Giant magnetoimpedance and field sensitivity in amorphous and nanocrystalline $(\text{Co}_{1-x}\text{Fe}_x)_{89}\text{Zr}_7\text{B}_4$ ($x = 0, 0.025, 0.05, 0.1$) ribbons

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We report on the giant magnetoimpedance (GMI) effect and its field sensitivity (η) in amorphous and nanocrystalline $(\text{Co}_{1-x}\text{Fe}_x)_{89}\text{Zr}_7\text{B}_4$ ribbons with $x = 0, 0.025, 0.05$, and 0.10 . The nanocrystalline samples were obtained by annealing amorphous ribbons in a field of 2 T applied perpendicular to the ribbon axis. We find that for the amorphous samples the GMI ratio tends to decrease with Fe doping, while the largest value of η is achieved for the $x = 0.025$ composition. For the nanocrystalline samples, the GMI ratio and η first decrease with increase of Fe-doped content from $x = 0$ to $x = 0.05$ and then increase for $x = 0.1$. The field annealing significantly enhances the GMI ratio and η in the nanocrystalline samples with $x = 0$ and 0.1 , but decreases the η in the nanocrystalline samples with $x = 0.025$ and 0.05 . The variations of the GMI ratio and the field-induced magnetic anisotropy field (H_K) upon Fe doping are correlated with the microstructural changes in the nanocrystalline samples. © 2011 American Institute of Physics. [doi:10.1063/1.3540403]

Research on soft ferromagnetic materials exhibiting giant magnetoimpedance (GMI) effect for high-performance magnetic sensors is an area of topical interest.^{1,2} GMI is a large change of the ac impedance of a ferromagnetic conductor in a static magnetic field.¹ The origin of GMI can be understood in terms of the skin effect characterized by the skin depth (δ), which is given by²

$$\delta = \sqrt{\frac{\rho}{\pi \mu f}} \quad (1)$$

where ρ is the electrical resistivity, f the excitation frequency of an ac current, and μ the magnetic permeability. According to the theory, GMI should exist in magnetic materials with large μ , small ρ , and small δ .^{1,2} Experimental studies have shown that soft ferromagnetic materials, such as Co-based amorphous and Fe-based nanocomposite wires/ribbons, are good candidates for GMI sensors.^{1–5} Continued efforts have been devoted to improving GMI effect in these materials either by fine tuning alloy compositions and/or heat treatment.^{6–8}

A new area of study in nanocomposite materials has been in Co-rich alloys in which 2- or 3-nanocrystalline phases have been seen simultaneously within an amorphous matrix.^{9–11} The mechanism for induced magnetic anisotropy in these alloys has been an area of active inquiry. Recently, Ohodnicki Jr. *et al.*¹² reported on the large field-induced anisotropies in $(\text{Co}_{1-x}\text{Fe}_x)_{89}\text{Zr}_7\text{B}_4$ ($0 \leq x \leq 0.1$) nanocrystalline ribbons, which were obtained by annealing the corresponding amorphous ribbons at 540 °C for 1 h in a large field of

2 T applied perpendicular to the ribbon axis. The measured anisotropy constants in these Co-rich alloys show a markedly different behavior than compositions closer to equiatomic Fe-Co ratios.¹² The mechanism behind the good response to field annealing in these alloys can be attributed to the devitrification of multiple crystalline phases during the annealing process.¹³ Unlike conventional pair ordering in amorphous materials, the induced anisotropy has been shown to be stable at high temperatures.¹⁴ Since the magnitude of the field-induced anisotropy field significantly varies with Fe doping, these materials are of potential interest for GMI studies.

In this work, we have systematically investigated the GMI effect and its field sensitivity (η) in amorphous and nanocrystalline ribbons $(\text{Co}_{1-x}\text{Fe}_x)_{89}\text{Zr}_7\text{B}_4$ with $x = 0, 0.025, 0.05$, and 0.10 .

The amorphous ribbons with a width of 2 mm and a thickness of 30 μm were synthesized by arc melting method to obtain the ingots of appropriate composition followed by the rapid solidification through melt-spinning process. The nanocrystalline samples were obtained by annealing the corresponding amorphous ribbons at 540 °C for 1 h in a field of 2 T applied perpendicular to the ribbon axis. X-ray diffraction (XRD) and transmission electron microscopy (TEM) confirmed the amorphous and nanocrystalline nature of the amorphous and nanocrystalline samples.¹⁵ The ribbon samples of 3 cm length were used for the present study. Magnetoimpedance measurements were conducted along the ribbon axis in dc magnetic fields up to 120 Oe over a frequency range of 0.1 ~ 10 MHz with a constant ac current of 5 mA. The detail of the measurement system has been reported elsewhere.⁷ The GMI ratio and the magnetic field sensitivity of GMI ratio are defined as

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$$\Delta Z/Z = 100\% \times \frac{Z(H) - Z(H_{\max})}{Z(H_{\max})} \quad (2)$$

and

$$\eta = \frac{d}{dH} \left(\frac{\Delta Z}{Z} \right), \quad (3)$$

where $Z(H)$ and $Z(H_{\max})$ represent the impedance in a magnetic field H and in the maximum field ($H_{\max} = 120$ Oe), respectively.

Figure 1(a,b) shows the field dependence of GMI ratio ($\Delta Z/Z$) at a representative frequency of 0.5 MHz for $(\text{Co}_{1-x}\text{Fe}_x)_{89}\text{Zr}_7\text{B}_4$ ($x = 0, 0.025, 0.05$, and 0.1) amorphous and nanocrystalline samples. It is observed that the values of $\Delta Z/Z$ are larger for the nanocrystalline samples than for their amorphous counterparts. The GMI ratio as well as the shape of GMI curve significantly varies with Fe doping in the amorphous and nanocrystalline samples. A double-peak structure is observed in both cases, but it is much more pronounced in the nanocrystalline samples due to the presence of large field-induced magnetic anisotropies.

Figures 2(a)–2(d) shows the frequency dependences of maximum GMI ratio ($[\Delta Z/Z]_{\max}$) and the maximum field sensitivity of GMI ($\eta_{\max}$) for the amorphous and nanocrystalline samples, respectively. It can be observed in Figs. 2(a) and 2(c) that in the frequency range of 0.1–10 MHz for the

FIG. 1. (Color online) Magnetic field dependence of GMI ratio ($\Delta Z/Z$) at 0.5 MHz for $(\text{Co}_{1-x}\text{Fe}_x)_{89}\text{Zr}_7\text{B}_4$ ($x = 0, 0.025, 0.05, 0.10$) (a) amorphous and (b) nanocrystalline ribbons.

amorphous samples the $[\Delta Z/Z]_{\max}$ tends to decrease with Fe doping, except for the larger values of $[\Delta Z/Z]_{\max}$ of the $x = 0.05$ sample when compared with the $x = 0.025$ sample for $f > 3$ MHz. For $f < 3$ MHz, the largest value of $\eta_{\max}$ is achieved for the $x = 0.025$ composition; however, for $f > 3$ MHz the $\eta_{\max}$ gradually decreases as the Fe-doping level is increased. For the case of the nanocrystalline samples, the $[\Delta Z/Z]_{\max}$ and $\eta_{\max}$ first decrease with increase of Fe-doped content from $x = 0$ to $x = 0.05$ and then increase for $x = 0.1$ [see Figs. 2(b) and 2(d)]. The field annealing is found to significantly enhance both the $[\Delta Z/Z]_{\max}$ and $\eta_{\max}$ in the nanocrystalline samples with $x = 0$ and 0.1 , but decrease the $\eta_{\max}$ in the nanocrystalline samples with $x = 0.025$ and 0.05 , when compared to their amorphous counterparts. The width of the peaks and the corresponding sensitivity in the $x = 0$ nanocrystalline samples are significantly less than the samples containing Fe. A narrow peak in the GMI response is attributed to low dispersion in the anisotropy direction. Interestingly, characterization of hexagonal close packed (HCP) grains in similar samples showed no signs of texture¹³ so presumably the well defined anisotropy direction in the $x = 0$ composition should be attributed to another source. Herzer modeled the effect of multiple crystalline phases and a uniform induced anisotropy of varying strength on the dispersion of the easy axis.¹⁶ Through symmetry arguments and numerical calculations predicted a wider dispersion in an array of randomly oriented uniaxial particles compared to a similar array of cubic particles for a given $K_u/\langle K_1 \rangle$, where K_u and K_1 are field induced anisotropy constant and magnetocrystalline anisotropy constant, respectively. Assuming $\langle K_1 \rangle$ is larger for the HCP phase compared to the cubic phases, the induced anisotropy K_u is necessarily larger in the HCP phase. Since the HCP phase is largely monatomic compared to the cubic phases and directional pair ordering is not possible, the larger induced anisotropy must arise from another mechanism. Furthermore, we note in Fig. 2(a) and (b) that in the frequency range of 0.1 to 10 MHz, $[\Delta Z/Z]_{\max}$ first increases with increasing frequency, reaches a maximum at a critical frequency ($f_0 \sim 2$ MHz), and then decreases for higher frequencies. This trend can be interpreted by considering the relative contributions of domain wall motion and magnetization rotation to the transverse permeability and hence the GMI effect.^{2,7}

Finally, the variation in the GMI peak location with Fe doping in the nanocrystalline samples is noteworthy. It has been shown that the magnetic field at which the GMI curve reaches a peak corresponds to the effective magnetic anisotropy field (H_K).² As compared to their amorphous counterparts, the larger values of H_K are achieved in the nanocrystalline samples as a direct consequence of the field annealing at 2 T (see Fig. 1). To correlate the GMI ratio and H_K , determined from the GMI data, with the microstructure in the nanocrystalline samples, we plot in Fig. 3 the $\Delta Z/Z$ and H_K as functions of Fe-doped content. The values of $\Delta Z/Z$ and H_K were taken at $f = 3$ MHz. The values of H_K estimated from the measured M-H curves are also included in Fig. 3 for comparison. Clearly, the $\Delta Z/Z$ and H_K vary with Fe doping in an opposite trend. While the $\Delta Z/Z$ decreases with Fe doping from $x = 0$ to 0.05 and increases for $x = 0.1$, the H_K increases from $x = 0$ to 0.05

FIG. 2. (Color online) Frequency dependences of (a,b) the maximum GMI ratio ($[\Delta Z/Z]_{\max}$) and (c,d) the maximum field sensitivity of GMI ($\eta_{\max}$) for $(\text{Co}_{1-x}\text{Fe}_x)_{89}\text{Zr}_7\text{B}_4$ ($x = 0, 0.025, 0.05, 0.10$) amorphous and nanocrystalline ribbons.

and decreases for $x = 0.1$. This is respectively related to the decrease and increase of μ as the Fe-doping level is increased from $x = 0$ to 0.1. Interestingly, the changing trend of H_K with Fe doping determined from the GMI data exactly follows that determined from the M-H data. XRD and TEM studies have revealed that $(\text{Co}_{1-x}\text{Fe}_x)_{89}\text{Zr}_7\text{B}_4$ ($x = 0, 0.025, 0.05$, and 0.1) nanocrystalline ribbons possess multiple nanocrystalline phases: face-centered cubic (FCC) and HCP for $x = 0$; body-centered cubic (BCC), FCC, and HCP for $x = 0.025$; BCC and FCC for $x = 0.05$; BCC for $x = 0.1$.¹⁵ By comparing the values of $\Delta Z/Z$ and H_K between the $x = 0.05$ sample with BCC and FCC phases and the $x = 0.1$ sample with BCC phase, we note that the absence of the FCC phase leads to the decrease of H_K and the increase of $\Delta Z/Z$ in the latter sample. The peak in the anisotropy field occurs in the middle of the single FCC field on

the equilibrium Fe-Co phase diagram. This result is of particular interest given the observation¹³ of both bcc and close-packed nanocrystals in what is ostensibly a single phase region in the equilibrium phase diagram. This was further correlated with Fe-depleted and Fe-rich nanoparticles as observed by atom probe field ion microscopy. This suggests that a cooperative exchange of Fe(Co) atoms through the amorphous matrix has occurred. To the extent that a magnetic field may bias the nanoparticle growth, and the angular correlations between Fe-rich and Fe-poor nanoparticles, this may influence the induced anisotropy. The increase of H_K and the decrease of $\Delta Z/Z$ with increase of x from 0 to 0.05 also suggest that the volume fraction of the FCC phase increases as the Fe-doping level is increased. These results indicate that GMI can be used as a useful research tool for probing variations of field-induced magnetic anisotropy and hence microstructural changes in nanocrystalline magnetic materials.

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FIG. 3. (Color online) The GMI ratio ($\Delta Z/Z$) and the field-induced magnetic anisotropy fields (H_K), determined from the GMI data and the M-H data, are plotted as functions of Fe-doped content for $(\text{Co}_{1-x}\text{Fe}_x)_{89}\text{Zr}_7\text{B}_4$ ($x = 0, 0.025, 0.05, 0.10$) nanocrystalline ribbons.

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