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First order reversal curves analysis of the temperature effect on magnetic interactions in barium ferrite with La–Co addition

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ABSTRACT

First order reversal curves (FORCs) distributions are a powerful tool for investigating hysteresis and interactions in magnetic systems and have been widely applied. La–Co substitution in barium hexaferrites has also been extensively studied. The most effective substitution to improve the magnetic properties (coercive field and energy product) is given by $x = y = 0.2$ in the formula $Ba_{1-x}La_xFe_{12-y}Co_yO_{19}$. In this work, this stoichiometry is initially used to obtain a state where more than one phase is present. The magnetic behavior as a function of temperature was studied in order to have an insight into the magnetic interactions that originate a decrease in the magnetic performance of Ba hexaferrite magnets. The sample was structurally characterized by X-ray diffraction (XRD) and magnetically studied in a SQUID magnetometer. FORC distributions were used to study the dependence of the magnetic interactions with the temperature. FORC diagrams performed on the sample at different temperatures exhibit similar characteristics, such as the spread in the h_c–h_u plane and a spread out of the h_c-axes. These features are interpreted in terms of exchange-interacting particles and dipolar interactions, respectively. As the temperature decreases, stronger interactions are noticed among hard and soft phases.

Magnetic ferrites are a commercial magnet material widely employed in technological applications because of their low-cost production as well as their good performance. In order to improve their magnetic performance, structural and magnetic properties have been studied and modeled. In general, these studies are devoted to understanding and/or explaining the coercive field, anisotropy field or the role of the interactions in the magnetization process [1–7]. Also doping and substitution with Ti [8], Al [9], La, Co among other elements have been tested [3,10]. The usually observed temperature dependence of the magnetic parameters plays an important role in the final products because these materials are used in many applications which cover a wide temperature range. When the temperature varies the magnetic interactions between the grains of sample also vary and different magnetic behaviors are noticeable.

When the stoichiometry is not precise or the fabrication process is not adequate the ferritic phase is accompanied by other phases that promote magnetic interactions which result in a decrease of the magnetic performance of the magnets.

In recent years, first order reversal curves (FORCs) became an efficient tool to investigate and describe the magnetic hysteretic behavior of systems of particles and their interactions by means of a FORC diagram [3,5,10–12]. In order to calculate a FORC diagram, a family of FORCs with different reversal fields is measured, being $M(H_\alpha, H_\beta)$ the resulting magnetization as a function of both the reversal H_α and applied H_β fields, as depicted in Fig. 1.

Using the equation shown at the bottom of Fig. 1 and changing variables to the local coercivity $h_c = (H_\beta - H_\alpha)/2$ and the local bias $h_u = (H_\beta + H_\alpha)/2$, the FORC distribution $\rho(h_c, h_u)$ can be obtained. A FORC diagram is a contour plot of a FORC distribution. A more detailed explanation of the FORC model and its experimental implementation has been done in previous works [3,5,10].

In the present work the aim was focused on the study of magnetic interactions among the phases which are present in doped barium hexaferrite, when secondary phases appear.

The conventional ball-milling process was employed to obtain the studied system. The precursor oxides to form $Ba_{0.8}La_{0.2}Fe_{11.8}Co_{0.2}O_{19}$—barium carbonate ($BaCO_3$), hematite ($Fe_2O_3$), lanthanum oxide ($La_2O_3$) and cobalt acetate—were mixed in stoichiometric proportions and milled for 100 h at 200 rpm in air atmosphere in a planetary ball-mill with a ball/powder ratio of 3.

The as-milled powder was heat-treated in air atmosphere for 6 h at 1000 °C. The characterization by X-ray diffraction (XRD) with CuK_α radiation shows that barium hexaferrite has not completely crystallized after the thermal treatment. Small amounts of hematite still remain without transforming and

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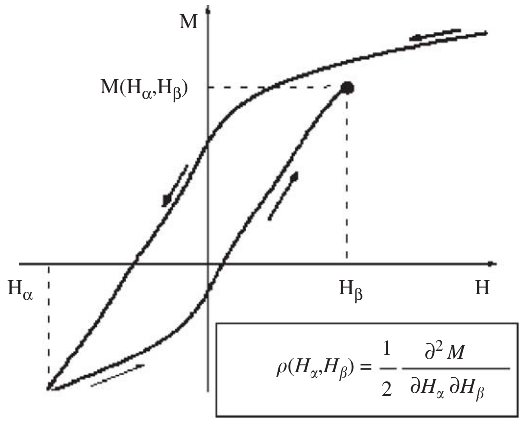

Fig. 1. Scheme of a FORC measurement and involved variables.

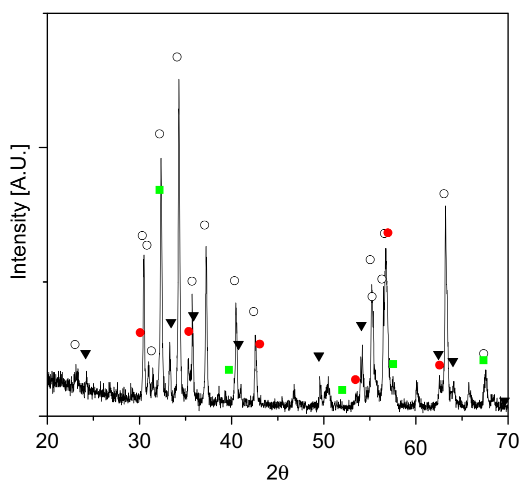

Fig. 2. X-ray diffraction pattern. Open circles: $BaFe_{12}O_{19}$, solid (red) circles: $CoFe_2O_4$, solid (black) triangles: Fe_2O_3 and solid (green) squares: $LaFeO_3$. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

segregation of secondary phases such as $CoFe_2O_4$ and $LaFeO_3$ is observed (Fig. 2). According to XRD data, the different phases appear in the following proportions: 59% hexaferrite, 14% cobaltite, 18% hematite and 9% La orthoferrite. From magnetic measurements at 5 K, saturation magnetization M_S of the sample was determined to be 70 emu/g. Considering the reported values of M_S at 0 K for Ba hexaferrite (100 emu/g [13]) and cobaltite (90 emu/g [13]) and their proportional contribution to the sample magnetization, a calculated $M_S^{calc} = 71.6\,\text{emu/g}$ was obtained. This value—obtained without considering neither hematite nor La orthoferrite contributions—is in good agreement with the measured M_S and supports the idea of disregarding weakly ferromagnetic Fe_2O_3 and antiferromagnetic $LaFeO_3$ in the analysis of magnetic interactions.

This sample was therefore chosen to study the magnetic interaction between particles of hexaferrite (hard phase) and cobaltite $CoFe_2O_4$ (softer phase, designated as 'soft' from now on).

The magnetic properties of the sample were studied in a SQUID magnetometer with a maximum applied field of 20 kOe. The coercive field H_c and the ratio of remanent to saturation magnetization M_r/M_S were obtained from the major hysteresis loops and are shown in Fig. 3. Both coercivity and M_r/M_S have a similar evolution with the temperature variation. When the sample is cooled down from room temperature both H_c and M_r/M_S decrease, reaching their lower values at 150 and 100 K, respectively. When lowering the temperature below 100 K these variables increase.

It is generally accepted that a decrease in coercivity is associated to an increase in exchange interactions (see, for instance, Ref. [15]). Having this in mind, the behavior of H_c vs. T shown in Fig. 3 would indicate an increase in exchange interactions from 300 to 150 K and a decrease from 150 to 5 K.

In the whole temperature range the sample exhibits small differences with the value 0.5 for the ratio M_r/M_S predicted by Stoner and Wohlfarth [14] for a system of non-interacting particles. Above 225 K the interactions are magnetizing and below this temperature a demagnetizing character is observed.

The FORC diagrams shown in Fig. 4 have peaks extended in the $h_c - h_u$ plane. The observed peaks in the $h_c - h_u$ plane are typical of samples with hard and soft magnetic interacting clusters with dipolar and exchange interactions [11,17]. For 300 and 200 K there

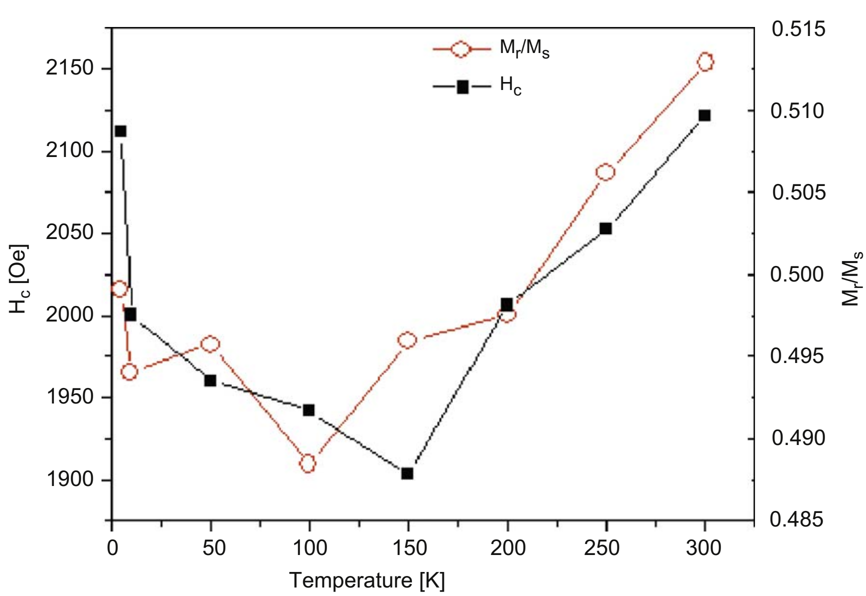

Fig. 3. Coercive field and M_r/M_S as function of temperature.