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GIANT MAGNETORESISTANCE EFFECTS IN CORRELATION WITH LOCAL MAGNETIC INTERACTIONS IN Fe-Cu AND Co-Cu GRANULAR THIN FILMS PREPARED BY THERMIONIC VACUUM ARC METHOD

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The simultaneous discharge of Fe (or Co) and, respectively, Cu metal sources via thermionic vacuum arc (TVA) was used as a new processing method to obtain granular magneto-resistive films. The nano-structured films with thicknesses of 120 ± 1 nm and 250 ± 1 nm were obtained in definite conditions and subsequently characterized via electrical measurements at room temperature, in applied magnetic field. The paper reveals the importance of different specific factors (from size and dispersion effects to annealing influence) on the magnetoresistance behavior. A special contribution of the Mössbauer Spectroscopy, concerning the local structure and interactions as well as the magnetic phase characterization, was emphasized.

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1. Introduction

The granular metallic thin films consisting of single domain ferromagnetic clusters embedded in a non-magnetic metallic matrix and presenting giant magnetoresistance (GMR), acquired in the last years a great interest, in challenge to the typical multilayer systems [1]. Granular systems are very convenient from the point of view of practical applications, because they are relatively easy to produce, present a good thermal stability and exhibit magnetoresistance effects in the usual current in plane (CIP) geometry, comparable or even larger than that of the multilayer systems [2]. Theoretical estimations suggest that the GMR effect is intimately related to the density and size distribution of the magnetic clusters and therefore it is strongly sensitive on the preparation conditions. For a low density of the super-paramagnetic clusters, the GMR effect increases with the amount of the magnetic phase up to the percolation threshold where the reciprocal interaction of the magnetic domains leads to the disappearance of the GMR effect. Not only the density but also the cluster dimensions play an important role to GMR effect related to the scattering of the conduction electrons by the magnetic moments of the clusters. In turn, the magnetic configuration of the clusters in applied magnetic field depends on their dynamical behavior at a certain temperature, which is directly influenced by their size distribution.

Using the simple random distribution of non-interacting magnetic moments of similar magnitude, it can be proved that the magnetoresistance displays a quadratic dependence on the relative magnetization. However, for many practical systems the quadratic law is not fulfilled in the

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range of medium and high values of the applied magnetic fields. In fact, the experimental data point to the complexity of this phenomenon in real systems and show that the origin of the spin-dependent scattering mechanism remains still challenging in many aspects. [4].

This paper reports on Fe-Cu and Co-Cu nano-globular thin films presenting GMR effects and obtained by a new procedure. The preparation method is briefly described. The obtained films are characterized in respect to both the magneto-transport properties and local magnetic interactions. The influence of subsequent thermal treatments on the magnetoresistive effects is also considered.

2. Experimental set-up and method

For the preparation of the granular nano-structured GMR films, an original method, which is placed between electron beam evaporation and electrical vacuum arc discharge, known as thermionic vacuum arc (TVA) was used [5 - 7]. The electron beam is emitted by an externally heated cathode (a tungsten grounded filament). A high anodic voltage accelerates the electrons. The electron beam can evaporate in a first step (for low electron energy and current intensity) the anode material as neutral pure particles. When the anode potential is increased up to a certain value, the evaporation rate increases as much as to allow an electrical discharge to be ignited in the evaporated pure material and the discharge is maintained even for discharge currents as low as a few hundreds mA. By using the TVA method, the metal deposition takes place in high or ultrahigh vacuum conditions, without the presence of any gas, excepting the material evaporated at the anode. This method allows the simultaneous deposition of different materials, providing the possibility of obtaining multi-component thin films, in this particular case the Fe-Cu or Co-Cu systems. Fig.1 presents, for exemplification, the experimental set-up suitable for preparation of Fe-Cu thin films with different Fe relative content. In this respect, the substrates were positioned at different distances from the Fe (d_{Fe}) and, respectively, Cu (d_{Cu}) anode. The temperature of the film deposition depends on both the discharge electrical parameters and the distance from the anodes. Therefore, the cluster size distribution can also be controlled via the preparation conditions. Consequently, the TVA method can process granular nano-composites with different concentration of the metallic constituents and different size of the magnetic clusters, allowing the possibility of a detailed investigation and control of microscopic-macroscopic correlations concerning GMR phenomena.

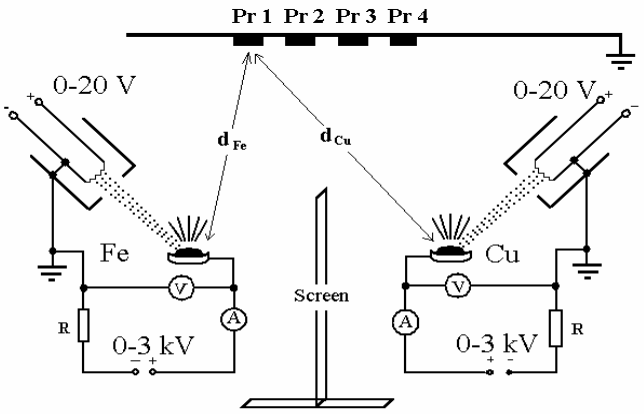

Fig. 1. The experimental arrangement

The above described method presents peculiar characteristics leading finally to some special properties of the obtained granular nano-structured thin films. Among these characteristics has to be mentioned: (i) the metal particles are evaporated from the anode by electronic (not ionic as in sputtering cases) bombardment, (ii) the evaporated atoms are partially ionized in a high voltage plasma (some hundreds of volts) generated near the anode, (iii) the localized plasma is surrounded by vacuum space in which the substrates are positioned, (iv) “real time annealing” during deposition

is induced, by both the intense thermal anode radiation and by energetic genuine ions bombardment, (v) the metal ions escaped from plasma reach the substrate without any collision in the vacuum space and (vi) the deposition rates are rather high, of the order of 1 nm.sec^{-1}. Consequently, the deposited films are characterized by high purity without any gas inclusion and a relatively narrow size distribution of the clusters. The both structural and morphological properties are very important in respect to the GMR behavior.

3. Experimental details

The Fe-Cu and Co-Cu films were prepared in a TVA system (2 x 10^{-6} torr base pressure) by simultaneous deposition of Fe (or Co) and Cu from two anodes bombarded with two TVA guns. The metal vapor "pressure" during the discharge process was about 5 x 10^{-1} torr (near the anode) decreasing towards 10^{-6} torr (in the proximity of the sample). The evaporation rate was of 10 ± 1 Å /s, stabilized and controlled with an accuracy of 10%. In situ thickness measurements were performed using a "Cressington" thickness-meter. The Kapton and Si substrates were used for Fe-Cu films and the glass for Co-Cu ones, without any heating during the deposition. The substrates have previously been ultrasonically cleaned in acetone and alcohol and dried in hot air. Three Fe-Cu samples (250 nm thickness) with different Fe content were prepared on Kapton substrates, displaying them at different distances from the Fe and Cu anodes: Pr1 (d_{Fe} = 28 cm, d_{Cu}=34 cm) Pr2 (d_{Fe} = 30 cm, d_{Cu}=32 cm) and Pr3 (d_{Fe} = 32 cm, d_{Cu}=30 cm). The Fe-Cu film (120 nm thickness) prepared on Si substrate was labeled as S1 (d_{Fe} = 32.5 cm, d_{Cu}=38.5 cm) and the Co-Cu film (120 nm thickness) prepared on glass substrate as G1 (d_{Co} = 41.5 cm, d_{Cu} = 34 cm). Subsequent annealing treatments at 410 °C for 150 min were performed in vacuum on samples S1 and G1. It is worth mentioning that the distance between the two anode sources was kept the same during all presented depositions.

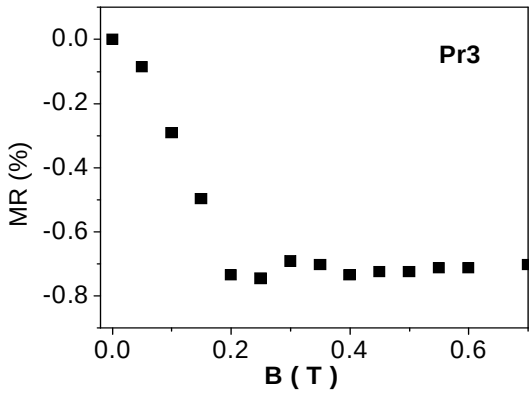

Fig. 2. MR effect for sample Pr3 in CIP geometry (room temperature measurement; a current of 10 μA was used).

The magnetoresistance ratio, MR(B)=(R(B)-R(0))/R(0), was measured vs. the applied magnetic field, B, by a dc method using a four-point configuration. The magnetic field (up to 0.8 T) was applied perpendicular to the sample plane. The ^{57}Fe Mössbauer spectra were collected at room temperature and 4.2 K only on the samples deposited on Kapton substrate, in transmission geometry, by using a constant acceleration spectrometer with a ^{57}Co source in Rh matrix.

4. Results and discussions

The magnetoresistance ratio MR(B) vs. the applied magnetic field, B, of the sample Pr3 (d_{Fe} = 32 cm, d_{Cu}=30 cm), obtained at room temperature in CIP geometry and using a current of

10 μA is presented in Fig. 2 . It has to be mentioned that the film resistance in zero applied field was 3.04 Ω. There is a negative magnetoresistive effect of about 0.8%, while a saturation process is observed starting from fields of 0.2 T.

The Fig.3 presents the MR effect of the sample S1 (d_{Fe} = 32.5 cm, d_{Cu}=38.5 cm) both in "as prepared" and annealed states. The MR effect of the sample G1 (d_{Co} = 41.5 cm, d_{Cu} = 34 cm), before and after the annealing treatment is shown in Fig. 4.

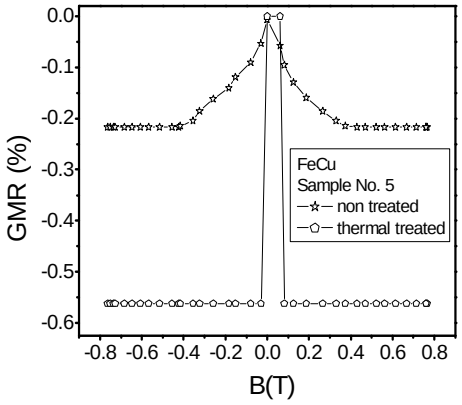

Fig. 3. MR effect for sample S1in CIP geometry, before and after the thermal treatment.

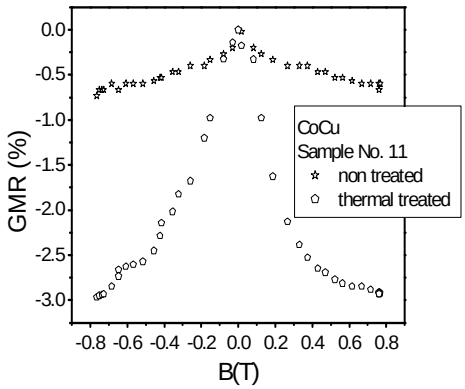

Fig. 4. MR effect for sample G1 in CIP geometry, before and after the thermal treatment.

One could observed that the ratio, d_{Fe}/d_{Cu} , in the analyzed Fe-Cu thin films are different, being higher for sample P3 as compared with sample S1. Therefore, a higher relative Fe amount has to be expected for the sample S1. The Fe deposited amount for both considered samples is under the percolation limit (as detailed in the following) and a higher effect would be expected in sample S1 as compared with Pr3 one. On the contrary, the MR effect is only 0.2 % in the sample S1 (Fig.3) and 0.8 %, i.e. four times larger, in the sample Pr3 (Fig.2). One could think on another parameter besides the Fe phase amount, dispersion or size, what can significantly influence the magnitude of the MR effect. Naturally, the most probably reason for the different MR behaviour has to be the type of the substrate. Following the same reasoning in respect to the ratio d_{Co}/d_{Cu}, the sample G1, deposited on isolating substrate, similar to one of the sample Pr3, has to present a relatively much lower MR effect than Pr3. The obtained value of about 0.7 % (Fig.4), close to MR value of sample Pr3, additionally suggests the influence of the specific type of magnetic metal from the 3d-transition period on the MR effect in the processed films.

On the other side, there is a remarkable aspect derived from the Fig. 3 and Fig. 4, that the magnitude of the MR effect can be strongly increased via a thermal annealing (about four times in Co-Cu sample and three times in the Fe-Cu sample, where the saturating field is also reducing). At a first glance this behavior might be related to the modification of size distribution of the magnetic clusters which could eventually suffer a nucleation / growing process under annealing. In fact, a significant modification of the crystallization process in the Fe-Cu alloys it is expected to appear at temperatures around 800° C and for an annealing time of 10-20 hours [8]. Therefore, due to the rather weak annealing conditions (150 minutes at temperatures of 410° C), the strong increase of the MR effects in the thermally treated samples, has to be related to structural relaxation phenomena rather than to nucleation/growing processes.

All the above observations plead that the observed MR behavior in the studied thin films is intimately connected with the local structure, phase composition and dispersion and size distribution of the magnetic clusters, with direct influence on their magnetic configuration.

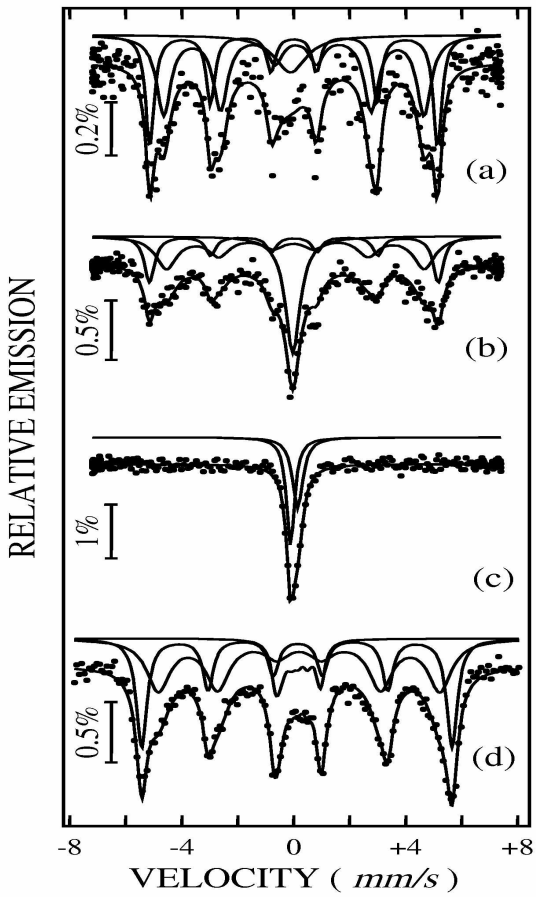

Fig. 5. Mossbauer spectra of samples Pr1 (a), Pr2 (b) and Pr3 (c) collected at room temperature and of sample Pr2 (d), collected at 4.2 K.

One of the sensitive methods giving simultaneously information about these aspects is the Mössbauer spectroscopy and here there are some relevant aspects. The room temperature Mössbauer spectra of samples Pr1, Pr2 and Pr3 are shown in Fig. 5 (a), (b) and (c) and put in evidence different magnetic behavior of the Fe phases in each sample. The spectra were fitted using two or three components, their relative area and characteristics being related, among other factors, on the d_{Fe}/d_{Cu} ratio. The Mössbauer spectra of samples Pr1 and Pr2 (Figs.5 (a) and 5 (b)) were fitted with two sextets and a central singlet. The two sextets are assigned, by their specific hyperfine parameters, to two nonequivalent magnetic Fe positions: one belonging to a bulk bcc α-Fe phase and the other one to interfacial/defected (Cu containing) α-Fe. The room temperature central singlet appearing in these samples will be further discussed. Samples with the highest relative amount of Fe (Pr1) showed the largest contribution (their relative area) of the magnetic phases, whereas sample P3 (the lowest Fe content) presents only paramagnetic singlets at room temperature (Figs.5 (a) and 5 (c)).

In order to elucidate the origin of the central components in the room temperature Mössbauer spectra, a low temperature (4.2 K) spectrum was collected for sample Pr2 and presented in Fig.5 (d). All the Fe atoms are subject to magnetic ordering (sextets) and can be assigned to bcc Fe clusters of different sizes. In this respect, the central singlets components appearing in the room temperature Mössbauer spectra are assigned to super-paramagnetic bcc Fe. The main differences among the non-annealed samples, with direct influence on the MR performances, consist therefore in different amounts of the magnetic phase and different size distribution of the magnetic clusters (due to both the different distances from support to anodes and the different film substrates).

5. Conclusions

It was presented the method of thermionic vacuum arc as a very suitable procedure for obtaining nano-granular magneto resistive films of a controlled quality and providing various MR properties.

Firstly, it is worth mentioning the role of cluster size and the nature of the substrate as well as the type of the magnetic atoms on the MR effects. It was proved that suitable thermal annealing substantially increased the MR effect. A relevant observation was related to the structural relaxation (induced by low temperature, 410 °C, annealing) increasing the MR effects via presumably electron-phonon interactions.

The ^{57}Fe Mossbauer results provide evidence for a composite Fe-Cu thin films consisting of α-Fe nanoparticles embedded in the Cu matrix. The size of the Fe clusters, their dispersion in the Cu matrix, as well as their magnetic configuration, can be analyzed starting from temperature dependent Mossbauer spectra and finally correlated with magnetoresistance effects.

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