Giant magnetoimpedance in the ferromagnetic alloy $Co_{75-x}Fe_xSi_{15}B_{10}$

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A room-temperature giant magnetoimpedance (GMI) effect has been observed in thin amorphous ribbons of the soft ferromagnetic alloy $Co_{75-x}Fe_xSi_{15}B_{10}$ at low fields, $H < 10$ Oe. The size- and field-dependence of the effect are very sensitive to the annealing conditions and concentration x. The peak value of the magneto-impedance ratio is strongly dependent on the frequency of the measuring ac current. Values as large as 130% have been measured at 1 MHz in a sample with the zero-magnetostriction composition ($x = 4.6$) properly annealed in a transverse field. We suggest that the frequency dependence of the GMI arises from electron scattering by ac current-induced domain-wall oscillations.

The discovery[1] of giant magnetoresistance (GMR) effects in magnetic multilayers has brought a great deal of excitement to the field of magnetism and magnetic materials. This discovery opened the possibility for important technological applications[2] and raised new challenging physical problems. Although GMR has been observed in many multilayer systems, its microscopic origin has yet to be elucidated.[3] Additional interest in the field has come from the recent observation of GMR phenomena in heterogeneous media containing ultrafine ferromagnetic particles.[4,5] Actually there are many similarities between the particulate materials and the multilayers, as well as between the GMR effects they present. Both systems are composed of ferromagnetic entities coupled by metallic nonmagnetic media. In zero external field they have no net magnetization (the GMR multilayers have antiferromagnetic alignment and the particulate media have frozen random particle moments) and present maximum resistivity. The application of an external field tends to align the moments parallel to one another resulting in a smaller resistivity (negative magnetoresistance). This effect is usually measured with direct current (dc) or low-frequency alternating current (ac) and is frequency independent. Magnetoresistance ratios between 0.1 to 150 % have been observed with fields that vary from a few Oersted to thousands of Oersteds.

In this paper we report the observation of room-temperature, very low-field GMR in a system completely different from the multilayers and the particulate media, namely, the amorphous ferromagnetic alloy $Co_{75-x}Fe_xSi_{15}B_{10}$. This new type of GMR effect is observed only for ac currents at relatively high frequencies. A striking feature of the voltage signal produced by the ac measuring current is the strong dependence of its amplitude and phase on the frequency, so this new effect is referred to as giant magnetoimpedance (GMI).[6]

The experiments were carried out with samples cut from amorphous ribbons of $Co_{75-x}Fe_xSi_{15}B_{10}$ prepared by Professor Frank Missell at the University of São Paulo using conventional melt-spinning techniques.[7] This is a well studied, high-permeability, amorphous alloy known to have low magnetostriction in the Co-rich side. In fact, the presence of Fe impurities with concentration $x = 4.6$ results in near-zero magnetostriction, making this alloy suitable for high-frequency applications.[7] Samples 1.5 cm long were cut from ribbons 40 μm thick and 1 mm wide, prepared with three different concentrations, $x = 2.5$, 4.6, and 6.5. Since the magnetic properties of this alloy are very sensitive to heat-field treatment, we investigated samples subjected to various annealing conditions. The results reported here were obtained with samples heated to a certain temperature in a He atmosphere in a nonmagnetic oven, and then cooled, either in zero magnetic field, $H_{an} = 0$, or in $H_{an} = 2$ kOe [zero-field-cooled (ZFC) or field-cooled (FC)] applied in the transverse direction (in the ribbon plane, perpendicular to its longitudinal direction). Since the magnetic properties of the alloy vary logarithmically in time after the heat treatment, we recorded the time t elapsed between the end of the annealing process and beginning of measurement. All data were taken at room temperature.

A four-probe ac technique was used to measure the magnitude Z and the phase θ of the magnetoimpedance, $\mathbf{Z} = Z \exp(i\theta)$, as a function of the field H applied along the ribbon length, i.e., parallel to the ac measuring current. The contacts were made with silver paint in carefully cleaned samples (typical contact resistance of the order of 1 Ω). Data were taken with current amplitude I in the range 1–25 mA and frequency f between 0.5 kHz and 1 MHz. The external field H was generated by a pair of Helmholtz coils with axis parallel to the Earth's magnetic field to allow field variation from 0 to ± 45 Oe. The phase and magnitude of the voltage signal were simultaneously recorded using a vector lock-in amplifier in the Z θ mode for $f < 120$ kHz. For larger frequencies the voltage amplitude in the samples with GMI was measured directly with a digital oscilloscope. The measured voltage versus field H was used to calculate the magnetoimpedance ratio $\mathrm{MIR} = 10^2[Z(H) - Z(0)]/Z(0)$. All data acquisition and analysis were completely automated.

Figure 1 shows the MIR versus field data at three fre-

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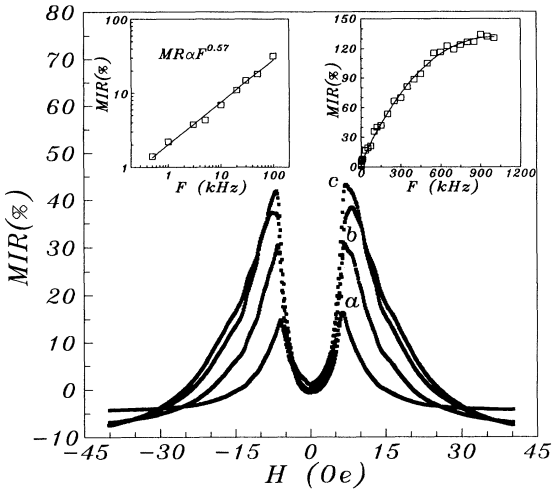

FIG. 1. Magnetoimpedance ratio $\{=10^2[Z(H)-Z(0)]/Z(0)\}$ (MIR) versus applied longitudinal magnetic field H in a transversely annealed (633 K) ribbon of $Co_{70.4}Fe_{4.6}Si_{15}B_{10}$ for three values of the measuring current frequency f: 30 (a), 80 (b), and 120 kHz (c). The upper right inset shows the dependence with frequency of the maximum of the MIR up to 1 MHz. At this frequency the MIR reaches 130%. The upper left inset shows the frequency dependence at low frequencies.

quency values, $f=30$, 80, and 120 kHz, obtained in the sample with largest GMI, using $I=10$ mA. This sample was cut from a zero-magnetostriction ribbon ($x=4.6$) and annealed for 15 min at $T_{an}=633$ K and $H_{an}=2$ kOe. The heating and cooling times were about 30 min. The data were taken immediately after the end of thermal treatment. In zero field the sample had a frequency-independent, purely resistive impedance of 0.5 Ω corresponding to a resistivity of 130 μΩ cm. The magnetic properties of similarly prepared samples have been previously reported.[7-10] Below its Curie temperature $T_c=555$ K there is a marked departure between the dc susceptibilities measured after cooling in a field (FC) or in zero field (ZFC). Only FC samples show behavior like that in Fig. 1. ZFC samples have small, positive, frequency-independent magnetoresistance.[11]

One of the striking features of the data in Fig. 1 is the strong frequency dependence of the MIR in the field range $5<H<20$ Oe. This is a completely new effect, not observed in the multilayers or the particulate materials. As shown in the upper left inset, at low frequencies the MIR maximum increases with frequency according to MIR $\propto f^{0.57}$. At higher frequencies MIR tends to saturate, approaching the fantastic value of 130% of 1 MHz, as shown in the upper right inset (above the lock-in frequency limit of 120 kHz the signal is seen directly on the oscilloscope screen).

The other remarkable feature of Fig. 1 is the field behavior of the MIR, also quite different from the data in other materials. Here we note that for the sake of clarity only the up-field sweep curves are shown for 30 and 80 kHz. When the field is swept in both directions we observe a small hysteresis, at all frequencies. The MIR, which is zero at $H=0$ by definition, initially increases with increasing H reaching a sharp peak at a field value which is slightly frequency dependent. Note that the value of H for which the magnetoresistance is maximum is extremely low compared to the saturation fields in the multilayers or the particulate media. At larger H the MIR decreases rapidly, becomes negative and reaches saturation at a frequency-dependent value that can still be considered "giant" (3–8 %).

The frequency dependence of the data prompted us to look at the phase of the signal. Figure 2, shows the field variation of the impedance phase for $f=10$, 30, 50, and 100 kHz, measured in the same sample as in Fig. 1. Note that the field behavior of the phase is similar to that of the MIR, except for a slight broadening of the peaks. On the other hand, the frequency dependence of the phase, shown in the inset, is less pronounced than for the MIR. It is important to note that the phase saturates at about 15°, and since $\cos 15°=0.966$, the magnetoimpedance is mostly resistive throughout the whole frequency range. This shows that the results reported here are quite different from the magnetoinductive effect reported by Mori *et al.* in amorphous wires.[6]

Figure 3 shows data for a second sample, cut from the same ribbon as the previous one and with the same dimensions, but with a different heat treatment. In this sample the annealing in a 2-kOe transverse field was done at 578 K for 15 min, with heating and cooling times of 18 min. In (a) we show the magnetization curve obtained with a mutual inductance hysteresis tracer operating at 120 kHz. (b), (c), and (d) show the MIR at 120 kHz measured with current amplitudes $I=24.5$, 10,0, and 2.7 mA.

Clearly, the MIR peaks occur near the same field values (6.3 Oe) where the magnetization deviates from a linear dependence and starts to saturate. In fact, in an earlier stage of this work we reported[12] data obtained in a

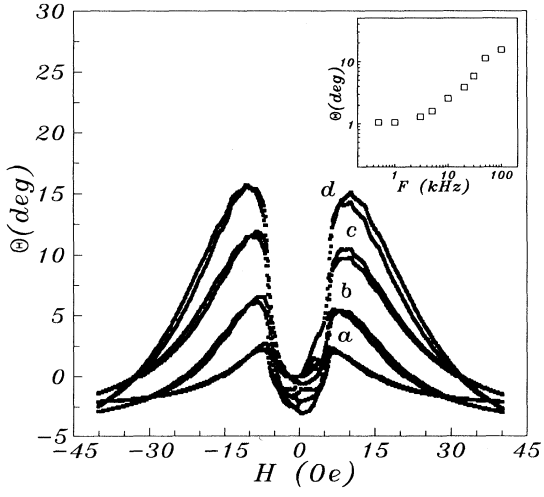

FIG. 2. Magnetic-field dependence of the magnetoimpedance phase for $f=10$ (a), 30 (b), 50 (c), and 100 kHz (d) for the sample of Fig. 1. The upper right inset shows the frequency dependence of the maximum in the MI phase.

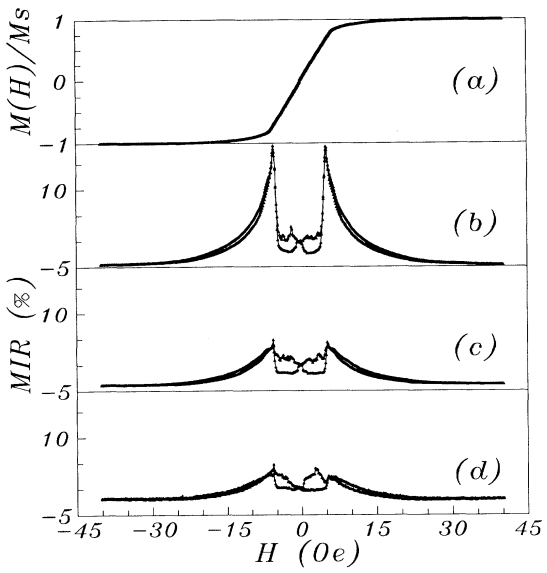

FIG. 3. Magnetization (a) and magnetoimpedance ratio versus field measured at $f=120$ kHz in a $Co_{70.4}Fe_{4.6}Si_{15}B^{10}$ sample annealed at $T_{an}=578$ K. The curves in (b), (c), and (d) were obtained with driving current amplitudes 24.5, 10, and 2.7 mA, respectively.

sample with thickness 50 μm, for which the MIR peak and the saturation of the magnetization occurred at a lower field $H\simeq 2.5$ Oe. (It is known[13] that the saturation field varies with sample thickness.) We also note that while hysteric effects are very small in the magnetization curve (the coercive field is only a few mOe), they are noticeable in the MIR. The correlation of the MIR with the magnetic properties and its hysteresis with field provide a strong indication that the mechanism responsible for the magnetoimpedance involves the domain structure in the sample.

It has been known for many years that a transverse anisotropy axis can be induced in melt-spun ribbons of amorphous magnetic alloys by annealing in a transverse field. This anisotropy results in the formation of a very fine domain structure in the transverse direction when the sample is unmagnetized.[13,14] In the alloy composition with substantial positive magnetostriction, the domain structure is very stress sensitive. However, in the near-zero magnetostriction alloy $Co_{70.4}Fe_{4.6}Si_{15}B_{10}$ the domain structure is mostly dependent on magnetic history. Moreover, in this alloy composition the domain walls are very mobile and can have an oscillatory motion driven by ac current of a few mA (Ref. 15) along the ribbon length. In fact, domain oscillations induced by currents or by ac fields have been observed by scanning transmission electron microscopy[14] and by longitudinal Kerr effect.[16,17] These considerations set up the scenario for a qualitative interpretation of the observations reported here.

In zero field there is a transverse domain structure, where neighboring domains about 100 μm wide have magnetization antiparallel to each other. The application of a longitudinal current produces a classical transverse field that displaces neighboring domain walls in opposite directions.[15] Hence an ac current produces domain-wall oscillations and consequently an increase in resistivity due to spin-dependent electron scattering by the moving walls.[18,19] When a small magnetic field is applied along the ribbon length, the magnetizations of both types of domains rotate towards the field.[20]

A simple rotation model[20] for uniaxial transverse anisotropy K gives for the angle α between the magnetization and the field $\cos\alpha=M_sH/2K$, where M_s is the saturation magnetization. Direct domain imaging by Kerr-effect microscopy[17] shows that as the field is increased the amplitude of the domain-wall oscillation increases, resulting in larger resistivity. A direct evidence that the increase in ac resistivity is due to domain-wall motion has been provided by longitudinal Kerr-effect experiments. The Kerr signal obtained in a sample cut from the same ribbon used here, under an ac current excitation, has the same field dependence as the MIR data of Fig. 1.[17] As the MIR, the Kerr signal reaches a peak at the field value for which the magnetization starts to saturate.

Further increase in field results in the disappearance of domain walls and hence the sources of magnetic scattering and the Kerr signal. In recent work with negative magnetostriction amorphous wires[21,22] the frequency dependence of the impedance has been explained in terms of skin-depth effects and classical electrodynamics. However, in the case of zero-magnetostriction ribbons the correlation between the Kerr signal and MIR data shows that domain-wall motions play a decisive role in the origin of the magnetoimpedance. Due to the complexity of the domain structure and the domain-wall motion in an applied longitudinal field, as well, as the lack of a good model for electron scattering on moving domain walls, we have not been able to describe quantitatively the frequency dependence of the MIR. A simple model with domain-wall pinning and a Lorentz force between electrons and walls[19] leads to a frequency-dependent resistivity[12] which is not sufficient to explain the data. However, we believe that the MIR frequency dependence is partly due to domain-wall motion, and at higher frequencies to skin-depth effects, which have proved important in other amorphous systems.[21,22]

Finally we mention that the shape of the MIR vs field curves and the size of the maximum MIR are very dependent on the temperature and times involved in the annealing process. This is consistent with the proposed mechanism because it is known that the induced anisotropy and consequently the domain structure are very sensitive to the annealing.[13] Furthermore, the amplitude of the MIR decays logarithmically in time after the end of the annealing process, as do the magnetic properties of the random alloys. The MIR is also very sensitive to changes in Fe concentration. In fact, data for $x=2.5$ and $x=6.5$ gave MIR maximum at least one order of magnitude smaller than measured in the zero-magnetostriction sample. Again this is consistent with the proposed domain scattering mechanism because in the zero-magnetostriction alloy the domain motion is stress free. We are currently undertaking a systematic

study of the influence of the thermal annealing process and driving current on GMI effects.

In conclusion, we have reported a room-temperature giant magnetoimpedance effect in the zero-magnetostriction amorphous alloy $Co_{70.4}Fe_{4.6}Si_{15}B_{10}$. The GMI effect depends strongly on the frequency of the measuring current. This observation presents new challenges for theorists trying to explain the magnetoresistance previously measured in other systems.

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