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Citation: [Journal of Applied Physics](#) **93**, 8140 (2003); doi: 10.1063/1.1541650

View online: <http://dx.doi.org/10.1063/1.1541650>

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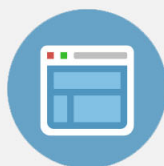
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# Magnetic interaction effects on the hard magnetic properties of ball-milled $\text{SmCo}_5 + \text{NiO}$ and $\text{SmCo}_5 + \text{CoO}$ composites: A $\Delta M$ plot study

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(Presented on 14 November 2002)

The effects of magnetic interactions on the hard magnetic properties of  $\text{SmCo}_5$  ball milled with NiO [antiferromagnetic (AFM) at room temperature] or with CoO [paramagnetic (PM) at room temperature] have been studied. The FM–AFM system exhibits improved magnetic properties (coercivity and squareness) for all compositions. The effects of magnetic interactions on the magnetic properties are analyzed in terms of classical  $\Delta M$  plots. The plots show that in both systems magnetizing-like FM–FM exchange interactions are predominant for fields  $\mu_0 H < \mu_0 H_C$ , while long-range dipolar interactions prevail for  $\mu_0 H > \mu_0 H_C$ . The different types of magnetic interactions are found to depend on the degree of  $\text{SmCo}_5$  dispersion in the AFM (NiO) or PM (CoO) matrices. Moreover, the role of the AFM appears to be to enhance both the dipolar and exchange-like interactions, although the exchange effects appear to be responsible for the improvement of the magnetic properties. © 2003 American Institute of Physics.

[DOI: 10.1063/1.1541650]

## I. INTRODUCTION

Recently, it has been demonstrated that by ball milling antiferromagnetic (AFM) and ferromagnetic (FM) materials it is possible to produce a microstructure of FM particles embedded in an AFM matrix. This microstructure favors exchange interactions between FM and AFM phases, which are found to induce coercivity,  $\mu_0 H_C$ , and squareness,  $M_R/M_S$ , enhancements.<sup>1,2</sup>

In FM materials, a procedure, known as  $\Delta M$  plots, was developed to evaluate the strength of the different types of magnetic interactions.<sup>3</sup> This technique differentiates between two types of remanence curves: (i) the isothermal remanent magnetization  $M_r(\mu_0 H)$ , which is obtained by measuring the remanent magnetization after progressively magnetizing a fully demagnetized sample, using increasingly larger positive magnetic fields; and (ii) the demagnetizing remanent magnetization  $M_d(\mu_0 H)$ , which is obtained by progressively demagnetizing a fully saturated sample in increasingly negative magnetic fields. Usually, both  $M_r(\mu_0 H)$  and  $M_d(\mu_0 H)$  are normalized to the value obtained after saturating the sample (i.e.,  $M_r(\infty)$ ), so that  $m_r(\mu_0 H) = M_r(\mu_0 H)/M_r(\infty)$  and  $m_d(\mu_0 H) = M_d(\mu_0 H)/M_d(\infty)$ . From the normalized remanence curves  $\Delta M$  is defined as

$$\Delta M(\mu_0 H) = m_r(\mu_0 H) - (1 - 2m_d(\mu_0 H)). \quad (1)$$

Positive  $\Delta M$  values are interpreted as magnetizing short-range exchange interactions, while negative  $\Delta M$  values cor-

respond to demagnetizing long-range dipolar interactions. However, note that the effects of FM–AFM exchange interactions on  $\Delta M$  plots have not been studied systematically so far.<sup>4</sup>

In this work, a comparative study of the magnetic properties of  $\text{SmCo}_5$  ball milled with NiO and CoO is presented. Since the Néel temperature of CoO is below room temperature, only in  $\text{SmCo}_5 + \text{NiO}$  composites some effects due to FM–AFM exchange interactions are expected. The results indicate that the observed enhancements of  $\mu_0 H_C$  and  $M_R/M_S$  in the FM–AFM system are related to the exchange-like interactions induced by the AFM–FM coupling.

## II. EXPERIMENTAL PROCEDURE

Ball milling of  $\text{SmCo}_5$  (99%,  $<500\mu\text{m}$ ) alone and together with NiO (99%,  $<44\mu\text{m}$ ,  $T_N=590\text{K}$ ) and CoO (99%,  $<44\mu\text{m}$ ,  $T_N=290\text{K}$ ) powders, in the weight ratios of 4:1, 3:2, 2:3, and 1:3, was carried out in a planetary mill, under argon atmosphere, at 500 rpm, using agate vials and balls, in a ball-to-powder weight ratio of 2:1. The milling time used in the present study was 16 h. Morphological and structural characterizations were performed by scanning electron microscopy (SEM) and x-ray diffraction (XRD), where microstructural parameters were evaluated using a full pattern fitting procedure (Rietveld method).<sup>5</sup> Magnetic hysteresis loops and remanence curves were carried out on tightly packed isotropic powders by means of an extracting magnetometer, with a maximum applied field of 20 T.

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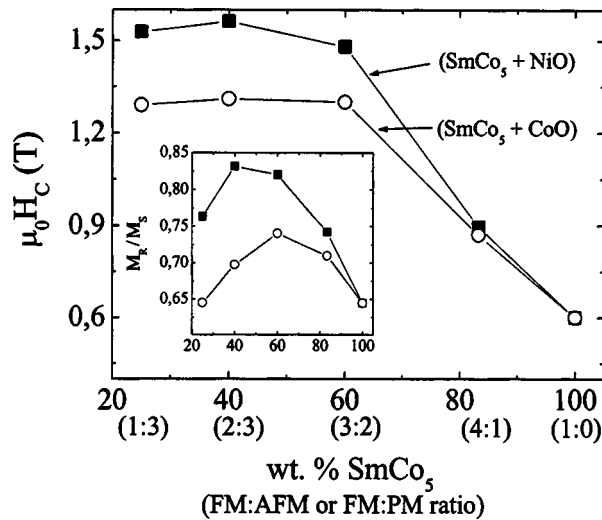


FIG. 1. Dependence of the coercivity  $\mu_0 H_C$  on the  $\text{SmCo}_5$  weight percentage [i.e., FM:AFM (or PM) ratio], for  $\text{SmCo}_5$  milled with NiO (-■-) or with CoO (-○-) for 16 h. In the inset, dependence of the squareness ratio  $M_R/M_S$  on  $\text{SmCo}_5$  weight percentage for  $\text{SmCo}_5$  milled with NiO (-■-) or with CoO (-○-) for 16 h. The lines are guides to the eye.

### III. RESULTS AND DISCUSSION

The SEM images of  $\text{SmCo}_5$  powders milled together with NiO or CoO reveal that, during the milling, both components tend to solder together, forming aggregates of a few  $\mu\text{m}$  in size, in which  $\text{SmCo}_5$  grains become embedded in the AFM (NiO) or paramagnetic (PM) (CoO) matrices.<sup>2,6</sup> Moreover, the analysis of the XRD spectra shows that for a fixed milling time (e.g., 16 h), the milling is more aggressive (i.e., more structural disorder) for mixtures with larger FM content.<sup>2,6</sup> For example,  $\text{SmCo}_5$  crystallite size,  $\langle D \rangle_{\text{SmCo}_5}$ , reduces progressively with the  $\text{SmCo}_5$  content, from 15.7 nm in  $\text{SmCo}_5(1):\text{CoO}(3)$  to 5.8 nm in  $\text{SmCo}_5$  milled alone. In addition, for all FM percentages, the difference of  $\langle D \rangle_{\text{SmCo}_5}$  between the  $\text{SmCo}_5 + \text{NiO}$  and the  $\text{SmCo}_5 + \text{CoO}$  systems are found to be small (i.e., less than 6%). This reveals that, in fact, the microstructure developed during ball milling is very similar in both cases.

Shown in Fig. 1, is the dependence of  $\mu_0 H_C$  with the FM:AFM (or PM) ratio (i.e., FM percentage) for  $\text{SmCo}_5 + \text{NiO}$  and  $\text{SmCo}_5 + \text{CoO}$ . As can be seen in the figure, both systems exhibit a similar behavior, i.e., there is a small maximum for intermediate FM percentages and a clear decrease of coercivity for increasing FM contents. However,  $\mu_0 H_C$  is larger for  $\text{SmCo}_5 + \text{NiO}$  than for  $\text{SmCo}_5 + \text{CoO}$ , especially for small FM contents. A similar behavior is observed for the squareness ratio,  $M_R/M_S$  (see Fig. 1 inset). These effects are the interplay between structural and magnetic effects (e.g., particle size, crystallite size, and packing density).<sup>7</sup> For example, diluting the FM in the AFM or PM matrix results in the reduction of the milling-induced structural disorder as well as the reduction of the dipolar and FM-FM exchange interactions.<sup>2,6</sup> However, due to the antiferromagnetic character of NiO, AFM-FM exchange interactions are also induced, which lead to the observed differences between  $\text{SmCo}_5 + \text{NiO}$  and  $\text{SmCo}_5 + \text{CoO}$ .<sup>2,6</sup>

Figure 2 shows an enlargement of the  $m_r(\mu_0 H)$  and

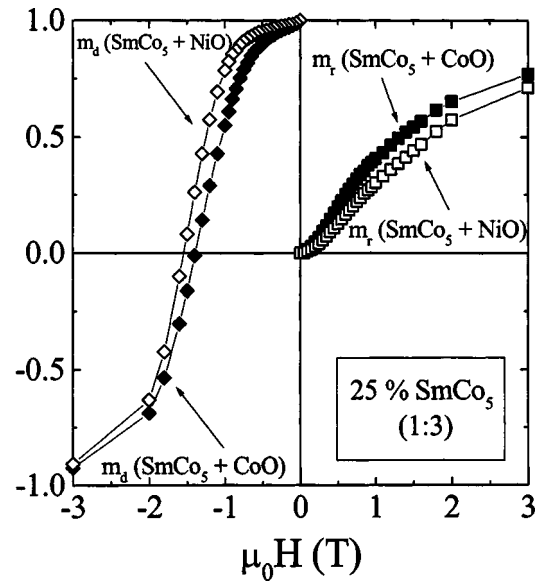


FIG. 2. Enlargements of the isothermal and demagnetizing remanence curves  $m_r(\mu_0 H)$  and  $m_d(\mu_0 H)$  in the field range  $(-3,3)$  T, for  $\text{SmCo}_5$  milled for 16 h with NiO (open symbols) or with CoO (filled symbols), in the FM:AFM (or PM) 1:3 weight ratio (i.e., 25% of  $\text{SmCo}_5$ ). The lines are guides to the eye.

$m_d(\mu_0 H)$  curves of  $\text{SmCo}_5(1):\text{NiO}(3)$  and  $\text{SmCo}_5(1):\text{CoO}(3)$  composites. Note that the curves were measured up to 20 T. Although both systems have very similar morphological and structural properties,  $m_r(\mu_0 H)$  and  $m_d(\mu_0 H)$  are found to be somewhat different. Namely, as can be seen in the figure,  $m_r(\mu_0 H)$  increases more rapidly with field in  $\text{SmCo}_5 + \text{CoO}$  than in  $\text{SmCo}_5 + \text{NiO}$ . This means that after applying a certain positive magnetic field the overall magnetization retained in the direction of the field, once the field is removed, is higher for FM:PM than for FM:AFM composites. Moreover,  $m_d(\mu_0 H)$  decreases more slowly in  $\text{SmCo}_5 + \text{NiO}$  than in  $\text{SmCo}_5 + \text{CoO}$ , i.e., the latter system is easier to demagnetize. A similar trend has also been observed for the other FM:AFM (or PM) weight ratios and even for particles obtained after milling for different times (e.g., 8 or 32 h). Actually, FM-AFM exchange interactions, which influence the magnetization reversal of  $\text{SmCo}_5 + \text{NiO}$  composites, are probably responsible for these differences. For instance, the AFM spins could exert, during magnetization and demagnetization, a microscopic torque on the spins of the FM, which would make them difficult to reverse.

The  $\Delta M$  plots corresponding to  $\text{SmCo}_5(1):\text{NiO}(3)$  and  $\text{SmCo}_5(1):\text{CoO}(3)$  are shown in Fig. 3. It can be seen that both types of interactions (e.g., dipolar and exchange) are present in both systems. Positive  $\Delta M$  values (exchange interactions) are obtained when the applied field is lower than  $\mu_0 H_C$ , while dipolar interactions are predominant for  $\mu_0 H > \mu_0 H_C$ . Similar  $\Delta M$  plots are obtained for the other FM:AFM (or PM) weight ratios, except for as-milled  $\text{SmCo}_5$  powders alone, where the positive peak due to FM exchange interactions is not observed. This is because demagnetizing (i.e., dipolar) interactions predominate in FM particles milled alone, probably due to the large stray fields that each FM particle creates on its neighboring particles. It can also be

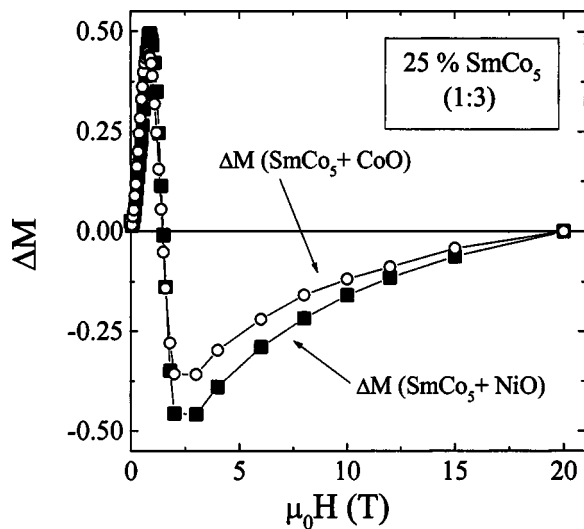


FIG. 3.  $\Delta M$  plots for  $\text{SmCo}_5$  milled for 16 h with NiO (-■-) and with CoO (-○-) in the 1:3 weight ratio (i.e., 25% of  $\text{SmCo}_5$ ). The lines are guides to the eye.

seen in Fig. 3 that the area as well as the height of the positive and negative peaks of the  $\Delta M$  plots are larger in  $\text{SmCo}_5 + \text{NiO}$  than in  $\text{SmCo}_5 + \text{CoO}$ . Analogous observations are made when comparing the  $\Delta M$  plots of other FM:AFM (or PM) percentages. Thus, it seems that the net effect of FM–AFM exchange interactions on classical  $\Delta M$  plots is to increase both the magnetizing-like and the demagnetizing-like interactions.

A single parameter commonly used to quantify the strength of magnetic interactions, i.e., the so-called interaction field  $\mu_0 H_{\text{INT}}$  has also been calculated. The interaction field is defined as<sup>8</sup>

$$\mu_0 H_{\text{INT}} = \frac{1}{2}(\mu_0 H'_r - \mu_0 H'_d), \quad (2)$$

where  $\mu_0 H'_r$  and  $\mu_0 H'_d$  correspond to the position of the maximum of the field derivative of the  $m_r$  and  $m_d$  curves, respectively.

The values of  $\mu_0 H_{\text{INT}}$  for the different systems are plotted in Fig. 4. This parameter exhibits a small maximum for intermediate  $\text{SmCo}_5$  percentages. A negative value of  $\mu_0 H_{\text{INT}}$  is obtained in  $\text{SmCo}_5$  milled alone, where dipolar interactions are found to predominate. Moreover, larger  $\mu_0 H_{\text{INT}}$  values are obtained in  $\text{SmCo}_5 + \text{NiO}$  (i.e., FM + AFM) composites. Thus, although AFM–FM coupling appears to induce an increase of both dipolar-like and exchange-like interactions, the trend of the interaction field indicates that the relative importance of the exchange interactions increases for the AFM–FM system. Thus, exchange-like interactions appear to be responsible for the improvement of the hard magnetic properties of the composites. Note that when comparing the normalized loops of  $\text{SmCo}_5 + \text{NiO}$  and  $\text{SmCo}_5 + \text{CoO}$  (inset of Fig. 4) the former has a squarer loop, which is also an indication of enhanced exchange coupling.

#### IV. CONCLUSIONS

A systematic study of the different types of magnetic interactions present in  $\text{SmCo}_5 + \text{NiO}$  (AFM) and  $\text{SmCo}_5$

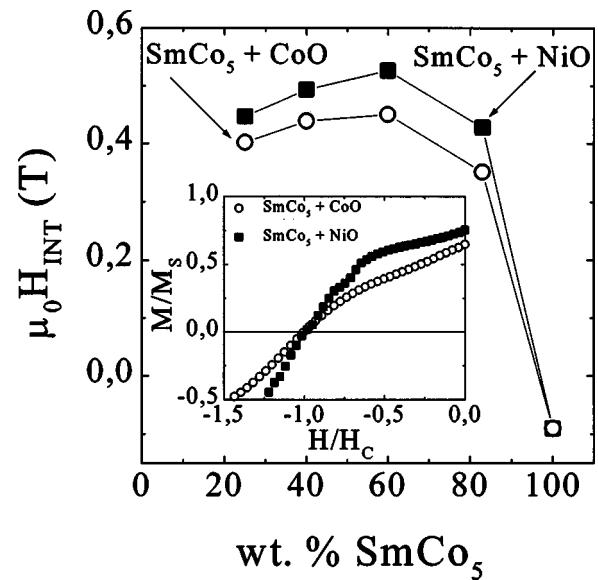


FIG. 4. Dependence of the interaction field  $\mu_0 H_{\text{INT}}$  on the  $\text{SmCo}_5$  weight percentage for  $\text{SmCo}_5$  milled with NiO (-■-) or with CoO (-○-) for 16 h. The inset shows the demagnetizing branch of the normalized ( $M/M_S$  vs.  $H/H_C$ ) hysteresis loops for  $\text{SmCo}_5$  milled for 16 h with NiO (-■-) and with CoO (-○-) in the 2:3 weight ratio (i.e., 40% of  $\text{SmCo}_5$ ). The lines are guides to the eye.

+ CoO (PM) composites, synthesized by ball milling and their effects on the magnetic properties, has been carried out by means of classical  $\Delta M$  plots. It has been shown that FM:AFM exchange coupling generates an increase of the interactions, both magnetizing and demagnetizing-like, in the system. However, the enhanced coercivity,  $\mu_0 H_C$  and remanence  $M_R/M_S$  observed for  $\text{SmCo}_5 + \text{NiO}$  (AFM) appear to be controlled by the increase of the exchange interactions.

#### ACKNOWLEDGMENTS

The authors wish to thank L. Lutterotti for providing the Rietveld XRD refinement program. J.S. thanks the DGU for its financial support. Partial support from CICYT (Grant No. MAT2001-2555), DGR (Grant No. 2001SGR00189), and the Human Potential Program (Grant No. HPRI-1999-CT00030) are also acknowledged.

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