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Magnetic properties and giant magneto-impedance in amorphous FeNiCrSiB films

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Abstract. The giant magneto-impedance effect has been observed at room temperature in amorphous FeNiCrSiB films prepared by RF sputtering followed by an annealing treatment. The frequency spectra and field dependence of the effect are studied in the frequency range from 100 kHz to 13 MHz and the magnetic field range from 0 to 70 Oe. The longitudinal, transverse and perpendicular effects are observed in detail. A transverse magneto-impedance effect with a magnitude similar to the longitudinal case has been obtained, and a large perpendicular MI effect is also obtained. A maximum GMI ratio of about 38% is obtained at 13 MHz in the longitudinal and transverse cases. The field sensitivity of the longitudinal GMI is 3.4% Oe⁻¹ at 13 MHz. A large perpendicular MI ratio of 11% is obtained at 11 MHz. The effect of annealing on the effective permeability and GMI is reported in this paper.

1. Introduction

The magneto-impedance (MI) effect is a phenomenon in which the voltage induced by a high-frequency ac current in a ferromagnet changes with the application of an external dc field [1]. In the low-frequency region, the voltage change is due to a decrease of the averaged internal inductance L, which relates to the transverse permeability with respect to the applied ac current. This is a magneto-inductive effect in which the inductance of the ferromagnet responds to the external field via the permeability of the material. At high frequency, the skin effect is evident. The external field sensitively affects the penetration depth via the permeability. So both resistive and inductive components in the impedance sensitively depend on the external field due to the skin effect. This effect is referred to as giant magneto-impedance (GMI) [2, 3].

The GMI effect is very attractive because of its high sensitivity at low fields, rapid response, lack of hysteresis and high temperature stability. This phenomenon has applications in field sensing and magnetic recording heads. Earlier works focused on zero—or slightly negative—magnetostrictive Co-based amorphous wires [4–6]. Now the investigations on the GMI effect have been extended to Co-based amorphous ribbons and films. Fe-based nanocrystalline or amorphous alloys with slightly positive magnetostriction have excellent soft magnetic properties, better than those of Co-based amorphous alloys [7], making them a good candidate material for GMI elements. Thin-film magnetic sensors are very useful in micromagnetic techniques. Thin films are also the preferred media for

exploring the MI phenomena, which remain to be understood in detail. However very few works deal with the GMI effect in films, especially at relatively low frequencies [8]. So far, most studies on the MI effects are on the longitudinal MI effect, in which the external magnetic field is applied along the direction of the probe current. Fewer transverse MI effects have been reported, where the magnetic field is applied perpendicular to the current direction in the film plane [9]. Very few reports to date are on the perpendicular MI effect, where the magnetic field is applied perpendicular to the film plane [10].

In this paper we report observations on the large MI effects in amorphous FeNiCrSiB films made by RF sputtering. It includes the longitudinal and transverse MI with a similar size of the effects and the perpendicular MI at relatively low frequencies.

2. Experimental details

The samples were prepared by RF sputtering. The target was an alloy of composition $(\text{FeNi})_{77.5}\text{Cr}_{0.5}(\text{SiB})_{22}$ (at.%). The distance between the target and the substrate holder was about 4 cm, both of which were water cooled. After the chamber system was evacuated to a high vacuum of about 5×10^{-6} Torr, 99.999% pure Ar gas was introduced and controlled at a pressure of 5×10^{-3} Torr during sputtering. The target surface was cleaned by pre-sputtering for 30 min before deposition. The substrates were glass slides of 0.2 mm thickness. The deposition rate was 1.4 Å s⁻¹. The as-prepared films of thickness 6.65 μ m were in the amorphous state. This was confirmed by x-ray diffraction. The films were then annealed in a high-vacuum system (about 1×10^{-5} Torr) at different temperatures for 20 min.

The magneto-impedance of the annealed films was measured at room temperature using an HP4192A impedance analyser, the frequency of which ranges from 5 Hz to 13 MHz. The sample was connected to the analyser with the accessory 16048B test lead which is a carefully designed unit with four coaxial cables. The measurements were carried out in the frequency range from 100 kHz to 13 MHz with a constant ac current amplitude of 10 mA. A pair of Helmholtz coils (30 cm in diameter) was used to generate an applied dc magnetic field in the range of 0–70 Oe. The coils are so placed that the applied field is perpendicular to the earth's magnetic field. Rectangular samples of dimensions 10 mm \times 3 mm have been used in the MI measurements with the probe current always in the long direction.

The magnetic properties of the films were measured by a microprocessor-controlled vibrating-sample magnetometer (VSM). The specimen used in the measurements has an area of $5 \times 5 \text{ mm}^2$. The applied magnetic field was generated by a pair of Helmholtz coils. The temperature dependences of the magnetic properties of the samples were measured using an oven from room temperature to $620 \,^{\circ}\text{C}$ with the temperature varied at a rate of about $11 \,^{\circ}\text{C} \, \text{min}^{-1}$. The sample in the oven was in a vacuum of 10^{-3} Torr in order to protect it against oxidation.

3. Results and discussion

The structure of the samples annealed at different temperatures is determined by x-ray diffraction. When the annealing temperature is lower than 400 °C, the sample is in the amorphous state. For the sample annealed from 400 to 450 °C, a smaller and broader peak around Fe(110) superimposed on a diffuse amorphous pattern can be observed in the x-ray diffraction pattern. This indicates that the nanocrystalline Fe is separated from the amorphous matrix in this film. When the annealing temperature increases, this diffraction peak becomes higher and narrower indicating the growing of Fe particles. The hysteresis



Figure 1. Typical hysteresis loops for as-prepared film (a), annealed at $350 \degree C$ (b) and annealed at $450 \degree C$ (c).



Figure 2. Dependence of saturation magnetization $4\pi M_s$ on the temperature *T* for an asprepared amorphous film. $T_1 = 447 \,^{\circ}\text{C}$ and $T_2 = 507 \,^{\circ}\text{C}$ correspond to the first and second crystallization temperatures, respectively.

loops of the films measured by VSM are shown in figure 1. The films annealed below 400 °C have good soft magnetic properties. The coercive force H_c is about 1.5 Oe and it is not sensitive to the annealing temperature below 400 °C as shown in figure 1(a) for the as-prepared film and in (b) for the film annealed at 350 °C, respectively. The inclination of the loop from the ordinate is probably caused by the demagnetization field in the transverse direction of the films and the perpendicular anisotropy in as-prepared films. The annealing process improved the uniformity of the magnetization rotation as seen by the hysteresis loop (b) in figure 1. Figure 1(c) is obtained from the film annealed at 450 °C. H_c of the film obviously increases. The saturation magnetization of the film is about 7100 G. Figure 2 shows the dependence of saturation magnetization M_s on the temperature T for an asprepared amorphous film. The Curie temperature determined from the M_s-T curve is about 346 °C. Two phase transitions T_1 and T_2 can be seen on the curve. T_1 is at about 447 °C, perhaps corresponding to the first crystallization temperature at which the nanocrystalline Fe separates from the amorphous matrix. T_2 is at about 507 °C, perhaps corresponding to the second crystallization temperature at which the amorphous matrix is crystallized.



Figure 3. The annealing temperature T_a dependence of longitudinal effective permeability μ_e at a frequency of 100 kHz.



Figure 4. The annealing temperature T_a dependence of longitudinal magneto-impedance ratio at a frequency of 13 MHz.

Figure 3 shows the dependence of the effective permeability μ_e of the film on the annealing temperature T_a . The ac current frequency used in the measurements is 100 kHz. The annealing effect is to eliminate the internal stress and to refine the magnetic domains in the film. It can be seen from figure 3 that 350 °C is the best annealing temperature at which the μ_e reaches its maximum value of about 1500. When T_a is higher than 400 °C, the crystal particles of Fe separate first from the amorphous matrix, and presumably grow larger with increasing T_a . The magnetic properties of the film are hardened by the crystallization process.

Figure 4 shows the dependence of the GMI ratio of the film, $\Delta Z/Z_{H=70} = (Z_{H=0} - Z_{H=70})/Z_{H=70}$, on the annealing temperature T_a , at a frequency of 13 MHz, where $Z_{H=0}$ and $Z_{H=70}$ are the film impedance at 0 and 70 Oe magnetic field, respectively. As one can see the film can almost be magnetized to saturation below the magnetic field of 70 Oe. Figure 3



Figure 5. Frequency dependences of the longitudinal effective permeability μ_e in the fields of 0 (circles) and 70 Oe (dots).

and figure 4 show that GMI ratio and μ_e have a good corresponding relationship, although the frequency used in the measurements is quite different. A large effective permeability corresponds to a large GMI ratio. 350 °C is an optimum annealing temperature for GMI, so we will focus our attention only on the films annealed at 350 °C. We have studied the GMI of the films annealed at 350 °C in three cases, i.e., the longitudinal GMI (LMI) with the applied magnetic field parallel to the ac current flow, the transverse GMI (TMI) with the field perpendicular to the ac current flow in the film plane and the perpendicular GMI (PMI) with the field along the film normal. Figure 5 shows the dependences of μ_e on the ac current frequency f, where the circles represent the results obtained at zero magnetic field and the dots serve for those obtained at a longitudinal field of 70 Oe. When the frequency increases, zero-field permeability decreases rapidly first below 3 MHz and then slowly at higher frequencies, but μ_e at 70 Oe is almost constant near zero in the entire frequency range. The change of permeability with the field yields the MI effect, but a big change of permeability does not certainly correspond to a large MI especially at low frequencies. Figure 6 shows the dependences of the maximum value of the LMI ratio (circles) and PMI ratio (dots) on the ac current frequency f. In the longitudinal case, the GMI ratio increases almost linearly with the increase of f, and the peak value does not appear until 13 MHz (the upper frequency limit in this measuring instrument). A maximum GMI ratio of 38% is obtained at 13 MHz. A much bigger peak value of 80% is obtained at 35 MHz by this film. We will publish these results elsewhere. Our results are quite different from those obtained by Sommer and Chien in amorphous FeCuNbSiB films [9], where the obvious GMI effects appear at very high frequencies up to several tens of MHz and the curvature of GMI ratio against f is positive. Our GMI results are obtained at much lower frequencies and have a negative curvature of GMI ratio as the frequency is over 2 MHz. The GMI effect with negative curvature has been observed usually in amorphous wires [10, 11] and ribbons [12] at lower frequencies. In the perpendicular case, GMI is much smaller than that in the longitudinal case because of the influence of a perpendicular demagnetization field; even so, a large GMI ratio up to 11% is observed. PMI reaches the maximum value at about 11 MHz.

Figure 7 shows the dependence of LMI ratio on the longitudinal applied magnetic field



Figure 6. Longitudinal (circles) and perpendicular (dots) magneto-impedance ratio versus frequency f.



Figure 7. Longitudinal magneto-impedance ratio versus external field H_L with the frequency f as a parameter.

 H_L at different frequencies. We only give the results obtained in positive field in figure 7. Similar results can be obtained when the magnetic field is negative, i.e., GMI curves are symmetric with respect to the magnetic field. This is a usual feature of GMI obtained in amorphous wires and ribbons [13, 14]. When H_L increases, the LMI ratio decreases monotonically. No peak appears on the curves when H_L increases. This is because the films are annealed at a temperature above the Curie temperature without a magnetic field. No anisotropies are induced, such as stress or magnetic field equal to the anisotropy field [13]. LMI ratio falls quickly below 15 Oe and slowly at higher field, and almost reaches saturation at 70 Oe for each frequency. The sensitivity of the GMI is defined as $S = \Delta Z/(2\Delta H_{1/2}Z_{H=70})$, where $\Delta Z = Z_{H=0} - Z_{H=70}$, $Z_{H=0}$ and $Z_{H=70}$ are the film impedance at 0 and 70 Oe magnetic field, respectively. $\Delta H_{1/2}$ is the value of field H_L at



Figure 8. Transverse magneto-impedance ratio versus external field H_T with the frequency f as a parameter.

which the MI ratio falls to half its maximum [12, 15]. Here we have used $Z_{H=70}$ as the value of film impedance at saturating magnetic field. Table 1 shows the sensitivity of the GMI versus frequency for longitudinal and transverse magneto-impedances. From table 1 one can see that when the frequency increases, the sensitivity increases and has a maximum of about 3.4% Oe⁻¹ at 13 MHz.

Table 1. Sensitivity (%) of the GMI at different frequencies, taken from figure 7 and figure 8.

	1 MHz	5 MHz	10 MHz	13 MHz
Sensitivity (LMI)	0.2	2.2	3.3	3.4
Sensitivity (TMI)	0.03	0.5	0.9	1.1

Figure 8 shows the dependences of TMI ratio on the transverse applied magnetic field H_T at different frequencies. The maximum TMI ratios for different frequencies at zero field are almost the same as those in the longitudinal case for the corresponding frequencies, but with much smaller sensitivity of GMI. This is caused by the influence of demagnetization field in the transverse direction of the film. The maximum sensitivity in the transverse case is about 1.1% Oe⁻¹ at 13 MHz (see table 1).

Figure 9 shows the dependences of PMI ratio on the perpendicular magnetic field H_P at different frequencies. The size and the sensitivity of PMI ratio are much smaller than those in the former two cases, because of the big influence of the perpendicular demagnetization field in the film. Even though the PMI ratio is far from the saturation state until $H_P = 70$ Oe, a larger MI ratio of about 11% is obtained at 11 MHz.

In summary, FeNiCrSiB amorphous films with good soft magnetic properties are obtained by RF sputtering followed by a suitable annealing process. When the films are annealed at 350 °C, a temperature above the Curie temperature and below the crystallization temperature of the films, for 20 min, a soft magnetic amorphous film is obtained. Its effective permeability μ_e is about 1500 at low frequencies, the coercive force H_c is 1.5 Oe, the saturation magnetization $4\pi M_s$ is about 7100 G, the Curie temperature is 346 °C. The longitudinal, transverse and perpendicular magneto-impedances of the film are studied



Figure 9. Perpendicular magneto-impedance ratio versus external field H_P with the frequency f as a parameter.

systematically at room temperature in the frequency range from 100 kHz to 13 MHz. A maximum GMI ratio of about 38% is obtained at 13 MHz in the longitudinal and transverse cases. The field sensitivity of longitudinal GMI is 3.4% Oe at 13 MHz. A large perpendicular MI ratio of 11% is obtained at 11 MHz.

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