Kondo-like effect in magnetoresistive CuCo alloys

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Electronic transport properties of twin roller melt spun $Cu_{100-x}Co_x$ ($x=10,15$) alloys, are investigated in the temperature range between 10 and 300 K. Negative magnetoresistance is observed up to 0.85 T in the as-cast state, which further increases with a treatment of 1 h at 923 K. Resistance exhibits a metallic behavior below room temperature and draws a minimum near 30 K in all the as-cast microstructures; this minimum diminishes when a magnetic field is applied and completely disappears after high-temperature annealing. A logarithmic dependence of the electric resistance on temperature is found below 30 K. A Kondo-like scattering mechanism involving small Co spin clusters is considered to explain the minimum.

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I. INTRODUCTION

After the discovery of giant magnetoresistance (GMR) in magnetic Fe/Cr multilayer,[1] a similar magnetoelectronic behavior was also found in different granular magnetic systems. For many years, $Cu_{1-x}Co_x$ ($x<0.3$) alloys have been investigated as model granular systems[2–4] to gain insight into the spin-dependent transport processes in metallic alloys containing a fine dispersion of nanometric magnetic particles.

Since the first works in CuCo alloys, the main issue has been the description of the actual mechanisms leading to the magnetotransport properties observed and the microstructure associated with their optimum values. At first, small magnetic Co clusters, embedded in the nonmagnetic Cu metallic matrix were considered as the main source for the electronic spin-selective scattering observed.[2–5] More recently,[6,7] GMR effects have been correlated with a lamellar spinodal decomposition in the form of parallel nanometric stripes, forming within the matrix grains. This self-organized microstructure,[8] appearing as a consequence of the natural segregation taking place during the melt spinning process, is proposed to be responsible for GMR in these materials. However, because of the microstructure complexity—a modulated Co composition leading to periodic strips and small Co nanoparticles dispersed in the matrix and grain boundaries—the predominant mechanism leading to the observed magnetoresistance remains still uncertain.

We have previously reported[9,10] the low-temperature magnetic, resistive, and magnetoresistive properties of $Cu_{90}Co_{10}$ twin roller melt spun ribbons, solidified at tangential wheel speeds of 5 m/s and 23 m/s, in both, the as-cast condition and after a heat treatment of 1 h at 923 K, respectively. This solidification method is known to promote a more uniform microstructure as compared to those obtained with single wheel devices, being possible to explore lower cooling rates. The x-ray patterns of the alloys, even in the as-cast state, were found to exhibit double peaks, corresponding to two different concentrations of Co in a Cu fcc matrix. These two CuCo peaks, better resolved in ribbons processed at low speeds (5 m/s), are consistent with the spinodal-like modulated composition revealed by Miranda *et al.*[6,7] in similar alloys. In the as-cast state, the mean sizes of these composition strips formed at different quenching rates were estimated using the Scherrer formula; for samples cooled at 5 m/s, values of 51 ± 4 nm and 55 ± 12 nm were estimated for the Co-rich and the Co-poor strips, respectively. For samples quenched at 23 m/s these sizes were 44 ± 4 nm (Co rich) and 85 ± 20 nm (Co poor), respectively. These values are comparable to those reported by other authors[6,7] for similar melt spun CoCu alloys, on the basis of transmission electron microscopy observations.

The hysteresis curves of these twin roller melt spun ribbons were all well fitted[9,11] by the sum of a small ferromagnetic contribution and a superparamagnetic one, the latter arising in Co clusters of about 3.5 nm and 2.3 nm in mean size, for ribbons quenched at 5 m/s and 23 m/s, respectively. Time dependence of the magnetization[11] was consistent with a mean fluctuations field of only 2 mT and an activation length of about 20 nm, certainly larger than the mean volume of small Co particles but comparable with the mean stripe width. Then, it is concluded that the samples processed in the twin roller contain both: a fine dispersion of Co nanoparticles in the fcc Cu matrix exhibiting alternating paramagnetic and ferromagnetic stripes, of about 50 nm mean width. The low-temperature electric resistance of these CuCo as-cast melt spun ribbons is found to decreases[10] almost linearly during cooling from 300 K and passes through a minimum at about 30 K. A similar resistance minimum has been previously reported in very diluted CuCo alloys[12,13] and also in more concentrated (0.5–2 %) alloys.[14] In the present work we report further results concerning this minimum in the electrical resistance measured in nanostructured $Cu_{90}Co_{10}$ and $Cu_{85}Co_{15}$ twin roller melt spun alloys. This minimum, found near 30 K in all the as-cast samples, is examined in connection with the microestructure and the observed magnetoresistance.

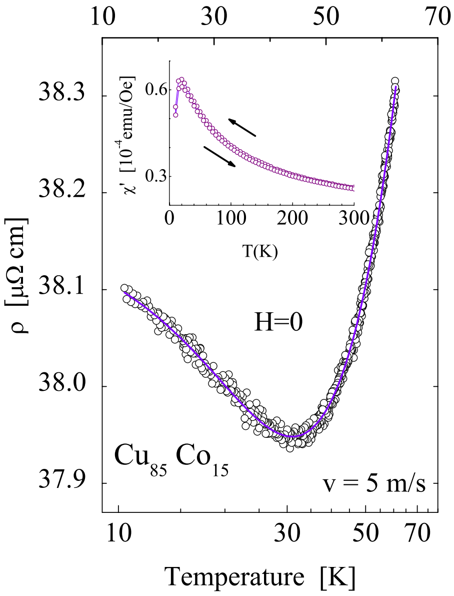

FIG. 1. (Color online) Resistance vs temperature for a $Cu_{85}Co_{15}$ as-cast ribbon. Inset: ac susceptibility measured during cooling and heating, showing the characteristic peaking effect at the Kondo temperature, T_K.

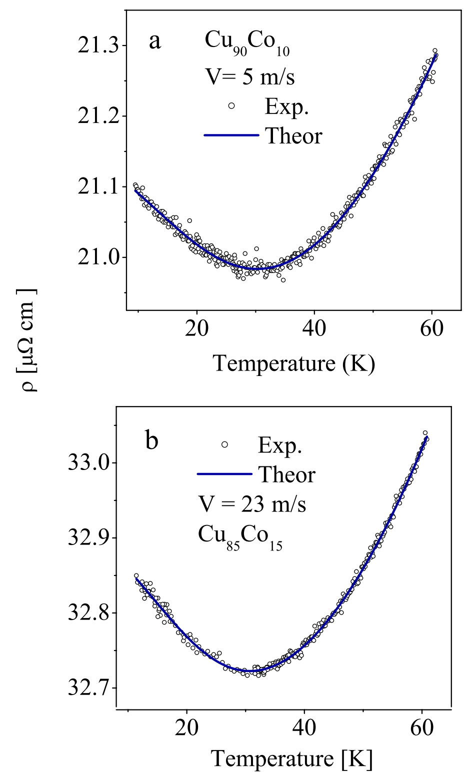

FIG. 2. (Color online) Resistance vs temperature for (a) $Cu_{90}Co_{10}$ ribbons quenched at 5 m/s and (b) $Cu_{85}Co_{15}$ ribbons, quenched at 23 m/s.

The $Cu_{90}Co_{10}$ (C10) and $Cu_{85}Co_{15}$ (C15) ribbons were produced by melt spinning in a twin roller device at tangential wheel speeds of 5 m/s (V5), 20 m/s (V20), and 23 m/s (V23), under an inert Ar atmosphere. The ribbons obtained for each quenching rate exhibited quite uniform magnetic properties and microstructure, as controlled by x-ray diffraction techniques and room-temperature magnetic measurements performed, on many ribbons. The as-cast samples were then heat treated for 1 h at 923 K. The electrical transport measurements were performed with a Keithley 220 Programmable Current Source and a Keithley 182 Nanovoltimeter, using the conventional four-point contact technique, between 10 and 300 K. These measurements were made on at least three ribbons and for many thermal cycles each.

II. RESULTS

The low-temperature dependence of the electric resistance under zero applied magnetic field, is illustrated in Figs. 1 and 2 for as-cast $Cu_{85}Co_{15}$ and $Cu_{90}Co_{10}$ ribbons, solidified at tangential wheel speeds of $\upsilon = 23.0 \pm 0.3$ m/s, $\upsilon = 20.0 \pm 0.3$ m/s, and 5.0 ± 0.1 m/s. Below 50 K a minimum in the resistance is detected during heating and during cooling, remaining unchanged during successive thermal cycles. At low temperature, a logarithmic dependence of the resistance fits well all the data measured [see Fig. 3(a)] while above the minimum, all the alloys show a metallic like behavior in the entire temperature range up to 300 K. This minimum effect slightly decreases when a magnetic field of 0.6 T is applied to the sample and clearly disappears after a thermal treatment of 1 h at 923 K—see Figs. 3(b) and 3(c).

The maximum values of MR, measured in the same alloys under an applied field of 0.85 T, are shown in Fig. 4(a) for both, the as-cast state and after a treatment of 1 h at 923 K. It is observed that the microstructures resulting for the higher quenching rate show higher MR values, which can be further increased by high-temperature annealing. This heat treatment leads to the appearance of relatively large Co precipitates (~7 nm mean size, detected by x-ray diffraction[9]) and to a larger difference in composition between stripes.[6] The depth of the resistance minimum, defined as $\mathrm{MD} = 100 \times [R(10\ \mathrm{K}) - R(T_{min})]/R(T_{min})$, is shown in Fig. 4(b) for as-cast ribbons, in the zero-field condition and under an applied field of 0.6 T. It is also observed that the effect of a magnetic field is to reduce the minimum depth in all the microstructures; the deepest minimum appears in samples quenched at a low rate and no minimum is observed, in any sample, after annealing at 923 K.

It is well known that electron-scattering mechanisms involving lattice vibrations and atomic defects yield to a residual resistivity at low temperature. Another contribution to this residual resistance in magnetic metallic systems is that described by Cabrera and Falicov[15] as arising in the scattering of conduction electrons by Bloch domain walls. In order to identify the scattering mechanism responsible for the low-temperature increase in the electric resistance (leading to the observed minimum) some other mechanisms must be considered. Spin mixing due to the spin-orbit interaction is little relevant as compared with extrinsic scattering due to defects. Valet and Fert[16,17] stressed the importance of the spin-flip scattering arising from spin fluctuations (magnons) in describing the GMR in multilayer but they showed that these quasiparticles undergo these inelastic spin-flip processes at relatively high temperature. Finally, another scattering

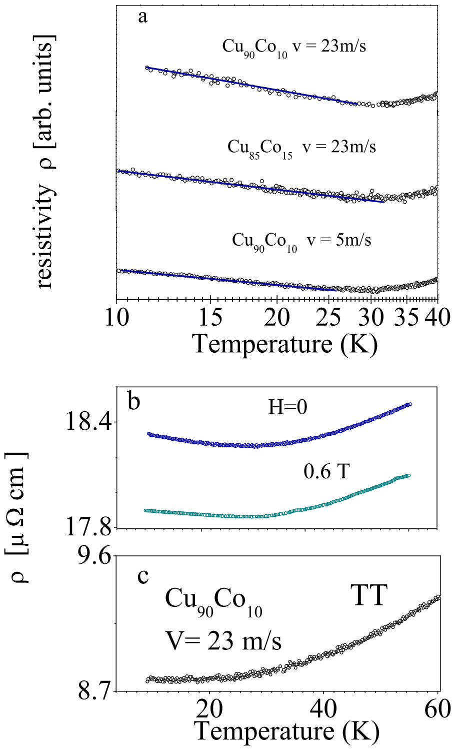

FIG. 3. (Color online) (a) Low-temperature logarithmic resistance behavior in all the as-cast samples. (b)The effect of a magnetic field (0.60 T) is to reduce the minimum depth; (c) after a thermal treatmet of 1 h at 923 K the minimum disappears.

mechanism largely investigated for several decades, is due to Jun Kondo;[18] it considers the scattering of conduction electrons by diluted magnetic impurities in the conducting matrix. This last mechanism is known to lead to a logarithmic

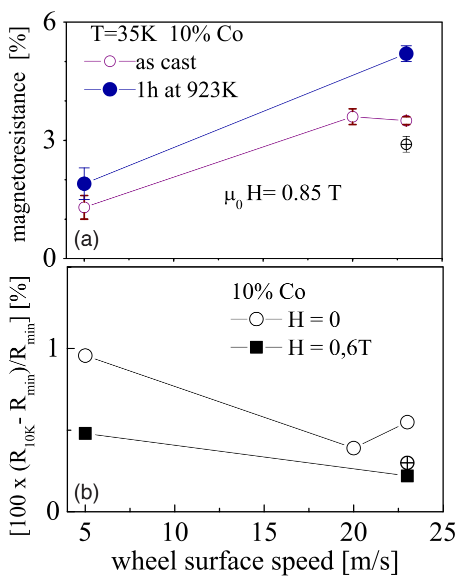

FIG. 4. (Color online) (a) The MR measured at 35 K in samples quenched at two different rates, in the as-cast state and after a heat treatment. (b) The effect of an applied field on the minimum depth, for the same quenching rates as in (a). In both plots, crossed circles correspond to Cu-15% Co.

TABLE I. Calculated values for ρ_0, ρ_{K0}, T_K, and S_{eff} from data in of Figs. 1 and 2.

Sample	ρ_0 ($\mu\Omega$ cm)	ρ_{K0} ($\mu\Omega$ cm)	T_K (K)	S_{eff}
V5 C10	20.5 ± 0.1	0.34 ± 0.03	30 ± 1	0.10 ± 0.01
V23 C10	17.9 ± 0.1	0.20 ± 0.01	27.5 ± 0.5	0.09 ± 0.01
V5 C15	33.5 ± 0.1	0.43 ± 0.06	30 ± 2	0.12 ± 0.01
V20 C15	33.6 ± 0.1	0.37 ± 0.02	28.5 ± 0.5	0.09 ± 0.01
V23 C15	32.3 ± 0.1	0.32 ± 0.05	26 ± 1	0.09 ± 0.01

dependence of resistance at low temperature. Within the assumption that the Matthiessen's rule is valid, the low-temperature resistance curves were fitted to the expression

$$\rho(T) = \rho_0 + aT^2 + \rho_K(T), \qquad (1)$$

where ρ_0 is the residual resistivity due to various temperature-independent scattering process, a is a constant, and $\rho_K(T)$ is the contribution of Kondo scattering in absence of magnetic field, given by[19]

$$\rho_K(T) = \rho_{K0} \times \left\{ 1 - \ln\left(\frac{T}{T_K}\right)\left[\ln^2\left(\frac{T}{T_K}\right) + S_{eff}(S_{eff}+1)\pi^2\right]^{-1/2} \right\}. \qquad (2)$$

Here T_K is the Kondo temperature and S_{eff} is the effective magnetic spin of the scattering units. The term aT^2 arises from electron-electron collisions.[20] The best-fitting curves are plotted together with the experimental data in Figs. 1 and 2 and the corresponding parameters values are listed in Table I. The values obtained for ρ_0 are in good agreement with those found by Yu *et al.*[21] ($\sim$10–30 $\mu\Omega$ cm) in similar CuCo alloys. The values for the coefficient a are in the range $(1.6 \pm 0.3) \times 10^{-4}$ ($\mu\Omega$ cm K^{-2}).

The nominal Co concentrations in the present samples seem too large to consider individual Co atoms as magnetic Kondo units. Instead, we suggest that a Kondo scattering mechanism operates but involving small Co spin clusters in an early stage of formation, in a similar way as proposed by Mei *et al.*[22] for the resistance minimum observed in nondiluted AuNi alloys. Then, the characteristics of the resistance minimum are expected to depend on the size and distribution of these magnetic Co clusters, which in turn depend on the quenching variables and on subsequent thermal treatments. The effective cluster spin value S_{eff}=0.08 obtained is similar to that found by Ustinov *et al.*[23] in superparamagnetic cluster-layered Fe/Cr nanostructures. This effective value for the spin of the scattering clusters (S_{eff}=0.08) is somewhat low, as compared with the expected value for the total cluster total spin S_T, which is known to increase with the cluster size.

It has been suggested[24] that the Kondo effect involving clusters should be due to quantum fluctuations in the cluster spin rather than to the cluster total spin itself. As these fluctuations are suppressed as $\frac{1}{S_T}$ when the magnetic clusters grow, the Kondo effect should vanish for larger cluster sizes. In this scenario small values of S_{eff} ($\ll S_T$) may be explained. The fact that the minimum depth increases when the quenching rate goes from 23 to 5 m/s but it vanishes after a heat treatment promoting further precipitate coarsening indicates that Kondo scattering operates under quite special conditions, that is, when clusters belonging to the small size tail of the size distribution are small enough to undergo fluctuations in their total spin S_T. This condition seems to be fulfilled by the size distributions with means about 2 to 4 nm found in the as-cast samples, but not in those obtained after the heat treatment, with a mean size of about 7 nm.[9] The Co precipitates involved in MR seem to be somewhat larger in size.

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