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# Switching field of partially exchange-coupled particles

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## Abstract

The magnetization reversal of partially exchange-coupled particles is studied in detail. The starting point is the observation of a complicated phenomenology in the irreversible susceptibility and FORC distribution functions of Ba hexaferrite samples obtained by means of different sintering conditions. Several peaks in the first-order reversal curve (FORC) distribution functions were identified and associated with clusters with different number of particles. The switching fields of these clusters were related to an effective anisotropy constant $K_{eff}$ that depends on the number of particles in the cluster. $K_{eff}$ is linked to the exchange-coupled volume between two neighboring particles and as a weighted mean between the anisotropy constants of the coupled and uncoupled volumes. By using the modified Brown's equation $\alpha_{ex} = 0.322$ is obtained.

In order to interpret these results, the switching field of a two-particle system with partial exchange coupling is studied. It is assumed that the spins reorientation across the contact plane between the particles is like a Bloch wall. The energy of the system is written in terms of the fraction of volume affected by exchange coupling and the switching fields for both particles are calculated. At small interaction volume fraction each particle inverts its magnetization independently from the other. As the fraction of exchange-coupled volume increases, cooperative effects appear and the two particles invert their magnetization in a cooperative way.

The proposed model allows to interpret for the first time the empirical factor $\alpha_{ex}$ in terms of physical arguments and also explain the details observed in the FORC distribution function.

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## 1. Introduction

Interparticle interactions such as dipolar and exchange interactions are key factors that affect the magnetic properties of granular or particulate

systems, especially in the nanosize scale. Single-phase hard magnetic systems consisting of nanosize isotropically distributed uncoupled particles have high coercivity $H_c$ and remanence $M_r$ that is half the saturation magnetization $M_s$. When they are strongly coupled the coercivity is lower but the remanence is enhanced above $M_s/2$ [1,2]. Such a behavior was studied based on micromagnetic modeling in order to reveal how exchange interactions and grain structure affect the magnetic properties [3,4].

Interactions also affect time effects in magnetic phenomena, such as magnetic viscosity and the magnetic storage of information. Magnetic viscosity degrades the magnetization as time passes and the magnetic storage of information depends on the time scale of the process that produces a given magnetization change: recording must be done very fast and the result must be stable for very long times. The density of recorded information on magnetic media has steadily increased in recent years and now the physics underlying the limit of density of recorded bits is of crucial importance. For a good signal-to-noise ratio, every bit should have many switching units. At today bit density in a storage media, the magnetic units are of the order of 10–100 nm and exchange interactions between them are becoming more and more important [5]. In this scenario, what is the limit of 'weakly' coupled ferromagnetic particles?

Strong exchange interaction between particles promotes magnetization reversal in a cooperative manner, while with a weak exchange interaction the magnetization reversal proceeds in a non-cooperative way. To our knowledge, the limit between the two regimes is not defined yet.

The objective of this work is to study how the magnetization reversal (the switching field) is affected by exchange interactions between nano-sized particles, the transition from non-cooperative to cooperative magnetization reversal as a function of the extent of exchange coupling and how the remanence is affected.

## 2. Experimental results

In previous works [6,7] it was observed that the irreversible magnetic susceptibility $\chi_{irr}$ of particu-

late Ba hexaferrite (phase M) samples is strongly dependent on the degree of sintering. Two samples are considered: sample I corresponds to a nanostructured (50 nm crystal size) sample obtained by milling the elemental oxides ($Fe_2O_3$ and $BaCO_3$) in stoichiometric proportions plus an addition of 20 wt% Fe and then performing a heat treatment at 1000 °C in air for 1 h. The resulting powder was then compacted into 1–2 mm thick platelets. Highly crystalline phase M and approximately 20% of hematite are present in this sample. Sample II has the same composition as sample I, but the powder was first compacted and then heat treated during 8 h at 1000 °C. The resulting mean crystal size is 60 nm.

Figs. 1 and 2 show the irreversible susceptibility $\chi_{irr}$ for the two mentioned samples. Sample I (Fig. 1) has two well-defined peaks in $\chi_{irr}$ and sample II shows an almost constant value of $\chi_{irr}$ for fields between 2 and 4 kOe (Fig. 2). The observed details have to be related to the microstructure in each sample. We can interpret $\chi_{irr}$ as due to the distribution of switching fields [8] with a richness of details that are hidden in $\chi_{irr}$.

There is a technique that can reveal the details of the switching fields distributions—this is the calculation of the first-order reversal curve distribution function (FORC distribution function). The FORC distribution function coincides with the Preisach distribution function if some general
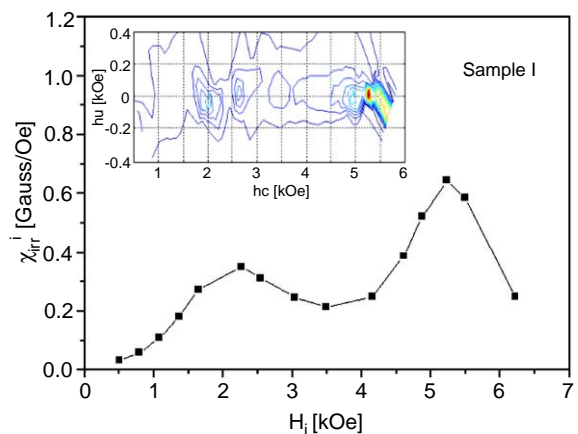


Fig. 1. Irreversible susceptibility $\chi_{irr}$ as a function of the internal field $H_i$ for sample I. The inset shows the FORC distribution function for this sample (Ref. [7]).
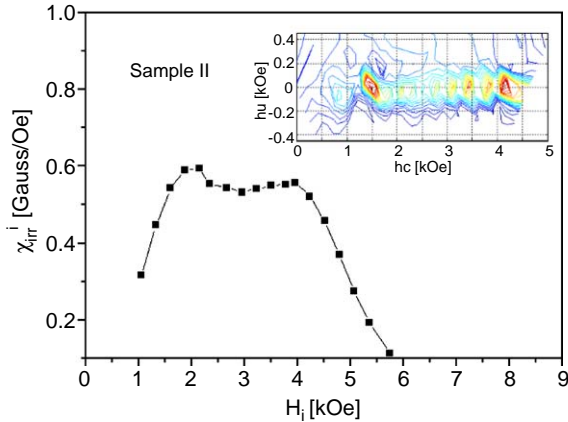
Fig. 2. Irreversible susceptibility $\chi_{irr}$ as a function of the internal field $H_i$ for sample II. The inset shows the FORC distribution function for this sample (Ref. [7]).

symmetry conditions are fulfilled [9–11]. The FORC distribution function describes the magnetic behavior of a system of particles considering both the switching field $h_c$ and the internal field $h_u$ acting on them. The whole system is described by the distribution function $f(h_c, h_u)$ which tells us how many elementary loops have switching fields between $h_c$ and $h_c + dh_c$ and interaction fields between $h_u$ and $h_u + dh_u$. The insets in Figs. 1 and 2 show the FORC distribution functions for samples I and II, respectively, calculated by following the outline given in Ref. [7]. The obtained distribution functions are more complex than usual FORC distributions reported in the literature for single magnetic-phase systems. For sample I (inset Fig. 1) the distribution has a narrow and high peak at 5.3 kOe and a number of small and overlapping peaks down to 2.0 kOe. Sample II (inset of Fig. 2) has a distribution function with a succession of almost equally spaced distinct peaks of roughly the same intensity, centered at different $h_c$ from 1.5 to 4.2 kOe with standard deviations not exceeding 250 Oe. In both cases the long-range interaction field has a zero mean value with standard deviation not greater than 300 Oe. This is a sign that no large inhomogeneities exist in these samples. A further confirmation of this is that the $\Delta M$ plots do not show large departures from the non-interaction

behavior: the samples are macroscopically homogeneous and no long-range dipolar interactions are present.

The multiple peaks observed in the FORC distribution functions suggest that they represent different groups of particle clusters that invert their magnetization at the fields $h_c$ of the maxima. These clusters have appeared by the sintering among different number of particles. The hematite phase surrounds clusters of phase M with different number of particles, preventing them from welding with each other during the sintering process. Sample I has more pores than sample II due to the fabrication process, and more clusters with few particles are expected in sample I.

Using the modified Browns equation [12] to describe the switching field due to an effective anisotropy constant $K_{eff}$, we may write

$$K_{eff} = K_1 \alpha_\phi \alpha_K \alpha_{ex} = \frac{M_s}{2}(H_c^{exp} + N_{eff} M_s), \qquad (1)$$

where $N_{eff}$ is the local demagnetizing factor [13], $\alpha_\phi = \frac{1}{2}$ due to random orientation of particles, $\alpha_K$ takes into account reduction in anisotropy near internal surfaces and $\alpha_{ex}$ describes the effect of exchange coupling between neighboring particles.

The next step is to assign the observed peaks in the FORC distribution function to particles that interact with different number of neighbors. As the exchange length in this system ($l_K = (A/K)^{1/2} = 5$ nm) is lesser than the particle size $D$ we deduce that there is partial exchange coupling between the particles of phase M inside each cluster and the volume fraction of particles affected by exchange interaction depends on the number of particles per cluster. Fig. 3 shows the result of such assignment, where $n$ is the number of neighbors interacting with a given particle, and varies from $n = 0$ (non-interacting particles) to 11.

We write $K_{eff}$ as a weighted mean between coupled ($K_{ex}$) and uncoupled ($K_{in}$) regions, as

$$K_{eff}(n) = K_{in}(1 - \beta n) + K_{ex}\, \beta\, n$$
$$= K_{in} + (K_{ex} - K_{in})\,\beta\, n, \qquad (2)$$

being $\beta$ the volume fraction per particle affected by exchange interaction. The linear relationship between $K_{eff}$ and number of neighbors is evident,
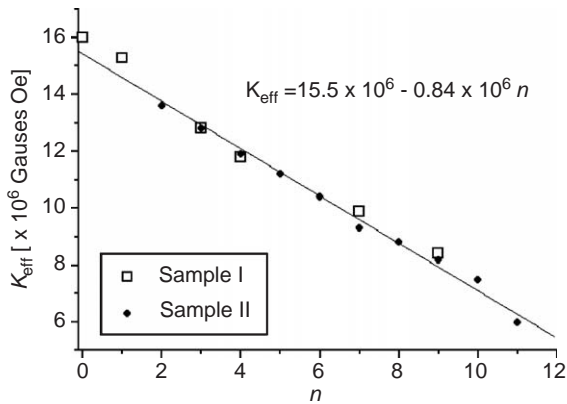
Fig. 3. Effective anisotropy constant $K_{eff}$ as a function of the number of neighbors that interact with a particle, $n$.

implying that $K_{eff}$ is a linear function of the coupled volume.

From the plot we obtain $K_{in} = K_1/2$; $\alpha_K = 1$ in the uncoupled regions and $K_{ex} = 5 \times 10^6$ GOe; $\alpha_{ex} = 0.32$ in the coupled regions.

## 3. Theoretical model

Considering the $\alpha$ factors are empirical, the next step is to account for the experimental results. With this purpose we study the magnetization reversal of a two-particle system with partial exchange coupling. We use the expression 'partial exchange coupling' not referring to the strength of the coupling but to the volume extent: 'fully coupled' means that all the volume of the particles is involved in the interaction and 'partially coupled' means that only a fraction of the volume is involved. We assume that the particles are discs and that the exchange coupling occurs through one of their plane faces, extending up to $5\, l_K$ into each particle. We assume that the spin reorientation across the contact plane is similar to that of a Bloch wall [9].

We calculated the total energy $E_T$ of the system (anisotropy and exchange interaction energies and potential energy under an applied magnetic field $H$) per unit area of contact plane at 0 K. We obtained the equilibrium angular positions $\theta_1$ and $\theta_2$ (corresponding to particles 1 and 2, respectively) for constant $H$. We identified the switching

fields $H_{sw}$ of each particle as a function of interacting volume fraction $\beta$.

All this information can be condensed in the switching paths, defined as the paths followed by $\theta_1$ and $\theta_2$ when $H = H_{sw}$ and the particles move from one stable position to another. The studied cases are labeled as $(\omega_1, \omega_2)$, where $\omega_1$ gives the orientation of the anisotropy axis of particle 1 and $\omega_2$ of particle 2 relative to the applied magnetic field $H$. The switching paths for $(\omega_1 = 0, \omega_2 = 30°)$ are shown in Fig. 4. The cases (0.30) and (0.60) have cooperative switching for $\beta > 0.66$ while the cases (20.45) and (20.60) show this behavior for $\beta > 0.3$. This is consistent with the fact that well-aligned particles have higher switching fields when they do not interact.

With this information we may calculate the demagnetizing curves as function of $\beta$. Some observations can be made

1. Fig. 5 shows that the switching behavior of cases (0.30) and (0.60) are almost identical. The switching field for particle 2 increases steadily with $\beta$ for both the non-cooperative and cooperative switching mode. The switching field of particle 1 first decreases when $\beta$ increases up to 0.66 (when the switching mode is non-cooperative) and then it increases with $\beta$ when the switching mode is cooperative. For the
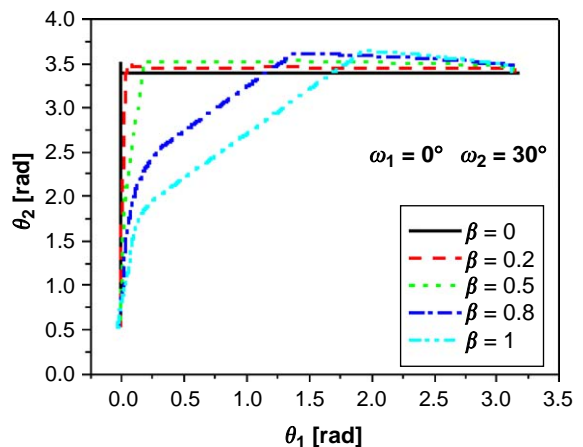


Fig. 4. Magnetic moments switching paths calculated for different $\beta$ when the energy barrier is zero for the case (0.30). $\theta_1$ ($\theta_2$) is the angle that forms particle 1 (2) with its anisotropy axis.

non-cooperative mode it is particle 2 that first inverts the magnetization and particle 1 follows independently, while for the cooperative mode the two particles invert their magnetization in a coordinated way. The coercivity of the two-particle system is equal to the switching field of particle 1.

2. For the (20.45) case the switching field is like the previous ones but for (20.60) if $\beta > 0.6$ it is particle 1 that first initiates the switching instead of particle 2.

3. The remanence of the two-particle system slightly increases with $\beta$, i.e., remanence enhancement is demonstrated and is due to an additional contribution from the coupled zone. Remanence enhancement is higher when one particle is well aligned with the applied field.

## 4. Comparison with the experiments: discussion

The studied model system is very simplified and we cannot expect it will reproduce in detail the observed behavior in Ba hexaferrite, but it is striking that the observed experimental result of $K_{eff}$ depending linearly with the fraction of exchange-coupled volume. Fig. 3 shows the same linear dependence found in the theoretical results of Fig. 5. They have the same limit in $\beta$ found for cooperative and non-cooperative switching mode ($\beta = 0.66$) and they differ at the value for $\beta = 0$: the experimental results give $K_{eff} = K_1/2$ while the calculations give $K_{eff} = K_1$ ($H_{sw}(\beta = 1) = H_K$). This is because we have not modeled the isotropic distribution in the orientation of anisotropy axis. The interpretation of the results presented in Fig. 3 include a factor $\alpha_\phi = 0.5$ that accounts for the isotropic distribution of the anisotropy axis.

The introduction of $K_{ex}$ as a mean value of $K_1$ in the exchange-coupled volume and the definition of $K_{eff}$ as a weighted mean between uncoupled and coupled regions mean that we are taking as the anisotropy and exchange energies of the particle the weighted anisotropy and exchange mean energies between the two regions. In this sense, $\alpha_{ex}$ represents the fraction of the anisotropy and exchange energies that is conserved in the exchange-coupled region.
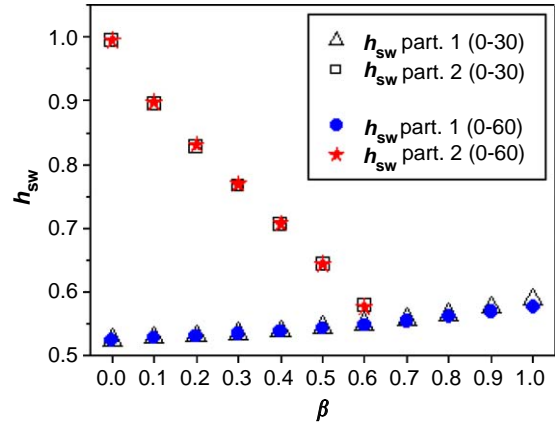


Fig. 5. Switching fields $h_{sw} = H_{sw}/H_K$ as a function of the interacting volume fraction $\beta$ for particles 1 and 2.

In Fig. 5 $h_{sw}$ is the ratio of the switching field to the anisotropy field, and this number is also $k_{eff} = K_{eff}/K_1$, being $K_{eff}$ the anisotropy constant related to $H_{sw} = 2K_{eff}/M_s$. In this figure $k_{eff}$ is linearly related to $\beta$ through the relationship $k_{eff} = 0.954 - 0.632\beta$. This relationship is just Eq. (2) divided by $K_1$; the slope is $(K_{ex} - K_{in})/K_1$ and the $y$-intercept is $K_{in}/K_1$. In our model there is not a parameter $\alpha_\phi$ involved and $K_{ex} = K_1 \alpha_{ex}$ which gives $\alpha_{ex} = 0.322$, exactly the same value found from the experimental results. Then, it seems that the deterioration of the structure produced by the exchange interaction does not depend on the orientation of the particle, at least when the angle between the particle and the field is between 0 and $30°$, but more on the angle between interacting particles. It is for this reason that Figs. 3 and 5 have the same slope but different $y$-intercept. This is the first time that such a detailed interpretation of the empirical factor $\alpha_{ex}$ has been given.

This model has also shed some light on a detail observed in the FORC distribution functions shown in the insets of Figs. 1 and 2: the peaks with lowest $h_c$ are wider and higher than the others, except for the highest peak observed at high fields. In Fig. 5, we see that particle 2 has a much lower switching field than particle 1 and when the exchange volume increases $h_{sw}$ also increases slowly, meeting the switching field of particle 1 when the cooperative mechanism for magnetization reversal begins. These are all the

particles that give rise to a much broader and higher peak in the FORC distribution function, because for every particle that behaves like particle 1 there is at least one particle that will behave as particle 2.

In conclusion, the magnetization reversal of partially exchange-coupled particles depends on the degree of coupling: for low exchange-coupled volume $\beta$ the mechanism of reversal is the non-cooperative inversion of particles while it is seen that well-aligned particles switch cooperatively for $\beta > 0.66$. This limit is lower if the particles are not well aligned with the applied field. The variation of the switching field with $\beta$ gives $\alpha_{ex} = 0.32$, in excellent agreement with the experimental results.

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