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Influence of Fe on magnetoimpedance effect of $Co_{72-x}Fe_xZr_8B_{20}$ alloys

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Abstract

The magnetoimpedance (MI) of as-spun amorphous $Co_{72-x}Fe_xZr_8B_{20}$ (CFZB, $x = 0, 2, 5, 7$ at.%) alloys has been measured at a frequency ranging from 310 to 1110 kHz and a dc magnetic field between 0 to 70 Oe. The maximum MI ratio $\Delta Z/Z(\%)_{\max}$ of the CFZB alloys increases with increasing frequency for each sample and decreases with the increment of Fe content at any frequency. The most drastic MI effect, the maximum value of about 90% at 1110 kHz accompanied by the field sensitivity of 18% Oe^{-1}, is observed in CFZB without Fe. Magnetic measurements reveal that coercivity of CFZB decreases first and then increases with increasing Fe content, and the minimum value of about 1.6 Oe is obtained in the sample with $x = 2$ at.%. The permeability of CFZB was evaluated from quasi-static hysteresis loops, indicating that the Fe-free CFZB is of the largest permeability. Measurements on electrical resistivity suggest that the Fe-free sample possesses smaller resistivity while the sample with $x = 7$ at.% has a much larger resistivity compared with CFZB with 2 and 5 at.% Fe. The correlation between MI effect and magnetic properties together with electrical resistivity for the CFZB alloy is discussed in this paper.

1. Introduction

The magnetoimpedance (MI) effect, which consists of a change in the total impedance of a magnetic conductor under application of a magnetic field, seems to be very promising and stimulating for technological applications in recording heads and magnetic sensors [1–4]. Generally speaking, the origin of the MI effect is ascribed to the classical skin effect. At a given frequency, the application of a dc magnetic field changes the transverse permeability (for ribbon) or circumferential permeability (for wire) and hence the electromagnetic penetration depth that in turn alters both the resistive and the inductive components of the total impedance. Since interest in the MI effect was triggered in 1992, when Mohri *et al* [5] discovered a large MI change in Co-based amorphous wires at relatively low frequencies, the MI effect has been investigated in a variety of Co- and Fe-based wires, ribbons, thin films, multilayers, etc [6–13].

It is well known that Co-based amorphous alloys possess excellent soft magnetic properties that play an important role in the MI effect. According to a number of previous studies, however, the MI effect observed in Co-based amorphous alloys in their as-quenched state is not very large. For example, the MI effect in as-quenched CoFeSiB microwires is very low for frequencies below 1 MHz and reaches the maximum of about 20% in the frequency range 5–10 MHz [10]. In another case, the largest MI ratio is only 60% in the as-cast condition of $Co_{68.25}Fe_{4.5}Si_{12.25}B_{15}$ amorphous ribbon at frequency $f = 2$ MHz [11]. The highest magnitude of MI value, 58% in microwires, is reported for the as-quenched

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CoMnSiB microwire at about 1.5 MHz [12]. With respect to Fe-based materials, a distinct MI effect has not been achieved in any as-quenched Fe-based amorphous alloys due to their large positive magnetostriction up to now, except for the $Fe_{71}Zr_5B_{20}Nb_4$ amorphous ribbon [14].

It has been established that the efficiency of MI elements made of low magnetostrictive amorphous materials can be improved by inducing transverse magnetic anisotropy, using an appropriate heat treatment (Joule-heating annealing, field annealing or stress annealing). Moreover, the MI effect of Fe-based materials can be enhanced through heat treatment by the formation and moderate growth of the nanocrystalline phase [15–18]. However, since the annealing temperature and time are not easy to master and control, as well as the annealed nanocrystalline alloys compared with the amorphous materials do not provide the necessary manufacturing flexibility, it is important to study the MI effect of alloys in their amorphous state, seek good MI materials in the as-cast condition and investigate the factors that play a key role in the effect.

Inoue *et al* have found that Co-based amorphous alloys with glass transition and supercooled liquid region are formed in the Co–Fe–M–B (M = Zr, Nb) systems and exhibit good soft magnetic properties with high permeability. In their work [19], $Co_{72-x}Fe_xZr_8B_{20}$ (x = 0–21 at.%) alloys, with permeability of 5500–18 300 in the frequency range 1–10^3 kHz and low magnetostriction between -1.5×10^{-6} and $+10\times10^{-6}$ including zero, have been intensively studied. Based on their reports, we prepared $Co_{72-x}Fe_xZr_8B_{20}$ (x = 0, 2, 5, 7 at.%) amorphous alloys (CFZB). In this article, our efforts have been devoted to investigating the influence of Fe content on MI effect of the as-quenched CFZB amorphous ribbons and highlighting the factors that affect the MI behaviour.

2. Experimental procedures

The CFZB ribbons, made by the melt-spinning technique in an argon atmosphere with a roll speed of about 25 m s^{-1}, had a reasonably uniform thickness (35–40 μm) and width (1.5 mm). The microstructure was studied by the x-ray diffraction (XRD) technique using Cu Kα radiation. Magnetization measurements at room temperature were carried out using a vibrating sample magnetometer (VSM). Electrical resistivity measurements were performed by the four-terminal method.

A Solartron 1260 impedance analyser was used for impedance measurements. A four-probe ac technique was used to measure the magnitude Z of the MI as a function of the field H applied along the ribbon length (about 50 mm), i.e. parallel to the ac measuring current. The current amplitude was kept constant (I = 3 mA) and the frequency f was between 310 and 1110 kHz. The external field H was generated by a solenoid with axis perpendicular to the Earth's magnetic field to allow field variation from 0 to 70 Oe.

3. Results

The XRD of all as-spun CFZB ribbons are shown in figure 1, each of which exhibits one broad diffused diffraction peak. This indicates that all the samples are amorphous alloys.

The impedance Z of as-spun amorphous CFZB, as a function of the external longitudinal dc field, was measured at f = 310, 510, 810, 910 and 1110 kHz. The MI ratio $\Delta Z/Z(\%)$ denotes $[Z(H) - Z(H_{max})]/Z(H_{max})$, in which $Z(H_{max})$ is the impedance value under the maximum magnetic field of 66 Oe. Figure 2 shows the field dependence of the MI ratio of the CFZB ribbon with x = 2 at various frequencies. It is found that the MI curves for CFZB with 2 at.% Fe show single-peak behaviour with the applied magnetic field under the measuring frequencies. In general, there are two types of MI curves, which depend on the effective anisotropy in the sample [18, 20, 21]. One is the single-peak curve, which is usually observed in samples with very small induced or intrinsic anisotropy and the very high MI ratio due to the magnetization rotation in a magnetic field applied along the easy magnetization direction. The other is the double-peak curve, which is usually obtained in samples with non-zero induced magnetic anisotropy, where the maximum MI ratio appears when an applied magnetic field is equal to about the value of the effective anisotropy field. In the present work, the MI curves for the as-quenched CFZB alloys belong to the

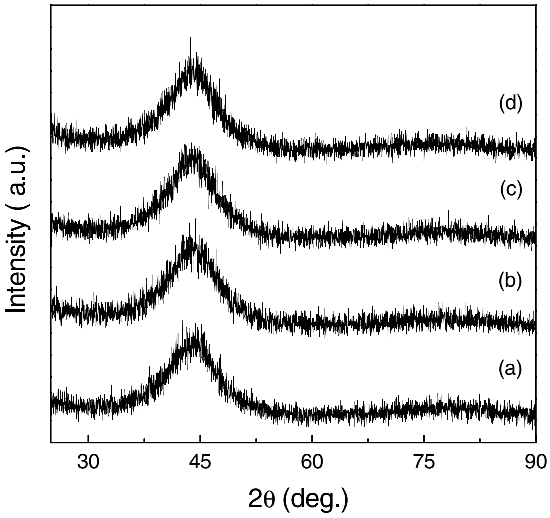

Figure 1. XRD patterns of $Co_{72-x}Fe_xZr_8B_{20}$ (x = 0, 2, 5, 7) melt spun ribbons with Fe content of (*a*) 0, (*b*) 2, (*c*) 5 and (*d*) 7 at.%.

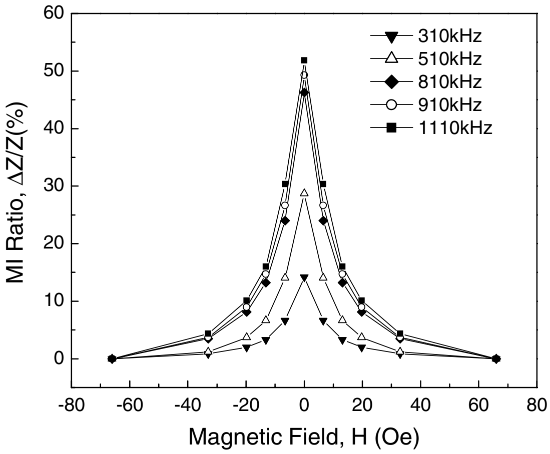

Figure 2. Field dependence of MI ratio, defined as $\Delta Z/Z(\%) = [Z(H) - Z(H_{max})]/Z(H_{max})$, for the sample with 2 at.% Fe at measuring frequency f = 310, 510, 810, 910 and 1110 kHz.

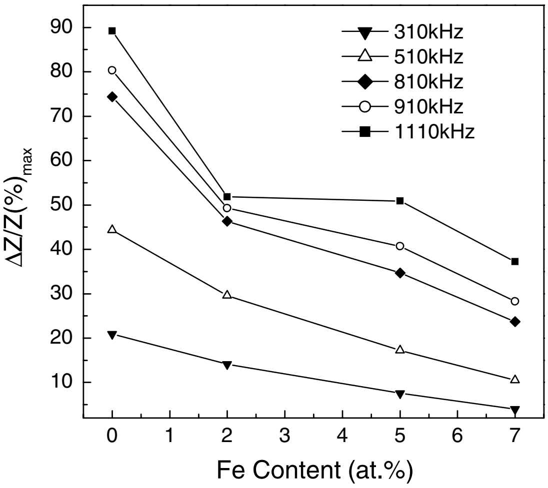

Figure 3. Dependence of the maximum MI ratio ($\Delta Z/Z(\%)_{max}$) on Fe content in CFZB at measuring frequency $f = 310, 510, 810, 910$ and 1110 kHz.

Table 1. Field sensitivity of MI effect for $Co_{72-x}Fe_xZr_8B_{20}$ ($x = 0$, 2, 5, 7 at.%) amorphous alloys at measuring frequency $f = 310$, 510, 810, 910 and 1110 kHz.

	ξ(% Oe^{-1})				
$Co_{72-x}Fe_xZr_8B_{20}$	310 kHz	510 kHz	810 kHz	910 kHz	1110 kHz
$x = 0$	5.2	10.7	16.3	16.6	17.9
$x = 2$	2.3	4.4	6.4	6.3	6.0
$x = 5$	1.2	2.7	4.2	4.2	4.7
$x = 7$	0.7	2.0	3.8	4.3	4.5

former case. In addition, as observed in figure 2, the MI effect is more obvious in the high frequency range ($f \geqslant 810$ kHz) than in the low frequency range ($f \leqslant 510$ kHz) and the MI ratio decreases rapidly and then slowly with increasing applied field at frequencies above 810 kHz. As the frequency decreases, the MI effect for the sample becomes very much diminished. The largest MI value observed in the sample is about 52% at a frequency of 1110 kHz. The MI effect in the CFZB samples with some other Fe content also displays a similar changing tendency.

The changes in the maximum MI ratio ($\Delta Z/Z(\%)_{max}$) with Fe content at various frequencies are plotted in figure 3. It can be noted that $\Delta Z/Z(\%)_{max}$ increases with frequency for each sample. At any frequency, $\Delta Z/Z(\%)_{max}$ decreases with increasing Fe content. Many previous research results have confirmed that the maximum MI ratio increases with increasing driving frequency and then decreases when the frequency exceeds a critical value [15,22]. The decrease in the MI effect at high frequencies is due to the strong suppression of the domain wall movement by the eddy current effect, which causes an extreme decrease in the permeability [15,16,23,24]. Unfortunately, the highest frequency can only reach 1110 kHz under our experimental conditions, so we are unable to give full $\Delta Z/Z(\%)_{max}$-frequency curves. Besides, it can be observed from figure 3 that the MI effect in CFZB alloy with $x = 0$ exhibits a distinct frequency response, while the effect in the samples with some other Fe content does not show this feature. Most prominently, the maximum MI value as large as 90% is obtained in the Fe-free CFZB at 1110 kHz, which is much larger than the values achieved in other as-cast Co-based amorphous alloys [10–12] and makes the alloy more promising for technical applications in different kinds of high sensitivity micromagnetic sensors.

To clarify the field response of the MI effect of CFZB alloys at measuring frequencies, we calculate the field sensitivity of the MI effect for the samples, which is expressed as [16]

$$\xi = \frac{2[\Delta Z/Z(\%)_{max}]}{\Delta H}, \tag{1}$$

where ΔH is the full width at half maximum of the MI response. The calculated results shown in table 1 indicate that ξ of almost all the studied alloys increases with frequency except for $x = 2$ and the Fe-free CFZB possesses the highest field sensitivity at any frequency, especially 18% Oe^{-1} at 1110 kHz.

4. Discussion

Many research results have confirmed that the MI effect is closely associated with soft magnetic properties of materials. Generally, an obvious MI effect can be observed in alloys with a relatively small coercivity H_c [8,15,25]. Moreover, near zero magnetostriction λ_s is also proved to be essential for the MI ratio [2,25,26]. Co-based amorphous alloys, which have been found to exhibit good soft magnetic properties and possess small negative magnetostriction, are excellent MI materials. As λ_s depends on alloy composition, both the sign and value of magnetostriction of Co-based alloys may change by the proper increment of some elements. For example, by a small addition of Fe or Mn, Co-based alloys with near zero magnetostriction were obtained [27]. Inoue *et al* [19] indicated that λ_s of $Co_{72-x}Fe_xZr_8B_{20}$ alloys shows a negative value of -1.5×10^{-6} at $x = 0$ at.% Fe, changing to positive values passing through zero around 2 at.% Fe and the further increment in Fe content causes an increase in the positive λ_s. Besides, according to our results of the magnetic measurements on CFZB, as shown in figure 4(*a*), the coercive force of CFZB with $x = 2$ is less than 1.6 Oe which is the smallest value among all the CFZB alloys, indicating CFZB with $x = 2$ exhibits excellent soft magnetic properties. However, CFZB alloy with 2 at.% Fe does not show the most prominent MI effect, whereas the sample with 0 at.% Fe, which shows a negative magnetostriction constant and coercivity of over 2.2 Oe, exhibits the largest MI ratio. Based on the above results and analysis, it is deduced that there must exist other factors dominating the MI effect of CFZB amorphous alloys.

For better understanding of the mechanism responsible for the drastic MI effect of CFZB with $x = 0$, longitudinal hysteresis loops of CFZB have been studied and plotted in figure 5. As shown in figure 5, the longitudinal hysteresis loop shape of CFZB changes evidently with Fe content, vividly indicating that the anisotropy field changes sensitively with Fe content. In addition, judging from the inclination grade of hysteresis loops of CFZB alloys, it is deduced that permeability (μ') also changes with the increment in Fe content. The inset in figure 5 shows Fe content dependence of permeability evaluated from quasi-static hysteresis loops, indicating that the permeability of CFZB begins to decrease at $x = 2$ and

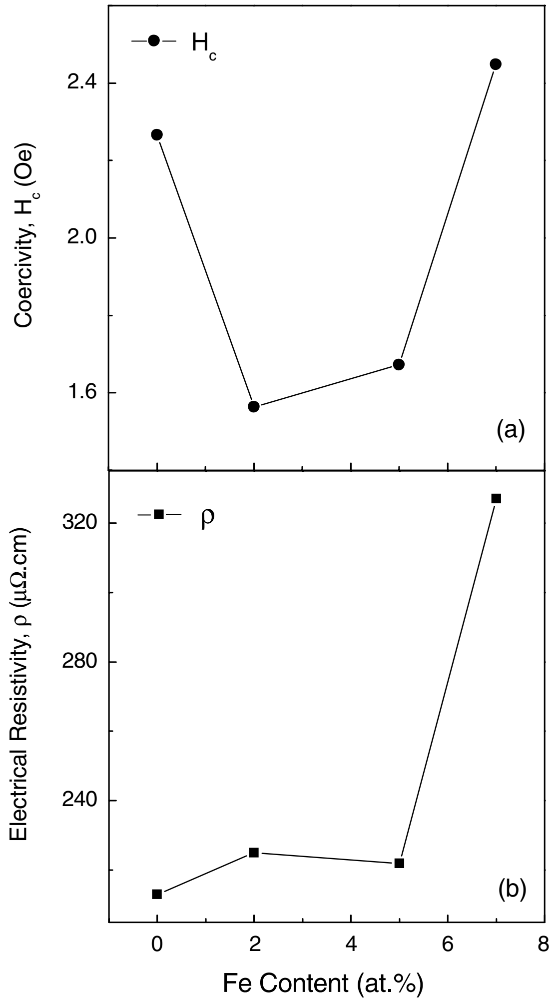

Figure 4. Dependence of coercivity H_c (*a*) and electrical resistivity ρ (*b*) on Fe content for CFZB alloys.

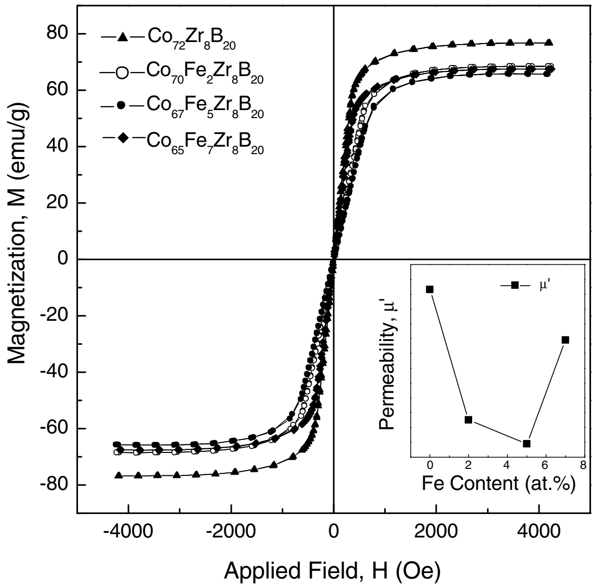

Figure 5. The longitudinal hysteresis loops of amorphous CFZB. The inset of the figure shows the change of permeability evaluated from hysteresis loops.

increase at $x = 7$ and the sample with $x = 0$ has the largest permeability. (Study of the transverse hysteresis loops of CFZB also suggests the sample without any Fe content possesses the largest permeability.)

Since the large MI effect has been observed in amorphous ferromagnets and not in ordinary crystalline alloys, the role of large permeability as a result of very small magnetic anisotropy inherent to amorphous materials is strongly suggested. During the rapid quenching process, variation in the super-cooling rate across the whole section of the ribbon results in internal stress and a difference in microstructure over the ribbon cross-section [17, 28]. Co-based amorphous alloys with $\lambda_s > 0$ or $\lambda_s < 0$ are observed in a maze domain structure, which indicates the existence of tensile stress regions for alloys with positive magnetostriction and compressive stress regions for alloys with negative magnetostriction. Complicated domain structure describes inhomogeneous stress distribution in different regions of material (called stress fluctuation) which, coupling with magnetostriction, produces local anisotropy that affects domain motion and thus leads to a decline in permeability and increment in coercivity. Therefore, magnetic permeability, which is the essential factor for the MI effect in CFZB amorphous alloy, is closely connected with the magnetostriction, stress distribution and magnetic anisotropy—and hence the domain structure—of the CFZB material. However, due to the complexity of the domain structures and the domain wall motion in an applied field, as well as the lack of a good technique for characterization, the microstructure still needs to be further studied. Furthermore, it has been noticed that the magnitude of the MI value and the changing tendency of the MI effect are closely associated with the variation of permeability [29,30], so it is inferred that CFZB with $x = 0$ may also be provided with a large change of permeability under an applied magnetic field at a different frequency, thus exhibiting the most drastic MI effect. Besides, according to the work of Hernando and Prida *et al* [16, 18], the highest field sensitivity of the MI effect corresponds to the samples with magnetostriction coefficients close to zero and slightly negative, so the highest sensitivity of 18% Oe^{-1} is obtained for CFZB with 0 at.% Fe, which possesses the small negative magnetostriction.

Moreover, the electrical resistivity (ρ) of CFZB alloys has been measured and Fe content dependence of ρ is plotted in figure 4(*b*). A lot of previous studies [16–18] have demonstrated that small resistivity is one of the general conditions that must be satisfied by MI material. It is due to the fact that the MI effect has a classical electromagnetical origin being related to the penetration depth δ, which is defined as (in CGS units) [31]

$$\delta = 5030\left(\frac{\rho}{\mu f}\right)^{1/2}, \qquad (2)$$

where ρ is the electrical resistivity, μ is the transverse magnetic permeability and f is the driving field frequency. To achieve large MI effect, the penetration depth should be very small in the absence of any applied field, which can be increased with the application of a dc magnetic field. High value of frequency, large transverse magnetic permeability and low value resistivity give rise to a small penetration depth in the absence of any external magnetic field. According to some research results [22,32], the smaller the electrical resistivity of the sample, the larger the MI value obtained. From figure 4(*b*), as expected, it is noted that the electrical resistivity of the sample with $x = 0$ is smaller while that of the sample with $x = 7$ is much larger compared with CFZB with 2 and 5 at.% Fe. This also interprets why a drastic MI effect is obtained in CFZB without Fe content whereas the effect in CFZB with $x = 7$ is very small although the permeability of the two samples is very close.

In summary, we attribute the notable maximum MI ratio of CFZB alloy with $x = 0$ to high permeability as well as its large change with applied field and smaller electrical resistivity.

5. Conclusions

The MI effect in $Co_{72-x}Fe_xZr_8B_{20}$ ($x = 0, 2, 5, 7$) amorphous alloys with good soft magnetic properties has been intensively studied in the frequency range $310\,kHz \leqslant f \leqslant 1110\,kHz$. The maximum MI ratio of CFZB increases with frequency for each sample, while it decreases with the increment in Fe content at any frequency. The sample with 0 at.% Fe content does not possess the smallest coercive force and near zero magnetostriction, but it shows the most prominent MI value of 90% at 1110 kHz coincident with the largest field sensitivity of 18% Oe^{-1}. The correlation of the MI effect with the magnetic properties provides a strong indication that the mechanism responsible for the effect involves the microstructure in the sample. Analysis of the effect conclusively demonstrates that high permeability determined by the domain structure of the material, large change of permeability with applied magnetic field and smaller electrical resistivity play the dominant roles in this MI effect.

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