

Internal magnetostatic potentials of magnetization-graded ferromagnetic materials

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The authors investigated the internal magnetic field induced by a spatially varying magnetization in a compositionally graded ferromagnet. The authors discuss results on a hexagonal ferrite sample, with a variation in saturation magnetization of 30 emu/g over a sample thickness of 2.5 mm. The direct current magnetization shows a small anisotropy depending on the direction of the external magnetic field relative to the grading direction. This contribution from a grading induced magnetic field is more pronounced in alternating current susceptibility measurements. The authors find a shift in magnetic properties corresponding to an internal magnetic field of 30 Oe, which is significantly lower than the predicted value of approximately 1900 Oe. The authors discuss reasons for this discrepancy. © 2007 American Institute of Physics. [DOI: 10.1063/1.2437721]

Magnetization-graded ferromagnetic materials have recently emerged as the magnetic counterparts of compositionally doped semiconductors, with internal magnetostatic potentials determined principally by the magnitude and direction of their magnetization gradients, through the relation¹

$$\nabla^2 \phi(\mathbf{r}) = \nabla \cdot \mathbf{M}(\mathbf{r}). \quad (1)$$

Here, ϕ is the magnetic scalar potential and \mathbf{M} is the magnetization.

As in the case of polarization-graded ferroelectrics, the magnetization gradients are achieved by compositionally grading the ferromagnetic material.² Consequently, the spatial symmetry of these ferroic materials is broken, yielding internal potentials that may be exploited to create both active and passive devices as has been done with semiconductors (e.g., diodes, transistors, and other components).³ This is illustrated schematically in the inset in Fig. 1, which shows the direction induced internal magnetic field relative to the grading direction. The internal stress fields set up by the compositional gradients are also expected to couple to the magnetization much as they do in graded ferroelectrics.⁴ Therefore, compositionally graded ferromagnets require at least two coupled order parameters to describe their Landau potentials.⁵ In addition, as has been demonstrated with polarization-graded ferroelectrics, it is anticipated that graded ferromagnetic materials may also be created from homogeneous systems having either temperature or stress gradients.⁴

In this letter we prepare magnetization-graded ferromagnets from compositionally graded barium hexagonal ferrites. However, unlike our previous study, where we analyzed in-

ternal fields produced from magnetization gradients of compositionally graded Ni–Zn ferrites using ferromagnetic resonance microscopy,² in this study we show explicitly that an internal magnetic field $\mathbf{H}_{\text{int}}(\mathbf{r})$ is produced as a result of the magnetization gradient via relation (1) and

$$-\nabla \phi(\mathbf{r}) = \mathbf{H}_{\text{int}}(\mathbf{r}). \quad (2)$$

The presence of $\mathbf{H}_{\text{int}}(\mathbf{r})$ is manifested as a shift in the magnetic hysteresis loop along the field axis in accordance with Eqs. (1) and (2). For the hexagonal ferrite samples we are investigating, the calculated internal field is approximately $H_{\text{int}}=1900$ Oe, although this value is significantly suppressed by domain effects. After annealing the sample in a sufficiently large static magnetic field, no evidence of an internal field is observed at low magnetic field measurements. This is consistent with recent analysis concerning the domain struc-

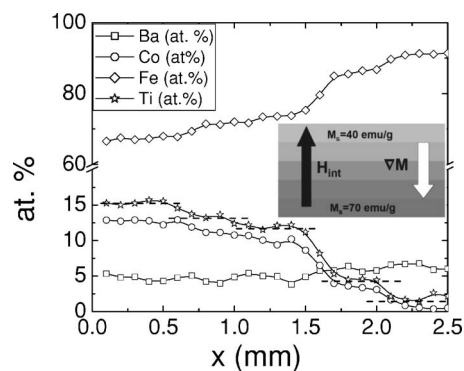


FIG. 1. Chemical composition of the hexagonal ferrite sample measured along the grading direction. Inset: Schematic representation of the compositionally graded hexagonal ferrite sample. The induced internal magnetic field is antiparallel to the grading in saturation magnetization, taken from Ref. 7.

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ture of graded ferroelectrics,⁶ which predict that these systems transform from a multidomain structure to a single domain structure when the applied field is larger than the largest coercive field of the graded materials.

Reagent grades of the carbonates of barium and cobalt as well as the oxides of titanium and iron were combined in appropriate ratio to produce hexagonal ferrite powders with the composition $\text{BaCo}_x\text{Ti}_x\text{Fe}_{12-2x}\text{O}_{19}$ with $x=0, 0.4, 1.2, 1.4,$ and 1.6 . The individual powders were calcined at 900°C for 3 h, ball milled for 16 h in isopropyl alcohol, then dried and screen meshed. Subsequently, 0.25 g of each of the five individual ferrites were poured (sequentially) and lightly tamped into a 13 mm diameter steel die (to yield separate compositionally uniform layers). The powder stackup was uniaxially pressed at 680 bars (10 000 psi) then sintered at 1200°C for 10 h so as to diffuse the components and make an approximately linear compositional gradient. The samples were annealed by slow cooling in air from 900 to 500°C at 25°C/h . Homogeneous samples were prepared in the same manner.

We experimentally verified the existence of the compositional grading in the hexagonal ferrite samples using energy dispersive x-ray analysis (EDX). A scanning electron microscopy (Hitachi S-2400 equipped with EDX detector) was used to acquire compositional spectra every $100\ \mu\text{m}$ along the cross section of the sample. The elemental concentrations (in at. %) were obtained by analyzing these spectra. The compositional profile along the grading direction is shown in Fig. 1. Because the Ba and Ti x-ray lines used for this study ($\text{Ti } K\alpha_1$ of $4.511\ \text{keV}$ and $\text{Ba } L\alpha$ of $4.466\ \text{keV}$) overlap, it is difficult to distinguish these two elements, leading to some small error in the Ba and Ti fractions, on the order of 2% – 3% (absolute). We found that the Fe content of the sample increased from approximately 68% to 92% as we measured along the grading direction, with a concomitant decrease in Ti and Co fractions. There are five clear steps visible (indicated by dashed lines), which are consistent with the compositions used to fabricate the original sample. The thickness of each compositionally distinct region ranges from about 300 to $500\ \mu\text{m}$, with interfacial regions of approximately $200\ \mu\text{m}$. The EDX data for the homogeneous hexagonal ferrite sample (not shown) showed a variation in chemical composition of less than 2% across the entire sample.

We measured the direct current (dc) magnetization of the graded sample using a Quantum Design superconducting quantum interference device magnetometer, at magnetic fields from -5 to $+5\ \text{kOe}$ and at a temperature of $300\ \text{K}$. These data are shown in Fig. 2. The open symbols show the magnetization measured perpendicular to the grading direction; the closed symbols show the magnetization parallel to the grading direction. The saturation magnetization for the graded sample is approximately $52\ \text{emu/g}$, consistent with the average magnetization for the hexagonal ferrite components.⁷ There is considerable anisotropy in the magnetization of this graded sample, which was absent in the homogeneous sample (not shown). We attribute this anisotropy to the large interfacial area in the layered sample, rather than any effects from the grading.

There are two other significant features observed in the $M(H)$ loop in the graded sample along the grading direction. There is a small feature in $M(H)$ close to $M=0$. This jump of approximately $4\ \text{emu/g}$ is repeatable, and we tentatively at-

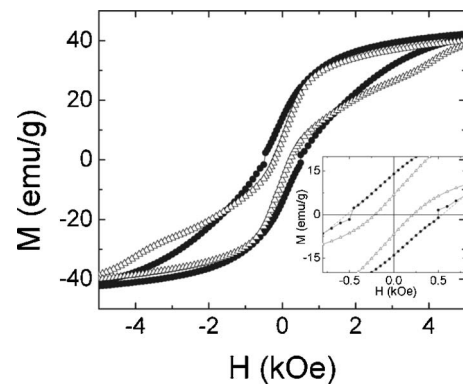


FIG. 2. Solid symbols: Magnetization measured with the magnetic field applied along the grading direction. Open symbols: Magnetization measured with the magnetic field applied perpendicular to the grading direction. These data were collected at $T=300\ \text{K}$. Inset: $M(H)$ loops near $H=0$. The dashed lines are a guide to the eyes showing the jump in magnetization measured along the grading direction.

tribute it to domain boundary effects at the grading interfaces. The second observation is that the coercivity has a slight asymmetry. The coercive field for positive H is $538\ \text{Oe}$, while the coercive field for negative H is $512\ \text{Oe}$. While this asymmetry has several possible origins, including some small residual field in the superconducting magnet or the different values of coercivity for the different compositions, this shift is consistent with the presence of an internal magnetic field produced by the compositional grading. Furthermore, the magnitude of asymmetry changed when the applied field was at 180° to the field direction used in measuring the data for Fig. 2 to a positive coercive field of $H=504\ \text{Oe}$ and a negative coercive field of $H=491\ \text{Oe}$. The key result of these dc investigations is not simply that there is an asymmetry in the coercive field but that the magnitude of this asymmetry depends on the direction of the applied magnetic field relative to the grading direction.

In order to more precisely investigate a possible grading induced shift in magnetization, we also measured the ac magnetic susceptibility of the hexaferrite samples at an excitation field of $2\ \text{Oe}$. We used the ac magnetic susceptibility option on a Quantum Design physical property measurement system to investigate the real and imaginary components of the magnetic susceptibility between $+2$ and $-2\ \text{kOe}$ over a range of temperatures. The upper panel of Fig. 3 plots the real part of the susceptibility with the (positive) magnetic field applied along the direction of decreasing saturation magnetization (BA), measured at $T=300\ \text{K}$. After these measurements, the sample was rotated through 180° so that the (positive) magnetic field was applied along the direction of increasing saturation magnetization (AB), still at $T=300\ \text{K}$. These data are plotted in the lower panel of Fig. 3. The insets to both panels show the peaks in susceptibility in more detail. These susceptibility measurements show a clear asymmetry, which depends on the orientation of the compositional grading relative to the external magnetic field.

In both cases, the amplitude of the susceptibility is larger when the external magnetic field is antiparallel to the grading direction. We can estimate the field producing a maximum in susceptibility for each direction of the external magnetic field relative to the internal grading by averaging over both sample orientations. Specifically, we average the peak field from the upper panel of Fig. 3 at positive fields with the peak

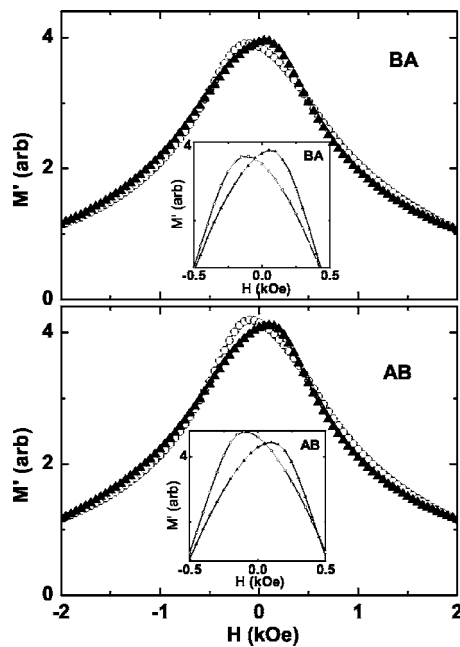


FIG. 3. Upper panel: Real part of the ac susceptibility measured with the positive magnetic field applied antiparallel to the grading direction. Lower panel: Real part of the ac susceptibility measured with the positive magnetic field applied parallel to the grading direction. The insets show the region near $H=0$ in more detail. For all panels the solid symbols were measured for the magnetic field swept from +2 to -2 kG and the open symbols for the field swept from -2 to +2 kG.

field from the lower panel of Fig. 3 at negative fields, and vice versa. We find that the peak field is smaller when the external magnetic field is antiparallel to the grading direction. The difference in peak field can be ascribed to an internal magnetic field of approximately $H_{\text{int}}=30$ Oe directed along the grading direction. The magnitude and direction of this field are substantially smaller than the predicted value of 1900 Oe, based on the saturation magnetization of each layer and Eqs. (1) and (2).⁷ While these observations suggest the presence of an internal magnetic field produced by the compositional grading, this internal magnetic field is roughly two orders of magnitude smaller than what is theoretically predicted. We understand this difference, however, in view of the fact that Eqs. (1) and (2) and the corresponding estimates are for a true compositionally graded material (Fig. 1 shows a smoothed, but stepwise composition gradient), without domain structures or local structural changes. The formation of magnetic domains is expected to substantially reduce the internal energy of the graded material, and these domains would be expected to dramatically suppress the internal field.

In order to further investigate whether these magnetic asymmetries could arise from causes other than compositionally induced field, we also measured the ac magnetic susceptibility for the *homogeneous* hexagonal ferrite sample for two sample orientations differing by a rotation through 180° . For the homogeneous sample, there was no shift in amplitude or peak field for the two different orientations of the

magnetic field relative to the sample. We also measured the ac magnetic susceptibility for the compositionally graded hexagonal ferrite sample with the magnetic field perpendicular to the grading direction, for two sample orientations differing by a 180° rotation. These measurements showed a small shift in amplitude and peak field consistent with an internal field of approximately $H_{\text{int}}=12$ Oe. While this shift may arise from some small misalignment of the sample, it is likely that there is also some small internal magnetic field produced tangentially to the grading direction because of some curvature at the interfaces. Finally, in order to probe effects due to magnetic domains, we also measured the susceptibility between +2 and -2 kOe after preparing the sample in a field of +1 T (starting from +2 kOe) or -1 T (starting from -2 kOe). We still observed an internal, grading induced magnetic field for these measurements, but the magnitude of the field was much smaller, approximately $H_{\text{int}}=3$ Oe. This suggests that it will be important to properly consider the role of magnetic domains in order to accurately model the grading field.

In conclusion, we have investigated the internal magnetic field induced by a static graded magnetization. Our experiments on a graded hexaferrite sample show evidence for an internal magnetic field of roughly 30 Oe, antiparallel to the magnetization gradient, which is significantly smaller than the predicted value of 1900 Oe. This difference likely arises from an internal relaxation mechanism (such as domains) that acts to substantially lower the overall internal energy state of the structure. The magnitude of the internal field is strongly suppressed when a large external magnetic field is applied to the sample, consistent with the expected changes in domain structure. These measurements highlight the importance of the domain structure in determining the internal field, as has been discussed in the context of graded ferroelectrics, but also suggest that very large internal fields (on the order of 2 kOe) may be attainable in samples in which domain formation can be suppressed; such structures, in thin film form, might find extensive use in communication devices as self-biased phase shifters and resonators.

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