Giant magnetoimpedance effects in the soft ferromagnet Fe_{73.5}CuNb₃Si_{13.5}B₉

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The components of the resistance *R* and the reactance *X* of the complex impedance $Z = R + iX$, as well as the relative effective permeability μ' , for as-quenched and nanocrystalline Fe_{73.5}CuNb₃Si_{3.5}B₉ ribbons have been studied as functions of drive current frequency $(1 \text{ kHz to } 13 \text{ MHz})$ and longitudinal magnetic field $(-75 \text{ to } 75$ Oe). For as-quenched samples, a mostly resistive feature was found over the whole frequency range. The magnetoimpedance effect was more sensitive for nanocrystalline samples: at 3 MHz, *R* and *X*, respectively, increased 10% and 16% at a longitudinal field of 4 Oe, or decreased 47% and 33% at a 75 Oe field. The magnetic responses of both *R* and *X* for nanocrystalline samples showed a maximum at about 4 Oe, whereas there was no maximum for as-quenched samples. The field dependence of effective permeability for nanocrystalline samples showed a sharp reduction at about 4 Oe; which is believed to be the anisotropy field. $[$ S0163-1829(96)04929-6]

Recent work on the strong effect of a dc magnetic field on the ac voltage induced in soft magnetic Fe-Co-Si-B amorphous wires and ribbons has generated considerable $interest$, $1-11$ since, from a practical point of view, this phenomenon is analogous to the giant magnetoresistance effect. The effect was reported to be the strongest for an amorphous wire of composition $Fe_{4.3}Co_{68.2}Si_{12.5}B_{15}$, having slightly negative magnetostriction on the order of -10^{-7} . In the lowfrequency range of 1–10 kHz, which is the typical region of the magnetoinductive effect, the inductive component of an ac wire voltage decreases by 50% at a longitudinal field of $2-5$ Oe. In the higher-frequency range of $0.1-10$ MHz, where the skin effect is essential, the giant magnetoimpedance (MI) effect occurs: the amplitude of the total wire voltage decreases by 40–60 % under the influence of a longitudinal field of $3-10$ Oe.³ The basic mechanism responsible for the MI effect is generally considered due to the skin depth, which is dominated by the effective circumferential permeability.3,9 The applicability of these and other views remains to be resolved.

Until now all reports about magnetoimpedance have been related to amorphous Fe-Co-Si-B alloys. Nanocrystalline Fe-Cu-Nb-Si-B soft ferromagnetic alloys are expected to have a considerable magnetoimpedance effect due to the excellent soft magnetic properties: high permeability and low magnetostriction.¹² However, nanocrystalline materials composed of small crystallites about 5–20 nm in size contain a large number of interfaces with random orientation and a substantial fraction of atoms located at these interfaces. This nanocrystalline structure may lead to different behavior in impedance spectra compared to that of amorphous alloys. In this report, we present the magnetoimpedance effect in both as-quenched and nanocrystalline Fe-Cu-Nb-Si-B soft ferromagnetic alloy ribbons. To investigate the influence of the permeability upon the MI effect, the dependence of the effective permeability on ac current frequency and on external dc magnetic field was also studied. The probable mechanism is discussed briefly.

The experiments were carried out with samples cut from amorphous $Fe_{73.5}CuNb_3Si_{13.5}B_9$ ribbons prepared using conventional single-roller rapid quenching in vacuum. The samples, 20 μ m thick and 10 mm wide, were first cut to 40 mm in length, and then heated to a chosen temperature between 520 and 650 °C in an oven without magnetic field. The oven was evacuated to the order of 10^{-5} Torr. After keeping the fixed temperature for 15 min, the sample was pulled out of the oven and cooled naturally in the vacuum system. All of the data presented here were obtained using a HP4192A impedance analyzer. Leads of metallic In were attached to the ribbons. The contact resistance was less than 1 Ω . The samples were connected to the analyzer with the accessory 16048B test lead which is carefully designed and contains four coaxial cables. The cables were 100 cm long and permitted the ribbons to sit within a Helmholtz coil (diameter 30 cm), which produced a dc magnetic field $-75 \leq H \leq 75$ Oe. The applied dc field was parallel to the ac current. The drive current amplitude used was 1 mA. All data were collected at room temperature.

X-ray diffraction indicated that the samples annealed at temperatures 520–600 °C have nanocrystalline structure, while the as-quenched sample is in the amorphous state. This is coincident with the results of Ref. 12. The measurements of the impedance spectra for nanocrystalline samples indicate that the impedance $Z(f,H) = R(f,H) + X(f,H)$ depends strongly on the frequency of the ac drive current and dc magnetic field. The resistance *R* and reactance *X* mostly increase with the frequency of the ac drive current. For the as-quenched samples, *R* increases sharply while *X* changes only slightly when the frequency increases from 5 kHz to 13 MHz. Figure 1 shows the spectra of the magnetoresistance *R* and the magnetoreactance *X* of the as-quenched sample and the 570 °C annealed sample. For the as-quenched sample, a striking feature is the strong resistive change over the whole frequency range. Similar results for Co-Fe-Si-B ribbons were observed by Machado *et al.*¹⁰ For an Fe-Co-Si-B ribbon,⁹

FIG. 1. Dependence of magnetoimpedance $Z = R + iX$ on the ac drive current frequency *f* in as-quenched and nanocrystalline $Fe_{73.5}CuNb_3Si_{13.5}B_9$ ribbons. The hollow symbols represent the data obtained without external field, and the black symbols represent the data measured under a maximum dc magnetic field of 75 Oe with the direction of the field parallel to that of the ac current.

Beach *et al.* reported that *R* had a positive curvature while *X* showed a negative curvature vs frequency below 3 MHz, indicating that *R* would increase more and more sharply while *X* would tend to saturate or decrease. Thus the mostly resistive feature may be common to amorphous ribbon alloys, which is quite different from the magnetoinductive effect reported by Mohri *et al.* in amorphous wires.¹ Another remarkable feature of Fig. 1 different from the amorphous case, is that *X* increases more sharply than *R* for the 570 °C annealed sample. This indicates that a different mechanism dominates in amorphous and nanocrystalline samples.

Figure 2 shows the responses of the magnetoresistance *R* and magnetoreactance *X* (in Ω /cm units) to the magnetic field, at drive current frequencies of 500 kHz and 3 and 10 MHz. We note that the resistance and the reactance decrease monotonically for amorphous samples. For nanocrystalline samples, when the field increases, both *R* and *X* first increase to a maximum at about 4 Oe, then decrease, and finally saturate. For $f=3$ MHz, the magnetoresistance ratio $(R_{H=4 \text{Oe}} - R_{H=0})/R_{H=0} = 10\%$ and $(R_{H=75 \text{Oe}} - R_{H=0})/R_{H=6}$ $R_{H=0} = -47\%$ and the magnetoreactive ratio $(X_{H=4}$ Oe $(X_{H=0})/X_{H=0} = 16\%$ and $(X_{H=75 \text{ Oe}} - X_{H=0})/X_{H=0}$ $=-33\%$.

The relative effective permeability μ' vs magnetic field has been studied for both samples in order to investigate the influence of the permeability upon the magnetoimpedance. The results are shown in Figs. 3 and 4. Figure 3 shows the frequency dependence of the relative effective permeability for 570 °C annealed and as-quenched samples. We can see that μ' for both samples decreases sharply in the lowerfrequency range, and then saturates in the higher-frequency range. Figure 4 shows the field dependence of μ' for both samples. We find that μ' first decreases slowly, then drops dramatically at about 4 Oe, and finally saturates for the

FIG. 2. The impedance $Z = R + iX$ of the as-quenched and the nanocrystalline Fe $_{73.5}$ CuNb₃Si_{13.5}B₉ ribbons vs external magnetic field *H* at $f = 500$ kHz, 3 MHz, and 10 MHz.

570 °C annealed sample. However, μ' for the amorphous sample decreases slowly in the whole field range $(0 \text{ to } 75)$ Oe).

Comparing Figs. 2 and 4, one finds a direct correlation between magnetoimpedance and effective permeability μ' . We note that the maximum shown in Fig. 2 occurs just at the field of about 4 Oe, where μ' has a rapid reduction, as shown in Fig. 4. The different behavior of as-quenched and nanocrystalline samples in MI effects comes from their different permeability properties, and indicates that they have different magnetic structure. As is well known, the as-quenched samples do not possess very soft magnetic properties, which are essential for MI effects. For the 570 °C annealed sample,

FIG. 3. Dependence of the permeability μ' on the applied ac current frequency f in the as-quenched and the nanocrystalline $Fe_{73.5}CuNb_3Si_{13.5}B_9$ ribbons.

FIG. 4. Dependence of the permeability μ' on the external magnetic field *H* in the as-quenched and the nanocrystalline $Fe_{73.5}CuNb_3Si_{13.5}B_9$ ribbons at $f=100$ kHz, 500 kHz, and 1 MHz.

the behavior of μ' in Fig. 4 shows some typical properties of the transverse anisotropy¹¹ and the anisotropy field is about 4 Oe. When an ac current flows through the ribbon, an easyaxis driving field will be generated. The external field *H*, being a hard-axis field with respect to the transverse anisotropy, suppresses the transverse magnetization by domain-

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wall motion. As *H* increases, the rotation portion of the remagnetization grows. When *H* reaches the anisotropy field, a rapid reduction of μ' occurs. For higher fields (*H*>4 Oe), μ' slowly decreases, which is consistent with pure rotational magnetization. The sensitive responses of μ' to *H* result in a large MI effect through the skin depth. However, the mechanism of the formation of the transverse anisotropy is not clear. Further work is needed to deal with all of the experimental results.

In summary, the impedance spectra of amorphous and nanocrystalline Fe_{73} ₅CuNb₃Si_{13.5}B₉ ribbons were obtained. A mostly resistive feature for the as-quenched samples over the whole frequency range was found. We also found that the MI effect is relatively larger for nanocrystalline samples than for as-quenched ones. The effective permeability μ' for nanocrystalline samples showed a rapid reduction when the external field was around the anisotropy field value, 4 Oe. The different MI responses between both samples indicate that they have different magnetic structures and we believe that transverse domains are responsible for the MI effects in nanocrystalline samples. The detailed relationship between the size of the nanocrystals and the magnetoimpedance, as well as the mechanism of magnetoimpedance in as-quenched and nanocrystalline alloys, are under investigation.

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