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Dependence of magnetomechanical performance of CoGa$_x$Fe$_{2-x}$O$_4$ on temperature variation

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The temperature dependence of the magnetoelastic properties of the CoGa$_x$Fe$_{2-x}$O$_4$ system (for x = 0.0, 0.2, and 0.4) has been studied. It has been shown that increase in temperature resulted in reduced magnetostrictive hysteresis. For both CoGa$_{0.2}$Fe$_{1.8}$O$_4$ and CoGa$_{0.4}$Fe$_{1.6}$O$_4$, the measured magnetostriction amplitudes were higher at 250 K than at 150 K. It was also shown that the temperature stability of magnetostriction in CoGa$_{0.2}$Fe$_{1.8}$O$_4$ is higher than that of the other compositions studied which is important for sensor applications. The highest strain sensitivity was obtained for CoGa$_{0.2}$Fe$_{1.8}$O$_4$ at 250 K. Results demonstrate the possibility of tailoring magnetomechanical properties of the material to suit intended applications under varying temperature conditions. © 2011 American Institute of Physics. [doi:10.1063/1.3540662]

I. INTRODUCTION

Cobalt ferrite and its derivatives are very promising materials for stress sensor and actuator applications. The need to control the magnetostrictive properties has resulted in several studies including the influence of vacuum sintering,[1] annealing and quenching heat treatment,[2] metal bonding,[3] and cation substitutions.[4–7] Of these, cation substitution has been found very useful for improving the strain response of cobalt ferrite to applied magnetic field. Substitution of nonmagnetic cations alters the exchange coupling in the ferrite and the site occupancy of the Co^{2+} and thereby alters the magnetocrystalline anisotropy and magnetostrictive properties. As a result, the desired properties for specific magnetomechanical applications can be controlled by selectively tailoring exchange coupling and cation site occupancy via cation substitution.

The influence of temperature variation on the properties of CoGa$_x$Fe$_{2-x}$O$_4$ for magnetomechanical applications is reported. At room temperature, CoGa$_x$Fe$_{2-x}$O$_4$ has over 145% higher magnetomechanical sensitivity than CoFe$_2$O$_4$ (Ref. 6) but little is known about how the response varies with temperature. In all reported cation substitution studies on CoFe$_2$O$_4$ for magnetostrictive applications, the highest strain sensitivity at room temperature was obtained for CoGa$_x$Fe$_{2-x}$O$_4$.[7] Since devices based on CoGa$_x$Fe$_{2-x}$O$_4$ would need to work under varying temperature conditions, understanding the dependence of performance on temperature is crucial. This study therefore reports on the dependence of the magnetostrictive properties of CoGa$_x$Fe$_{2-x}$O$_4$ on both cation composition and temperature.

II. EXPERIMENTAL DETAILS

Samples were pressed, calcined twice at 1000 °C and sintered at 1350 °C for 24 h in air. The crystal structures were studied by x-ray diffractometry (XRD). To investigate the microstructures of the samples, backscattered electron micrographs were obtained using an analytical scanning electron microscope equipped with an x-ray analyzer for energy dispersive x-ray spectroscopy (EDX). Magnetostriction (λ) was measured parallel to the applied field using resistive strain gauges attached to the samples, over a temperature range 50–350 K using a physical property measurement system. The derivative of the λ-H curve with respect to field (dλ/dH) was used to determine the strain sensitivity of the materials.

III. RESULTS AND DISCUSSIONS

X-ray diffraction patterns of the samples are shown in Fig. 1. The patterns have peaks which are consistent with the cubic spinel phase. They are not shifted relative to the unsubstituted (x = 0) sample as was the case of CoAl$_x$Fe$_{2-x}$O$_4$.[8] This indicates that the lattice parameters of the
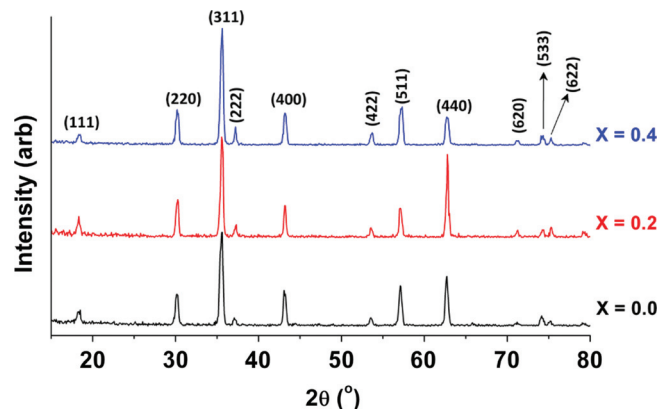

FIG. 1. (Color online) XRD patterns of the CoGa$_x$Fe$_{2-x}$O$_4$ samples (x = 0.0, 0.2, and 0.4).

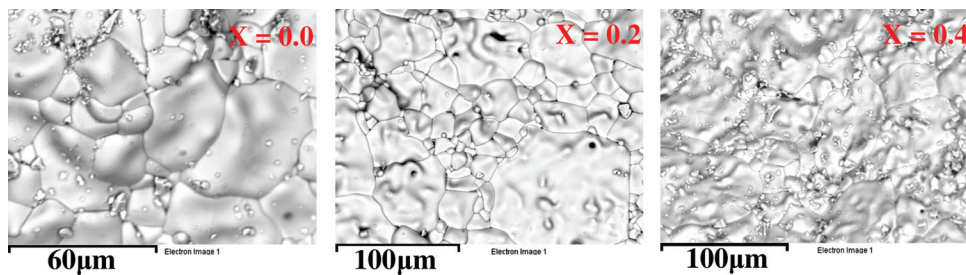Author to whom correspondence should be addressed. Electronic addresses nlebedim@iastate.edu and nlebedim@ameslab.gov.

FIG. 2. (Color online) Backscattered scanning electron micrographs for the $CoGa_xFe_{2-x}O_4$ system (x = 0.0, 0.2, 0.4).

$CoGa_xFe_{2-x}O_4$ group of samples were similar or even identical and therefore not sensitive to changes in chemical composition. The lattice parameter of all samples was found to be 8.38 Å. The preservation of the lattice parameter for $CoGa_xFe_{2-x}O_4$ in contrast with the $CoAl_xFe_{2-x}O_4$ system arises because the ionic radii of Ga^{3+} in tetrahedral and octahedral positions ($r_A = 0.47$ Å, $r_B = 0.62$ Å) are quite similar to those of Fe^{3+} ($r_A = 0.49$ Å, $r_B = 0.65$ Å). Therefore, substituting Ga^{3+} for Fe^{3+} is expected to have little effect on the lattice parameter compared with the substitution of the Al^{3+} with much smaller ionic radii ($r_A = 0.39$ Å, $r_B = 0.54$ Å). From EDX, the sample compositions were shown to be $Co_{1.02}Fe_{1.98}O_4$ (for x = 0), $Co_{1.04}Ga_{0.2}Fe_{1.74}O_4$ (for x = 0.2), and $Co_{1.03}Ga_{0.39}Fe_{1.58}O_4$ (for x = 0.4).

Figure 2 shows the backscattered electron images of the samples. The uniformity in contrast observed for all the samples further confirms that all had a single phase crystal structure. They also appear to be of uniform composition because the image contrast in a backscattered electron image is a function of composition.

The variation of room temperature magnetization with applied magnetic field is shown in Fig. 3. In contrast with Al^{3+} substitution,[7] the saturation magnetization of the Ga^{3+} substituted cobalt ferrite initially increased at x = 0.2 and later decreased at x = 0.4. The saturation magnetization at x = 0.4 is also higher than that at x = 0.0. The increase in saturation magnetization with Ga^{3+} substitution is in agreement with Ga^{3+} having tetrahedral site preference. In spinel ferrites the moments of the octahedral and tetrahedral sites couple antiparallel.[9,10] The net magnetization (M_s) therefore is the difference in magnetization (M) of the sites: $M_s = M_{oct} - M_{tet}$. Substituting nonmagnetic Ga^{3+} for Fe^{3+} in the tetrahedral

sites of cobalt ferrite will decrease the net magnetization of the tetrahedral site and at the same time increase the overall net magnetization. Moreover, the more Ga^{3+} is substituted, the lower the Curie temperature becomes[6] such that at higher Ga^{3+} concentrations, the room temperature becomes significant compared to the Curie temperature. This should also result in lower permeability and lower technical saturation magnetization at higher Ga^{3+} concentration.

As shown in Fig. 4(a) for unsubstituted cobalt ferrite samples, peak to peak magnetostriction amplitude decreased with increase in the measurement temperature. In the substituted samples, the slope of the high field region of the magnetostriction curve was initially low at 50 K, increased at 150 and 250 K, then decreased further with increase in measurement temperatures. The easy and hard magnetization directions in cobalt ferrite are ⟨100⟩ and ⟨111⟩, respectively. It has large negative λ_{100} and smaller positive λ_{111} contributions to its magnetostriction amplitude.[11] The initial part of the curve represents a region in which the contribution of λ_{100} is dominant. It continues until all magnetic domains align parallel to the easy axes ⟨100⟩. The contribution of λ_{111} is observed if the magnetic field is further increased. In this study, the contribution of λ_{111} to the resultant magnetostriction increased with increase in measurement temperature. It can also be seen from all the results that increase in temperature resulted in reduced magnetostrictive hysteresis. This can be explained in terms of the effect of increase in temperature on anisotropy and coercive field. A previous study[12] shows that increase in temperature results in lower anisotropy and coercive field which was expected to lead to less magnetostrictive hysteresis as observed.

In both $CoGa_{0.2}Fe_{1.8}O_4$ [Fig. 4(b)] and $CoGa_{0.4}Fe_{1.6}O_4$ [Fig. 4(c)], measurement at 250 K resulted in higher magnetostriction amplitude than at 150 K. A similar result was obtained for the magnetostriction of $CoAl_xFe_{2-x}O_4$ at x = 0.5 and 0.7.[8] This result shows that the substitution of Ga^{3+} for Fe^{3+} alters both magnetostriction and its variation with temperature as shown in Fig. 4(d). It could also be seen in Fig. 4(d) that under temperature variations, the magnetostriction of $CoGa_xFe_{2-x}O_4$ is more stable than that of $CoFe_2O_4$ which could be essential for sensor applications. In Fig. 4(e), it appears that the magnetostriction of $CoGa_{0.2}Fe_{1.8}O_4$ is more stable with temperature variation than $CoGa_{0.4}Fe_{1.6}O_4$.

Figure 4(f) shows the variation of $(d\lambda/dH)_{max}$ of $CoGa_xFe_{2-x}O_4$ with temperature. At all temperatures studied except 250 and 300 K, $(d\lambda/dH)_{max}$ decreased with increase in gallium concentration. As in the case of Al^{3+} substituted cobalt ferrite,[8] at 250 K, $(d\lambda/dH)_{max}$ initially increased from x = 0.0 to 0.2 and decreased afterwards. This is
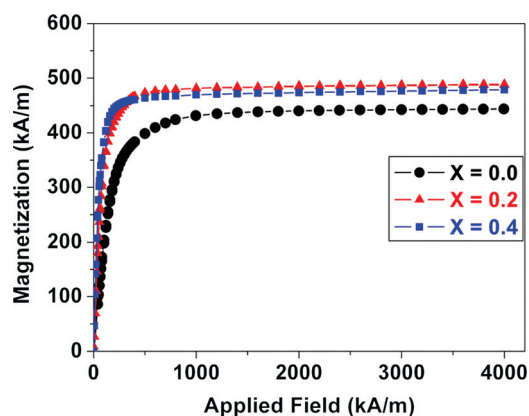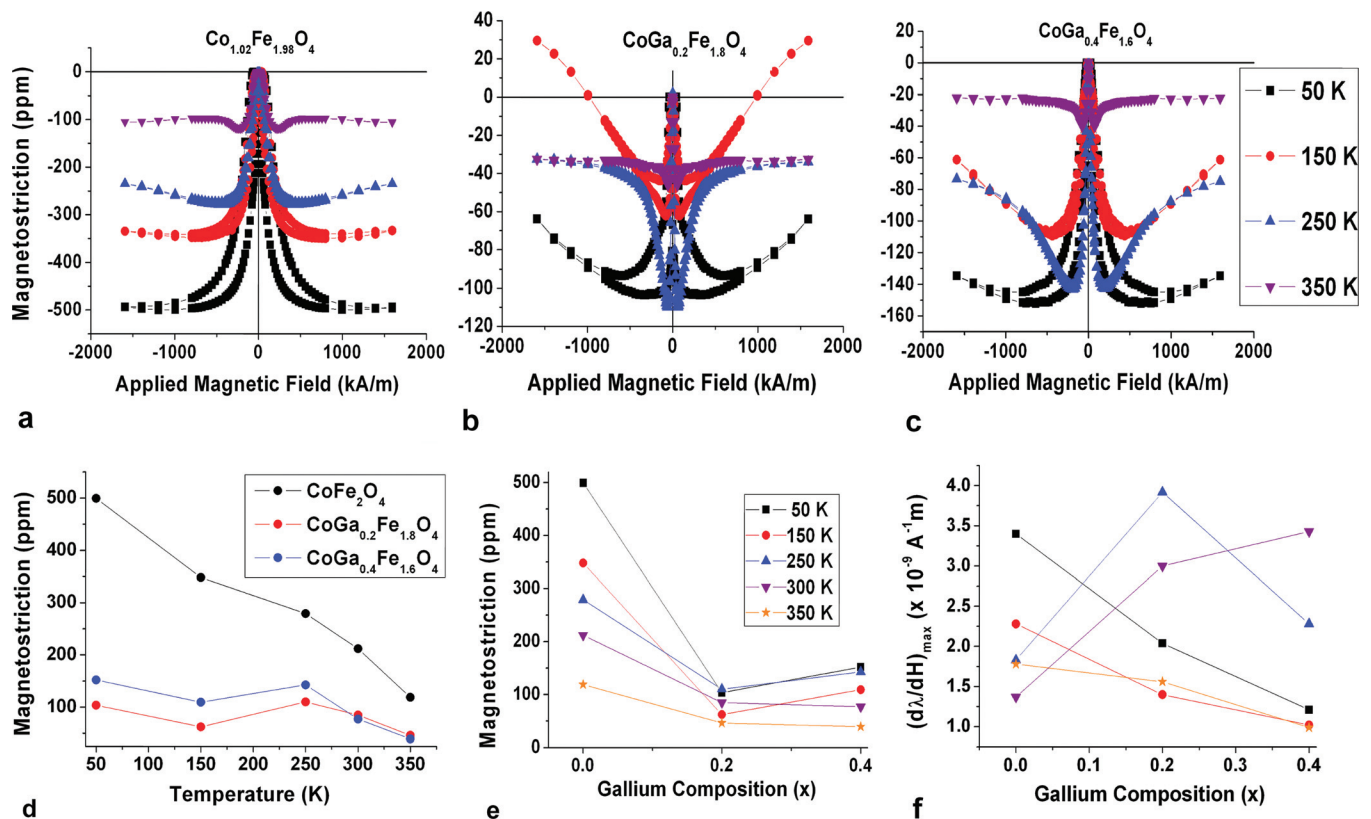

FIG. 3. (Color online) Variation of room temperature magnetization as a function of applied field for Ga^{3+} concentrations x = 0.0, 0.2, and 0.4.

FIG. 4. (Color online) Magnetostriction curves of the $CoGa_xFe_{2-x}O_4$ system (a) x = 0.0, (b) x = 0.2, and (c) x = 0.4. Variation of magnetostriction amplitude with (d) temperature, (e) Ga^{3+} composition, and (f) variation of strain derivative with temperature.

similar to the observation at 300 K except that the maximum $(d\lambda/dH)_{max}$ was at x = 0.4 rather than 0.2. A similar trend was observed in previous studies on cation substituted cobalt ferrite samples in which the $(d\lambda/dH)_{max}$ initially increased at lower concentrations of the substituted cations (x ≤ 0.2) and finally decreased at higher cation concentrations.[4] The figure also shows that with suitable cation substitution, the maximum strain sensitivity is in the temperature range 250–300 K. These results demonstrate the capability to alter magnetomechanical performance to suit intended applications by adjusting cation concentration at different temperatures.

IV. CONCLUSION

The study shows that magnetostrictive properties of cobalt ferrite can be altered by cation substitution. It was observed from XRD and SEM investigations that substitution of Ga^{3+} does not result in any observable changes in the crystal structure or microstructure of the material. On the other hand, magnetic and magnetostrictive properties have shown strong dependence on Ga^{3+} substitution. The highest strain sensitivity $(d\lambda/dH)$ was observed for $CoGa_{0.2}Fe_{1.98}O_4$ in the region 250–300 K.

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