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Enhanced high-frequency magnetoresistance responses of melt-extracted Co-rich soft ferromagnetic microwires

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We present the relationships between the structure, magnetic properties and high-frequency magnetoresistance (MR) effect in melt-extracted $Co_{68.2}Fe_{4.3}B_{15}Si_{12.5}$ microwires subject to thermal annealing. In order to release residual stresses to improve the magnetic softness while retaining the good mechanical property of an amorphous material, microwire samples were annealed at different temperatures of 100, 200, 350, 400 and 450 ^{o}C for 15 minutes. We have shown that relative to an as-cast amorphous microwire, annealing microwires at T_a = 100, 200 and 350 ^{0}C improved both the magnetic softness and the MR effect, while an opposite trend was observed for the microwires annealed at T_a = 400 and 450 ^{o}C. We have observed a distinct difference in the frequency dependence of MR response (ξ) for dc applied magnetic fields below and above the effective anisotropy field of the microwires. While the microwire annealed at 200 ^{o}C shows the largest MR ratio (~580%) at 100 MHz, the highest value of ξ (~43 %/Oe) has been achieved at 400 MHz for the microwire annealed at 350 ^{o}C. These results indicate that the optimally annealed $Co_{68.2}Fe_{4.3}B_{15}Si_{12.5}$ microwires are attractive candidates for high-frequency sensor applications.

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1. Introduction

Soft ferromagnetic amorphous materials have found many technological and industrial applications, such as in high-frequency transformers, magnetic shielding devices, and magnetic sensors [1-3]. Among them, Co-rich amorphous microwires exhibiting giant magnetoimpedance (GMI) effects have emerged as an excellent candidate for high-performance magnetic sensor applications [3-6]. These microwires are being used in smart phone devices and gauss magnetometers [7]. The microwires have also been used to fabricate magnetic biosensors for biomolecular detection [8] and for sensing weak electromagnetic fields in biological systems [9]. GMI refers to a large change in the ac impedance ($Z = R + j.X$, where R and X are the ac resistance and reactance, respectively) of a ferromagnetic conductor subject to a dc magnetic field, and its origin can be understood from the dc magnetic field-induced changes in circular permeability and electrical resistance that alter the magnetic penetration depth of an ac current and hence the ac impedance of the material [4].

Recently, there has been a growing interest in the development of melt-extracted Co-rich amorphous microwires due to their excellent magnetic softness and GMI responses [10-12]. While these works focused on exploring the GMI effects in the low and intermediate frequency ranges up to 100 MHz, the GMI response of the microwires in the high frequency range of 100 – 1000 MHz has not been yet exploited. Since the realization of large GMI values at high frequency ranges up to 1 GHz becomes increasingly important in high-performance magnetic sensor devices [4,5,13-16], there is a pressing need for improving the high-frequency GMI response of these microwires.

In this paper, we have performed the first systematic study of the high-frequency magnetoresistance (MR) effect and its field sensitivity and their relationships with the magnetic softness and microstructure of melt-extracted $Co_{68.2}Fe_{4.3}B_{15}Si_{12.5}$ microwires subject to thermal annealing.

2. Experiment

Soft ferromagnetic amorphous microwires of a nominal composition $Co_{68.2}Fe_{4.3}B_{15}Si_{12.5}$ were prepared by the melt-extraction technique, details of the sample fabrication has been reported elsewhere [10]. Microwire samples were annealed at 100, 200, 350, 400, and 450 ^{o}C for 15 minutes. The structures of the as-cast and annealed microwires were examined by X-ray diffraction using Bruker AXS D8 Focus Diffractometer. The surface topology of the samples was analyzed by Scanning Electron Microscope (SEM). Magnetic hysteresis measurements were performed at room temperature, using a vibrating sample magnetometer (VSM) probe that has been integrated inside the Physical Property Measurement System (PPMS) from Quantum Design. Magnetoimpedance measurements were carried out along the wire axis in applied dc magnetic fields of up to ±115 Oe over a frequency range of 100 MHz – 1 GHz, using a microtrip line method. Microwire specimens of the length 1.5 μm were used for these measurements. A Helmholtz coil (diameter ~30 cm) produced the dc magnetic field (H_{dc}) that is perpendicular to the ac magnetic field (H_{ac}) generated by an applied I_{ac} of 5 mA flowing along the wire. An impedance analyzer was used to measure the absolute value of the ac impedance of the sample at room temperature. All of the electronic instruments were controlled using LabVIEW. For the high-frequency impedance analysis, we are interested in the real component of the ac impedance, namely, the ac resistance (R). The change in R, defined as the magnetoresistance (MR) ratio, has been calculated as

$$\frac{\Delta R}{R}(\%) = \frac{R(H)-R(H_{\max})}{R(H_{\max})} \times 100(\%)\,, \tag{1}$$

where $R(H)$ and $R(H_{max})$ are the resistance values of the microwire in the measured magnetic field and the maximum field, respectively. ξ is the field sensitivity of R in response to the dc magnetic field, which has been calculated by

$$\xi\,(\%/\mathrm{Oe}) = \frac{d\left(\frac{\Delta R}{R}\right)}{d(H)}. \qquad (2)$$

2. Results and Discussion

We first studied the influence of thermal annealing on the microstructure of the $Co_{68.2}Fe_{4.3}B_{15}Si_{12.5}$ microwires. Figure 1 shows the XRD patterns of the as-cast and annealed microwire samples. It can be observed that the XRD pattern of the as-cast microwire exhibits only one broad peak around 2θ = 45°, which is often known as a diffuse halo, indicating that the microwire prepared by melt-extraction is amorphous in nature. Annealing microwires at T_a = 100, 200 and 350 °C did not alter the amorphous state. When annealed at T_a = 400 and 450 °C, however, the microwire samples show crystalline features, which are most pronounced in the 450 °C annealed sample. It is likely that the low temperature annealing ($T_a \leq 350$ °C) released residual stresses without altering the amorphous structure of the microwires, while the high temperature annealing ($T_a > 350$ °C) caused the surfaces of the microwires to crystallize partially [17].

To further clarify this, the surface topology of all the as-cast and annealed microwire samples was examined by SEM, and the obtained results are displayed in Figure 2a-f. It can be seen that the surfaces of the as-cast sample and those annealed at T_a = 100, 200 and 350 °C are quite smooth and uniform (Figure 2a-d), while noticeable modifications in the surface layer appear to occur in the microwire samples annealed at T_a = 400 and 450 °C (Figure 2e,f). We recall that the smooth surfaces of the microwires are favorable for obtaining the GMI effects [4,5]. For the microwire samples annealed at 400 and 450 °C, the surface modifications seen in SEM images

(Figure 2e,f) are correlated with the microstructural variations noted in the XRD patterns (Figure 1). Such changes in the microstructure and surface topology would affect the magnetic softness and the MR effect, as shown below.

Figure 3a shows the room-temperature magnetic hysteresis loops of the as-cast and annealed microwire samples. As one can see clearly in this figure, while the coercive field (H_C ~2.5 Oe) remained almost unchanged with annealing, the saturation magnetization (M_S) first increased with an increase in T_a up to 200 ^oC, reached a maximum at 200 ^oC, and then decreased for T_a = 350, 400, and 450 ^oC (Figure 3b). As compared to the as-cast amorphous microwire, the increase in M_S for the microwires annealed at T_a = 100, 200 and 350 ^oC is mainly attributed to the release of residual stresses or removal of surface defects, while the decrease in M_S for the microwires annealed at T_a = 400 and 450 ^oC is related to the crystallized surfaces of the microwires. Such crystallization caused the surface layer of the microwire to exhibit a harder magnetic characteristic, which is different from the soft magnetic inter layer. This would cause a significant reduction in the MR effect in these samples.

Figure 4a-c shows the magnetic field dependence of MR ratio ($\Delta R/R$) for the as-cast and annealed microwire samples at f = 100, 500, and 800 MHz, respectively. In general it is observed that with increasing T_a, $\Delta R/R$ first increased, reached a maximum for T_a = 200 ^oC, and then decreased for $T_a \geq 350$ ^oC. As compared to the as-cast microwire, the larger values of $\Delta R/R$ are achieved in the microwires annealed at T_a = 100, 200, and 350 ^oC. For the microwires annealed at 400 and 450 ^oC, however, the $\Delta R/R$ values are much smaller. This trend can be more clearly seen in Figure 4d, which shows the frequency dependence of maximum $\Delta R/R$ (denoted as $[\Delta R/R]_{max}$). It is worth mentioning that all the microwire samples show a double-peak feature near zero field, which has been attributed to the presence of circular magnetic anisotropy as a result of the circular magnetic domain structure formed during fabrication for typical Co-rich amorphous microwires [4,5]. More interestingly, the depth in MR profiles seen around zero field appears to develop with an increase in the measurement frequency. This feature is desirable for highly sensitive sensor applications as such field-induced changes in resistance can be achieved in a small magnetic field range [18].

To fully exploit these interesting features, we have plotted in Figure 5a the dc magnetic field dependence of derivative of $\Delta R/R$, which has been defined above in Eq. (2) as the field sensitivity of the sensor (ξ). It can be observed that owing to the double–peak feature of MR curves, ξ possesses two maxima (ξ_{max}) at two dc magnetic field regimes; $H > H_K$ and $H < H_K$, where H_K is an effective magnetic anisotropy field (Figure 5b). It is important to note here that for the case of $H > H_K$, ξ_{max} has a decreasing trend with increasing frequency for all samples investigated (Figure 5c). In the frequency range of 100-1000 MHz, the largest values of ξ_{max} are achieved for the microwire annealed at 200 ^{o}C, which showed the best magnetic softness (the highest value of M_S). Although the M_S and $\Delta R/R$ of the microwire annealed at T_a = 100 ^{o}C are greater than those of the as-cast microwire, the latter possesses higher values of ξ_{max} in the entirely investigated frequency range. In case of the microwire annealed at 350 ^{o}C, there exists a crossover frequency above which this sample shows higher values of ξ_{max} as compared to the as-cast microwire.

For the case of $H < H_K$, ξ_{max} (f) shows a different variation trend. As one can see clearly in Figure 5d, ξ_{max} first increased with frequency, reached a maximum at a characteristic frequency (~400 MHz), and then decreased for higher frequencies. The microwire annealed at 350 ^{o}C shows the largest values of ξ_{max} in the entirely investigated frequency range. ξ_{max} = 34%/Oe at f = 400

MHz for this sample. This value is greater and obtained at a much higher frequency as compared to the case of $H > H_K$.

4. Conclusions

Having systematically investigated the structure, magnetic properties and high-frequency magnetoresistance responses of melt-extracted $Co_{68.2}Fe_{4.3}B_{15}Si_{12.5}$ microwires subject to thermal annealing, the followings are worthy of notes:

i. Short-time and low-temperature annealing can release residual stresses or remove surface defects without altering the amorphous structure (the preserved good mechanical strength) and the circular magnetic domain structure, leading to overall enhancements of the magnetic softness and MR effect.

ii. High-temperature annealing can destroy the amorphous state, reducing the mechanical strength, and significantly alter the desirable circular magnetic domain structure, which, in effect, degrades both the magnetic softness and the MR response.

iii. For the case of $H > H_K$, the frequency dependence of ξ_{max} follows that of $[\Delta R/R]_{max}$, and both parameters appear to decrease with increasing frequency.

iv. For the case of $H < H_K$, however, the frequency dependence of ξ_{max} does not follow that of $[\Delta R/R]_{max}$, and there exists a certain frequency at which ξ_{max} reaches the highest value.

These findings are of practical importance in exploiting the low-field MR responses of melt-extracted Co-rich microwires for high-frequency sensing applications.

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Figure captions

Figure 1: XRD patterns of as-cast and annealed microwires of $Co_{68.2}Fe_{4.3}B_{15}Si_{12.5}$. The microwires were annealed at 100, 200, 350, 400, and 450 ^{o}C for 15 minutes.

Figure 2: SEM images of as-cast and annealed microwires of $Co_{68.2}Fe_{4.3}B_{15}Si_{12.5}$; (a) as-cast and annealed at (b) 100, (c) 200, (d) 350, (e) 400, and (f) 450 ^{o}C for 15 minutes.

Figure 3: Magnetic hysteresis loops taken 300 K for the as-cast and annealed $Co_{68.2}Fe_{4.3}B_{15}Si_{12.5}$ microwire samples.

Figure 4: The dc magnetic field dependence of magnetoresistance ratio ($\Delta R/R$) for the as-cast and annealed microwire samples at f = 100 (a), 300 (b) and 800 MHz (c). (d) the frequency dependence of maximum MR ratio ($[\Delta R/R]_{max}$) for the as-cast and annealed microwire samples.

Figure 6: The field sensitivity of MR (ξ) for the as-cast and annealed microwire samples at f = 200 (a), 600 (b), and 1000 MHz (c). (d) the maximum sensitivity of MR (ξ_{max}) as a function of frequency for the as-cast and annealed microwire samples.