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Estimate of the anisotropy field in isotropic SmCo 2:17 magnets with the Stoner-Wohlfarth CLC model

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Abstract. The Callen-Liu-Cullen (CLC) modification of the Stoner-Wohlfarth model was found able to describe properly the hysteresis curves of isotropic Sm(CoFeCuZr)_z magnets. The SW-CLC model uses three parameters, and all of them have physical meaning. One of the parameters is related to the saturation magnetization, another to the anisotropy field, and another is $1/d$, which evaluates the interaction between grains or particles. The model was applied for several magnets, indicating an anisotropy field of 6-7 T, which is compatible with other methods for anisotropy field determination. The model also gives insight into the abnormal temperature dependence of the coercivity found in SmCo 2:17 magnets. For compositions with a low z , the parameter $1/d$ is significant. These compositions with a low z are those showing the most abnormal coercivity behavior with temperature.

1. Introduction

It is expected that randomly distributed, non-interacting, single domain particles of a high uniaxial anisotropy phase, embedded in a paramagnetic matrix, might exhibit a behavior similar to that predicted by Stoner and Wohlfarth (SW) [1-5]. In real magnets, we expect interactions between grains. The SW model was modified by Callen, Liu, and Cullen (CLC) [6] in order to treat amorphous ferromagnetic alloys. The SW result can be presented in terms of a reduced magnetization $m = M/M_s$ as a function of a reduced magnetic field $h = H/H_a$. The experimental variables M and H are scaled by the saturation magnetization M_s and the anisotropy field H_a . CLC [6] added mean-field interactions to the SW model. Hadjipanayis and Gong [7] applied the SW-CLC in melt spun NdFeBAlSi with grain size below the single domain particle size. McCallum [8,9] employed the mean-field calculation of CLC to model the first quadrant demagnetization curve of several NdFeB magnets, thereby obtaining values for M_s and H_a as well as determining the mean-field interaction parameter $1/d$. This treatment leads to a straight forward mean-field equation:

$$h_{CLC} = h_{SW} + (1/d) m \quad (1)$$

This equation says that the reduced field h_{CLC} for the CLC model [6-9] is obtained for any magnetization state m by correcting the SW reduced field h_{SW} with a term proportional to m . In this equation, $1/d$ is the mean-field interaction coefficient, where d is the ratio of anisotropy to exchange energy. Equation (1) can be applied to fit experimental curves and was used by McCallum to obtain M_s and H_a for two NdFeB magnets [8]. To fit experimental curves with the CLC model, one must vary M_s and H_a , the scale factors, as well as the mean-field interaction parameter ($1/d$). Figure 1 shows the effect of varying $1/d$, while maintaining M_s and H_a constant.

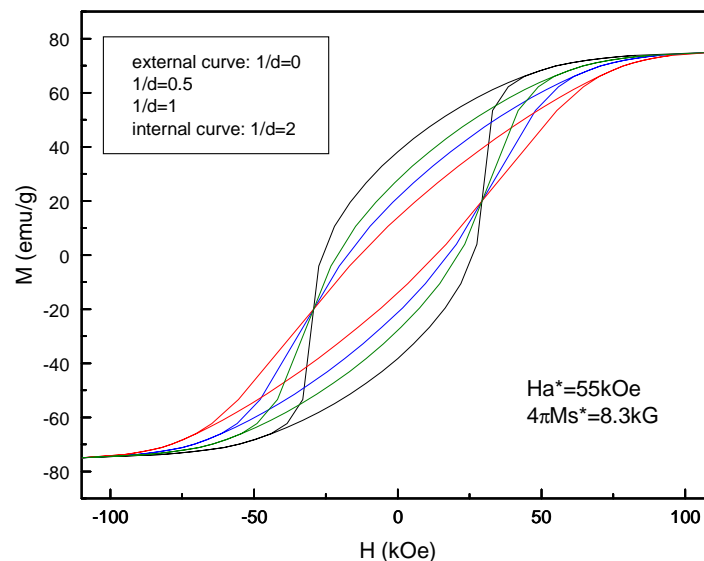


Figure 1. Hysteresis curves calculated from the SW-CLC model showing the effect of variations in the interaction parameter $1/d$. For the example shown, the anisotropy field was taken to be $H_a = 55$ kOe.

The SW model with the CLC modifications was recently used to study some compositions of SmCo 2:17 magnets. The hysteresis curves were well-described by the SW-CLC model and this indicated the presence of interactions [10]. The model also gave a very reasonable estimate of the anisotropy field. The reported anisotropy field at room temperature is around 6-7 T [11,12]. The present study again focuses on Sm(CoFeCuZr)_z magnets and we report the results for an alloy with higher z than the previous one [10]. This alloy exhibits almost perfect SW behavior, indicating coherent rotation as a reversal mechanism. But this is only possible if the grains are below the single domain particle size. In the case of the 2:17 phase, the single domain particle size is around 0.6 μm [12]. Previous TEM studies have indicated nanograin size around 100 nm [13,14] in these magnets.

Experimental

One set of samples was prepared from pure elements in an arc-furnace. Those samples have the chemical composition $\text{Sm}(\text{Co}_{\text{bal}}\text{Fe}_{0.2}\text{Cu}_{0.1}\text{Zr}_x)_8$ (bal=balance) with $x = 0.02$ and, 0.04. Those samples were encapsulated in quartz tubes under an Argon atmosphere and underwent a solubilization heat treatment at 1175°C for 4h. The samples were quenched in water, reheated up to 820°C, remaining at this temperature for 7h, and were then slow cooled ($-1^\circ\text{C}/\text{min}$) to 400°C, remaining for an additional 3 h at this temperature. Another set of samples was produced from a commercial alloy powder with composition $\text{Sm}_{0.104}\text{Co}_{0.60}\text{Fe}_{0.195}\text{Cu}_{0.072}\text{Zr}_{0.027}$ and received the same heat treatment.

The samples for magnetic measurement were parallelepipeds with dimensions 5 x 1 x 1 mm , for minimizing demagnetizing field effects. Magnetic measurements were performed at room temperature in a 9 T superconducting coil, using an EG&G Princeton Applied Research model 4500 vibrating sample magnetometer (VSM).

Results and Discussion

In Figure 2, we present the experimental room-temperature hysteresis curve (dashed line) for the isotropic magnet obtained from the commercial alloy. We note that the curve obtained from the SW-CLC model (solid line) is in good agreement with the experimental curve when we use the values of the scale factors given in the insert. The saturation magnetization $4\pi M_s = 10.5$ kG and the anisotropy field $H_a = 69$ kOe, are in good agreement with experimental determinations of these quantities. We note also the slight curvature of the initial magnetization curve. In the SW model, the initial magnetization curve is the average between 1st and 4th quadrant. This was already predicted by Stoner and Wohlfarth in 1948 [1]. Thus the initial magnetization curves serves as additional confirmation of coherent rotation as the coercivity mechanism. The same was observed in our previous study [10], where the Callen-Liu-Cullen modification was employed [6].

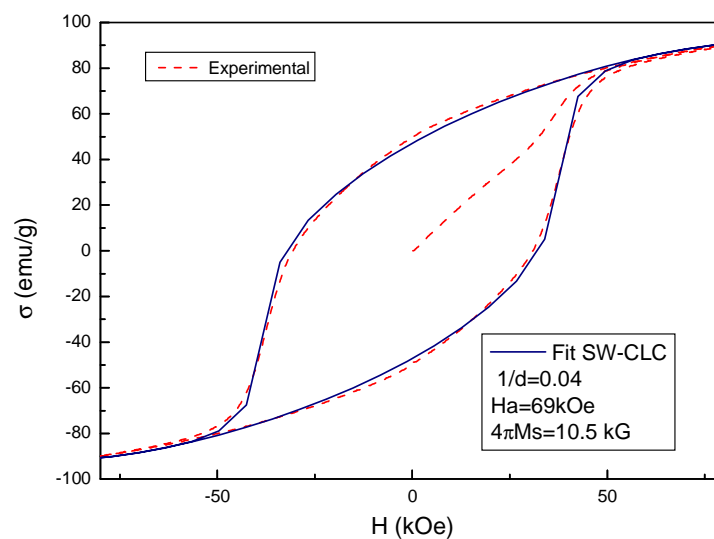


Figure 2. Experimental room-temperature hysteresis curve (dashed curve) fitted to SW-CLC model (solid curve). The small value of the interaction parameter ($1/d$) indicates nearly non-interacting Stoner-Wohlfarth behavior.

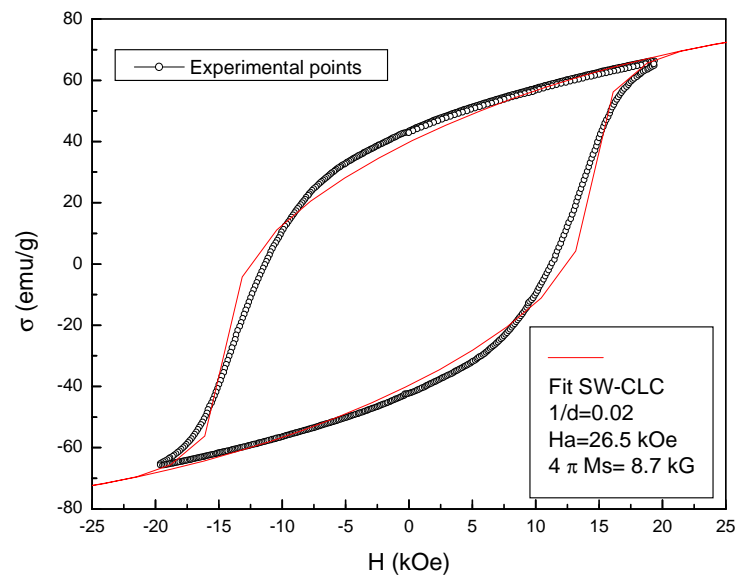


Figure 3. Experimental hysteresis curve compared to the prediction of the SW-CLC model for the sample of Figure 1, now measured at 300°C. The sample was initially magnetized by a pulsed field of 7T.

There is an additional difficulty in high temperature measurements, because the samples are small (5x1x1 mm). The samples were well protected for this high temperature measurement. However, when samples are measured at high temperatures, a Sm-depleted zone forms near surface. Sm has high vapor pressure [15] and vaporizes easily and also oxidizes easily. This Sm-depleted layer has lower coercivity and may explain the slight disagreement between experiment and the model, in Figure 3.

The single domain particle size decreases when temperature increases. However, the behavior displayed in Figure 3 shows that D_c at 300°C is above the nanocrystalline particle size of the samples, which should be in the range 100-200 nm [13]. The critical size for a single domain is given by Eq. (2), and depends on the domain wall energy given by Eq. (3). All these parameters are function of the temperature.

$$D_c = 9 / 2 \pi (\gamma / M_s^2) \quad (2)$$

$$\gamma = 4 (A K_1)^{1/2} \quad (3)$$

In these equations γ is the domain wall energy, M_s is the saturation magnetization, K_1 is the first order anisotropy constant, A is the exchange constant. Hysteresis curves for the magnets of Table I at room temperature are shown in Figure 4. The fits obtained using the model coincide almost exactly with the experimental curves.

Table I. Chemical compositions and SW-CLC parameters for the samples studied

Sample	z	1/d	H_a (kOe)	$4\pi M_s$ (kG)	Measurement temperature
$\text{Sm}_{0.104}\text{Co}_{0.60}\text{Fe}_{0.195}\text{Cu}_{0.072}\text{Zr}_{0.027}$	8.6	0.04	69.0	10.5	300 K
$\text{Sm}_{0.104}\text{Co}_{0.60}\text{Fe}_{0.195}\text{Cu}_{0.072}\text{Zr}_{0.027}$	8.6	0.02	26.5	8.7	573 K
$\text{Sm}(\text{Co}_{\text{bal}}\text{Fe}_{0.2}\text{Cu}_{0.1}\text{Zr}_{0.02})_8$	8	0.85	59.5	8.8	300 K
$\text{Sm}(\text{Co}_{\text{bal}}\text{Fe}_{0.2}\text{Cu}_{0.1}\text{Zr}_{0.04})_8$	8	1.05	57.0	8.3	300 K

Note: Zirconium replaces samarium in phases like 1:3, 5:19 or 2:7. Thus zirconium frees samarium, which leads to an increase of the $\text{Sm}(\text{Co,Cu})_5$ volume fraction.

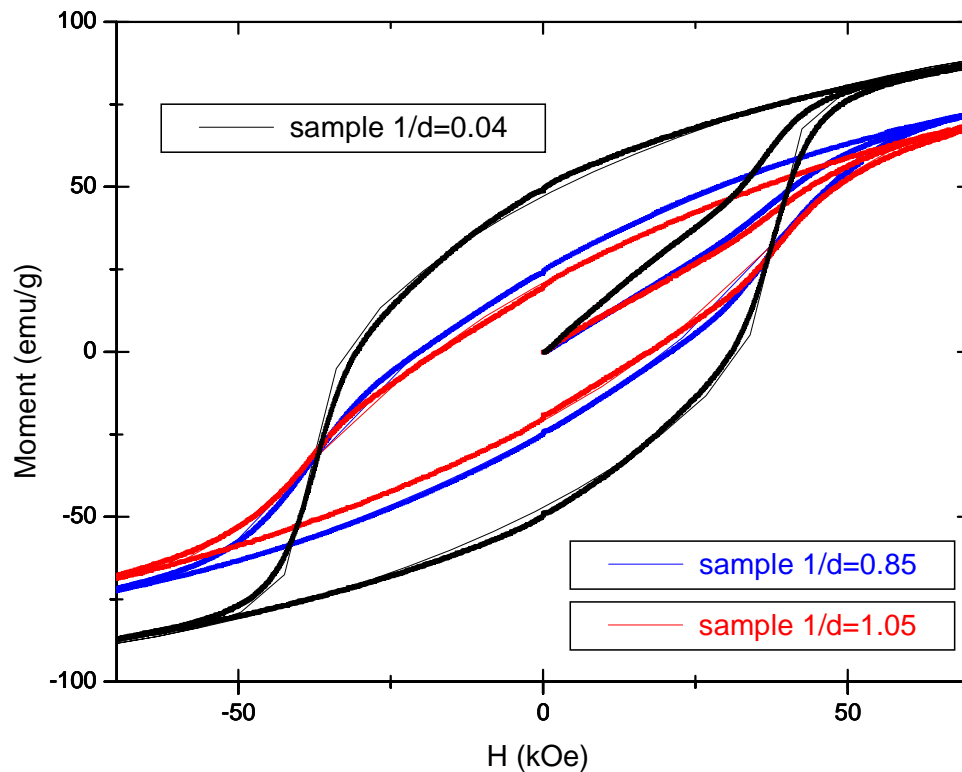


Figure 4. A comparison of some measured hysteresis curves at 300K where the SW-CLC model was applied. These magnets exhibited different values of $1/d$ (see Table I). Heavy curves are experimental. The fine solid lines represent the calculated model curves.

It is possible that the higher values of $1/d$ are associated with abnormal coercivity behavior. The abnormal coercivity behavior takes place when the $\text{Sm}(\text{Co,Cu})_5$ phase is not paramagnetic (or when there is not perfect isolation of the 2:17 grains). If z is small in $\text{Sm}(\text{CoCuFeZr})_z$, this increases the volume fraction of 1:5 phase. Increasing the volume fraction of the 1:5 phase, the Co content in 1:5 also increases, and the 1:5 is no longer paramagnetic, leading to abnormal coercivity behavior. This was experimentally verified in a previous study [13].

It is worthy of note that the model can be applied for non-isotropic magnets, since the texture is taken into account. Thus oriented magnets can also be modeled. The isotropic Stoner-Wohlfarth model is the average of the hysteresis curves of all orientations in the magnet. An oriented magnet can be modeled, for example, considering most of grains in a small angle between the applied field and the easy axis, let us say 0-15 degrees from the easy axis, as previously determined with SEM-EBSD for SmCo_5 magnets [16]. As already reported, SEM-EBSD can also be used to study crystallographical texture in 2:17 magnets [17,18].

Conclusions

As the values found with the SW-CLC are very close to experimental determinations using other methods, for example, the singular point detection method, this model can be considered another method for estimating the anisotropy field in 2:17 nanocrystalline magnets.

Our results further indicate that coherent rotation is the coercivity mechanism in these materials.

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