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ORIGINAL PAPER

Hysteresis Modeling of Nanocrystalline NdFeB Magnets

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Abstract It is discussed the modeling of hysteresis curves of nanocrystalline exchange coupled NdFeB magnets. The hysteresis was measured with an applied field of 9 T, using a vibrating sample magnetometer with superconducting coil. The demagnetizing curve of the first quadrant of the hysteresis was fitted using the Stoner-Wohlfarth (SW) model with the Callen-Liu-Cullen (CLC) modification. Interaction effects as exchange coupling of $Nd_2(FeCo)_{14}B$ phase with the alpha iron phase were determined. The volume fraction of the soft phase can be accounted with the CLC model. Feasible methods for improving the maximum energy product (BH_{max}) of rare-earth transition metal magnets with the help of the SW-CLC model data are discussed.

Keywords NdFeB · Magnets · Hysteresis · Exchange coupling · Stoner-Wohlfarth

1 Introduction

The Callen-Liu-Cullen (CLC) [1] alteration of Stoner-Wohlfarth (SW) hysteresis [2] is successful for the modeling of nanocrystalline SmCo magnets [3, 4]. In the present study, it is shown how and when the SW-CLC model may be applied, and the information that the model provides. As discussed along the text, some microstructural parameters, as for example, the volume of the exchange coupled soft phase can be estimated with the SW-CLC model. The Kneller-Hawig model [5] for exchange coupling can be used in combination with the SW-CLC model. Suitable nanostructures for application of SW-CLC model are also discussed with basis on the Kneller-Hawig criterion [5]. The presented model was tested for a $Nd_2(FeCo)_{14}B$ isotropic sample, but it can be easily extended for anisotropic samples, using equations discussed in previous papers [4].

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2 Theory: The Callen-Liu Cullen Modification

According the SW-CLC model, the applied field H is replaced by another field $H' = \mathrm{H} + \Psi\ \mathrm{M}$, where M is the magnetization and Ψ is the coefficient of the CLC interaction parameter [1]. The energy of magnetocrystalline anisotropy E_A can be written as $E_A = K_1 (1 - \cos^2 \varphi)$, where K_1 is the first-order anisotropy constant and φ is the angle between the easy axis and the magnetization vector. If an exchange energy term is expressed as $E = Z \cos \alpha$, then the mean field Z is given as $Z = \Psi\ M$. α is the angle between spin and applied field [1]. The angle β between magnetization and applied field is $\beta = \alpha + \varphi$. The SW-CLC model can be expressed as shown in (1), when only K_1 is relevant, using A for denoting anisotropy energy and μ as magnetic moment:

$$E = -\mu\ (H + Z) \cos \alpha - A \cos^2(\alpha - \beta) \qquad (1)$$

In the Stoner-Wohlfarth model [2], the first derivative $\partial E/\partial \alpha = 0$ gives the equations $m(h)$, and $\partial^2 E/\partial \alpha^2 = 0$ the field of irreversible rotation. h is reduced field ($h = H/H_A$) and m is the reduced magnetization ($m = M/M_s$). H_A is the anisotropy field and M_s is the magnetization of saturation. The only mathematical consequence of the exchange

energy term is a correction of the original SW curve, given according (2).

$$h_{\mathrm{CLC}} = h_{\mathrm{SW}} + (1/d)\, m \tag{2}$$

where $(1/d)$ is the interaction parameter. There are two possibilities: positive or negative $(1/d)$. It was found that several SmCo 2:17 magnets can have the entire hysteresis modeled with positive $(1/d)$ [3, 4]. However, for negative $(1/d)$, there is remanence enhancement and only part of the first quadrant has to be fitted with the model [6]. Examples of the modification given by (2) are presented in Fig. 1. It is clear that for negative $(1/d)$, there are some unphysical results, and the interpretation is that only the demagnetizing curve of the first quadrant can be fitted in this case.

3 Experimental

Hysteresis curves were measured in a vibrating sample magnetometer (VSM) EG&G Princeton Applied Research model 4500, using a superconductor coil up to 9 T. The examined magnet is a bonded magnet that follows the specification MQP B+, based on the $Nd_2Fe_{14}B$ compound with some alloying elements, especially cobalt. X-ray diffraction data was obtained with CrKα radiation. Rietveld analysis was performed with Topas Academic 4.1.

4 Results and Discussion

The necessary field for the measurements should be above the anisotropy field of the hard magnetic phase of the magnets, which is the present case for the sample $Nd_2(FeCo)_{14}B$. The soft phase is bcc alpha iron. A magnet with specification MQP B+ was modeled, see Fig. 2. The sample is a bonded magnet, made from flakes. Each flake is a group of many grains. For this situation, the epoxy volume fraction will affect the remanence proportionally, but not the coercivity. Microstructural analysis (XRD Rietveld) and transition electron microscopy (TEM) have shown that the crystals of alpha iron phase are very small, less than 5 nm, and the nanocrystalline hard phase is ~ 30 nm, see Figs. 3 and 4. As the domain wall thickness (δ) of $Nd_2Fe_{14}B$ is ~ 5 nm [7], this situation agrees well in the Kneller-Hawig [5] criterion, where the diameter of soft phase should be equal or less than twice the domain wall thickness of the hard phase. A relevant detail is that the bcc iron single-domain particle size is ~ 15 nm, and that both the exchange coupled soft phase and the hard phase need to be single domain size (SW assumption).

After fitting the first quadrant demagnetizing curve of hysteresis, the exchange coupling interaction parameter $(1/d)$ and the remanence increase $(\Delta\ J_r)$ were determined. $J_{s\,\mathrm{total}}$ is $J_{s\,\mathrm{tot}} = J_{s\,\mathrm{hard}} V_{\mathrm{hard}} + J_{s\,\mathrm{soft}} V_{\mathrm{soft}}$ and $J_r = {}^1/_2 J_{s\,\mathrm{hard}} V_{\mathrm{hard}} + J_{s\,\mathrm{soft}} V_{\mathrm{soft}}$. J is polarization, J_s is polarization of saturation, J_r is remanence, and V is volume. $V_{\mathrm{soft}} = (2\ J_r - J_{s\,\mathrm{tot}})\ /\ J_{s\,\mathrm{soft}}$ since all soft phase is exchange-coupled

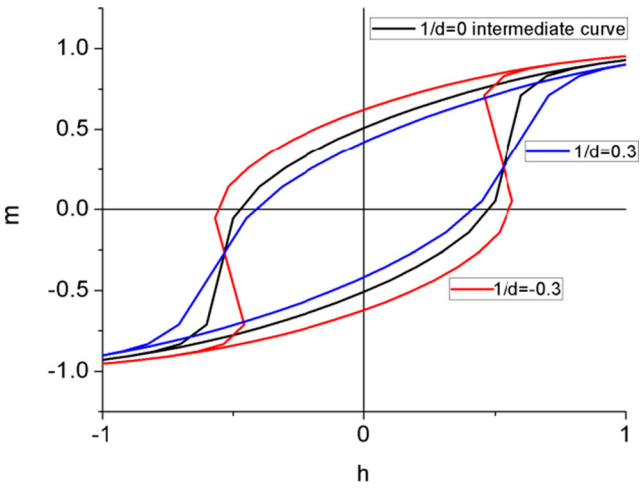

Fig. 1 SW-CLC modification for positive and negative interaction parameter (1/d)

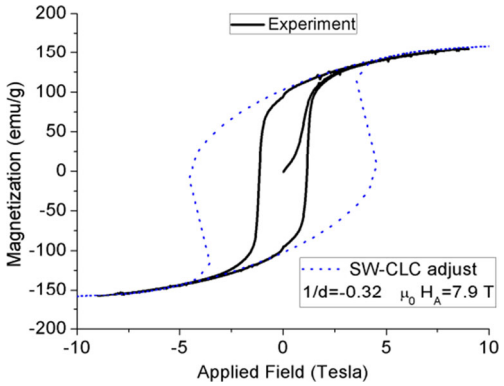

Fig. 2 Experimental curve of an exchange-coupled nanocrystalline NdFeCoB magnet modeled with the SW-CLC. Only the demagnetizing curve of the first quadrant has to be fitted. For negative (1/d), the SW-CLC model does not give any realistic prediction about coercivity

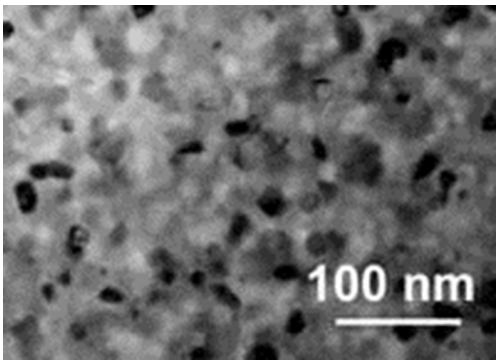

Fig. 3 TEM microstructure of the examined sample

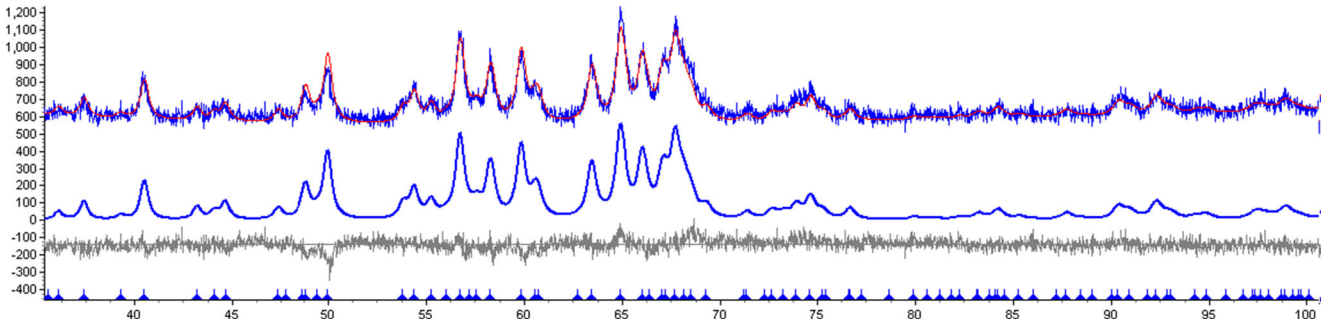

Fig. 4 Rietveld fitting of the X-ray diffraction experimental data (*above*). CrKα radiation. The *blue curve* (*below*) is the calculated intensity for $Nd_2Fe_{14}B$ structure with 30 nm and also alpha iron (bcc) nanograins < 5 mm, with ~ 3–5 % volume fraction

and also aligned parallel to the magnetization. ΔJ_r can be found with the SW-CLC fit, and as $\Delta J_r = J_{r\,CLC} - {}^1/_2 J_{s\,tot}$, then the V_{soft} can be found from the results of the SW-CLC, since $J_{s\,soft}$ is known. In other words, ΔJ_r depends only on the $(1/d)$ parameter, as seen in Fig. 1. $\Delta J_r = 0.13$ if $(1/d) = -0.32$, then $V_{soft} = 0.26\ J_{s\,tot}/\ J_{s\,soft}$. For sample density = 5.9 g/cm^3, a volume of 5.5 % for alpha iron is estimated as lower limit (because the soft phase was first assumed as completely aligned). The probable volume of soft magnetic phase is ~ 6.6 % obtained from 5.5/0.832 because the average remanence for isotropic bcc alpha iron is 0.832 J_s [7]. The modeling was performed considering only K_1 relevant. McCallum [6] previously also only used K_1 for applying SW-CLC in NdFeB exchange coupled magnets. Besides, it is not clear in the literature how cobalt affects the second-order constant K_2 in $Nd_2Fe_{14}B$ [8].

5 Suitable Nanostructures

The nanostructure of a two-phase magnetic material with exchange coupling was exhibited in Fig. 3. Reversion in the soft phase can affect the hard phase, especially if $J_{s\,soft} > K_1/\ J_{s\,hard}$. However, if the soft phase is exchange-coupled with the hard phase, this can be avoided. Kneller and Hawing [5] used the fcc lattice for discussing suitable nanostructures, resulting that the maximum possible volume fraction of soft phase is (1/8) $(\pi/3\sqrt{2}) = 9.25$ %, if the soft phase consists in spherical inclusions inside the matrix phase. Some suggested sandwich structures [5] may have the problem that the hard phase should be single-domain particle size, and the diameter of soft phase should be less than 2δ. A viable alternative comes from the double shell structure [9], presented in Fig. 5. The formula for total saturation is $J_{s\,tot} = J_{s\,hard} V_{hard} + J_{s\,soft} V_{soft}\ /\ (V_{hard} + V_{soft} + V_{para})$, where V_{para} is the volume of the paramagnetic shell that completely surrounds the soft phase (see Fig. 5). example, assuming spherical grains with radius of 25 nm, and soft phase shell layer with $2\delta = 10$ nm, plus 1 nm of paramagnetic layer, the resulting volumes is 33.5 % of hard phase, 58.4 % of exchange coupled soft phase, and 8.1 % of paramagnetic phase. Considering $J_s = 1.61$ T for $Nd_2Fe_{14}B$ and J_s bcc iron = 2.16 T, a magnet with saturation of 1.8 T could be obtained, implying in a possible high-maximum energy product (BH_{max}), because BH_{max} is $(J_{s\,tot})^2/_4$ for a magnet with perfectly oriented crystals. This nanostructure of Fig. 5 may be feasible in practice for $Nd_2Fe_{14}B$ alpha iron exchange-coupled magnets. A possibility is the vapor deposition process [10] for producing the iron layer and the paramagnetic layer.

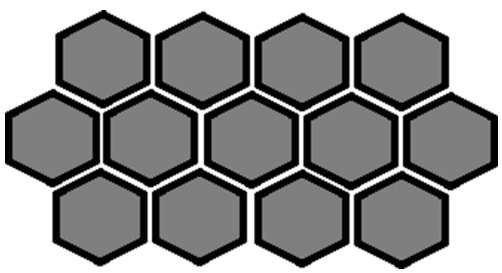

Fig. 5 The double shell model of exchange coupling. The hard phase (*gray*) is surrounded by a layer of the soft magnetic phase (*black*). There is a paramagnetic phase separating all grains (*white*). Schematic

6 Conclusions

The SW-CLC model was applied for an exchange coupled $Nd_2Fe_{14}B$ alpha iron magnet. The volume fraction of the soft magnetic phase can be estimated with the SW-CLC model. Suitable nanostructures for applying the SW-CLC model were discussed.

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