Magnetic susceptibility of Fe/Cu multilayers: Ferromagnetic, antiferromagnetic, and spin-glass phases

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Citation: Journal of Applied Physics **83**, 7372 (1998); doi: 10.1063/1.367800
View online: http://dx.doi.org/10.1063/1.367800
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/83/11?ver=pdfcov
Published by the AIP Publishing

Spin Glasses and Frustrated Systems Mark Rubinstein, Chairman

Magnetic susceptibility of Fe/Cu multilayers: Ferromagnetic, antiferromagnetic, and spin-glass phases

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In this work we present low-field magnetic measurements on multilayers of Fe 1.5 nm and Cu 1.5 nm layers, repeated 10 times, grown on Si(111) and Fe 5 nm/CaF_2 40 nm/Si(111) substrates. Magnetic characteristics of a spin-glass phase are exhibited by samples directly grown on Si(111), for which the glass temperatures (T_G) depend strongly on the magnetic field. Above T_G those samples exhibit a ferromagnetic behavior. In contrast, samples grown on Fe/CaF_2 buffer structure also show the spin-glass characteristics, but are antiferromagnetic above T_G. This work deals with the fundamental role played by the interface between Fe and Cu for these effects, which is clearly emphasized by the experimental results.
[S0021-8979(98)46011-0]

Research and development of metallic magnetic multilayers is an area of increasing interest, not only for the fundamental physics involved with films and layers, but especially due to its great potential for a variety of applications. Fe/Cu multilayers are very interesting systems exhibiting a strong correlation between magnetic and structural properties. For example, it is well known that face centered cubic (fcc) γ-Fe and body centered cubic (bcc) Cu metastable phases can be obtained in epitaxial or highly textured films.[1–10] Experimental results and first-principle magnetic structure calculations confirmed that a rich variety of magnetic configurations can be stabilized in fcc γ-Fe with a lattice constant range near that of fcc Cu.[11–15] It has also been shown that the magnetic anisotropy energies are rather different for the ferromagnetic and various antiferromagnetic configurations in fcc Fe.[11,12] Fe/Cu multilayers also present antiferromagnetic exchange coupling which is accompanied by moderate magnetoresistive and magneto-optic effects for chemical modulation periods in the nanometer length scale.[16] Despite of the great interest on magnetic properties of Fe/Cu multilayers, its low-field magnetic susceptibility behavior has not been thoroughly explored in the literature.

The present article reports on the irreversible behavior exhibited by zero-field-cooled (ZFC) and field-cooled (FC) magnetization curves of highly oriented Fe/Cu multilayers. Our experimental results are discussed assuming that a magnetic metastable phase occurs at the interfaces due to the existence of some interdiffusion between Fe and Cu.

Previous studies[17–19] have shown that highly oriented Fe(110)/Cu(111) multilayers consisting of successive deposition of Cu 1.5 nm and Fe 1.5 nm layers repeated 10 times can be grown directly either on Si(111) or on Fe 5 nm/$CaF_2$40 nm/Si(111) substrates. X-ray diffraction and electron diffraction analyses confirmed that the Fe/Cu multilayers grown on Fe/CaF_2 buffer structure have a better structural quality than those grown on Si(111) surfaces. Besides, x-ray reflectivity data reveal that each of these multilayers exhibits rather different interfacial roughness. The samples investigated in this study belong to a series widely explored so far. Despite the wide range of layer thicknesses in the series, we found no meaningful way to explain the very low magnetoresistance in these multilayer systems.

The dc magnetic susceptibility (χ_{dc}) measurements, performed in the temperature range 5–400 K and fields up to 1 kOe, were taken with a superconducting quantum interference device (SQUID) magnetometer (MPMS-5S, Quantum Design). Each measurement resulted from two sample scans over a length of 4 cm, with 40 point readings per scan. Low-field susceptibility measurements as a function of the temperature for different values of the applied field, were carried out using a routine combining a ZFC warming run followed by a FC experiment. To start the procedure, the sample was first heated up to 400 K and then cooled down to 5 K in the absence of magnetic field. Magnetization loops were also measured for many temperatures in between 5 and 400 K. Remnant demagnetization cycles were done at 400 K to prevent spurious irreversible effects associated with magnetic history of the sample in the ZFC measurements. Real and imaginary parts of the low-field ac susceptibility ($\chi_{ac}=\chi' + j\chi''$) of the samples were also measured using the SQUID

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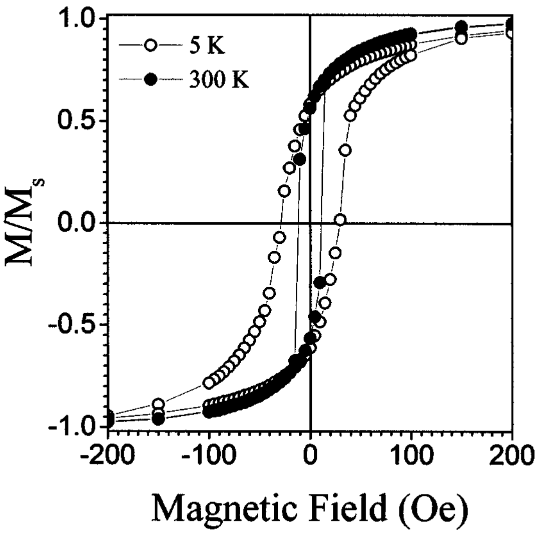

FIG. 1. Hysteresis loops measured for sample 2, at temperatures (○) 5 and (●) 300 K.

magnetometer with an excitation field $h_{ac}=3.0$ Oe. The temperature dependence χ_{ac} at different frequencies was measured while heating the samples.

Figure 1 shows the central portion of hysteresis loops, measured at 5 and 300 K, for one of the Fe/Cu multilayers investigated, grown on a Fe/CaF_2 buffer structure (sample 2). Notice that, at 200 Oe, the relative magnetization is about 95% of its saturation value. Hysteresis loops for the other multilayer studied here, grown on a Si(111) surface (sample 1), are similar and will appear in another publication.[17] Both samples exhibit typical ferromagnetic behavior, saturating at relatively low magnetic fields. It is worth mentioning that the remnant magnetization values measured at 5 and 300 K for sample 2 are smaller than those observed for sample 1.

Figure 2 shows measurements of χ_{dc} for those two multilayers. One can see that both exhibit a spin-glasslike behavior at low magnetic fields, manifested by several typical signatures, including a field-dependent irreversible magnetization, a frequency dependent χ_{ac}, and the occurence of metastable states characterized by a logarithmic time relaxation upon application or suppression of external fields of low magnitude.

The irreversible behavior is relatively weak, being more pronounced for sample 2. Besides, the irreversibility temperature associated with the bifurcation in the ZFC/FC curves is quite high and strongly dependent on the applied magnetic field. A glass temperature T_G, associated with a hump in the ZFC curve, is also observed for low fields. Another important feature when comparing these multilayers is that the FC curves for low fields are clearly different. While the FC response for sample 1 resembles those of a ferromagnet, sample 2 exhibits a more complex behavior, including antiferromagnetism for higher temperatures, bounded by a local maximum of χ_{dc} on the vicinity of 380 K. Since the crystal structure of both multilayers is similar, exhibiting identical orientation along the film growth axis, one would conclude that irreversibility comes from the occurence of dissimilar interfaces in different samples. This magnetic behavior seems to be related to a certain roughness at the Fe/Cu interfaces, which manifests itself as a local intermixing of adjacent layers. Our results suggest that the width of the

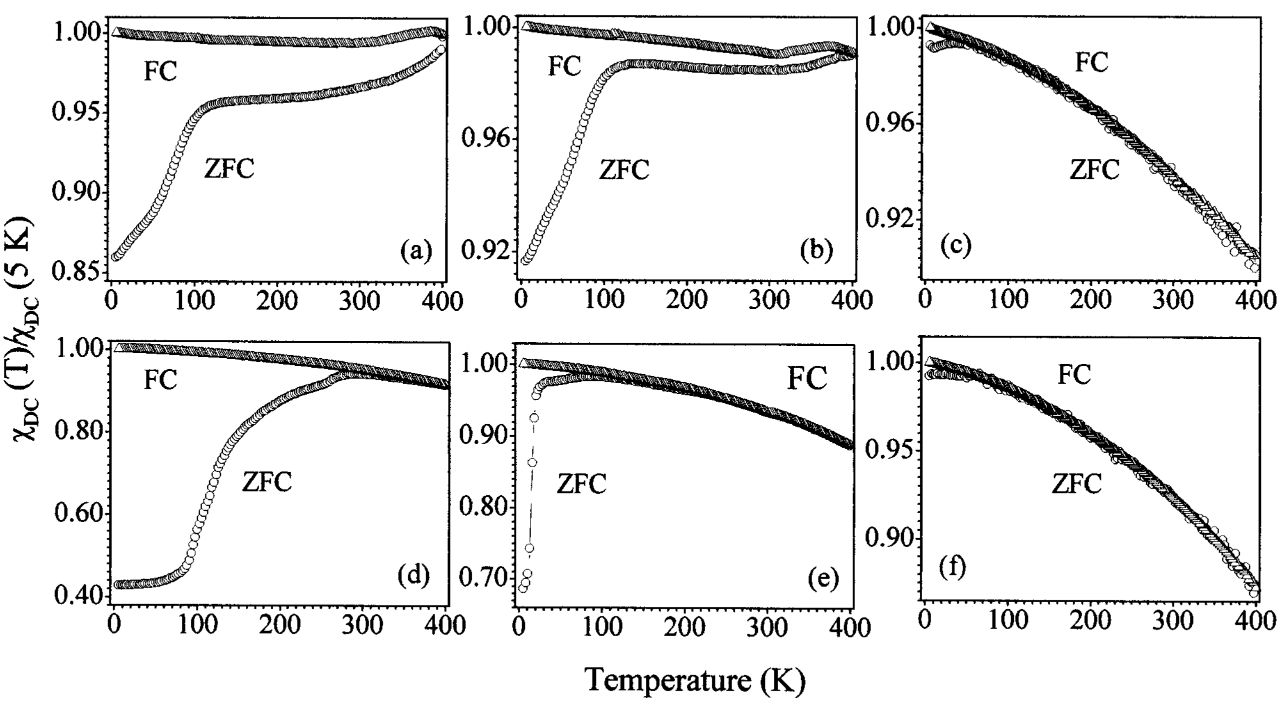

FIG. 2. Temperature dependence of χ_{dc} for sample 2 at fields: (a) 10, (b) 50, and (c) 1000 Oe; and for sample 1: (d) 10, (e) 50, and (f) 1000 Oe. Two curves are shown for each value of the field: the lower (○) is for the multilayer zero-field cooled to 5 K while the upper (△) is for the film cooled in the measuring field. Irreversibility temperatures associated with a bifurcation in ZFC/FC curves are observed.

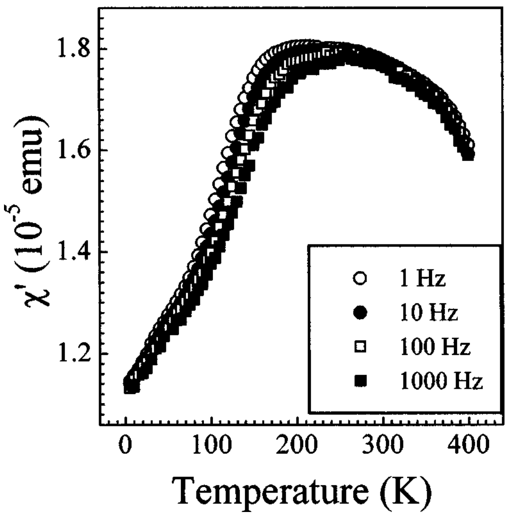

FIG. 3. Temperature dependence of χ' for sample 2. A small memory effect, dependent on the frequency, is present.

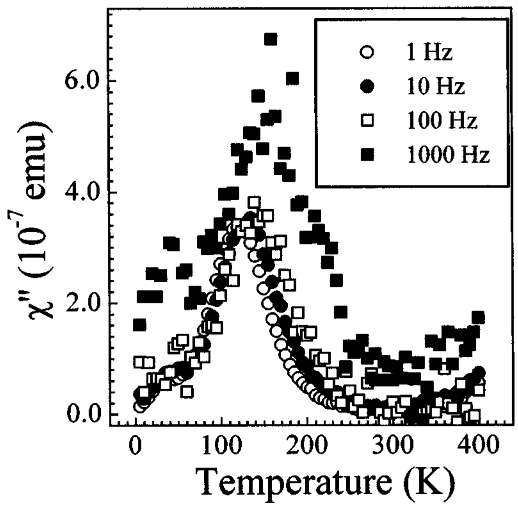

FIG. 4. Temperature dependence of χ'' for sample 2. Typical frequency shifts of χ'' peaks are observed.

mixed interfacial region should be small, involving only a few number of monolayers. Otherwise, one could not explain the ferromagnetic and antiferromagnetic behavior of FC curves.[20,21]

Figure 3 shows $\chi'(T)$ for sample 2, whereas $\chi''(T)$ appears in Fig. 4. Both exhibit the expected frequency dependence for a spin-glass system.[22] χ' shows a typical frequency shift, while χ'' has a maximum which also shifts towards high temperatures. An interesting feature of Fig. 3 is the flatness of χ' between 200 and 350 K which might be evidencing a possible overlapping of spin-glass and antiferromagnetic phases. Also noticeable is the increase in χ'' dispersion for higher frequencies, which could be attributed to an enhanced activity of magnetic domains induced by the rapidly varying excitation field.

In summing up, we have reported clear experimental evidences of a spin-glass behavior overlapping with ferro- and antiferromagnetism in Fe/Cu multilayers. As a matter of fact, a detailed investigation of the microscopic character of the interfaces in the samples of interest is a crucial task in the study of the mechanisms leading to the behavior reported here. In the past we have employed x-ray reflectivity with a double-crystal setup and also conventional θ–2θ low-angle diffraction, in an attempt to clarify those questions. Other powerful techniques, like *in situ* transmission, tunneling, and atomic force microscopy, could represent an expressive step forward in this subject, but neither is available at the moment. We are now conducting an alternative study, to appear soon elsewhere, in which we change the number of interfaces and the surface/volume ratio.

Furthermore, there is an still unpublished work resulting from near-edge extended x-ray absorption fine structure experiments conducted at LURE (Orsay, France) on these multilayers, which indicates the existence of highly disordered Cu sites at the interfaces, corroborating the proposed scenario of local intermixing and roughness at adjacent layers.

As we see, a considerable amount of information on the structure of these systems has been collected in this and in previous work, indicating that the Fe/Cu interface peculiarities are to be responsible for the presently reported behavior, as well as for the absence of significant giant magnetoresistance effects in these multilayers.[19]

Financial support from FAPESP, FAPERGS, CNPq, and FINEP/PRONEX is acknowledged.

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