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Magnetic and structural behavior of FeCo/Cu multilayers submitted to Kr irradiation

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Abstract

We have studied the effects of ion irradiation on the structural and magnetic properties of FeCo/Cu multilayers. The films were irradiated at room temperature with 600 keV Kr ions, with fluences ranging from 1 to 5×10^{15} ions/cm^2. X-ray absorption spectroscopy (XAS) was used to characterize the structural changes around Fe and Co atoms. The XAS data have shown a phase transformation of FeCo alloy from bcc for the as-deposited sample to fcc after irradiation. The structural effects induced by irradiation changed the saturation magnetization (M_S), the coercive field (H_C) and the magnetic particle size. The interplay between structural and magnetic effects indicates that some degree of mixing took place even being the elements immiscible.

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Keywords: Ion irradiation; Magnetic thin films; Structural effects

1. Introduction

Over the last years ion irradiation has emerged as a technique suitable to modify the magnetic properties of thin films, for example, Co/Pt multilayers [1]. The ability of controlling magnetization reversal while keeping the surface of the films flat is the main advantage of the technique. Surface flatness was preserved due to the choice of He as irradiating ion, which induces small density of defects in the material. The irradiation effects on thin films are quite diverse depending on the thermodynamic properties of the particular system (heat of mixing), mass and energy of the incident ions. 1 MeV Si^+ irradiation of Co/Ag multilayers e.g. characterized by strong positive heat of mixing ($\Delta H_m = +26$ kJ/mol), produces a complete segregation of Co from Ag matrix [2]. Films with ultra-thin Co layers show a superparamagnetic behavior in the as-deposited state. After irradiation, the ferromagnetic component becomes relevant and increases with the fluence. As the Co segregates and grows in size with irradiation, the giant magnetoresistance (GMR) decreases, showing the inverse dependence on the Co particle size.

Systems with low positive heat of mixing like Co/Cu ($\Delta H_m = +10$ kJ/mol) behave differently when submitted to 1 MeV Si^+ irradiation [3]. At low fluences (10^{14} ions/cm^2), interfacial mixing is observed, with a suppression of interlayer antiferromagnetic coupling and a consequent decrease of GMR. However, with post-irradiation annealing the initially sharp interface is recovered and, as a consequence, the GMR magnitude increases again. This can be explained by means of a backdiffusion process chemically driven, activated by the positive heat of mixing. Irradiation with higher fluences ($\sim 10^{16}$ ions/cm^2) generates metastable Co–Cu alloys, which have similar magnetic properties of alloys obtained by other techniques [4].

FeCo/M (M = Ag, Cu) magnetic multilayers have attracted great scientific interest because of considerable GMR values observed in such systems, even at room temperature [5]. The GMR effect is strongly influenced by the

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very local environment around the magnetic atoms. As ion irradiation affects small local volumes in a material, it seems to be suitable for changing the near-neighbor environment around FeCo. The FeCo alloy plays an important role in magnetic storage industry. It is used for a long time in recording heads due to its very high saturation magnetization associated with low coercivity [6]. Nowadays, FeCo alloys are used in magnetic heterostructures for spin-polarized current injection [7].

In recent studies [8,9] we have shown that Kr irradiation on FeCo/Cu multilayers produces interesting structural effects. Irradiation triggers strain release, induces an increase of grain size and enhances the crystalline quality of Cu. Irradiation also promotes a structural phase transformation around FeCo, changing its magnetic properties. Using a focused ion beam (FIB) the FeCo/Cu films can be nano-patterned, forming adjacent regions with quite different magnetic behavior. The goal of the present paper is to investigate the effects of ion-beam-induced mixing of the normally immiscible elements Fe, Co and Cu, using magnetic measurements combined with X-ray absorption spectroscopy (XAS).

2. Experimental techniques

The $[Fe_xCo_{1-x}/Cu]_{10\times}$ multilayers were fabricated by alternate e-gun evaporation in ultra-high vacuum, being deposited on SiO_2(2000 Å)/Si(1 1 1) substrates at room temperature. The thicknesses, monitored by a quartz micro-balance (with usual error of 5%), were 15 Å for Fe_xCo_{1-x} and 50 Å for Cu samples, using two concentrations of Fe, i.e. 30 and 70%. Irradiation was performed at room temperature with 600 keV Kr ions and beam current density of 100 nA/cm^2. This very low current assures that samples' temperature during irradiation does not suffer any considerable enhancement. The fluences ranged from 1 to 5×10^{15} ions/cm^2. The energy loss profile of the ions, as calculated by SRIM [10], is rather uniform in the thin films and the projected range of the ions is localized deep in the substrate.

XAS measurements were done at XAFS beam lines of LNLS [11] and ELETTRA in fluorescence mode, at Fe and Co *K*-edges. Cu and Fe references spectra were also collected for analysis and comparison purposes. The magnetic behavior of the samples was verified by magnetization versus applied field curves, measured with an alternate gradient field magnetometer (AGFM). The temperature dependence of the samples' magnetization was accompanied by field-cooled/zero-field-cooled (FC/ZFC) curves from 10 to 300 K, measured with a SQUID magnetometer. It is important to note that all samples were cut in the same disk-like shape (with 3 mm diameter) in order to be able to compare the results.

3. Results

The EXAFS signals [$\chi(k)$] at Fe *K*-edge for FeCo/Cu multilayers are presented in Fig. 1. The Co *K*-edge curves show a similar behaviour as the Fe *K*-edge ones. For the sake of simplicity, we have chosen the two most representative samples, i.e. as-deposited and irradiated with 3×10^{15} ions/cm^2. The signals for the as-deposited samples (for the two Fe concentrations) have qualitatively the same features. Comparing these signals with the Fe reference signal, it is possible to assert that the FeCo alloy in the as-deposited samples has a bcc structure [8]. The bcc structure can be recognized, e.g. by the α and β peaks, as indicated in the figure for the samples and the Fe reference. The amplitudes of the samples' signals are smaller than the Fe reference signal. This is an effect of structural disorder due to defects in the films, which comes from the film deposition process. After irradiation, the EXAFS signals change their shape, indicating a structural transformation around Fe. By comparison with the Cu reference signal (see the γ feature in figure), it is clear that the local order around Fe is transformed to fcc, confirmed by fitting results [8].

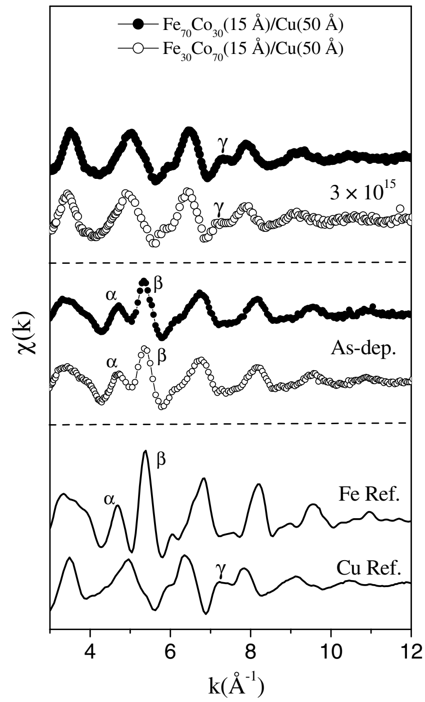

Fig. 1. EXAFS signals at Fe *K*-edge for Fe_xCo_{1-x}/Cu multilayers before and after irradiation with 3×10^{15} ions/cm^2. The Fe and Cu reference signals are also shown for comparison.

Fig. 2 shows the hysteresis loops for the samples presented in Fig. 1. The curves for the as-deposited samples have a ferromagnetic-like shape, which indicates that the films are constituted of large coupled grains. Sample (a) has a larger saturation magnetization than sample (b), 1.8×10^6 A/m and 1.4×10^6 A/m, respectively. This agrees with the Slater–Pauling curve of FeCo alloy [12], where the specific moment has a maximum for 70% of Fe. Sample (b) presents an appreciably larger in-plane anisotropy when compared to sample (a). This is characterized by the coercive field (H_C), more than two times higher for sample (b). Such an effect indicates the role of Fe concentration,

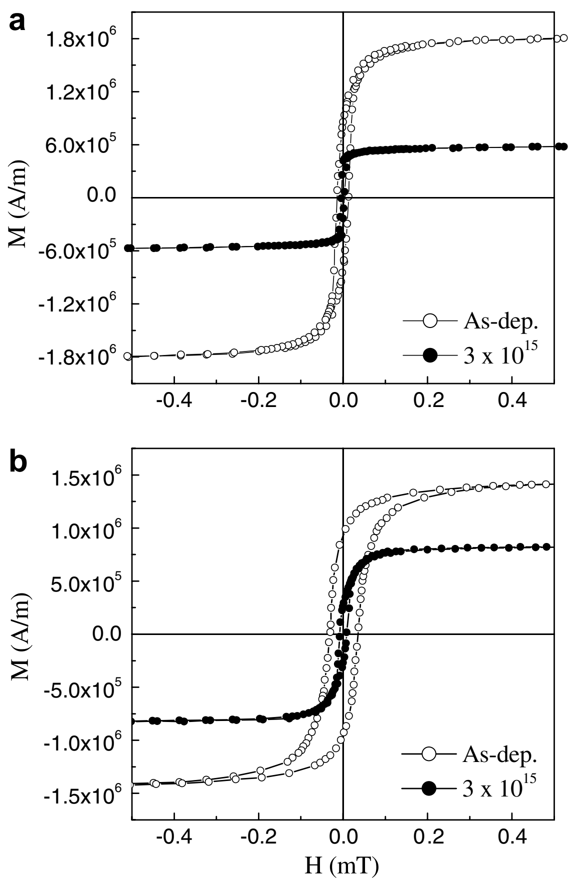

Fig. 2. Hysteresis loops for the as-deposited and irradiated multilayers. (a) $Fe_{70}Co_{30}$/Cu and (b) $Fe_{30}Co_{70}$/Cu, for DC applied field paralell to the film surface.

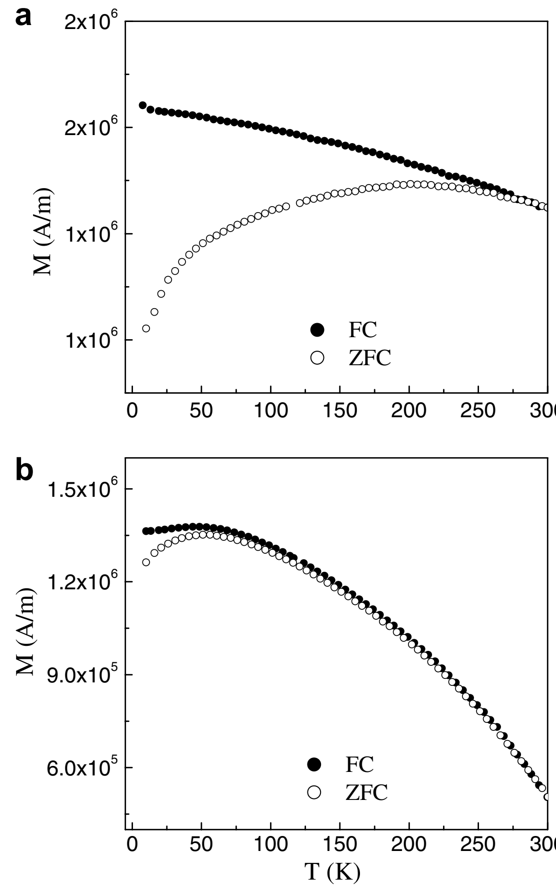

Fig. 3. Magnetization dependence on temperature (FC/ZFC curves) for (a) as-deposited and (b) irradiated with 3×10^{15} ions/cm^2 multilayers. The $Fe_{70}Co_{30}$/Cu sample curves were chosen since they represent well the irradiation effects. The $Fe_{30}Co_{70}$/Cu sample shows a quite similar behavior.

that is, alloys with smaller concentration are magnetically harder.

Notably after irradiation, the saturation magnetization (M_S) of the samples experiences a significant decrease, diminishing 70% for sample (a) and 43% for sample (b). As M_S is an intrinsic magnetic property [12], such a considerable decrease can be a consequence of a change in the chemical environment of the magnetic atoms, in our case Fe and Co. The FeCo alloy suffered a structural phase transformation and, possibly, its nearest-neighbor atoms have changed.

The magnetization dependence on the temperature is shown in Fig. 3. The FC/ZFC curves for the as-depsosited sample (Fig. 3(a)) are characteristic of a ferromagnetic film with coupled grains. The temperature for the onset of irreversibility is quite high, around 250 K, indicating that the effective volume of the magnetic grains is big. The curves also indicate that the Curie temperature (T_C), another intrinsic magnetic property, is considerably higher than room temperature.

The FC/ZFC curves after irradiation change a lot (Fig. 3(b)). The temperature for the onset of irreversibility goes down to values around 50 K, indicating that irradiation promoted an important reduction of the magnetic grain size. At the same time there is a clear indication that Curie temperature also decreases. This effect is consistent with a change in the chemical environment of FeCo induced by the irradiation.

4. Discussion and conclusions

EXAFS data revealed that irradiation has induced a phase transition around FeCo (in FeCo/Cu multilayers). The FeCo structure evolves from bcc in the as-deposited sample to fcc after irradiation. The magnetic measurements have shown decrease of M_S, T_C and magnetic particle size after irradiation. Despite the reduction of magnetic grain size, there is no apparent superparamagnetic behavior observed, since the hysteresis loops keep a large remanence to saturation ratio after irradiation (see Fig. 2). The conclusion that magnetic particle size decreases after irradiation comes from the reduction of irreversibility temperature. At the same time that data indicates the decrease of magnetic particle size, the Cu grain size increases with fluence, evolving from 50 Å in the as-deposited sample to 250 Å after 3×10^{15} ions/cm^2 [9]. Taking into account the magnetic and structural effects, it is possible to conclude that irradiation induced mixing between the elements. One can understand the reduction of magnetic grain size as a "dissolution" process of FeCo into the Cu lattice.

The highly energetic incident ions break up the Fe–Co bindings and mix them to Cu, forming in this way a metastable ternary Fe–Co–Cu substitutional alloy with the same structure of Cu, i.e. fcc. The Cu imposes its structure during the relaxation period of the ion-solid interaction process, when the new structure can become stable. One may query about the influence of the positive heat of mixing, $\Delta H_{\mathrm{m}}^{\mathrm{Fe–Cu}} = +19\ \mathrm{kJ/mol}$ and $\Delta H_{\mathrm{m}}^{\mathrm{Co–Cu}} = +10\ \mathrm{kJ/mol}$. In this case, samples formed by about 77% of Cu (a Cu-rich system), the heat of mixing has a minor relevancy. However, for samples with less Cu concentration, the thermodynamic factor assumes a dominant role [9], imposing a higher energetic barrier for the transformation. It is important to note that up to now Fe–Co–Cu alloys have only been obtained by ball milling experiments [13,14]. As long as the authors know, this is the first time that such alloys are obtained using ion irradiation.

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