TITLE: Anisotropy Determinations in Exchange-Spring Magnets

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ABSTRACT: Ferromagnetic nanocomposites, or "exchange spring" magnets, possess a nanoscaled microstructure that allows intergrain magnetic exchange forces to couple the constituent grains and alter the system's effective magnetic anisotropies. While the effects of the anisotropy alterations are clearly seen in macroscopic magnetic measurement, it is extremely difficult to determine the detailed effects of the system's exchange coupling, such as the interphase exchange length, the inherent domain wall widths or the effective anisotropies of the system. Clarification of these materials parameters may be obtained from the "micromagnetic" phenomenological model, where the assumption of magnetic reversal initiating in the magnetically-soft regions of the exchange-spring magnet is explicitly included. This approach differs from that typically applied by other researchers[1] and allows a quantitative estimate of the effective anisotropies of an exchange spring system. Hysteresis loops measured on well-characterized nanocomposite alloys based on the composition $Nd_2Fe_{14}B$ + α-Fe at temperatures above the spin reorientation temperature were analyzed within the framework of the micromagnetic phenomenological model. Preliminary results indicate that the effective anisotropy constant in the material is intermediate to that of bulk α-Fe and bulk $Nd_2Fe_{14}B$ and increases with decreasing temperature. These results strongly support the idea that magnetic reversal in nanocomposite systems initiates in the lower-anistropy regions of the system, and that the soft-phase regions become exchange-hardened by virtue of their proximity to the magnetically-hard regions.

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ANISOTROPY DETERMINATIONS IN EXCHANGE-SPRING MAGNETS

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A modified micromagnetic phenomenological model is applied to nanocomposite permanent magnetic alloys with the goal of determining the system's effective anisotropy. Hysteresis loops were obtained from well-characterized nanocrystalline alloys based on the composition $Nd_2Fe_{14}B$ at temperatures above the spin reorientation temperature. The resultant data are analyzed within the framework of the micromagnetic phenomenological model. Preliminary results indicate that the effective anisotropy constant in the material is intermediate to that of bulk α–Fe and of bulk $Nd_2Fe_{14}B$ and increases with decreasing temperature. These results support the idea that magnetic reversal in magnetic nanocomposite systems initiates in the lower-anisotropy regions of the system, and that the soft-phase regions become exchange-hardened by virtue of their proximity to the magnetically-hard regions.

1 Introduction

Ferromagnetic nanocomposites, or "exchange spring" magnets [1], possess a nanoscaled microstructure that allows intergrain magnetic exchange forces to couple the constituent grains and alter the system's effective magnetic anisotropies. While the effects of the anisotropy alterations are clearly seen in macroscopic magnetic measurement, it is extremely difficult to determine the detailed effects of the system's exchange coupling, such as the interphase exchange length, the inherent domain wall widths or the effective anisotropies of the system. Clarification of these materials parameters may be obtained from the "micromagnetic" phenomenological model, where in the present work the assumption of magnetic reversal initiating in the magnetically-soft regions of the exchange-spring magnet is explicitly included. This approach differs from that typically applied by other researchers [2,3] and allows a quantitative estimate of the effective anisotropies of a ferromagnetic exchange spring system.

2 Modification of the Phenomenological Micromagnetic Model

To date, efforts to determine in nanocomposite magnets the physical parameters described above have been restricted to first-principles-type numerical micromagnetic calculations [4] and phenomenological models of magnetic reversal. These phenomenological models are based either on the formation of a magnetization inhomogeneity, known as the "global" model [5], or on overcoming energy barriers to reversal, known as the "micromagnetic" model [6]. The latter model examines magnetic reversal against an energy barrier that is proportional to the magnetocrystalline anisotropy energy. In this formalism reverse domain nucleation occurs at the first local departure from magnetic saturation, and thus invokes the Stoner-Wohlfarth [7] (S-W) model of magnetic reversal to describe the local reversal. The "micromagnetic" phenomenological model mathematically states that the coercivity $H_C(T)$ is always reduced from the ideal anisotropy field $H_A(T)$ via materials defects and demagnetization effects from surrounding grains (Eq. (1)). The unitless parameter α describes the effect of materials defects on magnetic reversal, and N_{eff} is the effective demagnetization factor that encompasses the self-demagnetization field and the dipolar interactions of neighboring grains. $J_S(T)$ is the measured temperature-dependent saturation polarization of the system.

$$\mu_0 H_c(T) = \alpha\mu_0 H_A(T) - N_{eff} J_s(T) \quad (1)$$

In the present work the static micromagnetic model, as developed by Kronmüller, was modified for application to magnetic nanocomposites in which the magnetic reversal nucleates, in a Stoner-Wohlfarth-type manner, in the magnetically-soft phase component. This viewpoint is a departure from previous applications of the micromagnetic model to magnetic nanocomposites[2, 3] in which the reversal is presupposed to occur in the magnetically-hard component of the material, against the bulk anisotropy field of the hard phase. To this end, Eq. (1) was modified to explicitly include the materials parameters of the $Nd_2Fe_{14}B$ + α-Fe system as below:

$$\mu_0 H_c(T) = \alpha\mu_0 \frac{\beta K_{eff}}{J_s(Fe,T)} - N_{eff} J_s(T) \quad (2)$$

The modification includes the substitution of the expression $\beta K_{eff}/M_S(Fe,T)$ for the effective anisotropy field of cubic α-Fe. The factor β is equal to 2 if the easy magnetization directions in the cubic phase are along the <100> directions as is the case for Fe [8].

In this modified approach it is desired to experimentally determine the value of the anisotropy in the region of the initial magnetic reversal, which is understood to be the magnetically-soft phase component of the nanocomposite. In the current work the α-parameter will be taken as a constant, not as an experimental unknown to be determined through application of a model. The determination of the α-parameter necessitates a revisit to the Stoner-Wohlfarth model as applied to the reversal of an isolated particle with cubic anisotropy, instead of the typical uniaxial anisotropy. The assumption that the initial magnetic reversal may be modeled as an isolated cubic Stoner-Wohlfarth particle is valid in the limit that the soft-phase regions are sufficiently isolated by the hard-phase regions in the sample, and the condition of uniform magnetization is assumed to exist in the region that gives rise to the reverse magnetization nucleus. By symmetry there are two energy extrema in the cubic system in the range $0 \le \varphi \le 90°$, analogous to the uniaxial situation with two energy minima in the range $0 \le \varphi \le 180°$. The critical field H_K describes the operative switching field necessary to reverse the magnetization at a given angle φ between the between the applied field and the magnetization vector. The expression for the normalized critical field H_K for the cubic system in the Stoner-Wohlfarth formalism is approximated by analogy to that for the uniaxial system, Eq. (3a), and is provided in Eq. (3b):

$$\text{(uniaxial)} \quad H_{K,U} = \frac{2K}{M_S}\left(\cos^{2/3}\varphi + \sin^{2/3}\varphi\right)^{-3/2} \quad (3a)$$

$$\text{(cubic)} \quad H_{K,C} = \frac{K}{M_S}\left(\cos^{2/3}(2\varphi) + \sin^{2/3}(2\varphi)\right)^{-3/2} \quad (3b)$$

In both cases the minimum normalized critical field takes on a value of 0.5:

$$i.e., \left(\frac{H_{K,U}}{\left(2K/M_S\right)}\right)_{\min} = 0.5 \text{ and } \left(\frac{H_{K,C}}{\left(K/M_S\right)}\right)_{\min} = 0.5.$$

Therefore $H_{K,U} = 0.5 \cdot \left(2K/M_S\right) = K/M_S$ and $H_{K,C} = 0.5 \cdot \left(K/2M_S\right)$. It is appropriate to utilize the α-parameter that corresponds to the minimum (φ_{min}) values under the condition that the soft-phase regions are distributed more-or-less isotropically throughout the nanocomposite; thus it is assumed that there are always cubic soft phase regions in which a portion of magnetization reversal will nucleate at the minimum reduced coercive field of $K/2M_S$ that provides a value of the minimum α-parameter equal to 0.5.

Inclusion of the minimum α-parameter of 0.5 allows Eq. (2) to be rewritten as:

$$\mu_0 H_c(T) = \mu_0 \frac{0.5\beta K_{eff}}{J_s(Fe,T)} - N_{eff} J_s(T) \tag{4a}$$

and rearranged to yield the equation:

$$\mu_0 \frac{H_c(T)}{J_s(T)} = \mu_0 \frac{K_{eff}}{(J_s(Fe,T) * J_S(T))^2} - N_{eff} \tag{4b}$$

where the value of $\beta = 2$ has been explicitly included as appropriate for iron. Evaluation of Eq. (4b) provides a linear slope equal to effective anisotropy constant of the nanocomposite K_{eff} and an axial intercept that provides the effective demagnetization constant N_{eff}. In the case of single-phase nanocrystalline $Nd_2Fe_{14}B$ the analogous expression of the modified phenomenological model is

$$\mu_0 \frac{H_c(T)}{J_s(T)} = \mu_0 \frac{2K_{eff}}{(J_S(T))^2} - N_{eff} \tag{4c}$$

that reflects the appropriate expression for anisotropy of $2K_1/M_S$ in uniaxial materials with uniaxial anisotropy constants $K_{1,U} >> K_{2,U}$. In the present work we describe preliminary results that demonstrate the utility of application the modified phenomenological approach to permanent magnet nanocomposites and to state the need for further work on this topic.

3 Experimental Details

Determinations of the intrinsic coercivities H_{ci} and saturation magnetizations J_S as functions of temperature were made on two melt-spun nanocrystalline samples: one nominally-stoichiometric

sample of $Nd_2Fe_{14}B$ (2-14-1) and one nanocomposite sample of 2-14-1 plus 27 wt% α-Fe. The alloys were made from commercial-grade materials by standard melt-quenching techniques at Magnequench International, Inc.[8] and then post-annealed to optimize their hysteretic magnetic properties. The amount of excess iron in the nanocomposite sample was determined from the ferromagnetic hysteretic response measured at 750 K.

A nanocrystalline microstructure was confirmed by x-ray diffraction to exist in the two samples studied. The average grain size in the single-phase nanocrystalline Nd-Fe-B was found to be 32 nm. A more detailed investigation of the microstructure of the nanocomposite sample that separated strain effects from size-broadening effects in the Bragg reflections provided grain size determinations of 44 nm for the 2-14-1 phase and 38 nm for the α-Fe phase [9]. The samples were mounted in silica tubes and evacuated to P ~ 1 x 10^{-6} Torr to avoid oxidation during magnetic measurement. Hysteresis data was obtained with a MPMS SQUID magnetometer after field cooling in a 5 T field from T = 380 K to the measuring temperature. The temperature range selected for study is 340 K ≤ T ≤ 160 K; the lower end of the temperature range is above the spin reorientation temperature for bulk $Nd_2Fe_{14}B$ of T = 135 K [10]. The saturation magnetization $J_S(T)$ was obtained from extrapolation to the abscissa of the graphed data trend $(1/\mu_0 H^2(T))$ *vs* $J_S(T)$. It was necessary to express the coercivity as a negative number in the calculations to be consistent with well-known result that the anisotropy constants in $Nd_2Fe_{14}B$ above the spin reorientation temperature are non-negative.

4 Results

The measured trends of saturation magnetization J_S and coercivity H_{ci} of both samples studied are provided as functions of temperature in Fig. 1. Consistent with the results of other researchers, the coercivity of the nanocomposite sample is significantly lower, but more thermally stable, than that of the single-phase nanocrystalline sample [11]. The saturation magnetization J_S of the nanocomposite sample is close to 30% higher than that of the single-phase nanocrystalline sample, reflecting the high-magnetization α-Fe content.

Graphical representations of Eqs. (4b) and (4c) are shown in Figs. 2 and 3. The figures include linear fits to the data that provide estimates of the effective anisotropy constants K_{eff} and the effective demagnetization constants N_{eff} in the systems. Regression R factors that characterize the goodness of the linear fit are included in the figure legends. The data of the single-phase nanocrystalline Nd-Fe-B sample are shown in Fig. 2 and provide a calculated K_{eff} of 8.1 MJ/m^3, which compares moderately favorably with the literature value of 5 MJ/m^3 at room temperature [12]. The calculated N_{eff} factor for the single-phase nanocrystalline sample is 0.89, which is somewhat less than the value of 1 typically found for exchange-decoupled nanocrystalline and sintered magnets [2] and possibly signals the presence of moderate intergranular exchange present in the system.

Figure 3 shows results of the micromagnetic model applied to the nanocomposite sample. The data trend can be divided into two linear regions: a higher-temperature and a lower-temperature region. At the higher temperatures both the calculated effective anisotropy constant K_{eff} of 6.35 MJ/m^3 and the effective demagnetization factor N_{eff} = 0.18 are reduced from those found for the case of the nominally-stoichiometric sample. In the lower temperature region for T < 300 K the effective anisotropy of the nanocomposite sample is calculated to be approximately

17 mJ/m^3. This value is consistent with an increase in the anisotropies with decreasing temperature of both $Nd_2Fe_{14}B$ and α-Fe, and is close to the NdFeB phase K_1+K_2 value of approx-

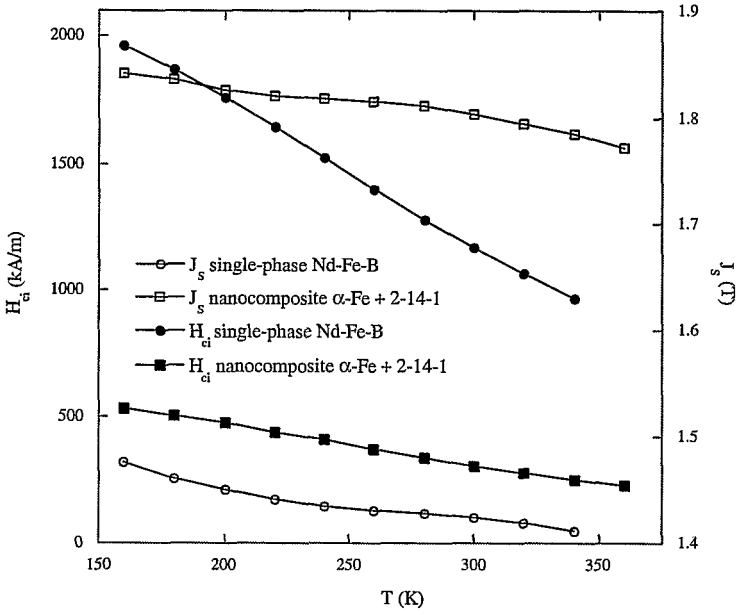

Figure 1. Measured coercivity H_{ci} and saturation magnetization J_S trends of the two samples studied as functions of temperature.

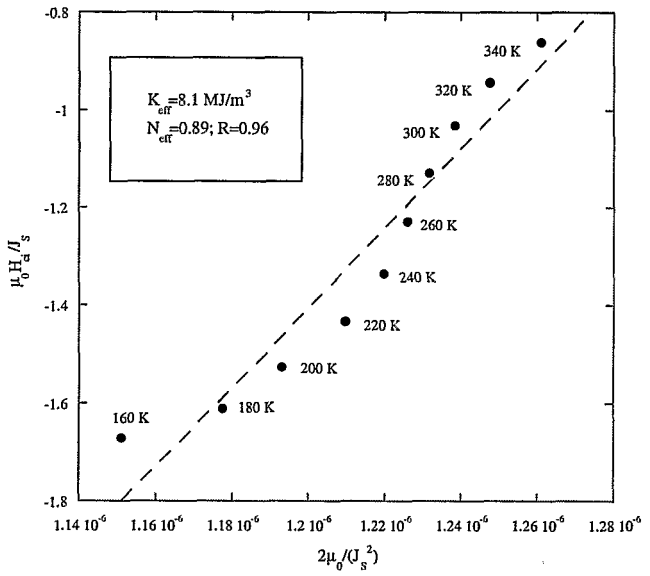

Figure 2. Graphical representation of the modified phenomenological model of magnetic reversal in single-phase nanocrystalline $Nd_2Fe_{14}B$ described by Eq. (4c). The linear fit to the data is signified by the dashed line, and the resultant parameter determinations are provided in the legend.

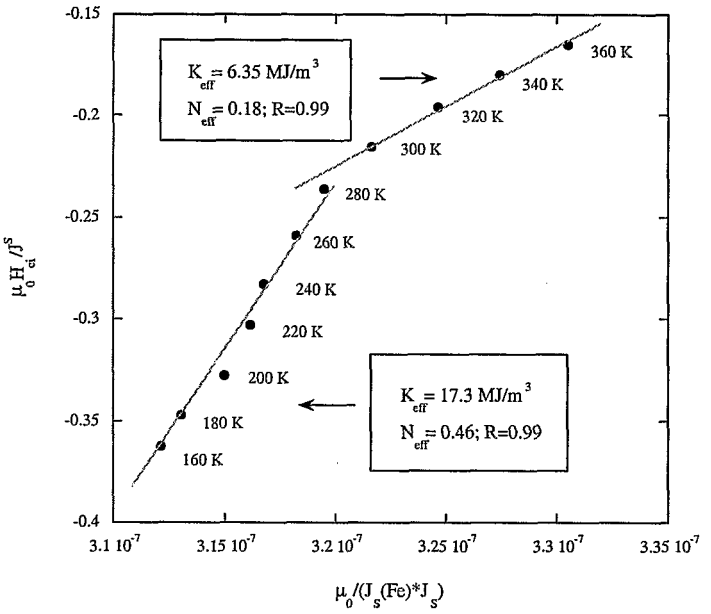

Figure 3. Graphical representation of the modified phenomenological model of magnetic reversal in nanocomposite $Nd_2Fe_{14}B$+α-Fe described by Eq. (4b). The linear fits to the data are signified by dashed lines, and the resultant parameter determinations are provided in the legends.

imately 10 MJ/m^3 in the temperature range 150 K ≥ T ≥ 200 K [12]. The calculated N_{eff} value in the lower-temperature region of the nanocomposite data has increased relative to that characterizing the higher-temperature region to a value N_{eff} = 0.46, but remains smaller than the value found for the single-phase nanocrystalline counterpart.

5 Discussion and Summary

The goal of application of the modified phenomenological model, as described in Section 1, is to experimentally determine the value of the effective magnetic anisotropy in the region of the initial magnetic reversal in permanent magnetic nanocomposites. To date this type of information has not been obtained in magnetic systems. The results reported here, while not extremely close to experimental reports, are encouraging because they provide anisotropy values of the correct order of magnitude. This is a notable accomplishment upon consideration of the simplicity of the model and the complexity of the nanocrystalline microstructures. An additional factor that complicates the model is that the simple linear equation provides only a single value of anisotropy over the studied temperature range. Therefore application of the model to the single-phase Nd-Fe-B system may not be appropriate, as the anisotropy is rather temperature-dependent in the region studied. It is perhaps more appropriate to apply the model to the nanocomposite system, where the anisotropy present in the nucleating soft phase is less sensitive to temperatures significantly less than its Curie temperature. Perhaps more valuable is comparison of the results obtained from the two types of samples, the single-phase material and the nanocomposite material. At higher temperatures a moderate decrease in the effective anisotropy of the nanocomposite system relative

to that determined in the single-phase system is noted. If, as anticipated, the reverse magnetization nucleation occurs in the magnetically-soft α-Fe regions of the specimen, this result indicates that the α-Fe phase has been significantly exchange-hardened well beyond its room-temperature anisotropy value of 0.04 – 0.05 MJ/m^3 [13]. The reduction in effective anisotropy in the nanocomposite system is accompanied by a significant decrease in the effective demagnetization constant. The effective demagnetization constants determined in this study agree with those of Bauer *et al.*[2], where they find effective demagnetization constants in the approximate range 0.10 – 0.20 for nanocomposite samples. As the effective demagnetization constants characterize the internal stray fields acting on the nanograins, low N_{eff} values are consistent with the internal magnetic flux threading its way through the magnetically-soft regions of the exchange-coupled nanocomposite. As temperature is lowered in the nanocomposite the domain wall widths of the system, which ultimately communicate the interphase exchange coupling, decrease as the phase anistropies increase. An increase in the N_{eff} values at lower temperatures, as found for the α-Fe + Nd-Fe-B sample, is expected to be concurrent with the reduction in the operative exchange coupling.

The elbow in the experimental trend at lower temperatures found for the nanocomposite system illustrated in Fig. 3 is somewhat puzzling, as there is no known report of a change in the materials parameters of either α-Fe or $Nd_2Fe_{14}B$ for T < 300 K. Such a result may signify transfer a portion of control of reverse nucleation in nanocomposite samples to the Nd-Fe-B phase from the α-Fe phase as temperature is lowered.

In summary, modification of the current micromagnetic model to include the phenomena of reverse nucleation in the magnetically-soft region of nanocomposite magnets seems to be a promising procedure that can yield insight into the effective anisotropies and internal demagnetization conditions in this class of advanced magnetic material.

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