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Magnetic and transport properties of Co–Cu microwires with granular structure

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ABSTRACT

Magnetic, transport and structural properties of granular $Co_x–Cu_{100-x}$ (5<x<40 at.%) glass-coated microwires were studied. Co–Cu microwires exhibited giant magnetoresistance (GMR) effect. For x = 5% we observed the resistivity minimum at 40 K associated with the Kondo effect. For x > 10 partial evidences of granular structure have been observed. For x ≥ 30 anisotropic contribution to GMR has been observed giving rise to non-monotonic dependence of GMR on the field. Temperature dependence of magnetization measured during a cooling regime without external magnetic field and in the presence of the field shows considerable difference at low temperatures, being attributed to the presence of small Co grains embedded in the Cu matrix. By X-ray diffraction we found, that the structure of the metallic nucleus is granular consisting of two phases: fcc Cu appearing in all the samples and fcc α-Co presented only in microwires with higher Co content. For low Co content (x ≤ 10%) X-ray diffraction technique indicates that Co atoms are distributed within the Cu crystals. The quantity and the crystallite size of the formed phases strongly depend on the geometry of the microwire. The structure, magnetic and transport properties were affected by the glass coating inducing the internal stresses and affecting the quenching rate.

1. Introduction

Since the observation of giant magnetoresistance (GMR) in Co–Ag nanogranular alloys in 1992 [1,2], numerous studies of granular materials have been done (see, for example [1–4], and references therein). Structure of materials consists of ferromagnetic nano-sized grains, usually of Co or Fe, embedded in a nonmagnetic metallic matrix (typically Cu, Ag, Au or Pt). At zero magnetic field, the magnetic moments of the single-domain particles are not aligned (e.g. oriented randomly) so the resistivity of material is high. The application of an external field aligns these moments, decreasing the resistivity. It is widely believed that GMR in granular materials and multilayers is related to the spin-dependent scattering of conduction electrons within the magnetic entities as well as at the interfaces between magnetic and nonmagnetic regions (it is, however, supposed that the interfacial effect contributes to a dominant extent to GMR [1–3]).

Melt spinning techniques, allowing fabrication of metastable metallic alloys using rapid quenching from the melt, are quite suitable to obtain granular structures [5,6]. In most cases, structure consisting of nano-sized grains embedded in nonmagnetic matrix can be obtained in as-quenched state or after adequate annealing of metastable alloys obtained by melt quenching of alloys from immiscible elements. It is worth mentioning that there are few rapid quenching techniques allowing fabrication of planar (ribbons), cylindrical (wires) and even composite (glass-coated microwires) with rather different quenching conditions and heat extraction geometry during rapid solidification. Therefore one can expect that the structure of such alloys as well as GMR behavior must depend on parameters of particular melt spinning technique (quenching rate, internal stresses, oxidation…). Recently great attention has been paid to the studies of glass-coated microwires produced by Taylor–Ulitovsky technique allowing the fabrication of long glass-coated metallic microwires (typical metallic nucleus diameters 1–30 μm, the thickness of the glass coating 0.5–20 μm). This method allows achieving high quenching rate and therefore producing microwires with amorphous, nanocrystalline, microcrystalline or granular structure [6–9]. Amorphous and nanocrystalline microwires are quite suitable for many technological applications owing to quite peculiar magnetic properties such as the magnetic bistability [10] and the giant magnetoimpedance effects [6–9,11]. On the other hand, existence of the outer glass-coating with physical properties completely different from metallic nucleus alloys (different thermal conductivity, thermal expansion coefficients…) in some cases drastically affects structure and magnetic properties of glass-coated microwires [6–8]. Thus, crystalline structure, crystallization temperature and magnetic properties of nanocrystalline microwires are rather different from

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nanocrystalline ribbons of the same composition [6,7]. Aforementioned rapid solidification is a way to further expand the limits of investigation and properties for magnetic materials, through the formation of textures, amorphous to crystalline transitions and nanocrystalline structures [6–11]. Consequently, recently Taylor–Ulitovsky technique has been proposed for preparation of granular microwires with different composition [12–14], microwires with magnetocaloric effect [15], and Heusler type microwires [16–18].

One the other hand, the structure of the granular material is rather complex and the relationship between its microstructure and magneto-transport properties is still not fully understood. For example, the role of spin-dependent scattering of conduction electrons on single magnetic ions dissolved in matrix is still under debate [2,3].

In the case of Co–Cu system, granular structure usually has been observed for moderate Co content (below 20 at.%) [1,2]. In this work, we present a review of our last experimental results on magnetoresistance (MR) in Co_xCu_{100-x} ($5 \leq x \leq 40$ at.%) glass-coated microwires.

2. Experimental details

Co_x–Cu_{100-x} ($5 \leq x \leq 40$ at.%) glass-coated microwires (typical total diameters, D, from few till 50 μm) consisted of metallic nucleus (typical diameters, d, 1–30 μm) covered by outer glass shell (Pyrex glass, typical thickness from 0.5 μm till 20 μm) have been prepared by the Taylor–Ulitovsky technique (see details on composition and diameters of studied microwires in Table 1) [6–18]. Within each composition of metallic nucleus we also produced microwires with different ratios of metallic nucleus diameter and total diameter, i.e. with different ratios $\rho = d/D$. This allowed us to control residual stresses, since the strength of internal stresses is determined by ratio ρ [8,19]. On the other hand up to 723 K the Pyrex glass used for microwire preparation exhibit high mechanical and thermal stability, therefore thermal annealing should be limited by 723 K.

Usually quenching rate achieved by the Taylor–Ulitovsky method is limited by 10^7 K/s, depending on the quenching medium (gaseous or liquid) [6]. Additionally, the quenching rate is determined by the thickness of the glass-coating and type of used glass: the thinner the glass-coating the higher the quenching rate [6].

X-ray diffraction (XRD) measurements were carried out at room temperature by means of a D8 Advance (BRUKER) diffractometer in reflexion configuration (Bragg-Brentano geometry). The experimental set-up was provided with automatic divergence and receiving slits as well as a graphite monochromator. Cu Kα (λ = 1.54 Å) radiation was used in all the patterns. The XRD scans were collected in the $40° \leq 2\theta \leq 100°$ diffraction angle region, where all the diffraction peaks corresponding to Cu and α-Co phases appear, by steps of 0.04° with accumulation times of 5 s. When the statistics of the result was not good enough, the counting time was 10 s and subsequently the diffracted intensity values renormalized.

Table 1
Compositions and geometric properties of studied Co_xCu_{100-x} microwires.

Microwires composition	Microwires geometry: metallic nucleus diameter, d, total diameter, D and ratio ρ = d/D
Co_5Cu_{95}	d = 13.3 μm; D = 16.9 μm; ρ = 0.787
$Co_{10}Cu_{90}$	d = 11.6 μm; D = 15.4 μm; ρ = 0.753
	d = 14.7 μm; D = 20.2 μm; ρ = 0.728
$Co_{20}Cu_{80}$	d = 13.3 μm; D = 20.5 μm; ρ = 0.649
	d = 14.4 μm; D = 20.5 μm; ρ = 0.702
	d = 12.9 μm; D = 18 μm; ρ = 0.717
$Co_{30}Cu_{70}$	d = 9 μm; D = 14.7 μm; ρ = 0.612
	d = 11.2 μm; D = 16.9 μm; ρ = 0.663
	d = 18.4 μm; D = 24.1 μm; ρ = 0.763
$Co_{40}Cu_{60}$	d = 15.4 μm; D = 27.7 μm; ρ = 0.57
	d = 9.4 μm; D = 15.1 μm; ρ = 0.62

Magnetic and magneto-transport properties have been measured at various temperatures within 5–300 K using suitable options of the Quantum Design magnetometer. MR has been defined as:

$$\Delta R/R(\%) = (R(\mathrm{H}) - R(0)) \times 100/R(0). \quad (1)$$

Whenever the other was not stated the MR was measured for the field directed along the axis of the microwire, parallel to the current.

3. Results and discussion

In the majority of as-prepared Co–Cu glass coated microwires considerable MR was observed. Thus, Fig. 1(a, b) shows $\Delta R/R$ (H) dependences for $Co_{20}Cu_{80}$ and $Co_{30}Cu_{70}$ microwires respectively. In addition, MR response for $Co_{20}Cu_{80}$ microwires is negative and tends to saturate in high fields. These features clearly show that, when x = 10–30, GMR is a major contribution in total MR of the studied microwires. As previously reported elsewhere [8,19] rapid quenching from the melt and strong internal stresses induced during the simultaneous rapid solidification of the thin wire surrounded by the glass coating when using Taylor–Ulitovsky method are essential for the formation of magnetic properties and structure of glass coated microwires. The strength of internal stresses is determined by the ρ-ratio, increasing with decreasing of ρ-ratio [6,19]. Consequently, we investigated MR of Co_xCu_{100-x} microwires as a function of chemical composition and ρ-ratio. To compare different samples we plotted maximum values of MR as a function of temperature (Fig. 2a–c). At low temperatures the largest values of MR were observed for the composition with x = 20. It agrees with the concentration dependence found in other granular materials [1,2]. For $Co_{30}Cu_{70}$, $\Delta R/R$ (H) dependences showed non-monotonic behavior: at low H-values (up to 1 T) we observed an increase of the $\Delta R/R$ with increasing of the magnetic field, H, as also previously reported for $Co_{29}Ni_{25}Mn_1Cu_{45}$ microwires [12,20]. $\Delta R/R$ (H) dependences was measured in $Co_{30}Cu_{70}$ samples when the field is oriented both along and perpendicular to the current. Negative MR observed for the transversal configuration (Fig. 3). Therefore we guess that positive MR contribution in the $Co_{30}Cu_{70}$ samples should be associated with the anisotropic MR (Fig. 3). In this case the anisotropic

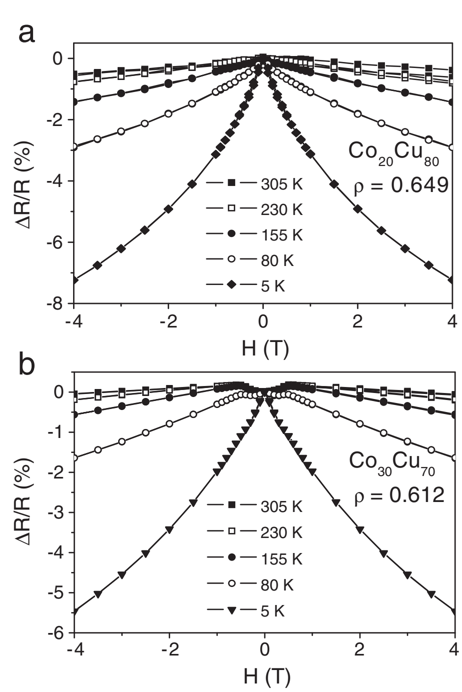

Fig. 1. ΔR/R (H) of Co_xCu_{100-x} microwires for: (a) x = 20, ρ = 0.649 and (b) x = 30, ρ = 0.612.

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