq. (21) is used obtain total dipolar field. If we consider the magnetization for the non-saturated states to be $m = \frac{1}{2}(1m)$ and the demagnetizing field to be H_d, which is equal to the NM_s [9], then Eq. (21) can be rewritten as:

$$(H^D_T) = NM_s + \frac{1}{18} NM_sP \pm \frac{1}{18} NM_sPm \quad (22)$$

If we consider Brown's theorem $K_s = \frac{1}{2}(M_sH_s)$ is the anisotropy constant, where M_S is the saturation magnetization, and the shape anisotropy constant is equal to $K_s = \frac{(\Delta NM_s^2)}{2}$

Then, it can be shown that H_s is equal to ΔNM_s. Moreover, we can define the switching field (H_{sw}) as follows:

$$H_{sw} = H_a - H_s \quad (23)$$

where is the anisotropy and is the shape anisotropy fields. Combining Eq. (22) and (23), we get the following Eq:

$$H_T = H_a + \frac{1}{18} NM_sP \pm \frac{1}{18} NM_sPm \quad (24)$$

Equation 9 represent total anisotropy energy where the first term represent the uniaxial anisotropy, the second term signify the magnetostatic energy and the third term characterise the interaction energy. Fig. 4 shows the ΔM curve versus magnetic field curve for the nanocrystalline Sm–Fe–Ti alloys. Fig 4 (a) shows that exchange and dipolar interactions are present in the sample. Here, is noted that the weak exchange and improved the dipolar interactions. Fig. 4 (b) shows the ΔM versus magnetic field for nanocrystalline Sm–Fe–Ti alloys with different values of interaction field coefficient. Fig. 4 (c) shows the equivalent ΔM plots normalized to their maximum values for nanocrystalline Sm–Fe–Ti alloys [6].

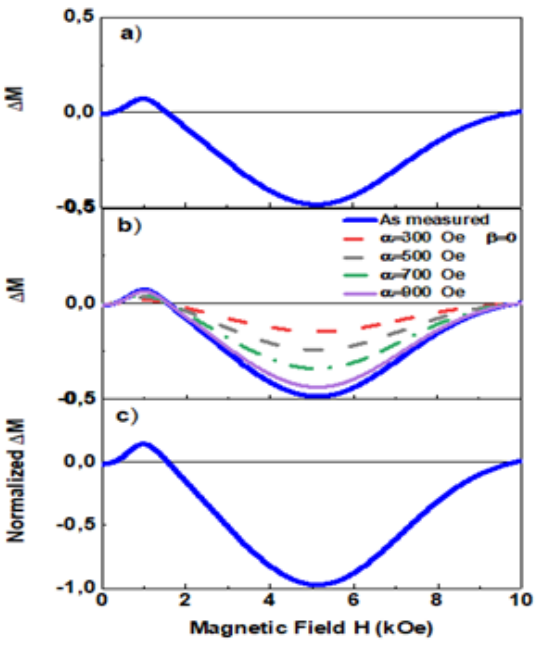


Fig.4. ΔM versus magnetic field for nanocrystalline Sm–Fe–Ti alloys

Fig. 5 shows the Henkel plot for nanocrystalline Sm–Fe–Ti alloys. The Henkel plot here shows that the experimentally

measured values are below the Wohlfarth line, where the effects due to the presence of a local disorder are dominant [4]. Here, the disorder is considered as fluctuations in the particle size, shape, arrangement, as well as relative position; defects of the lattice; and rearrangement in the domain structure.

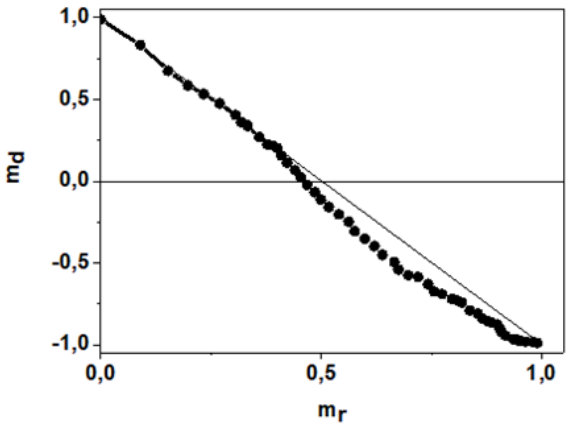


Fig.5. Henkel plot for nanocrystalline Sm–Fe–Ti alloys.

DISCUSSION

From the section describing the (m_r) and normalized (m_d) curves for the nanocrystalline Sm–Fe–Ti alloys, the curve is below the 1–2 curve. This can be explained by the dominance of dipolar interactions in nanocrystalline Sm–Fe–Ti alloys. Conversely for the model used in the section discussing the ΔM plot for nanocrystalline Sm–Fe–Ti alloys with different values of interaction field coefficient. Additionally, the mean field approximations are significant owing to the interaction field coefficient summary of demagnetizing effects, such as others in other interaction forms [13]. Furthermore, the mean field model shows that each atomic moment interacts equally with other atomic moments, and this is valid for a paramagnetic phase. However, for the ferromagnetic phase, the idea of a mean field occurring only in one domain and that the interaction between the atomic moment with distance is applied. Here, the positive values of the interaction field coefficient (α>0) for the nanocrystalline Sm–Fe–Ti alloys are secondhand, which proved to be a good approximation as is shown in the ΔM plot section. Conversely, in the Ising model the interaction field coefficient is used for ferromagnetic phases too, where the interaction is only between the nearest neighbors. Additionally, understanding if the interaction between the magnetic moment exists is crucial because in calculating the exchange energy, and interaction field is (H_{in}= αNm). Consider the situation where only nearest neighbor interaction is meaningful and that (on the wall domain) the directions along the wall domain of the magnetic moment vary from domain to domain, in this case the approximation of the mean field is not valid. Introducing the mechanical exchange constant is necessary in the afore mentioned case, the constant of quantum mechanical exchange, J, which is an alternative representation of the interactions between the nearest neighbors and decides the coupling of the mean field of the Weiss α. Therefore, the mean-field parameter can be associated with the exchange coupling in the domain and can be used in the mean field approximation [13]. Additionally, other authors use the interaction field coefficient, which defines the effects of the mean field to a large scale. Another model important for understanding the properties of nanocrystalline Sm–Fe–Ti alloys is the Heisenberg model, which considers that exchange coupling between the spins of the two ions with quantum number of spin (S) and no orbital moment. In this model, the principal magnetic interaction in rare earth and transition metal alloys can be attributed to the atoms of transition metals owing to their overlapping 3d shell above neighboring sites. The interaction is normally ferromagnetic, but this depends rationally on the interatomic spacing. For alloys that have Fe

the sign can change from negative to positive when the interatomic spacing is greater than 25 nm. For the nanocrystalline Sm–Fe–Ti alloys, the crystal size is of 18 nm next of annealing at temperature of 840 °C through 30 min. Conversely, the exchange interactions can be modeled using the molecular field approximation. If the interaction -2JijSiSj of the Heisenberg model is substituted and added above each couple of atoms, the interaction of each moment with an effective molecular field actuating in the site can be obtained. However, in this model the compound appeared to have two magnetic sublattices comprising rare earth and transition metal atoms [7]. Therefore, the model proposed in this section describing ΔM plots is a model wherein the interaction field coefficient, α explains the effects attributed to the presence of dipolar and exchange interactions, as in the mean-field model, the quantum mechanical treatment, the Heisenberg model, and as modeled for-exchange interactions using the molecular field approximation. In this model for the nanocrystalline Sm–Fe–Ti alloys α shows the strength of the interactions considers the importance of the interatomic spacing and that the principal magnetic interactions in the rare earth and transition metal alloys can be attributed to the transition metal atoms owing to an overlap in the 3d shell sites above neighbors. Conversely, in section, referring to the Henkel plot for the nanocrystalline Sm–Fe–Ti alloys, it is concluded that the experimentally measured values are below the Wohlfarth line, where the effects due to the presence of a local disorder are dominant. However, in section before modeling the magnetic interactions with the ΔM plot for nanocrystalline Sm–Fe–Ti alloys, it was observed that the positive values of the interaction field coefficient can be used for describing the magnetic interactions in the nanocrystalline Sm–Fe–Ti alloys.

CONCLUSIONS

In conclusion, through the model proposed based on IRM and DCD remanence curves, m_r and m_d for nanocrystalline Sm–Fe–Ti alloys, the type of magnetic interactions and if exist more than one interaction in the material can be inferred. ΔM plots for nanocrystalline Sm–Fe–Ti alloys showed negative and positive values, and these values are associated with the existence of dipolar and exchange interactions, respectively, wherein dipolar interactions are dominant [19]. Decisively, the model proposed for the ΔM plots was tested and confirmed using different values of the interaction field coefficient. These results are important in the study of magnetic interactions because they help researchers understand the effects of the interaction field above the ΔM plots for nanocrystalline Sm–Fe–Ti alloys. These results show that ΔM plots can be modeled using large values of the interaction field coefficient. We have found via Henkel plots for nanocrystalline Sm–Fe–Ti alloys that the effects due to the presence of a local disorder are dominant.

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