High-Temperature First-Order-Reversal-Curve (FORC) Study of Magnetic Nanoparticle Based Nanocomposite Materials

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ABSTRACT

First-order-reversal-curves (FORCs) are an elegant, nondestructive tool for characterizing the magnetic properties of materials comprising fine (micron- or nano-scale) magnetic particles. FORC measurements and analysis have long been the standard protocol used by geophysicists and earth and planetary scientists investigating the magnetic properties of rocks, soils, and sediments. FORC can distinguish between single-domain, multi-domain, and pseudo single-domain behavior, and it can distinguish between different magnetic mineral species [1]. More recently, FORC has been applied to a wider array of magnetic material systems because it yields information regarding magnetic interactions and coercivity distributions that cannot be obtained from major hysteresis loop measurements alone. In this paper, we will discuss this technique and present high-temperature FORC results for two magnetic nanoparticle materials: CoFe nanoparticles dispersed in a SiO_2 matrix, and FeCo-based nanocrystalline amorphous/nanocomposites.

Keywords: magnetometry, first-order-reversal-curves, FORC, nanoscale magnetic materials

INTRODUCTION

The most common measurement that is performed to characterize a material's magnetic properties is measurement of the major hysteresis loop M(H). The parameters that are most commonly extracted from the M(H) loop are: the saturation magnetization M_s, the remanence M_r, and the coercivity H_c.

First-order-reversal-curves (FORCs) [2] can give information that is not possible to obtain from the hysteresis loop alone. These curves include the distribution of switching and interaction fields, and identification of multiple phases in composite or hybrid materials containing more than one phase [3,4]. A FORC is measured by saturating a sample in a field H_{sat}, decreasing the field to a reversal field H_a, then measuring moment versus field H_b as the field is swept back to H_{sat}. This process is repeated for many values of H_a, yielding a series of FORCs. The measured magnetization at each step as a function of H_a and H_b gives $M(H_a, H_b)$, which is then plotted as a function of H_a and H_b in field space. The FORC distribution $\rho(H_a, H_b)$ is the mixed second derivative, i.e., $\rho(H_a, H_b) = -(1/2)\partial^2 M(H_a, H_b)/\partial H_a \partial H_b$.

The FORC diagram is a 2D or 3D contour plot of $\rho(H_a, H_b)$. It is common to change the coordinates from (H_a, H_b) to $H_c = (H_b - H_a)/2$ and $H_u = (H_b + H_a)/2$. H_u represents the distribution of interaction or reversal fields, and H_c represents the distribution of switching or coercive fields.

EXPERIMENT

To demonstrate the utility of the FORC measurement and analysis protocol for characterizing high-temperature magnetic properties of materials, measurements were conducted for two different magnetic nanoparticle materials: CoFe nanoparticles dispersed in a SiO_2 matrix [5], and FeCo-based nanocrystalline amorphous/nanocomposites [6,7]. All magnetic measurements were performed using a Lake Shore Cryotronics MicroMag™ vibrating sample magnetometer (VSM) with a high-temperature furnace, enabling variable temperature measurements from room temperature to 800 °C. All measured magnetization data are presented in terms of the magnetic moment (emu) as a function of field (Oe) and temperature (°C). There are a number of open source FORC analysis software packages such as FORCinel [8] and VARIFORC [9]. In this paper, FORCinel and custom analysis software were used to calculate the FORC distributions and plot the FORC diagrams.

DISCUSSION

CoFe Nanoparticles Dispersed in an SiO_2 Matrix

Figure 1 shows the room temperature hysteresis loops with SiO_2 volume fractions ranging from 40% to 80%, and show that the magnetic properties tend towards superparamagnetism with increasing SiO_2. Figure 2 shows the room temperature 2D FORC diagrams at SiO_2 volume fractions of 40%, 50% and 60%. There is a single peak in each FORC distribution that is centered at H_c, and there is a distribution of both interaction (H_u, vertical axis) and switching fields (H_c, horizontal axis), the former due to interparticle interactions, and the latter due to different particles switching at different applied field strengths. Note that the peaks in the FORC distributions are shifted towards negative interaction fields (H_u, vertical axis) which is a feature that is typically associated with exchange interactions occuring between magnetic particles [10].

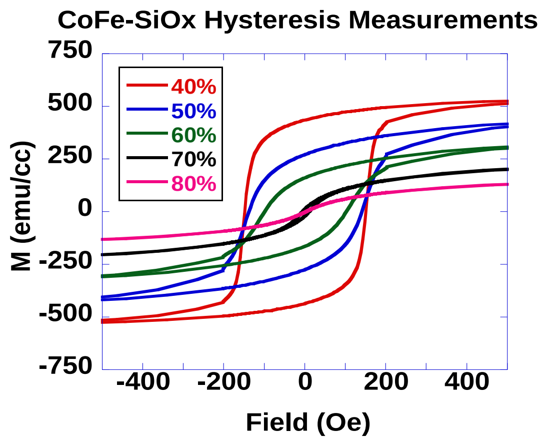


Figure 1. Room temperature hysteresis loops for CoFe nanoparticles dispersed in SiO_2 with volume fractions ranging from 40% to 80% (data are volume normalized and presented in emu/cc).

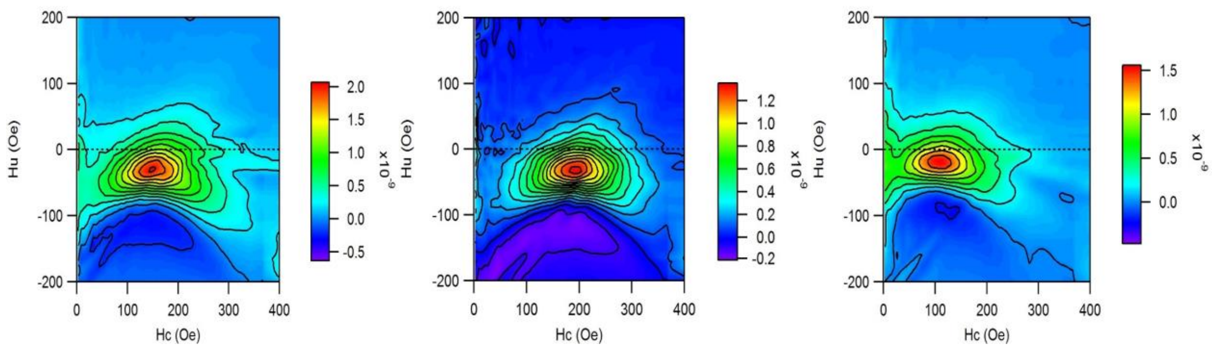


Figure 2. Room temperature 2D FORC diagrams for CoFe nanoparticles dispersed in SiO_2 volume fractions of 40% (left), 50% (middle) and 60% (right).

The hysteresis M(H) loops and FORCs were measured at temperatures of T = 25, 100, 200, 300, and 400 °C for the sample containing 40% volume fraction of CoFe nanoparticles dispersed in a 60% volume fraction of SiO_2. Transmission electron microscope (TEM) images show that the CoFe nanoparticles are approximately 10 nm in diameter and separated by an intergranular SiO_2 phase [5]. Figure 3 shows the hysteresis loops at each temperature and the measured FORCs at 25 °C. Figure 4 shows the 2D FORC diagrams at temperatures ranging from 25 to 400 °C. There is a single peak in the FORC distribution at each temperature that is centered at H_c and that shifts towards lower switching fields as temperature increases, coincident with the decrease in H_c with increasing temperature. In a FORC diagram, entirely closed contours are typically considered to be a fingerprint of single-domain (SD) particles, whereas entirely open contours that diverge towards the H_u axis are a fingerprint of multi-domain (MD) behavior [1]. The results shown in figure 4 demonstrate closed contours at lower temperatures and open contours at increasing temperatures, suggesting a transition from SD to MD behavior based upon the conventional interpretation of FORC diagrams. Further studies are required to better understand if this traditional interpretation is valid for such densely packed nanoparticle aggregates. At all temperatures there is a distribution of both interaction (H_u, vertical axis) and switching fields (H_c, horizontal axis), the former owing to interparticle interactions, and the latter due to different particles switching at different applied field strengths.

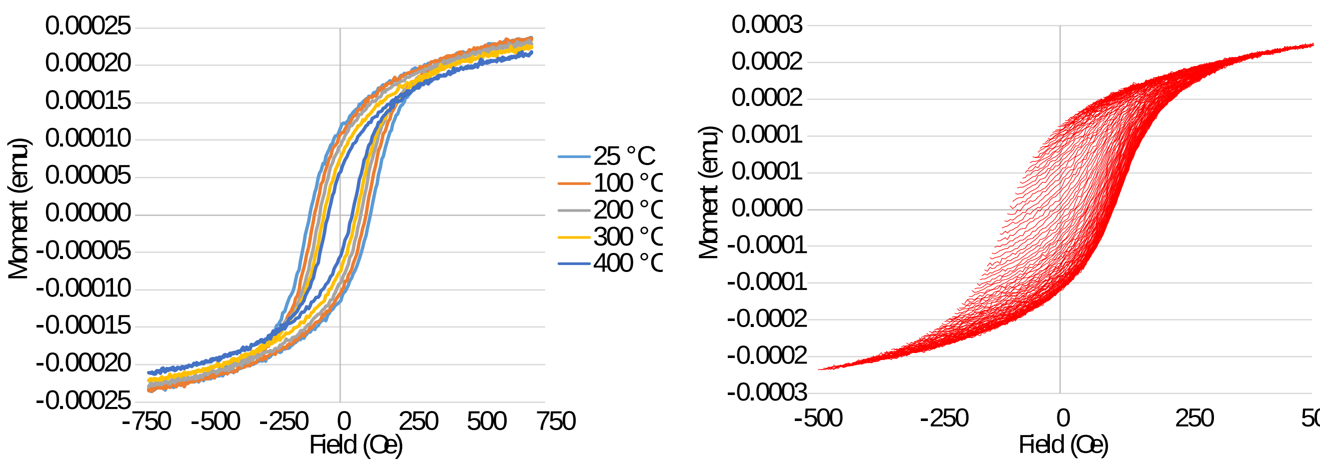


Figure 3. Hysteresis loops at T = 25, 100, 200, 300 and 400 °C (left) and measured FORCs at 25 °C (right) for CoFe nanoparticles (40%) dispersed in SiO_2 (60%).

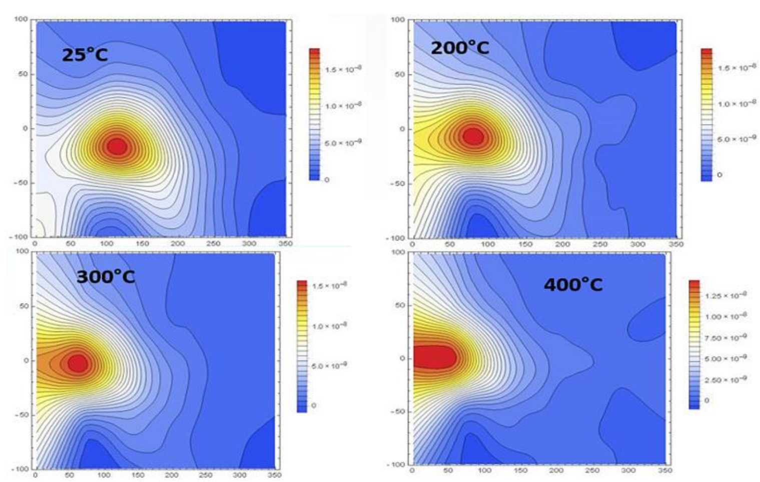


Figure 4. 2D FORC diagrams at T = 25, 200, 300 and 400 °C.

FeCo-based Nanocrystalline Amorphous/Nanocomposites

$Fe_{56}Co_{24}Si_2B_{13}Nb_4Cu_1$ samples were fabricated using a conventional melt spinning process, and TEM images show that the samples are fully amorphous or composed of nanocrystal diameters <~10 nm embedded in an intergranular amorphous phase after being subjected to a primary crystallization treatment at approximately 520 °C (‘crystallized’). The hysteresis M(H) loops and FORCs were measured for a disc-shaped sample (0.3 cm diameter) with the applied field oriented parallel to the ribbon plane, and at temperatures of T = 25, 400, and 780 °C, which is above the primary crystalization temperature, and then at 25 °C after cooling from 780 °C. Figure 5 shows the M(H) loops at each temperature and the measured FORCs at 780 °C. Figure 6 shows the corresponding 2D FORC diagrams and show a wishbone-like feature at 780 °C and at 25 °C after cooling. Wishbone FORCs typically mean that there are dipolar (or magnetostatic) interactions between particles. Because the Curie temperature of the amorphous phase is approximately 520 °C, at elevated temperatures dipolar interactions could indeed become an important interaction, whereas at lower temperatures the nanoparticles and amorphous precusor should be fully exchange coupled. The peak in the FORC distribution is shifted towards positive reversal or interaction fields (H_u, vertical axis) as temperature increases, which can suggest that dipolar interactions are becoming increasingly important with increasing temperature near and above the Curie temperature of the amorphous precursors.

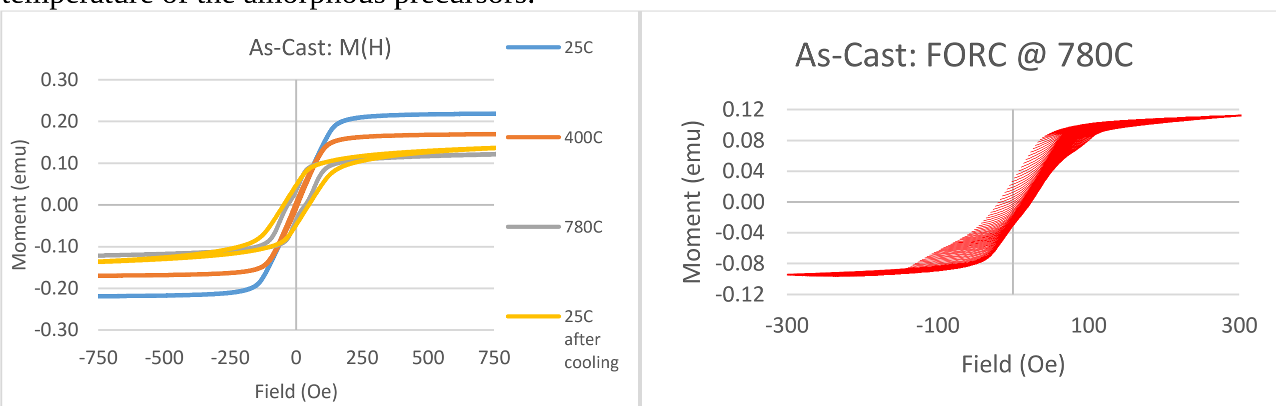
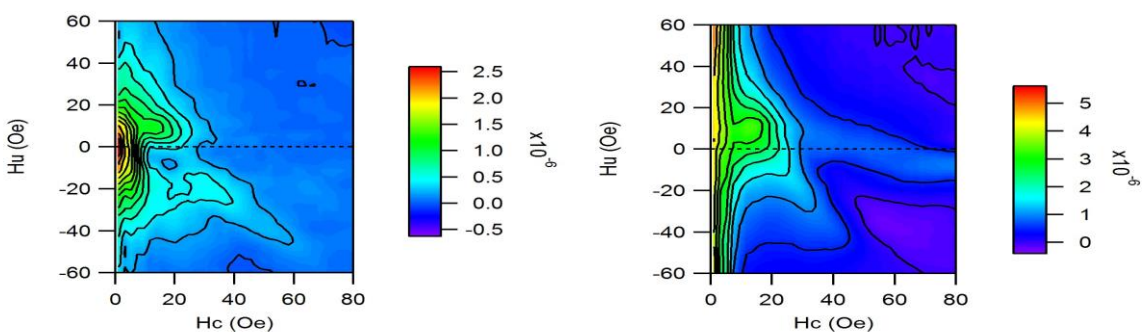


Figure 5. M(H) (left) at 25, 400, 780 and 25 °C after cooling, and the measured FORCs (right) at 780 °C for as-cast FeCo-based nanocrystalline amorphous/nanocomposite sample.



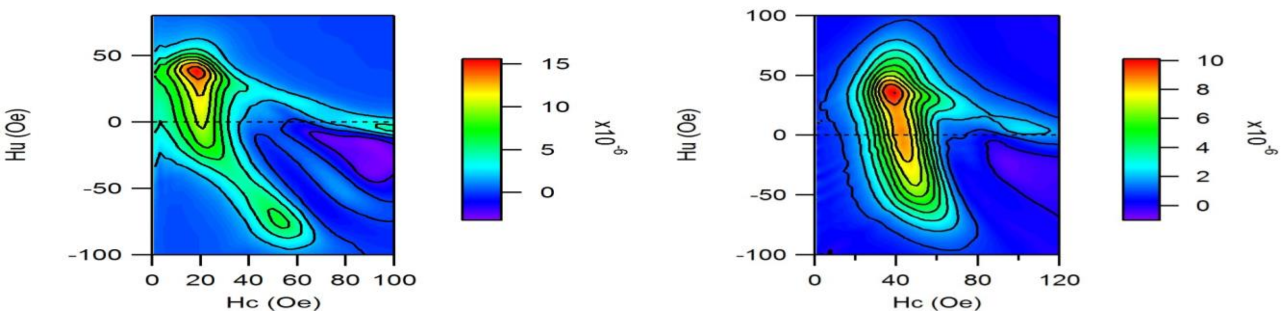


Figure 6. 2D FORC diagrams at 25 (upper left), 400 (upper right), 780 (lower left) and 25 °C after cooling (lower right) for a as-cast FeCo-based nanocrystalline amorphous/nanocomposites.

CONCLUSIONS

FORC analysis is indispensable for characterizing interactions and coercivity distributions in a wide array of magnetic materials, including; natural magnets, magnetic recording media [11,12], nanowire arrays [13], permanent magnets [14], and exchanged coupled magnetic multilayers [15]. In this paper, we have shown the evolution at high temperatures of the distribution of switching and interaction fields as determined from FORC analysis for CoFe nanoparticles and FeCo-based nanocrystalline magnetic materials

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